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BETA AND GAMMA DECAY HEAT MEASUREMENTS BETWEEN 0.1s - 50,000s FOR NEUTRON FISSION OF ^{235}U, ^{238}U AND ^{239}Pu

Progress Report

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ABSTRACT

In the investigations reported here, a helium-jet/tape-transport system was used for the rapid transfer of fission products to a low-background environment where their aggregate beta and gamma-ray spectra were measured as a function of delay time after neutron induced fission of ^{235}U, ^{238}U and ^{239}Pu. Beta and gamma-ray energy distributions have been deduced for delay times as short as 0.2 s and extending out to 100,000 s. Instrumentation development during the initial phase of the project included: 1) assembly and characterization of a NaI(Tl) spectrometer for determining aggregate gamma-ray energy distributions, 2) development and characterization of a beta spectrometer (having excellent gamma-ray rejection) for measuring aggregate beta-particle energy distributions, 3) assembly and characterization of a Compton-suppressed HPGe spectrometer for determining gamma-ray intensities of individual fission products to deduce fission-product yields. Spectral decomposition and analysis codes were developed for deducing energy distributions from measured aggregate beta and gamma spectra. Our aggregate measurements in the time interval 0.2 - 20 s after fission are of special importance since in this region data from many short-lived nuclei are missing and summation calculations in this region rely on model calculations for a large fraction of their predicted beta and gamma decay heat energy spectra. Comparison with ENDF/B-VI fission product data was performed in parallel with the measurements through a close collaboration with Dr. T. England at LANL, assisted by one of our graduate students. Such aggregate measurements provide tests of the Gross Theory of beta decay used to calculated missing contributions to this data base. Fission-product yields deduced from the HPGe studies will check the accuracy of the semi-empirical Gaussian dispersion model used presently by evaluators in the absence of measured yields. These studies, which involve several hundred identified gamma-ray lines, also provide numerous microscopic tests of nuclear databases at the individual nuclide level. These measurements and their analysis for ^{235}U and ^{238}U fission products will be completed by the end of the current three-year project period. ^{239}Pu measurements were recently begun and should also be completed during this period. During this past year a computer-based multiparameter, event-mode recording and analysis system was purchased, and its feasibility for future aggregate fission product studies demonstrated by using it to perform internal-conversion-electron/characteristic x-ray coincidence measurements. A number of internal conversion transitions in individual nuclides have been identified by sorting on characteristic x-rays.

DISCLAIMER

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1. RESEARCH

A. INTRODUCTION

We report here our progress on the separate measurements of aggregate beta and gamma-ray spectra as a function of time following the fission of ^{235,238}U and ^{239}Pu. A primary goal of this project, which began in mid-1992, is the determination of the distributions of beta and gamma energies for delay times as short as 0.1s and extending to >50,000s following fission. Of particular interest are spectra measured within the first 20s after fission. Few other aggregate measurements have been made in this time interval although well over one third of the energy released by the decay products occurs during this interval [Ref.1]. Furthermore, decay heat predictions based on summation calculations of individual fission-product nuclides display their largest uncertainties at short delay times [Ref.2]. These uncertainties are due to the incompleteness of the data base (ENDF/B-VI) for the beta and gamma energy spectra of short-lived nuclei which places a greater reliance on theoretical calculations, based in part on the "Gross Theory" of beta decay [Refs.3-5], to supplement the data base for short delay times.

Fission Is produced in our measurements using neutrons from the ^{7}Li(p,n) reaction initiated by our 5.5-MV Van de Graaff accelerator and also from our 1-MW research reactor. The geometries for these two experimental systems are depicted in Figs. A1 and A2. Fission fragments produced in a fission chamber are rapidly transported through a plastic capillary by means of a helium jet to a low background counting room, where they are deposited onto a moving tape. Depending on the capillary length, this transfer can be made as short as 0.1s. The tape carries the fission fragments/products to either the beta or gamma-ray spectrometer, the developments of which are described in Section 1B. Measurements of the transfer efficiency (Section 1H) have demonstrated that the fission fragments are transferred to the spectrometers with essentially equal probability over the full mass range, with the exception of the noble gases xenon and krypton for which corrections for their contributions can be readily applied. By varying the tape speed or the detector position along the tape, the delay time after fission can be varied. In measuring gamma-ray spectra, beta-gamma coincidences are used to gate the gamma spectrometer and this leads to a reduction in background by some two orders of magnitude. This feature also greatly enhances the sensitivity to high energy gamma rays, and in addition suppresses the sensitivity to beta-ray bremsstrahlung, which is primarily forward-peaked and thus heads away from the gamma-ray detector.

To date, we have completed measurements of the gamma-ray spectra for ^{235}U and (except for the longest delay times) ^{238}U, using the NaI(Tl) spectrometer. Beta spectra have also been measured for all but the very shortest and longest time intervals for ^{235,238}U and ^{239}Pu. Essental in these latter measurements was the special design of our beta spectrometer which provided very effective discrimination against gamma rays. Response functions were measured for both

spectrometers and parameterized in a new unfolding program (Section 1G) which was developed to extract the true energy distributions from the measured spectra. Analysis of these spectra is well under way and most remaining measurements will be completed during the remainder of this project period. Fourteen papers have been presented on these two studies at APS meetings and an international conference. This work is reviewed is Sections 1D and 1E.

Additional tests of the fission product data base are provided by high-resolution measurements of the aggregate gamma spectra. Using our HPGe spectrometer with Compton suppression, several hundred gamma lines have been observed in ^{235}U and ^{238}U for delay times ranging from 0.1s to $>100{,}000$s. Many of these lines have been identified with specific fission products through their energies, relative strengths and time dependences and from these the relative production probabilities determined and compared with ENDF predictions. A number of isomers have also been observed in the spectra and ratios determined for several isomeric-to-ground state productions. Isomeric states have been cited [Ref.5] as causing substantial changes in decay heat values. Ten papers on this high-resolution study have been presented at various meetings and the work is described in Section 1F.

The summation calculations based on ENDF/B-VI individual precursor data, which were carried out for comparison with our aggregate measurements, were performed on a Cray computer at Los Alamos National Laboratory in collaboration with Dr. Talmadge England. As part of this collaborative effort, a graduate student member of our group (Joann Campbell) has spent more than eight months at LANL so far during the project period under the supervision of Dr. England, supported in part by UMass Lowell and in part by LANL. Through this collaboration we have been able to make detailed comparisons between ENDF-predicted summation calculations of beta and gamma-ray decay heat spectra and our experimental results. These calculations are described in Section 1C.

During this period, 4 refereed papers were published, 3 have been submitted for publication, and 23 papers were presented at APS meetings, all on topics relating to this project. A listing of publications together with abstracts of presented papers and graduate theses can be found at the end of this report.

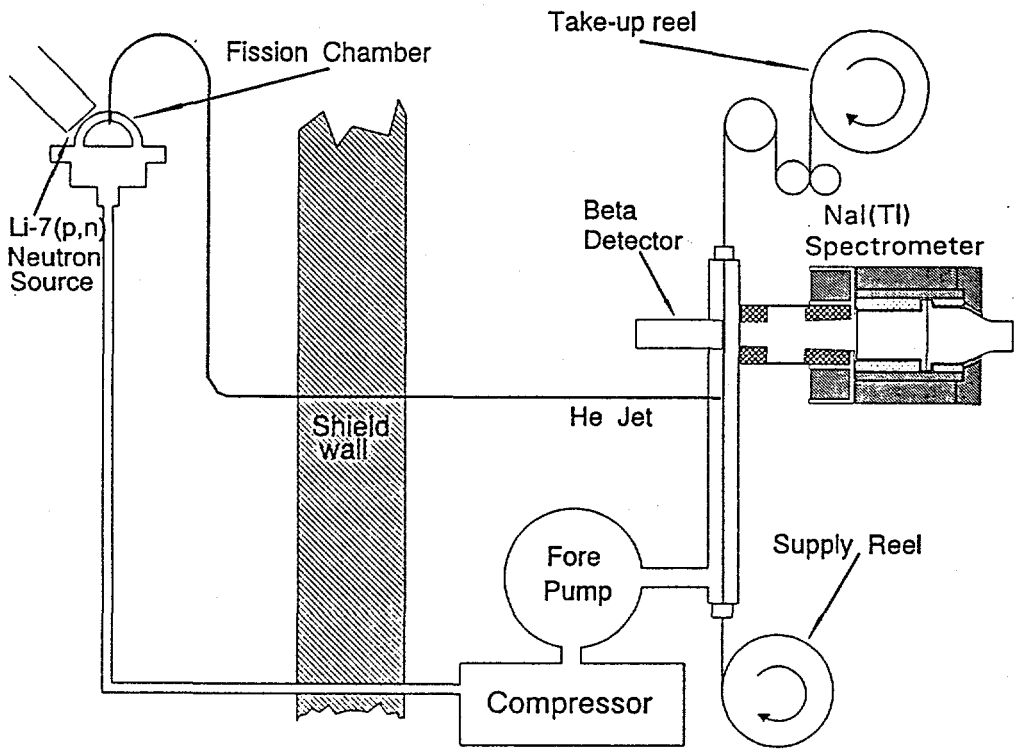

Figure A1. Helium jet/tape transport system with NaI spectrometer on accelerator.

(a): 1rem/hr γ, 0.4 mrem/hr n
(b): 0.3mrem/hr γ, 0.2 mrem/hr n
(c): 1 rem/hr γ, 0.4 mrem/hr n
(d): 1 rem/hr γ, 0.2 mrem/hr n

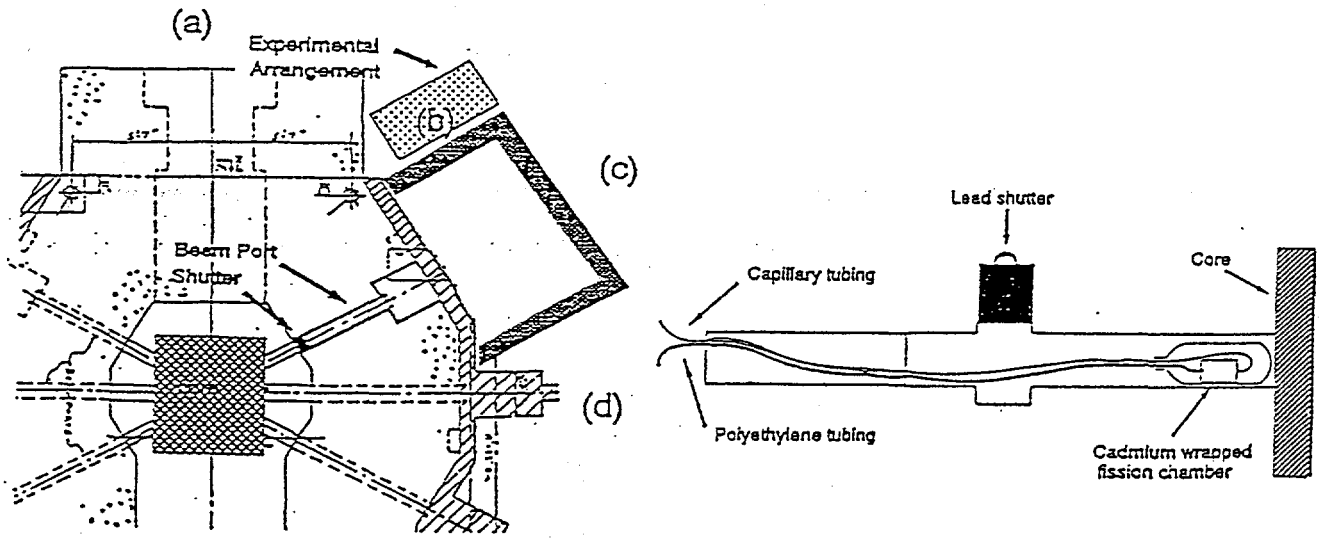

Figure A2. Reactor fast neutron beam port configuration and fission chamber positioning.

B. INSTRUMENTATION DEVELOPMENT

i) NaI(Tl) Gamma-Ray Spectrometer and Response Functions

The gamma-ray component of decay heat was measured using a 5" x 5" NaI(Tl) detector [Ref.6]. This detector was housed in a massive shield consisting of a combination of lead, brass and tungsten as depicted in Fig. B1. The tape bearing the fission products traveled perpendicular to the plane of the figure. Tungsten was used in the front snout which defined the solid angle of the detector. Edge effects in the detector were minimized by restricting the diameter of the tungsten aperture adjacent to the scintillator to just 3 inches. A magnet in the nose of the shield also served to deflect beta particles away so that mainly only the gamma component of the tape activity was detected. A thin beta detector was located on the opposite side of the tape and beta-gamma coincidences were used to gate the measured spectrum. Although the NaI(Tl) detector was well shielded and viewed only a limited portion of the tape, the beta detector which was tightly collimated truly defined the region of the tape viewed, and thus the delay time interval following fission. Equally important, the use of beta-gamma coincidence also reduced background (including that due to any betas which might have got through the magnetic deflection system) by approximately two orders of magnitude.

A typical gamma-ray spectrum obtained from the aggregate of fission products results from the superposition of several thousand gamma lines and thus has few clearly resolvable peaks in the NaI(Tl) spectrum. The measured spectrum is therefore the superposition of several thousand detector response functions, where each response function is itself a spectrum from a monoenergetic gamma ray. In order to extract the "true" energy distribution of the emitted gammas, the measured aggregate spectra must, therefore, be corrected for these response functions and the process of decomposing a gamma spectrum requires knowledge of such response functions as a function of energy.

Fifteen response functions were measured in the energy range 0.081 - 6.13 MeV, using monoenergetic gamma rays from radioactive sources and, for the highest energies, from selected reactions (see Table I). The shorter-lived sources were produced by neutron activation using our 1-MW research reactor, whereas the 6.13-MeV line from ^{16}O was produced in the $^{19}F(p,\alpha\gamma)^{16}O^*$ reaction using our smaller 2-MV Van de Graaff accelerator. The response functions are shown normalized in Fig. B2, which illustrates the variation of detector response with gamma-ray energy. The parameterization of these response functions and their use in the decomposition of the aggregate spectra is discussed in Section G.

The measurements and parameterization of NaI(Tl) detector response functions formed part of the M.S. thesis of Hung Nguyen.

Table I.

Sources of Gamma Lines Used in Measurements of NaI(Tl) Response Functions

Isotopes	E_γ(MeV)	$T_{1/2}$	Decay
^{133}Ba	0.0810	10.7 y	EC
^{139}Ba	0.1659	82.9 m	β^-
^{113}Sn	0.3917	99.5 d	EC
^{7}Be	0.4776	53.3 d	EC
! ^{22}Na	0.5110	2.6 y	EC
^{137}Cs	0.6616	30.1 y	β^-
^{54}Mn	0.8348	312 d	EC
! ^{60}Co	1.1732	5.27 y	β^-
! ^{22}Na	1.2746	2.6 y	β^+, EC
! ^{60}Co	1.3325	5.27 y	β^-
! ^{24}Na	1.3685	15.02 h	β^-
^{42}K	1.5246	12.36 h	β^-
^{28}Al	1.7787	2.24 m	β^-
! ^{24}Na	2.7541	15.02 h	β^-
$^{12}C^*$	4.4340	*****	$^{9}Be(\alpha,n\gamma)^{12}C^*$
$^{16}O^*$	6.1292	*****	$^{19}F(p,\alpha\gamma)^{16}O^*$

! Two gamma transitions.

ii) Beta Spectrometer and Response Functions

A beta spectrometer was designed for use in measuring the aggregate beta energy spectra at various delay times following fission [Ref.7]. The spectrometer was very insensitive to the gamma-rays accompanying the fission products, had good linearity, adequate energy resolution for the continuous beta energy distributions, and a relatively simple response function to monoenergetic electrons. Response functions were measured below 1 MeV and a trial set for higher energies was tested with beta spectra of known shape.

The spectrometer is shown in Fig. B3. A thin scintillator disk, 1.5 inches diameter and 0.020 inch thick, was mounted on the surface of a 3-in x 3-in plastic scintillator coupled to a 3-in photomultiplier and optically isolated with a thin aluminum foil. The disk was viewed by two 2-in photomultipliers. Betas passing through the scintillator disk gave rise to a signal which was used to gate pulses from the main scintillator. Gamma rays seldom interacted with the thin disk and were, therefore, very effectively discriminated away. Distortion of a beta spectrum due to edge effects was also minimized. This was due largely to the small disk diameter used, which resulted in most secondary electrons in a detection event being kept well away from the

cylindrical wall of the main detector. The only wall which could be seen by secondary electrons was the flat surface through which the beta enters. A quick-coupling O-ring joint at the mouth of the housing allowed easy attachment to vacuum systems, light-tight chambers, and sliding-tape jaws.

The excellent gamma-ray discrimination of the beta spectrometer was tested using a thin ^{24}Na source produced in the UML reactor and placed in a small chamber coupled to the mouth of the spectrometer. This source has a continuous beta spectrum of known allowed shape with 1.392-MeV end point energy and two gamma rays of energy 1.369 and 2.754 MeV. An intense (100μCi) encapsulated ^{22}Na gamma source (0.511 and 1.274 MeV) was placed on the outside of the chamber and near the ^{24}Na source. The spectra measured with and without disk gating are shown in Fig. B4. Without disk gating, the spectrum was dominated by the Compton bands of the 0.511- and 1.274-MeV gamma-rays from the ^{22}Na source. With disk gating active, however, these gamma pulses were essentially eliminated to reveal the ^{24}Na beta spectrum. The probability of valid beta-gamma coincidences from the two gamma-ray lines of ^{24}Na was kept small by maintaining a sufficiently large distance (approximately 5 inches) between the source and face of the scintillator.

Radioactive sources which produce electron groups through internal conversion were preferred over Monte Carlo simulations for determining response functions, since measurements take proper account of beta scattering from the spectrometer housing as well as losses due to edge effects. Such events produce low energy tails on the response functions. Only a limited number of IT sources were available for such measurements (Table II) and their corresponding electron energies were mostly <1 MeV. These IT sources were also accompanied by betas that

Table II

Internal Conversion Electron Sources

Source	Half-life	Conversion Electron Energy (MeV)
^{207}Bi	30y	0.481, 0.554, 0.976, 1.048
^{113}Sn	115d	0.365, 0.389
^{137}Cs	30y	0.624, 0.656

obscured the low-energy tail of the response function. In a measurement of the bare ^{137}Cs IT source, however, the beta spectrum was removed by performing an x-ray coincidence measurement. The ^{137}Ba 0.662-MeV level populated in the beta decay of ^{137}Cs is an isomeric state with 2.6-m halflife. Decay of this state and electron emission from an electron shell is accompanied by an x-ray. By gating on the K_α-x rays in this measurement, the low energy tail of the response function could be revealed, as is shown in Fig. B5.

No such monoenergetic electron sources were readily available above about 1 MeV. At these energies, the spectra of beta sources having known energy distributions were measured. These were then unfolded using trial sets of response functions and the result compared with the known energy distribution. This is discussed further in Section G(ii).

The measurements of beta spectrometer response functions form part of the Ph.D. thesis of Shengjie Li.

iii) HPGe Gamma-Ray Spectrometer

To observe discrete gamma-ray lines in the aggregate fission-product spectra requires premium energy resolution and this was provided by a high purity germanium detector [Ref.8]. A NaI(Tl) annulus was also used with the detector to suppress contributions from Compton-scattered gamma rays in the spectra. The system geometry is shown in Fig. B6. To maintain satisfactory count rates during measurements it was desirable to position the HPGe detector close to the fission product activity on the tape. To accomplish this the detector was designed with a longer cryostat pipe than is usual in order to bring the detector to the front face of the annulus rather than the center. Although this geometry results in reduced annulus efficiency, the peak-to-background ratio in the gamma-ray spectra was nevertheless greatly improved.

The effect of the annulus can be seen in the spectra of Fig. B7, measured for $^{235}U(n_{th},f)$ with and without Compton suppression. Beta-gamma coincidence gating was employed in both cases. The peak-to-background ratio was improved by a factor 2.1 over the energy region <1.5 MeV and by nearly 4 at energies >3 MeV. This improvement not only allowed the measurements to extend to weaker gamma-ray peaks but also extended the time range over which an individual peak could be followed. This aided considerably in identifying the precursor nuclides.

iv) Data Acquisition and Analysis System Upgrade

A major upgrade of both our data acquisition and data analysis systems was begun during this project period, made possible by matching DOE and University equipment grants.

New Multi-Parameter Data Acquisition System

The increasing complexity of our experimental program makes multiparameter event-mode data recording a desirable goal and in anticipation of this a new computer-based CAMAC

data acquisition system was purchased for our experimental facility. This comprises an 11-slot CAMAC minicrate with LeCroy FERA Driver and FERA Memory units, and Ortec FERAbus Histogramming Memory and Quad 8K ADC. The choice of FERAbus rather than CAMACbus provides faster data processing and allows significantly higher count rates in multi-parameter mode. The crate allows expansion of the acquisition system to meet future needs of the experimental program.

The CAMAC crate controller is interfaced with a Macintosh Centris 650 computer through a Kinetic Systems NUBus interface. The system is supported by Kmax software from Sparrow Corp. which incorporates many standard programs for data acquisition and sorting. A 21-in color monitor allows easy viewing of multiple spectra. Archiving or backup of data (either list mode or histogram) is provided by an external EXABYTE 8mm tape unit and Kmax backup software.

In a feasibility study of future aggregate fission-product investigations conducted last summer, this system was used to perform internal-conversion-electron/characteristic-x-ray coincidence measurements. A number of internal conversion transitions in individual nuclides were identified by sorting on the characteristic x rays and the advantages of the new system in our future studies were well demonstrated.

Microvax/PC Data Analysis Network

A new data analysis system for our research group was installed in a room dedicated to this system in the Physics Department. The system comprises a model 4000/60 microvax server (with 32MB RAM, 600MB CD-ROM and 1GB hard drive) and five DEC 486DX2/66Mhz PC clients, each with 16MB RAM, 245MB hard drive and 17-in monitor. The server and clients are linked together in an ethernet LAN and the microvax is connected through a fiber optics link to the University-wide computer network. In this way, the server (with address name PANDA.UML.EDU) can directly exchange data with laboratories elsewhere via Internet. Data files are now routinely up/downloaded between PANDA and Los Alamos which has greatly facilitated the ENDF/B-VI calculations for comparison with our measurements.

In addition to our own UML codes previously developed, we have installed the latest version of GELIFIT2, a spectral analysis code widely used in other research laboratories, as well as ENDF/B-VI fission-product data files. Also installed are common utilities such as X-Windows, tape archive utility TAR, compression/decompression codes ZIP and UNZIP, file transfer code KERMIT, as well as others.

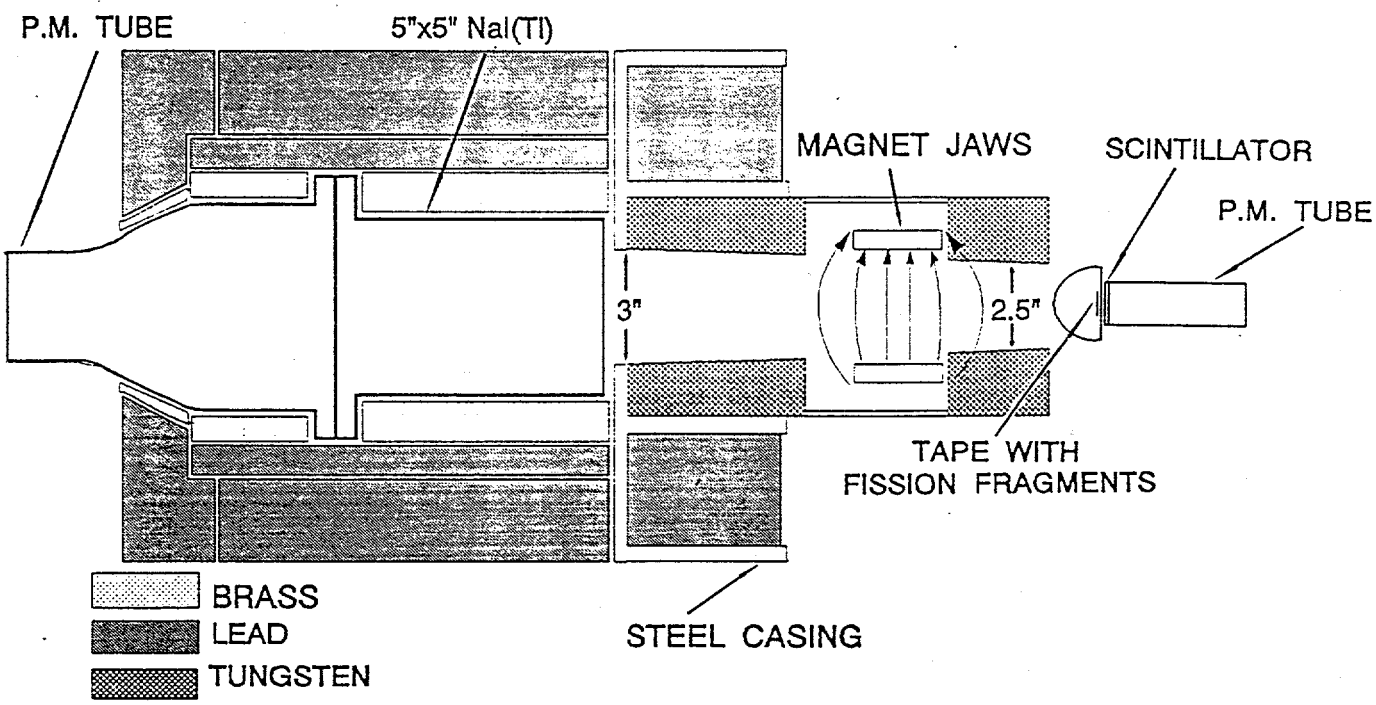

Figure B1. NaI(Tl) spectrometer with collimator, shield, and beta detector.

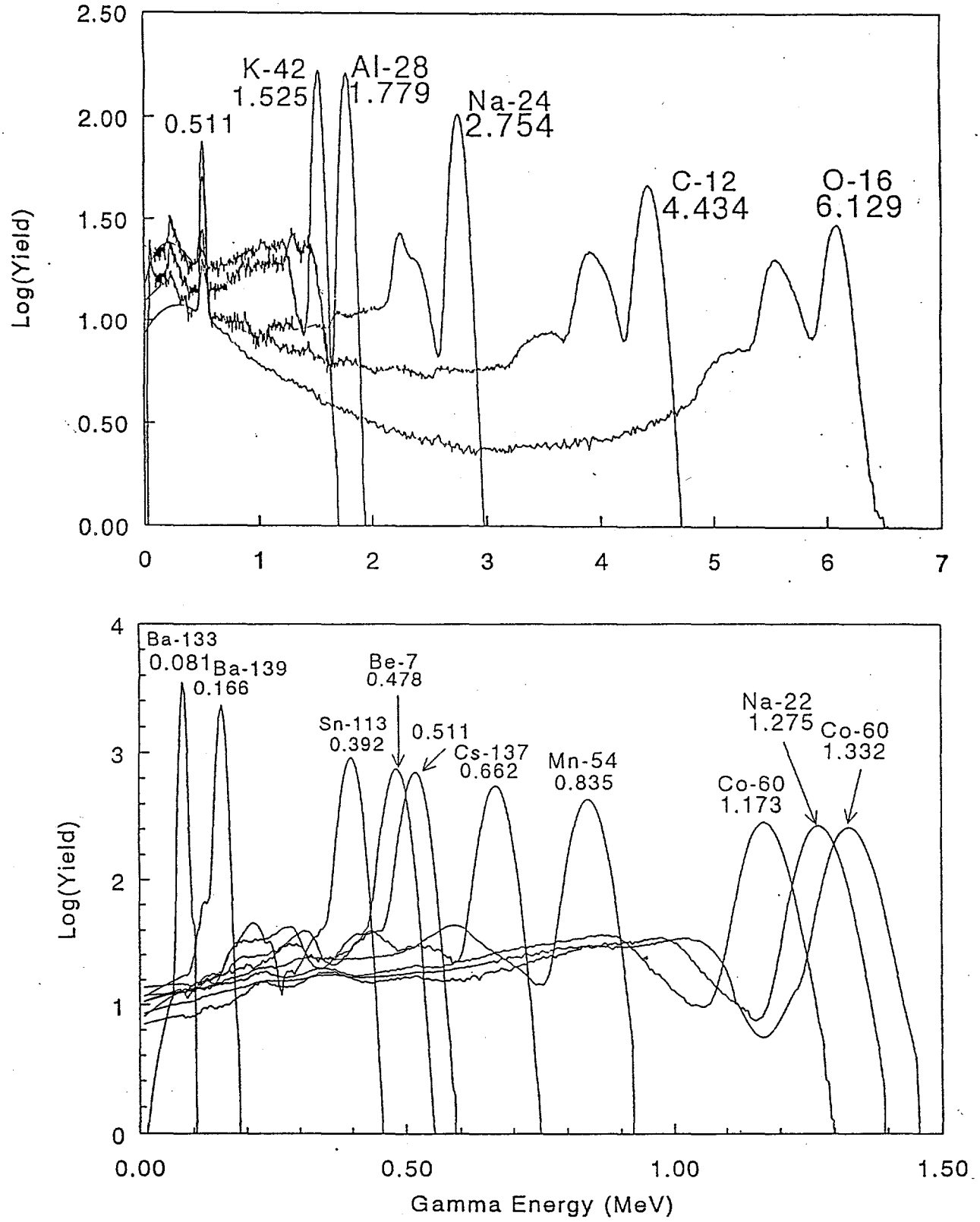

Figure B2. *Normalized response functions for the 5" x 5" NaI(Tl) spectrometer.*

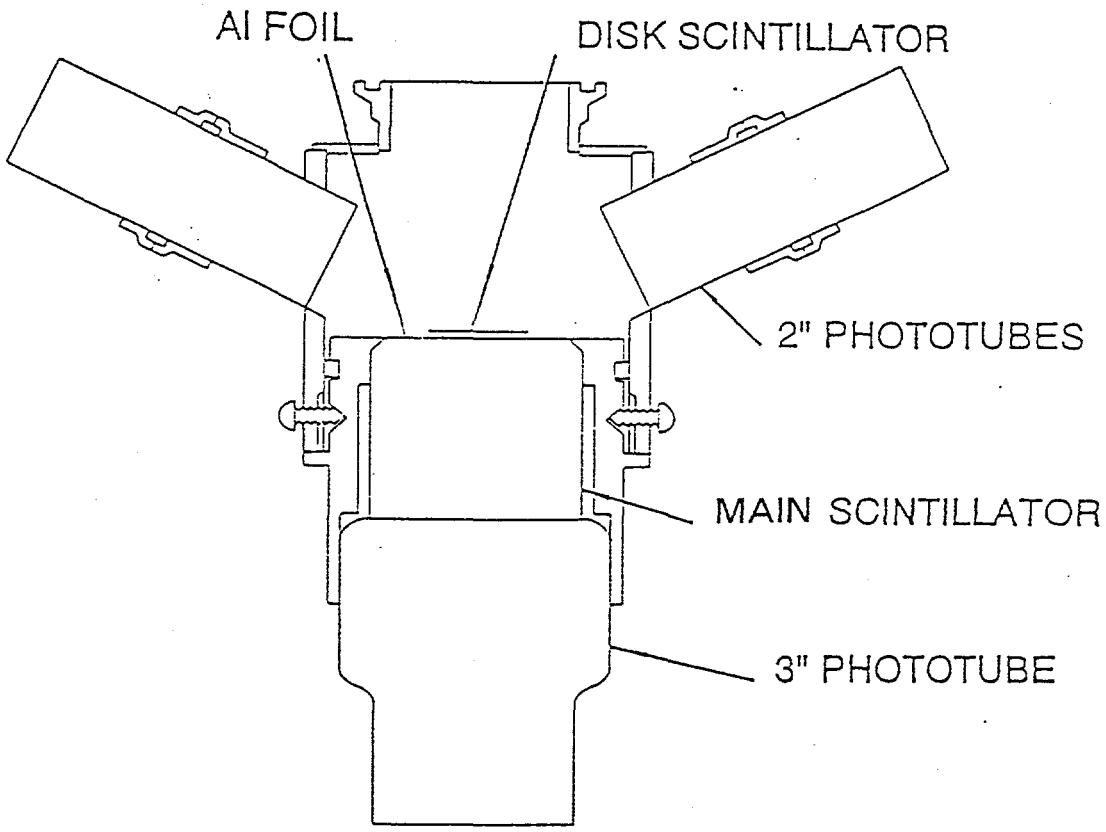

Figure B3. Cross section of the beta spectrometer.

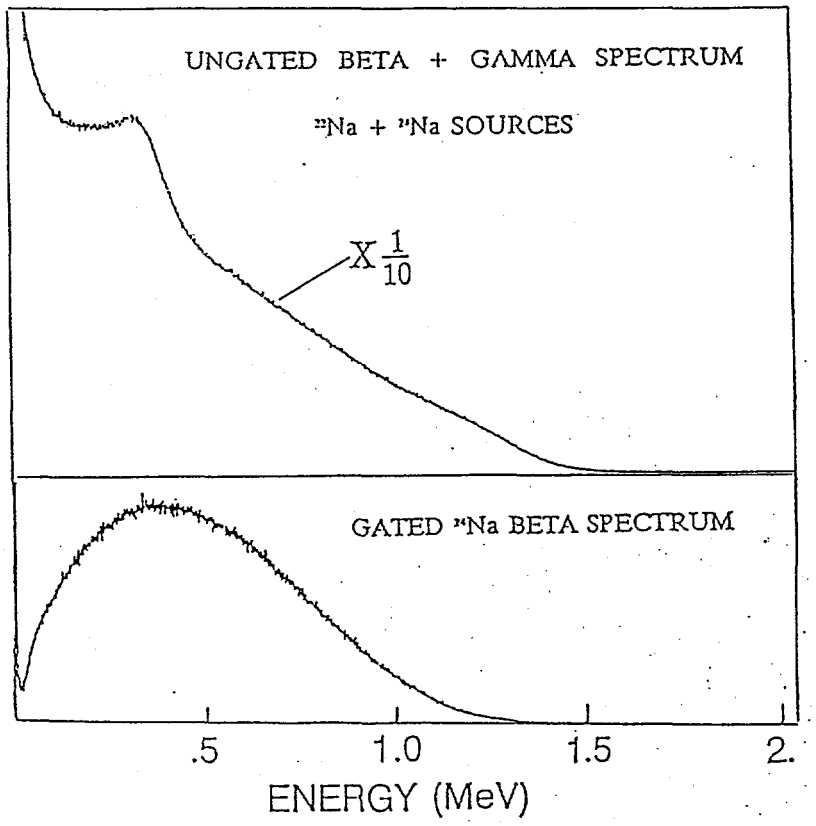

Figure B4. Demonstration of the rejection of severe gamma-ray background.

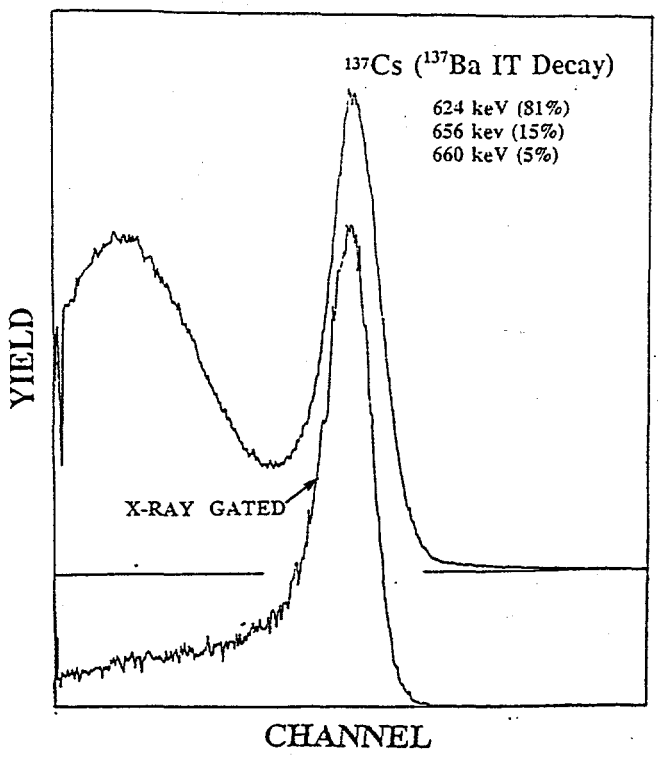

Figure B5. Measured response functions from a bare and an x-ray-gated ^{137}Cs source.

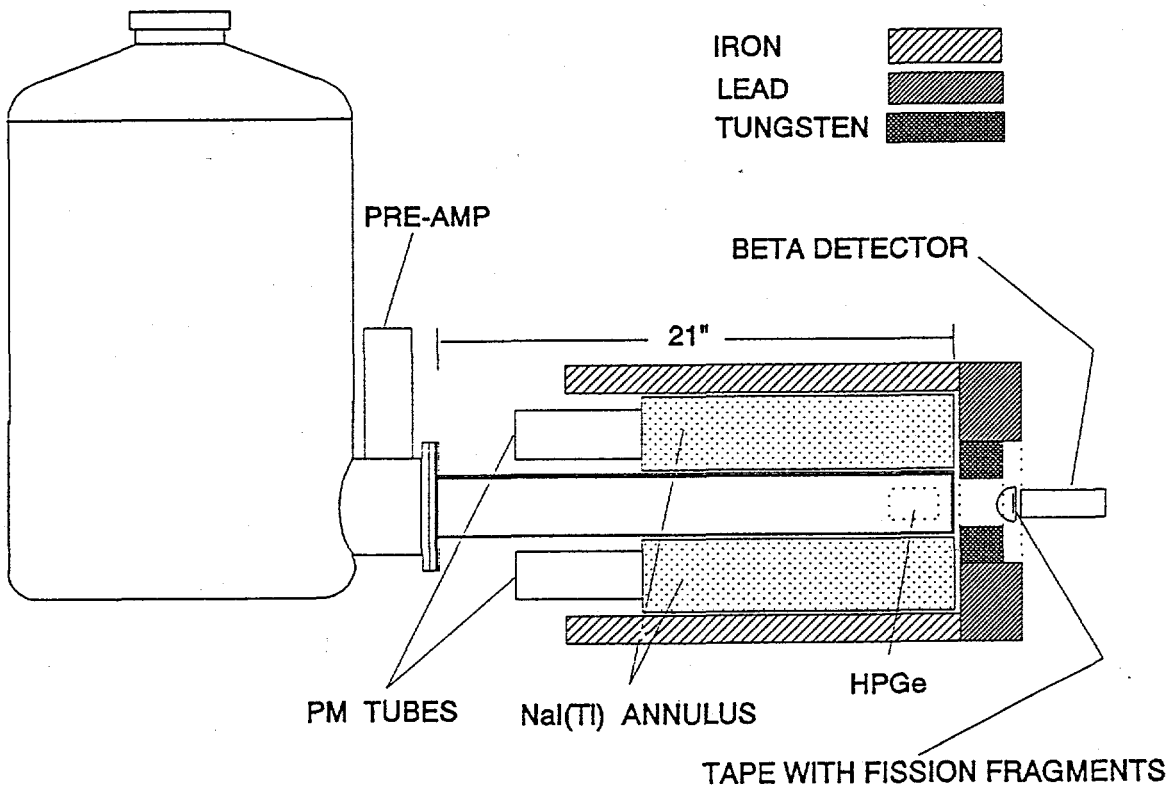

Figure B6. HPGe spectrometer with Compton supression annulus, shield and beta detector.

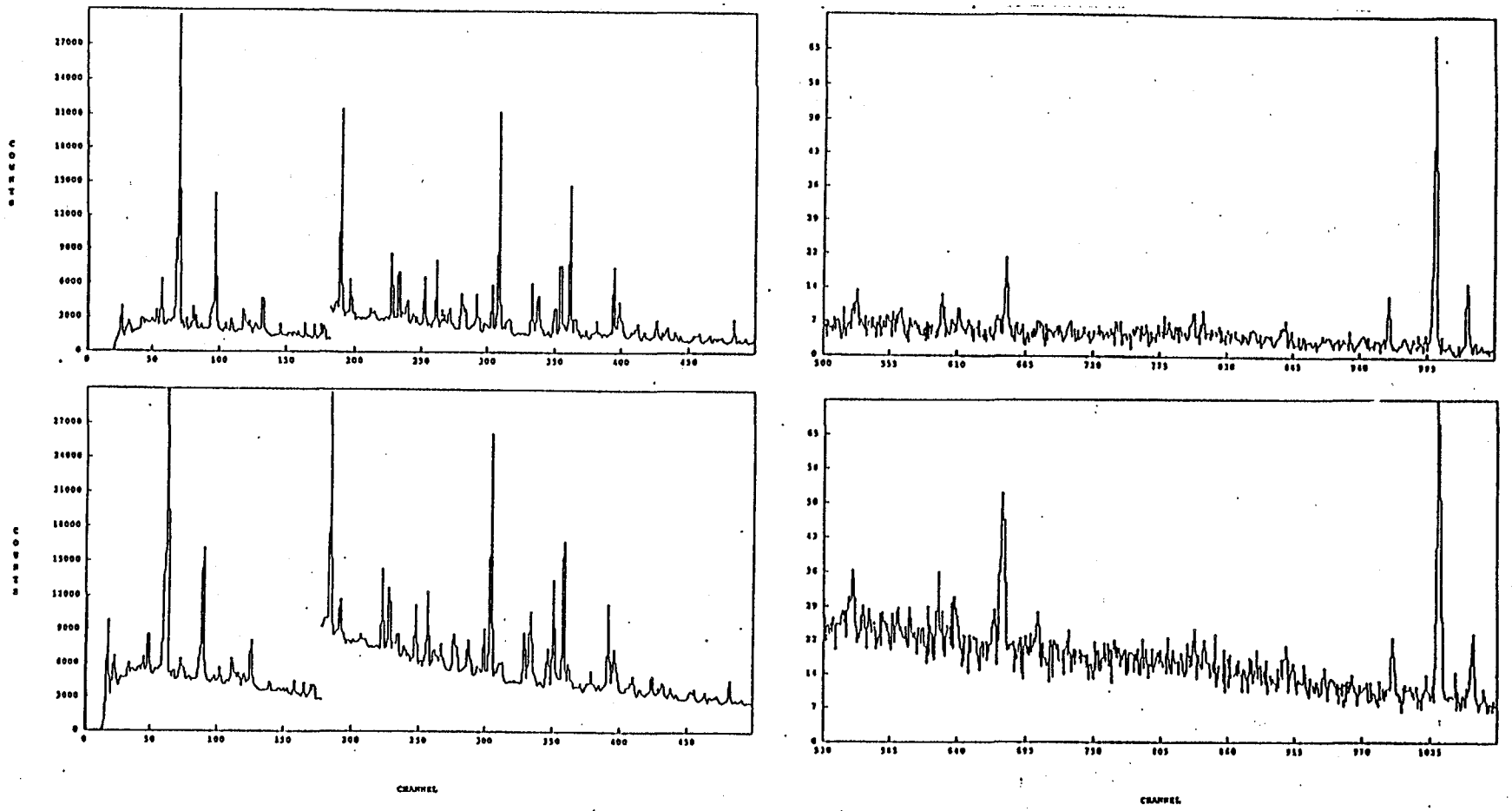

Figure B7. *Gamma-ray spectra measured for $^{235}U(n_{th},ff)$ with and without Compton suppression and using beta/gamma coincidence for the time interval 0.58 - 0.90s following fission. The spectra on the left cover the gamma energy range 0 - 1.5 MeV and those to the right are for 3 - 4.5 MeV. The upper spectra were measured with Compton suppression and the lower ones without suppression.*

C. ENDF/B-VI CALCULATIONS: UML/LANL COLLABORATION

In collaboration with Dr. Talmadge England at Los Alamos National Laboratory, our measured aggregate spectra have been compared with predictions based on individual precursor data. As part of this collaborative effort, one graduate student from our group (Joann Campbell) has spent over eight months so far during the project period at LANL under the supervision of Dr. England, supported in part by UMass Lowell and in part by LANL. Through this collaboration, detailed comparisons were made between our measurements and the spectra calculated using fission product data compiled in the Evaluated Nuclear Data File, ENDF/B-VI. Such comparisons with measured spectra are valuable in testing ENDF, which contains both experimentally-determined information based on individual nuclide data as well as theoretical model predictions (e.g., from "Gross Theory" of beta decay [Ref.9]) where experimental data are absent or suspect [Refs.10,11].

Three computer programs, CINDER10 [Ref12], SPEC5 [Ref.13], and TIMESPEC [Ref.14], were used to process ENDF data into a form with which our experimental measurements could be compared. These programs were run on a Cray YMP computer at LANL, with further processing of the resulting data performed by several auxiliary programs developed by our group.

CINDER10 is a well-established fission-product and depletion code. In this study it was used to calculate time-dependent concentrations, activities, and energy releases for individual fission products resulting from fission of ^{235}U, ^{238}U, and ^{239}Pu in a fission pulse. CINDER10 solved the differential equations describing the concentrations of actinides and fission products coupled by fission, radioactive decay, and neutron absorption in a specified neutron flux. Required input were branching ratios, halflives, cross sections, and fission yields obtained from the ENDF compilation.

SPEC5 was provided by Dr. England to calculate normalized beta and gamma-ray energy spectra for individual fission products over an arbitrary (and adjustable) energy grid. This program required ENDF spectral data as input.

The program TIMESPEC, which was written for the present study, combined the time-dependent activity calculated by CINDER10 with the normalized spectra determined by SPEC5 to produce time-dependent beta and gamma-ray energy spectra for individual fission products. From the calculated gamma-ray spectra of individual fission products, the time evolution, lifetimes, branching ratios, production probabilities and ratios of isomeric-to-ground-state productions were determined and compared with measured values found from our high-resolution gamma-ray study.

Additional utilities were also developed to further process the calculated individual spectra. The program DKSPEC was used to sum the spectra for individual fission product nuclides to determine aggregate beta and gamma-ray spectra for each experiment delay-time interval. The average energy as a function of time was also determined from the calculated aggregate spectra using program AVGENG. The resulting spectra and their average energies were then compared with those measured with our beta and NaI(Tl) gamma spectrometers. A UNIX shell script EXTRLAM was written to extract the total activity (n*lambda) of specified nuclides at each measured time interval to determine the time evolution of the nuclides for comparison with measurements.

In another useful form of processing, the gamma lines at each time produced by TIMESPEC were sorted first by intensity, to search for the 500 most intense lines during each interval, and then by energy, using shell scripts SORTBYI and SORTBYE and Fortran program GTIME2. This produced a listing of the most intense lines predicted by ENDF at each time, which aided in the identification of lines in the high-resolution gamma-ray study. The LANL 1993 ENDF/B-VI fission-product yields file was also downloaded over the Internet to UML for use in comparisons with experimentally-determined production yields.

With access now provided to the CINDER10 and SPEC5 programs on the LANL Cray computer, all calculations can now be performed from UMass Lowell using our recently upgraded data analysis system.

The methods used to convert ENDF data into a form with which our experimental data could be compared was part of the MS thesis of Joann Campbell. Further program developments applied to the study of high-resolution gamma spectra following fission form part of her Ph.D. thesis.

D. AGGREGATE DELAYED GAMMA SPECTRA FOLLOWING FISSION MEASURED USING NaI(Tl) SPECTROMETER

i) $^{235}U(n_{th},ff)$

Aggregate gamma-ray spectra following the thermal fission of ^{235}U were measured for 19 delay-time intervals out to 15,440s. Measurements for the three shortest delay times (0.14-0.32s, 0.25-0.66s, and 0.85-1.86s) are the first to be made of gamma-ray spectra for ^{235}U for times below 1s. Representative spectra, after background subtraction and for the first 2.5 MeV, are shown plotted on a linear scale in Fig. D1. This figure demonstrates the good statistical quality of the data and the changing structural character of the spectra with delay time. The shift of the end point energies of the spectra to lower energies at longer delay times is evident in Fig. D2, which shows the same data plotted on a logarithmic scale over the full measured energy range out to 8 MeV. (The counts in the high energy region constitute a statistically zero value and are therefore shown truncated.) The prominent peak at 4.45 MeV seen at shorter times is due to ^{98}Y ($T_{1/2}$=0.64s) and there is a clear increase in gamma yield at energies above this line with decreasing delay time.

The energy distribution of the aggregate gamma spectrum at short delay times is of particular interest, since the "Gross Theory" of beta decay [Ref.9] predicts a higher yield in the high-energy region than is supported by experimental studies of the gamma-ray spectra from individual fission products. We have unfolded several of the measured spectra in a preliminary fashion using FERD-PC [Ref.15] and a library of 79 response functions to span the energy 0.1 - 8.0 MeV. The resulting energy distributions can be seen in Fig. D3. Also shown in this figure are the results of summation calculations using the ENDF/B-VI data base supplemented with the predictions of beta decay "Gross Theory". Reasonable agreement is had with the unfolded measured aggregate spectra. A more efficient decomposition of the measured spectra to obtain the true energy distribution and average energies is now under way using code CRSUP recently developed in our group. The decomposition codes are discussed in Section G(ii).

The ^{235}U gamma-ray spectrum measurements and their analysis comprise part of the Ph.D. thesis of Hung Nguyen.

ii) $^{238}U(n,ff)$

Aggregate gamma spectra following the fission of ^{238}U were measured using neutrons from the fast port of our 1-MW swimming pool reactor and for 12 delay-time intervals ranging from 1.0 - 21,400s. No previous studies have been reported of ^{238}U fission-product gamma

decay below 20s. These measurements will shortly be extended to include delay times < 1s as well as longer times out to 50,000s.

Representative background-subtracted spectra are shown plotted in Figs. D4 and D5. Similar softening of the spectra and structural dependence on delay time is evident in these figures as for ^{235}U. Energy distributions for four spectra unfolded using FERD-PC are represented by the solid curves in Fig. D6. The dashed curves represent ENDF/B-VI predictions.

The ^{238}U gamma-ray spectrum measurements and their analysis comprise part of the Ph.D. thesis of Edward Seabury.

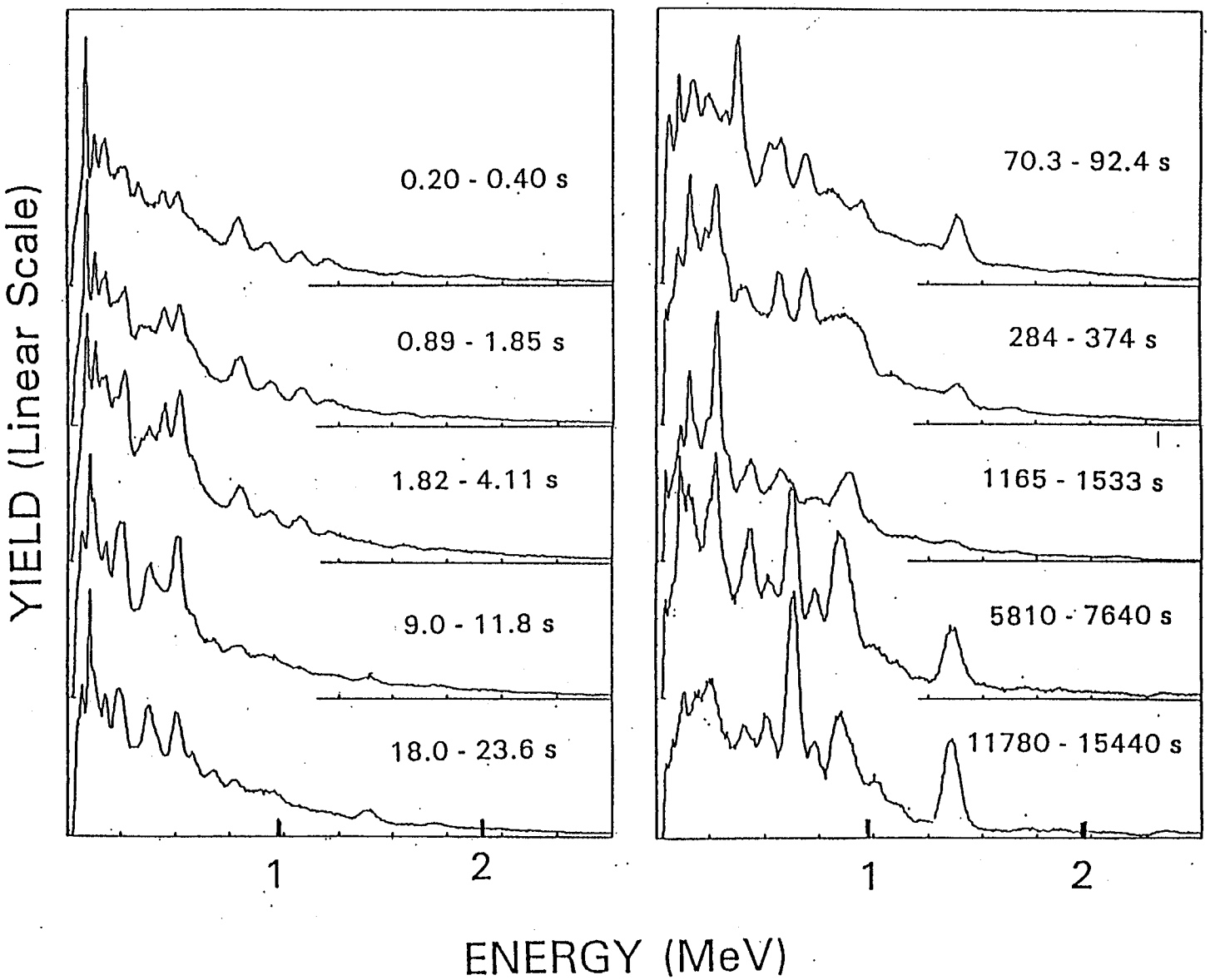

Figure D1. *$^{235}U(n_{th},ff)$ gamma-ray spectra measured at representative delay times using the NaI(Tl) spectrometer.*

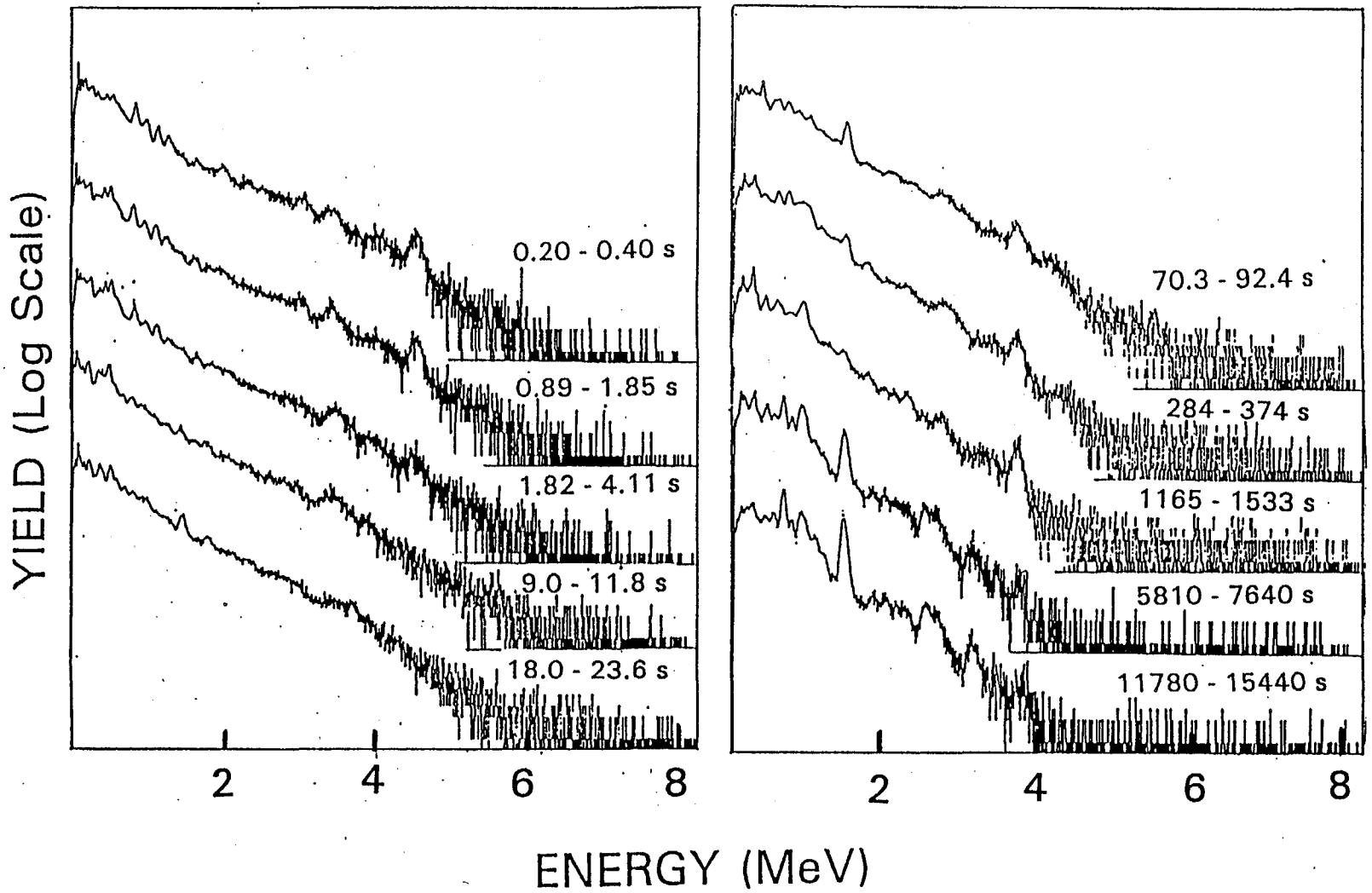

Figure D2. *Same gamma-ray spectra as in Fig. D1 replotted on logarithmic scale to view high energy gammas.*

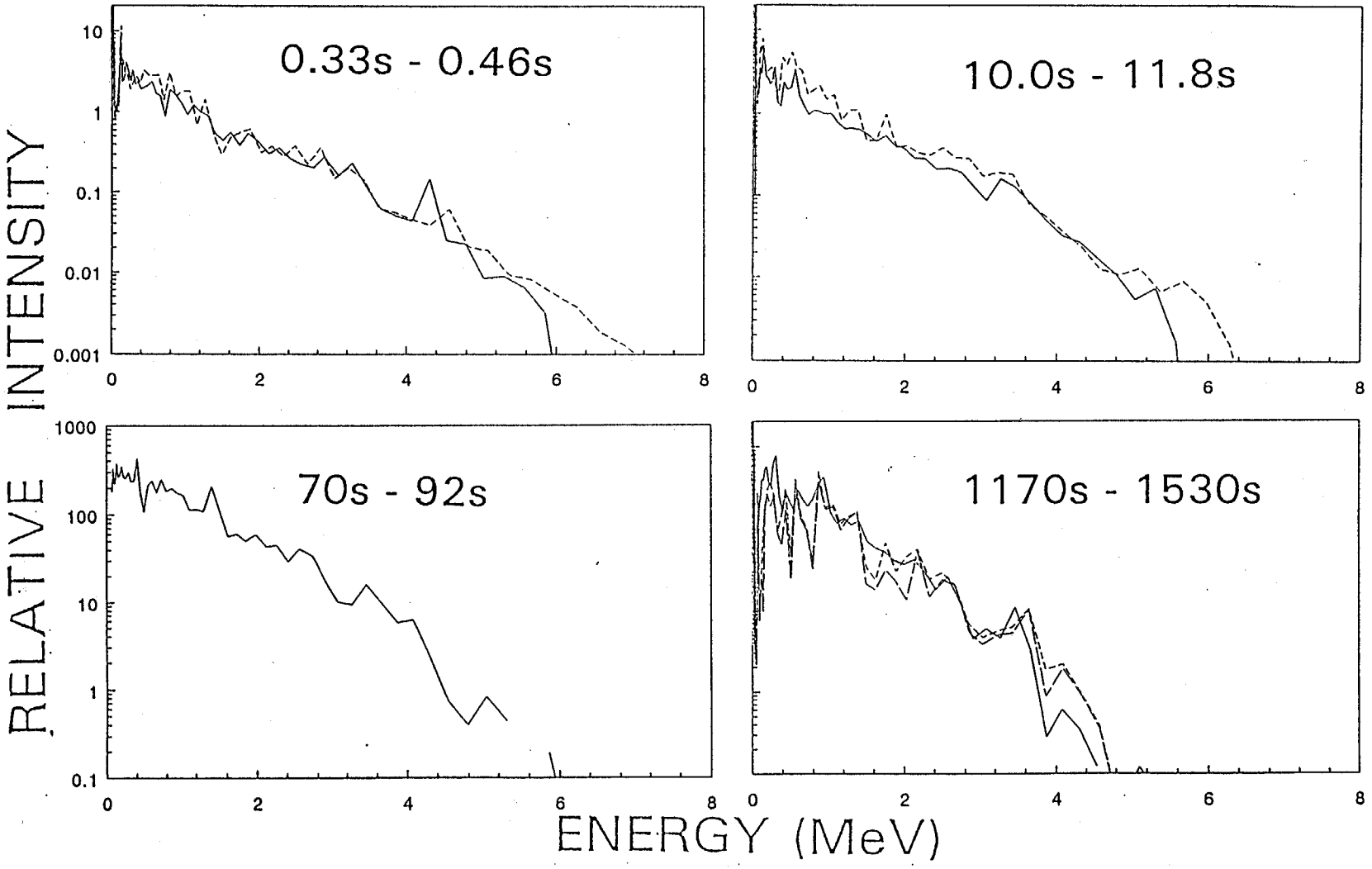

Figure D3. *Present gamma-ray energy distribution measurements for selected delay times following $^{235}U(n_{th},ff)$ (solid lines) compared with ENDF/B-VI (dashed lines).*

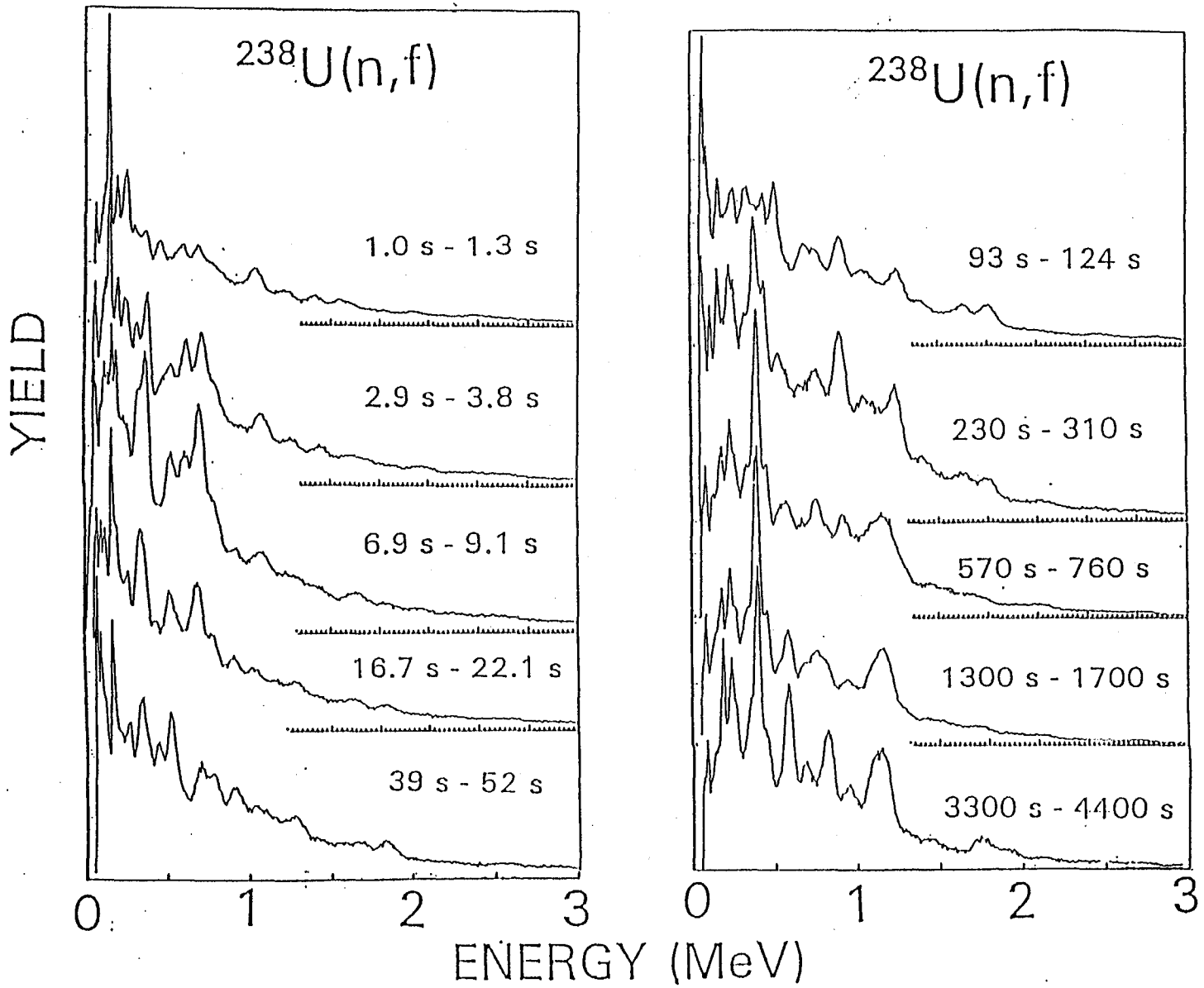

Figure D4. $^{238}U(n_{th},ff)$ *gamma-ray spectra measured at representative delay times using the NaI(Tl) spectrometer.*

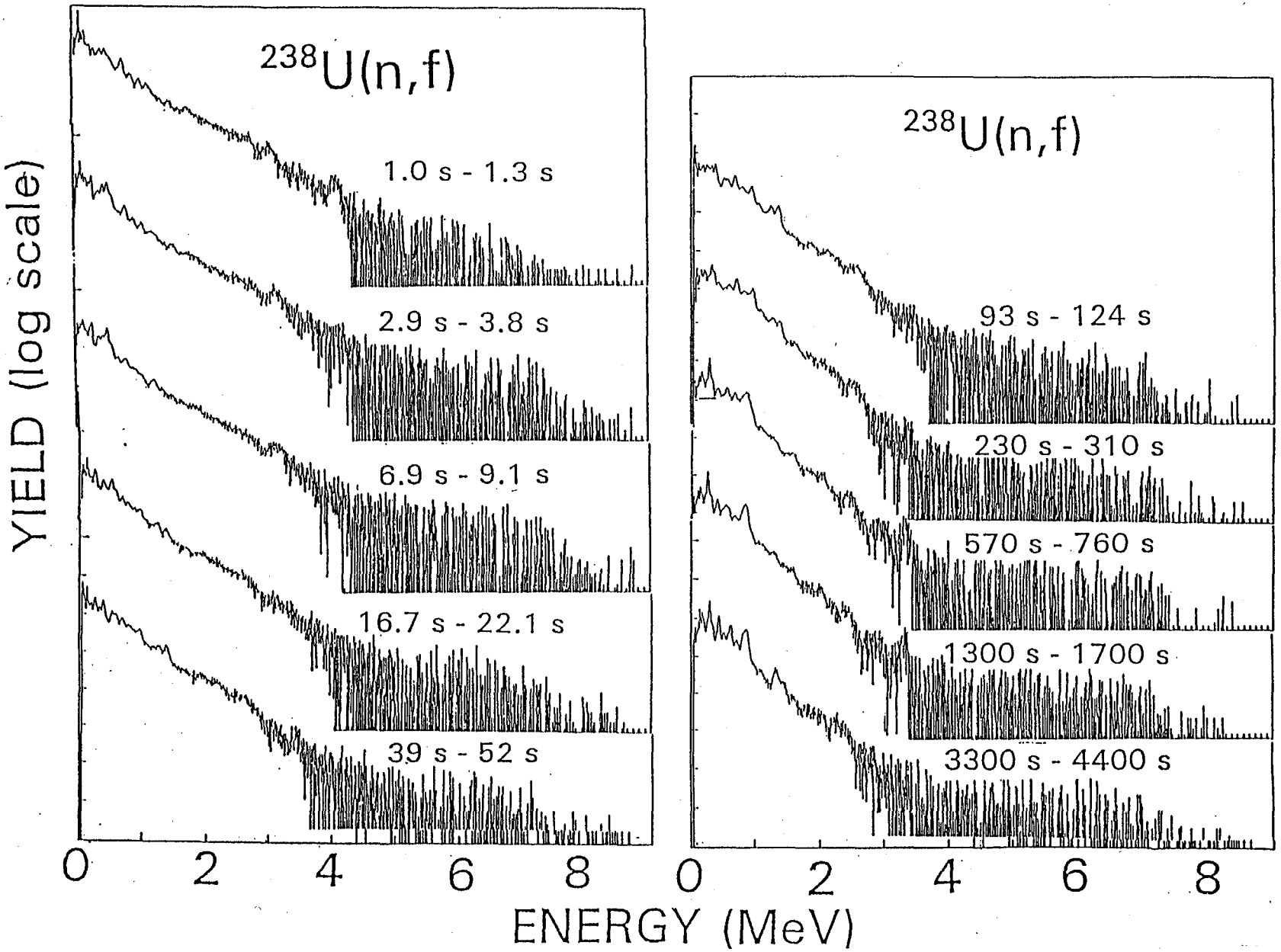

Figure D5. *Same gamma-ray spectra as in Fig. D4 replotted on logarithmic scale to view high energy gammas.*

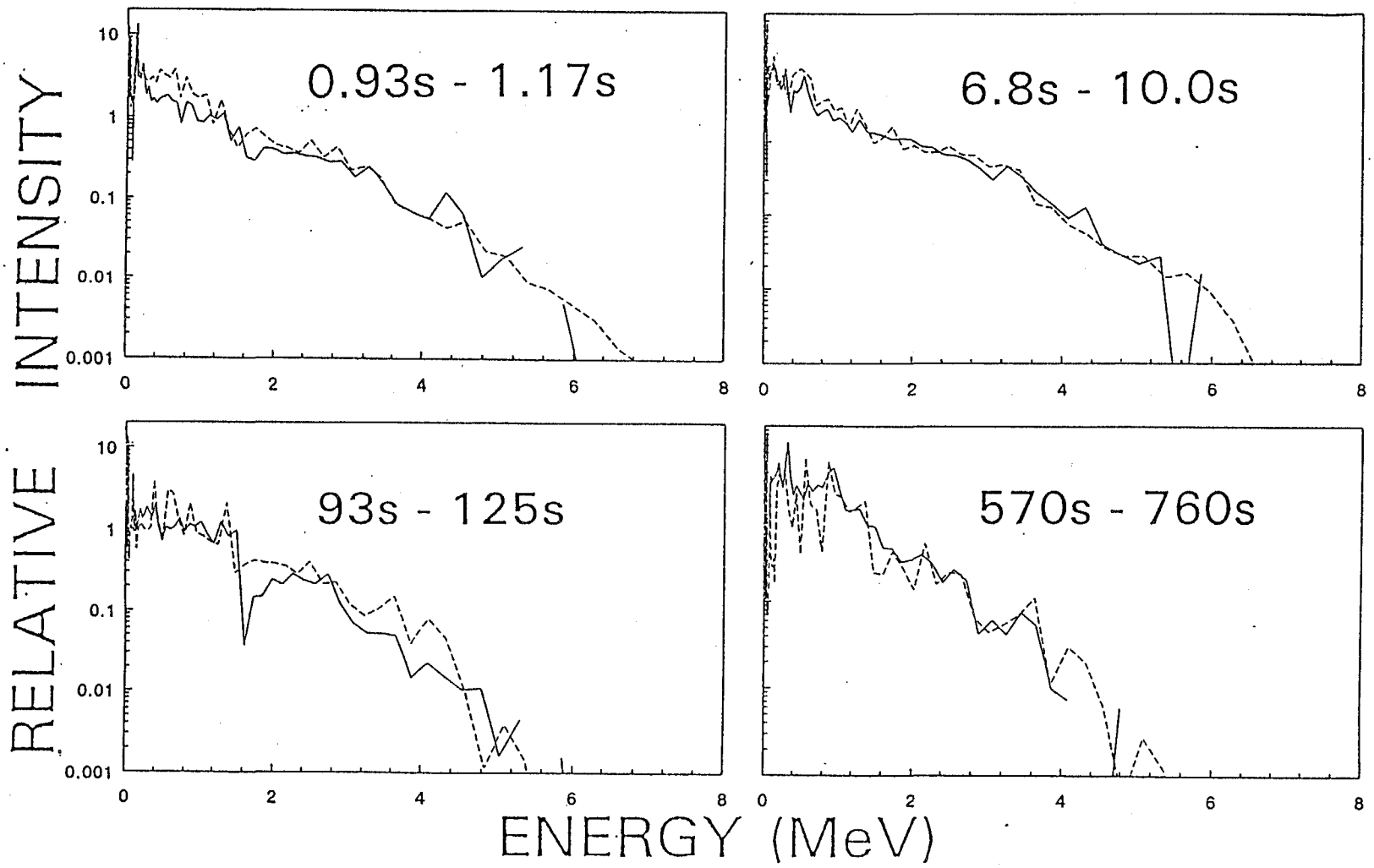

Figure D6. *Present gamma-ray energy distribution measurements for selected delay times following $^{238}U(n_{th},ff)$ (solid lines) compared with ENDF/B-VI (dashed lines).*

E. AGGREGATE DELAYED BETA SPECTRA FOLLOWING FISSION

i) $^{235}U(n_{th},ff)$

Aggregate beta spectra following the thermal fission of ^{235}U were measured out to 8 MeV for 17 delay time intervals spanning the range 0.25s to 11,960 s. They include the first aggregate beta spectra measurements at delay times below 2s. The beta spectra were corrected for background as well as for random coincidences arising from the gamma-ray discrimination system used to make the beta spectrometer insensitive to the incident gamma radiation (see Section B(ii)). Some representative spectra are shown in Figs. E1 and E2. In the linear plots (Fig. E1) the width of the Maxwellian-type distribution is seen to decrease with shorter delay times and in the logarithmic plots (Fig. E2) a considerable hardening of the spectra at shorter delay times is evident. Spectra at short delay times also display electron lines from internal transitions superimposed on the aggregate continuous beta spectra. These spectra have recently been remeasured and their analysis for obtaining the beta-energy distributions is in progress.

ii) $^{238}U(n,ff)$

Beta spectra were measured for 9 delay time intervals from 1s to 1025s. The specta were analyzed to extract energy distributions after correcting for detector response functions. These distributions are shown for four time intervals in Fig. E3 (solid curves). The beta energy distributions from $^{235}U(n_{th},ff)$ previously measured by Dickens [Ref.16] are also indicated here (dashed curves), together with the results of ENDF/B-VI summation calculations (dash-dot curves). ENDF predicts that the beta spectra for ^{235}U and ^{238}U should be nearly identical and this is borne out in this comparison. However, our spectra are generally softer than is predicted by ENDF/B-VI.

iii) $^{239}Pu(n_{th},ff)$

During the current intersession we measured the beta spectra for plutonium at 7 delay times, ranging from 0.8s to 213s. These measurements are currently being analyzed. This study will be extended to both shorter and longer delay times during the remainder of this project period, when the plutonium gamma measurements will also be completed.

In calculating average beta energy from the ENDF spectra to compare with measured values, a low-energy cutoff value has been used to match the energy threshhold of the beta spectrometer. The ENDF calculations nevertheless consistently show more energetic spectra

than our measured ones and those of Dickens [Ref.16]. In an effort to better understand this discrepancy we are currently studying the various contributions to the calculated spectra. The normal calculated spectrum contains summed contributions from both discrete spectra (i.e., due to internal conversion) and continuous spectra (as predicted by Fermi theory based on experimental evidence and modelling and also on the Gross Theory of beta decay). Modifications have been made to the SPEC5 program to separate the discrete, experimental and Gross Theory contributions to the beta spectrum and these are currently being studied.

The measurements and analysis of beta spectra for $^{235,238}U$ and ^{239}Pu beta spectra are part of the Ph.D. thesis of Shengjie Li.

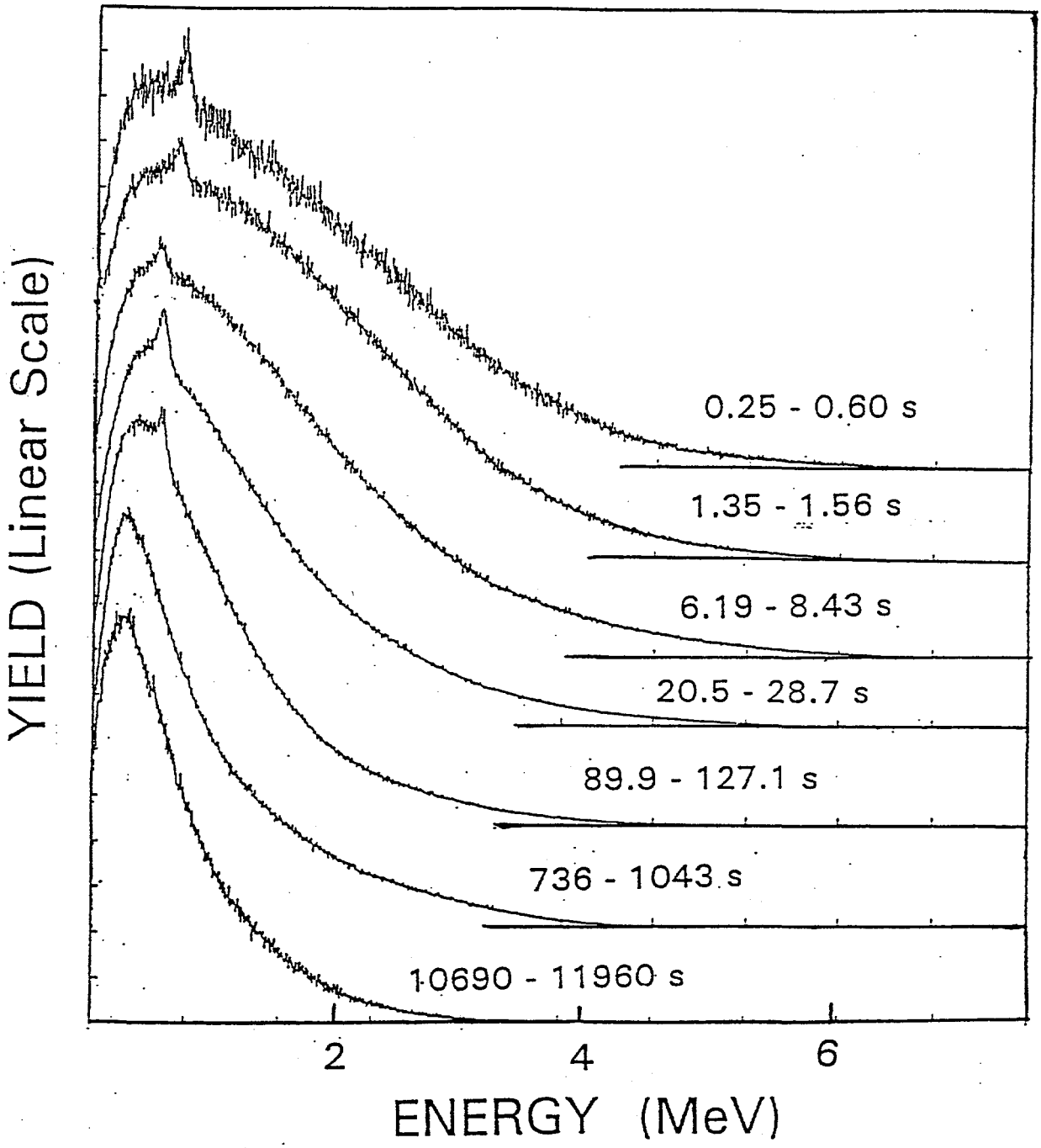

Figure E1. *$^{235}U(n_{th},ff)$ beta spectra measured at several representative delay times.*

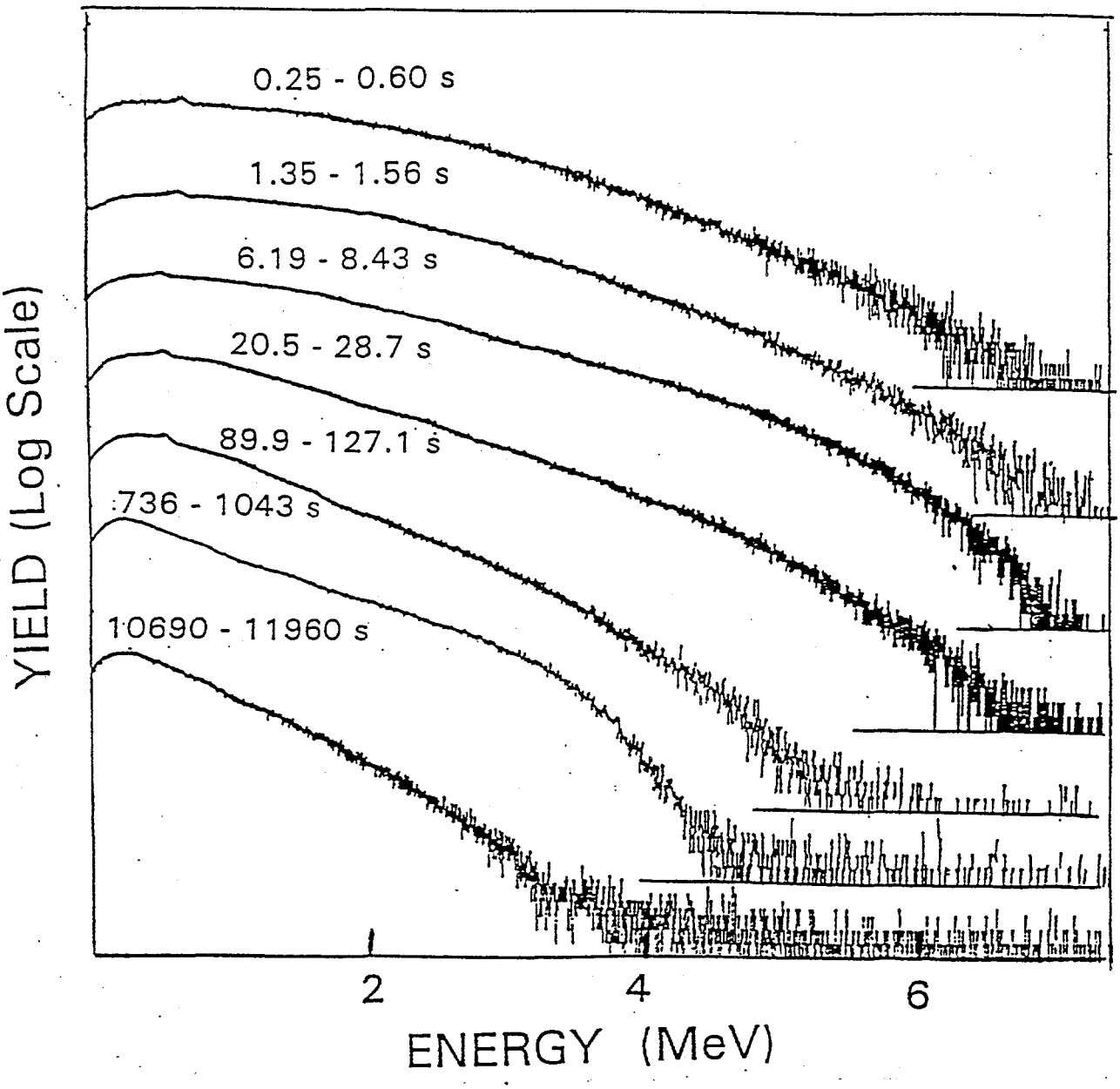

Figure E2. Same beta spectra as in Fig. E1 replotted on logarithmic scale to reveal end-point energies.

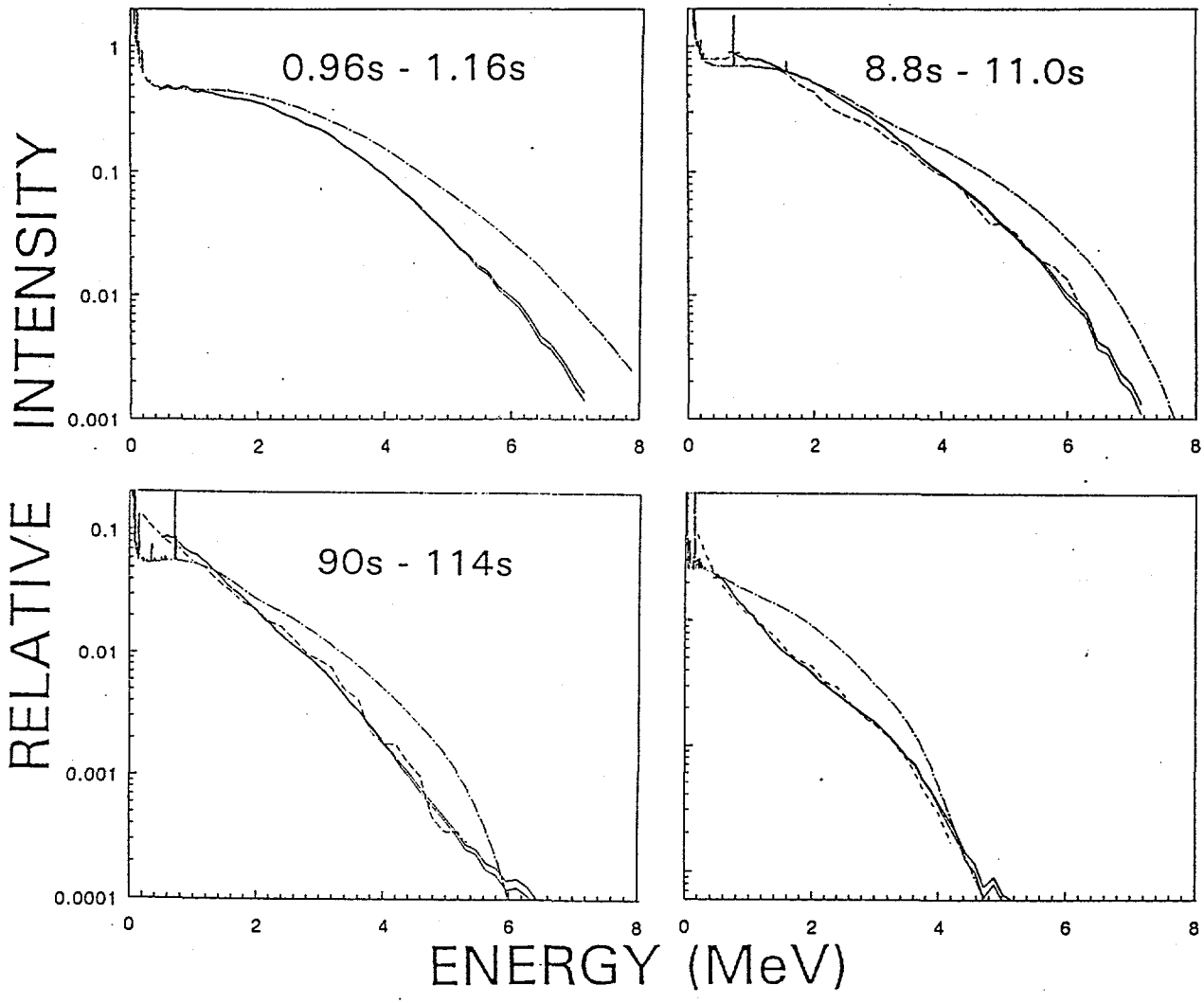

Figure E3. *Measured beta energy distributions for selected delay times following $^{238}U(n_{th},ff)$ (solid curves) compared with ENDF/B-VI (dash-dot curves). Dashed curves are from $^{235}U(n_{th},ff)$ measurements at similar delay times [Ref. 16].*

F. AGGREGATE BETA AND GAMMA DECAY HEAT MEASUREMENTS

Average energies determined from our beta and gamma energy distributions have been used to obtain decay heats as a function of time after fission. Absolute normalization of the results was achieved using the aggregate beta activity calculated using CINDER and ENDF/B-VI fission-product data. The beta decay heat was taken to be the product of the beta activity and our measured average beta energy, and the gamma decay heat was taken to be the product of the beta activity, the number of gamma-rays/beta determined from our measurements, and the average gamma-ray energy. These results are preliminary since the average energies have not yet been corrected for the loss of noble gases, true-coincidence pulse summing and (for the beta spectra) contributions from internal conversion electrons. Figure F1 shows these preliminary beta and gamma decay heats for fast neutron induced fission of ^{238}U. The error bars reflect our experimental uncertainties plus estimated uncertainties due to the missing corrections. Also shown for comparison are the earlier measurements of Akiyama et al. [Refs.17-19] (YAYOI). The solid curves labeled by flags are CINDER calculations based on ENDF/B-VI data [Ref.10]. They explicitly depict decay heat contributions due to measurements versus theoretical calculations. The designated flags refer to the following components: Flag 0 - average energy obtained from decay schemes [Ref.11]; Flag 1 - average energy "directly" measured at Studsvik [Ref.20]; Flag 2 - average energy from Gross Theory complemented by incomplete measurements; Flag 3 - average energy from Gross Theory.

In general our results are in good agreement with the YAYOI measurements as well as the theoretically augmented ENDF/B-VI calculations. Significantly, the ENDF/B-VI beta predictions for the short delay times are remarkably similar to our measurements. However, our results suggest a considerably larger gamma decay heat component at these times, a discrepancy that merits further investigation. With the completion of our study, these measurements will be extended down to times as short as 0.2 s after fission. Similar decay heat comparisons will soon be available from the energy distributions determined from the studies of thermal neutron fission of ^{235}U and ^{239}Pu.

Measurements of beta decay heat are part of the Ph.D. thesis of Shengjie Li and measurements of the gamma decay heat for ^{238}U are part of the Ph.D. thesis of Edward Seabury.

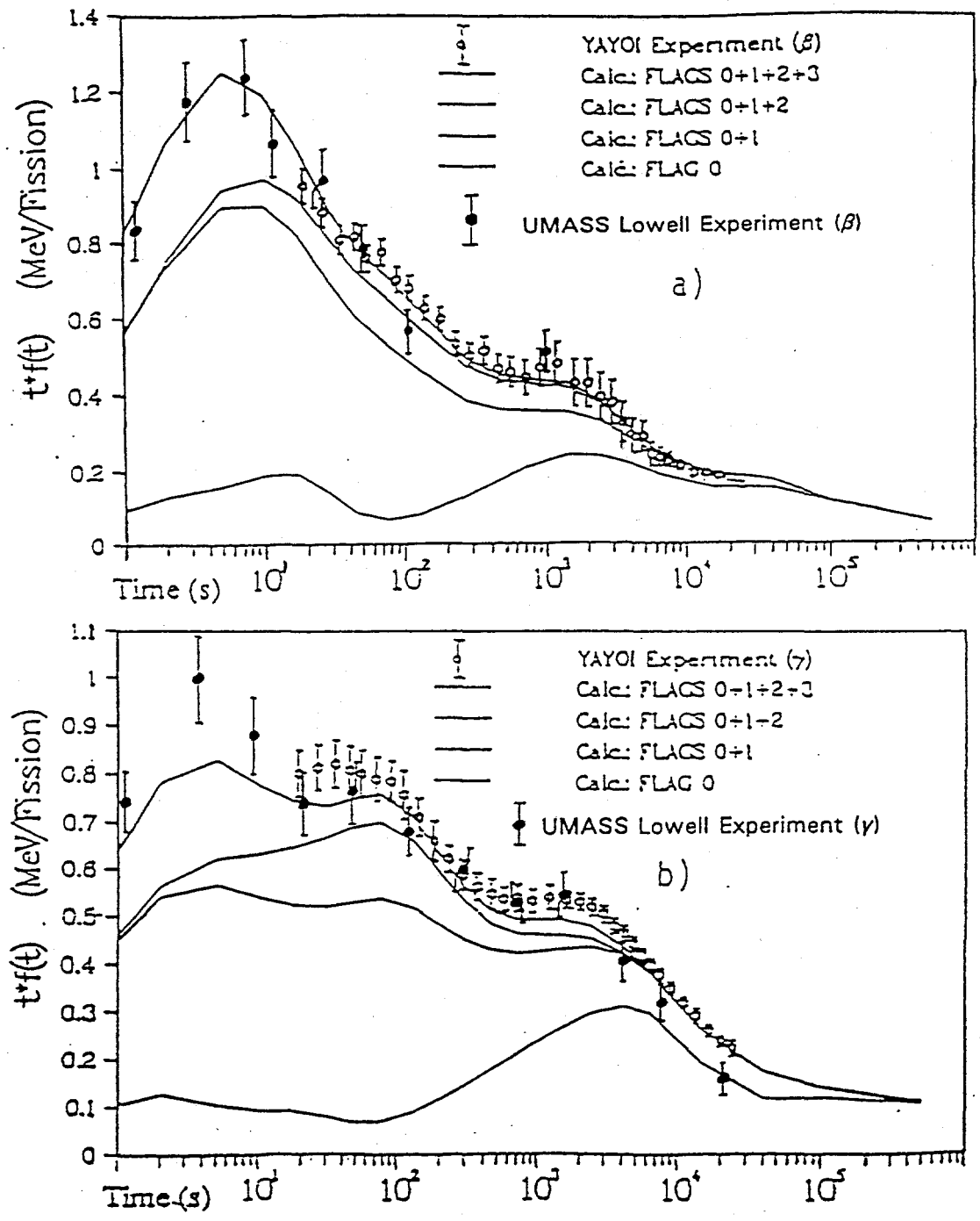

Figure F1. Comparison of our beta and gamma decay heat measurements (UMassLowell) to those of Akiyama et al. [Ref.17-19] (YAYOI) and calculations based on ENDF/B-VI.

G. INDIVIDUAL NUCLIDE STUDY USING HPGe SPECTROMETER

i) $^{235}U(n_{th},ff)$

High-resolution gamma-ray spectra were measured for 21 delay time intervals covering the range 0.2s to nearly 100,000s following fission. Analysis of the spectra with the code NDAA (see Section H(i)) has so far been carried out to nearly 1000s and from these over 30 fission-product nuclides have been identified. From relative gamma line intensities and their time evolutions, relative yields for fission products, isomeric/ground-state production ratios, precursor halflives, and beta-branching ratios have been calculated for comparison with ENDF/B-VI.

Figures G1(a) and G1(b) show a typical spectrum, measured between 0 and 6 MeV and for a time interval of 0.58 - 0.90s, with a number of prominant lines identified by their precursors. The density of clearly resolvable gamma lines in this spectrum is evident in Table III, which lists the energies of lines extracted from the spectrum. In determining these, only those peaks whose areas exceeded the local background area by at least two standard deviations were included, which in this example results in 286 such peaks (including single and double escape peaks). Almost half the total number of gamma lines lie below 1 MeV and no gamma lines are observed above 4.5 Mev. Although very rich in gamma lines, the spectra are nevertheless quite selective in the nuclides which are the predominant contributors. This selectivity results from the enhancement which occurs for those nuclides having beta-decay halflives which approximately match the delay interval following fission. Shorter-lived precursors have largely died away whereas longer lived ones do not decay appreciably during the selected interval. For example, the principal contributors identified in Fig. G1 have halflives of 0.40s (^{97}Sr), 0.65s (^{98}Y), 0.7s (^{98}Sr), 1.1s (^{96}Sr), 1.21s (^{98m}Y), and 2.1s (^{98}Zr) which are all comparable to the measurement interval of 0.58 - 0.90s.

The measured time dependences of individual lines were compared with predictions based on calculations using the LANL code CINDER10 [Ref.12]. These are shown in Figs. G2 - G5 after correcting for background and detector efficiency. The spectra were relatively normalized between time intervals by normalizing the measured total gamma energy to ENDF gamma decay-heat values. Lines drawn through the data points for individual nuclide gammas represent the gamma activities calculated for various times using data from ENDF/B-VI and code CINDER10. They are normalized to provide best fits to the measurements. The fits are generally very satisfactory and the figures demonstrate the sensitivity of the measurements to short-lived fission products (e.g., ^{98}Y: $T_{1/2}$ = 0.64s, and ^{97}Sr: $T_{1/2}$ = 0.42s) as well as to the direct-vs.-beta chain production, depicted by the curvatures apparent in a number of plots (e.g., ^{99}Zr and ^{145}La).

Table III. Gamma-ray Lines extracted from Spectrum of Fig. G1.

E (MeV)	Peak Area	E (MeV)	Peak Area	E (MeV)	Peak Area	E (MeV)	Peak Area	E (MeV)	Peak Area	E (MeV)	Peak Area
0.0584	6273	0.3560	312	0.7104	298	1.2284	62	1.9185	44	3.2503	30
0.0670	3386	0.3596	2808	0.7135	90	1.2351	129	1.9248	61	3.2848	177
0.0741	266	0.3634	622	0.7240	994	1.2442	396	1.9569	114	3.3086	287
0.0765	295	0.3680	1224	0.7298	180	1.2478	144	1.9825	110	3.3686	17
0.0817	1843	0.3752	536	0.7364	256	1.2578	322	1.9958	151	3.3875	19
0.0856	1282	0.3792	234	0.7462	112	1.2794	385	2.0094	101	3.3987	124
0.0926	1072	0.3876	2132	0.7558	402	1.2908	267	2.0353	55	3.4143	24
0.0973	1959	0.4005	2992	0.7597	262	1.3088	250	2.0798	48	3.4557	32
0.1028	6630	0.4069	686	0.7706	551	1.3215	59	2.0864	33	3.5457	30
0.1104	1167	0.4102	479	0.7822	343	1.3265	197	2.0932	85	3.5713	23
0.1147	1550	0.4147	963	0.7937	113	1.3316	80	2.1203	68	3.5966	33
0.1191	15699	0.4181	258	0.8010	336	1.3373	47	2.1727	55	3.6615	18
0.1186	9344	0.4289	1512	0.8094	7066	1.3646	57	2.1838	74	3.7582	12
0.1215	45513	0.4325	1027	0.8310	154	1.3847	241	2.2106	204	3.7632	23
0.1257	891	0.4401	406	0.8367	2302	1.3997	421	2.2253	49	3.8155	23
0.1302	1182	0.4447	1560	0.8471	396	1.4215	60	2.2462	45	3.8644	23
0.1377	3119	0.4561	492	0.8643	123	1.4275	285	2.2710	65	3.8852	20
0.1403	1910	0.4616	1974	0.8749	125	1.4389	92	2.2870	64	3.9285	15
0.1465	657	0.4630	9164	0.8782	155	1.4552	51	2.3051	82	3.9390	51
0.1513	311	0.4738	398	0.8841	107	1.4686	79	2.3248	92	3.9777	13
0.1610	22125	0.4796	1092	0.8912	315	1.4716	118	2.3422	35	4.0143	15
0.1725	1211	0.5039	1799	0.8952	105	1.4924	49	2.3576	35	4.1624	21
0.1792	2293	0.5109	2745	0.9069	135	1.5016	89	2.3749	41	4.1745	13
0.1872	797	0.5215	157	0.9145	346	1.5154	103	2.4203	141	4.2730	12
0.1925	4852	0.5293	1845	0.9251	92	1.5239	56	2.4301	47	4.3653	13
0.1988	2207	0.5359	4554	0.9314	1176	1.5484	47	2.4351	61	4.3964	29
0.2048	2210	0.5460	6779	0.9382	62	1.5562	77	2.4402	55	4.4155	11
0.2127	7350	0.5513	1085	0.9490	246	1.5676	64	2.4633	34	4.4500	188
0.2185	280	0.5547	422	0.9535	915	1.5772	454	2.5143	38	4.4897	47
0.2323	1402	0.5600	130	0.9693	2659	1.5904	514	2.5733	112	4.5504	9
0.2374	298	0.5639	255	0.9801	144	1.5970	58	2.6014	36		
0.2425	1096	0.5705	234	0.9864	187	1.6055	43	2.6146	60		
0.2472	895	0.5754	673	0.9994	98	1.6124	82	2.6793	41		
0.2514	892	0.5785	336	1.0046	188	1.6165	46	2.6883	36		
0.2588	1712	0.5835	222	1.0112	256	1.6326	304	2.6974	27		
0.2629	438	0.5895	241	1.0227	242	1.6538	55	2.7168	51		
0.2688	1768	0.5940	3384	1.0639	287	1.7227	38	2.7273	53		
0.2761	1922	0.6007	2463	1.0722	98	1.7361	85	2.7425	64		
0.2791	2555	0.6136	377	1.0895	380	1.7440	191	2.7681	62		
0.2847	358	0.6203	723	1.0956	122	1.7500	305	2.7747	49		
0.2918	859	0.6276	319	1.1030	4685	1.7864	49	2.8825	26		
0.2961	8583	0.6405	1014	1.1173	97	1.7904	52	2.8897	41		
0.3030	255	0.6472	307	1.1305	56	1.8011	133	2.8990	25		
0.3068	1654	0.6517	824	1.1419	71	1.8100	157	2.9281	28		
0.3103	256	0.6605	432	1.1485	56	1.8377	80	2.9307	31		
0.3257	186	0.6661	165	1.1531	79	1.8537	45	2.9412	369		
0.3285	517	0.6856	384	1.1670	85	1.8677	69	2.9531	30		
0.3325	559	0.6936	155	1.1756	120	1.8869	77	3.0626	39		
0.3408	124	0.6971	322	1.1848	97	1.8969	65	3.0630	43		
0.3459	263	0.7028	224	1.1945	281	1.9042	480	3.2010	54		
0.3520	2938	0.7067	223	1.2827	2135	1.9102	79	3.2277	79		

Relative gamma-line intensities following beta decay were extracted for several nuclides. Results for one such case, ^{98g}Y ($T_{1/2}$ = 0.64s), are presented as an example in Table IV and compared with the values published in the Nuclear Data Sheets [Ref.21]. The overall agreement is quite satisfactory and indicates that the majority of lines are not appreciably intermixed with lines from other precursors in the spectrum.

Table IV

Present measurement and NDS relative gamma-line intensities following the beta decay of ^{98g}Y ($t_{1/2}$ = 0.64 s).

Level (MeV)	Gamma (MeV)	NDS [Ref.17]	Measured Relative Intensities
4.451	3.228	1.4	1.1
	4.450	3.4	3.3
4.164	2.306	0.7	0.7
	2.421	1.6	1.4
	2.574	0.8	1.2
	2.941	5.8	4.7
	3.310	2.4	4.0
1.859	0.269	2.6	2.7
	0.636	0.5	-
1.744	0.522	0.7	0.7
	0.890	0.4	1.5
	1.744	1.4	1.6
1.437	0.214	1.5	-

From these measurements cumulative (and several independent) fission-product yields have also been determined for 28 nuclides. These results are presented below where they are compared with ENDF/B-VI predictions. Further analysis of the ^{235}U measurements is continuing out to longer time intervals to provide further tests for the ENDF data base.

The ^{235}U high-resolution gamma spectra measurements and their analysis are part of the Ph.D. thesis of Sameer Tipnis.

ii) ^{238}U(n,ff)

High-resolution aggregate gamma spectra were measured for 13 different delay times, from 0.59s to 24,100s, following fast fission of ^{238}U. Background subtraction and peak area calculations were performed on our VAX using the GELIFIT2 spectral analysis code. A

FORTRAN conversion file was written to transform data from our MCA which were stored in PC data files into a form readable by GELIFIT2. A separate program was also written to convert from GELIFIT2 format to an ASCII text file.

In a preliminary analysis a total of 34 gamma lines from eight nuclides (^{146}La, ^{144}Ba, ^{96m}Y, ^{94}Rb, ^{97m}Y, ^{96}Sr, ^{99}Zr, and ^{142}Cs) have so far been identified and representative time evolutions for two of these are shown plotted in Fig. G6. Measurements at very short times (<.5s) and and at longer times out to 50,000s will be completed during the remainder of this period.

The ^{238}U high-resolution gamma spectra measurements and their analysis are part of the Ph.D. thesis of Joann Campbell.

iii) Fission Product Yields

From the measured gamma intensities and known gamma activities [Ref.21], the beta activity was calculated for each parent nuclide identified in the ^{235}U study and used to determine the relative fission-product yields for 28 nuclides. The relative cumulative yields are compared in Table V with the values calculated from ENDF/B-VI. In the case of rubidium and cesium isotopes, we are able to measure the relative independent yields (i.e., from fission alone) since their krypton and xenon feeders in the beta chains are missing in our measurements. A comparison of these relative independent yields to those from ENDF/B-VI is presented in Table VI. The uncertainties in the measurements are estimated at no more than 10% and the assigned pre-ENDF/B-VI uncertainties (%) are in parentheses.

A comparison of isomeric to ground-state production ratios was also made where different gamma transitions were involved. Table VII shows such comparisons for ^{97m}Y/^{97}Y, ^{98m}Y/^{98}Y and ^{90m}Rb/^{90}Rb. The ^{97}Y and ^{90}Rb ratios are in fair agreement with ENDF whereas there is substantial disagreement in the case of ^{98}Y.

Measured fission product yields can be used to derive parameters required by empirical dispersion models used to characterize yields as a function of charge and mass distributions [Ref.22]. Once these parameters are established for a fissionable nuclide, the models can be used to estimate the yields for fission products that have not been measured. Since the majority of fission products fall into this category, it is important to know how reliable these estimates are. The measurements are particularly sparse in the case of isomers and very short-lived fission products, for which the present study should provide valuable tests of the models.

The determination of fission product yields from ^{235}U are part of the Ph.D. thesis of Sameer Tipnis.

TABLE V

Relative Cumulative Fission Product Yields From $^{235}U(n_{th},f)$

NUCLIDE	HALFLIFE (s)	ENDF/B-VI	PRESENT
^{93}Sr	445	2.99 (1.0)	3.6
^{94}Sr	75.3	3.07 (1.4)	3.83
^{96}Sr	1.06	1.17 (6.0)	1.41
^{97}Sr	0.42	0.83 (2.8)	0.56
^{98}Sr	0.65	0.39 (6.0)	0.39
^{96m}Y	9.6	0.97 (45)	0.61
^{97m}Y	1.23	0.88 (?)	1.29
^{97g}Y	3.5	1.46 (23)	1.42
^{98g}Y	0.64	0.53 (23)	1.91
^{98m}Y	2.0	0.63 (32)	0.33
^{99}Y	1.47	1.00 (6.0)	0.79
^{99}Zr	2.1	2.69 (2.8)	2.69
^{100g}Y	0.735	0.15 (64)	0.13
^{141}Ba	1096	2.78 (2.8)	3.52
^{143}Ba	14.5	2.65 (2.8)	3.62
^{144}Ba	11.5	2.10 (2.0)	2.95
^{144}La	0.8	2.61 (1.4)	3.69
^{145}La	24.8	1.85 (6.0)	3.54
^{146}La	6.7	1.15 (4.0)	0.81

TABLE VI

Relative Independent Fission Product Yields From $^{235}U(n_{th},f)$

NUCLIDE	HALFLIFE (s)	ENDF/B-VI	PRESENT
^{89}Rb	909	0.12 (11)	0.51
^{90m}Rb	258	0.29 (>64)	0.12
^{90g}Rb	158	0.13 (32)	0.69
^{91}Rb	58.4	1.06 (2.0)	1.56
^{94}Rb	2.702	0.78 (2.8)	0.71
^{95}Rb	0.377	0.37 (4.0)	0.35
^{140}Cs	63.7	0.99 (8.0)	1.78
^{141}Cs	24.9	1.39 (2.0)	3.36
^{142}Cs	1.7	1.09 (4.0)	0.97

Table VII

Isomeric/Ground State Production Ratios

NUCLIDE	ENDF/B-VI	PRESENT
^{97m}Y / ^{97g}Y	0.60	0.91
^{98m}Y / ^{98g}Y	1.19	0.17
^{90m}Rb / ^{90g}Rb	0.25	0.18

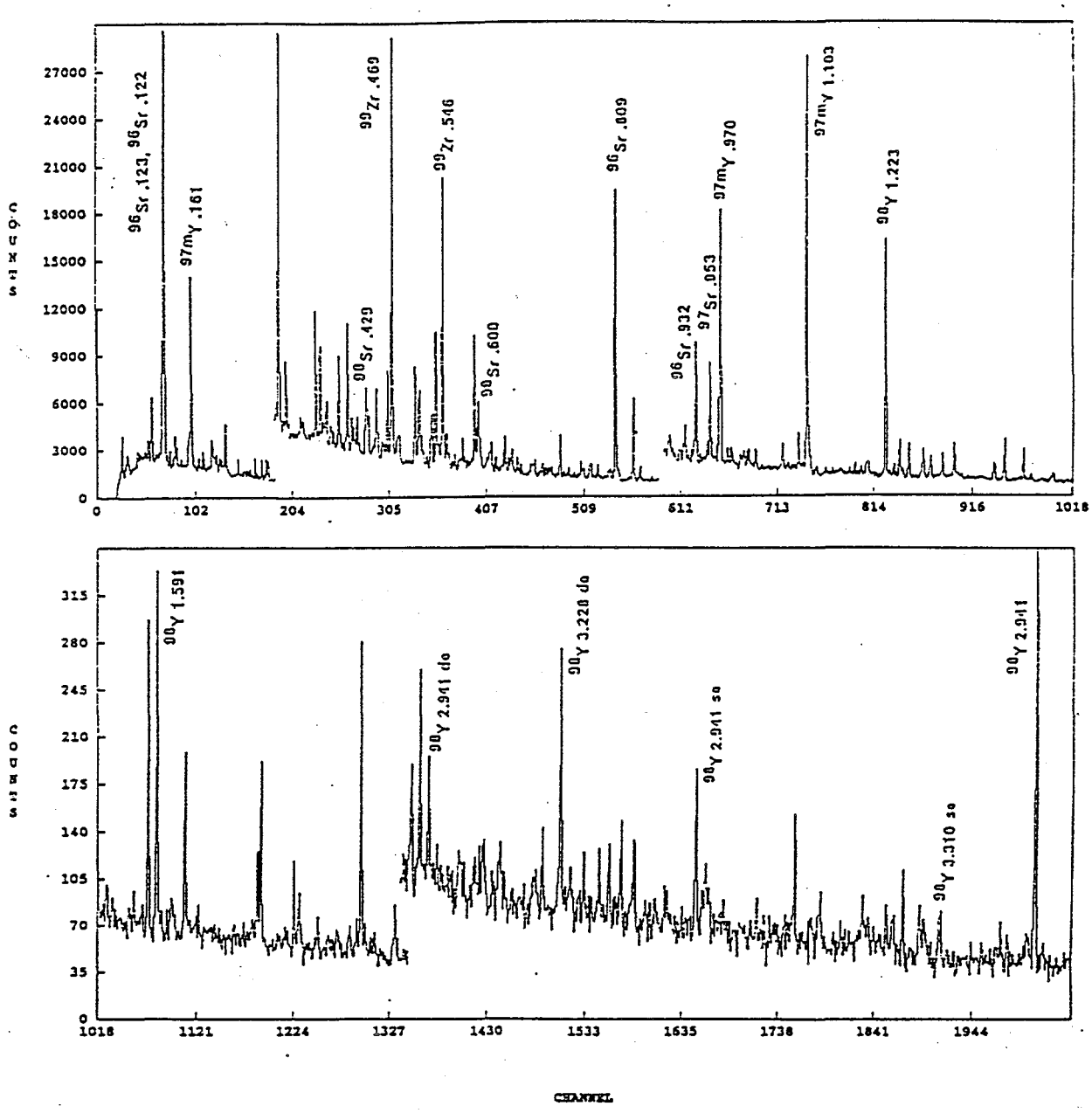

Figure G1(a). Gamma-ray spectrum from $^{235}U(n_{th},ff)$ measured using HPGe spectrometer for the delay time interval 0.58 - 0.90s. The spectrum covers the energy range 0 - 3 MeV and selected lines are identified by isotope.

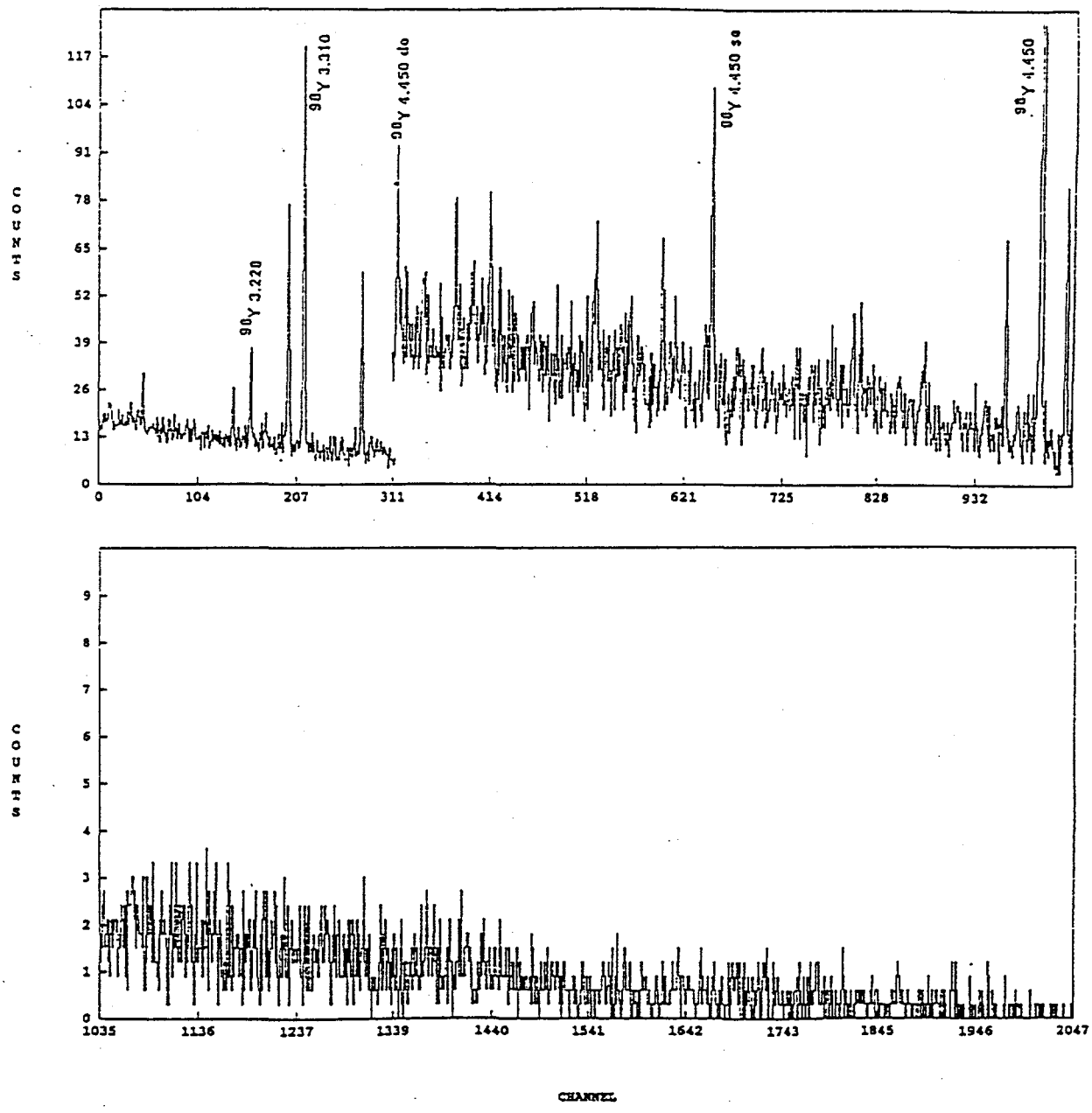

Figure G1(b). Continuation of the gamma-ray spectrum of Fig. G1(a) showing the energy region 3 - 6 MeV.

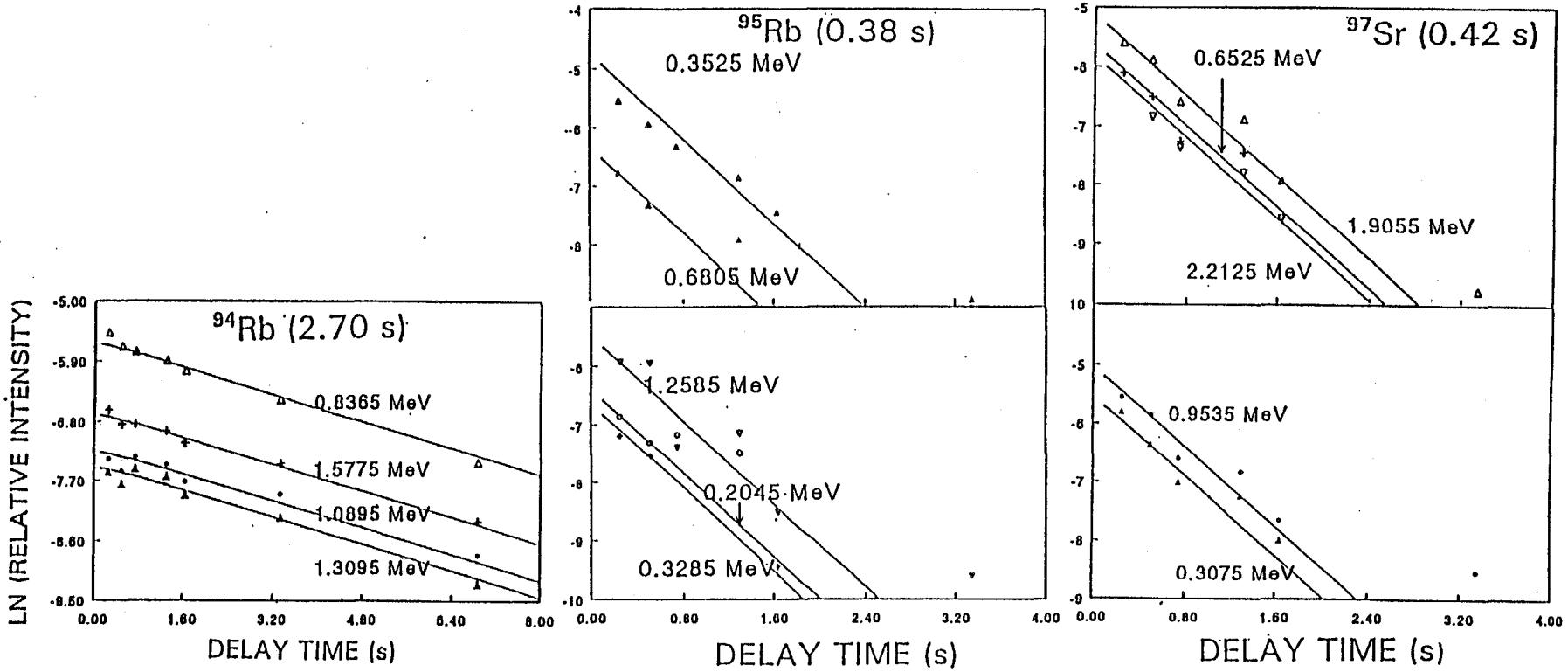

Figure G2. *Time evolution for selected gamma lines from $^{235}U(n_{th},ff)$ compared with ENDF/B-VI (normalized curves).*

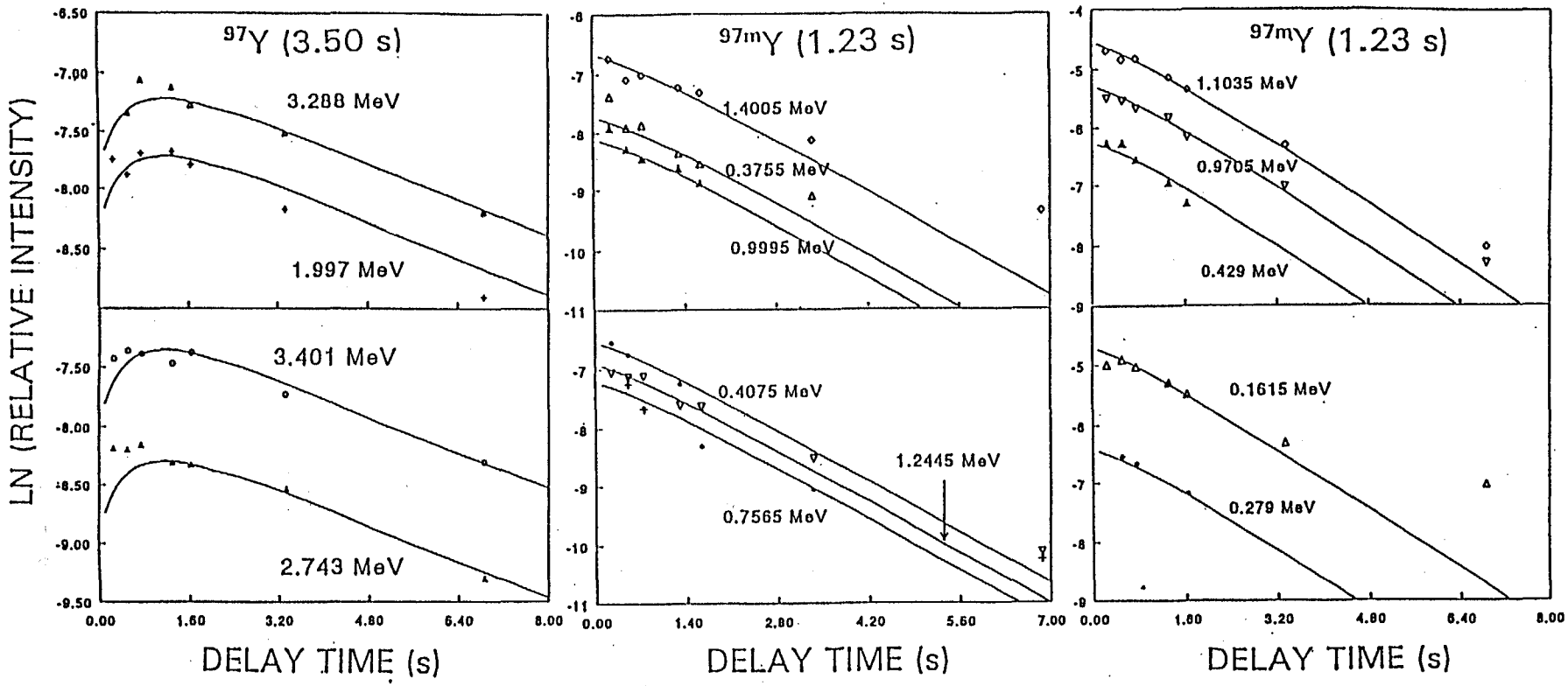

Figure G3. *Time evolution for selected gamma lines from $^{235}U(n_{th},ff)$ compared with ENDF/B-VI (normalized curves).*

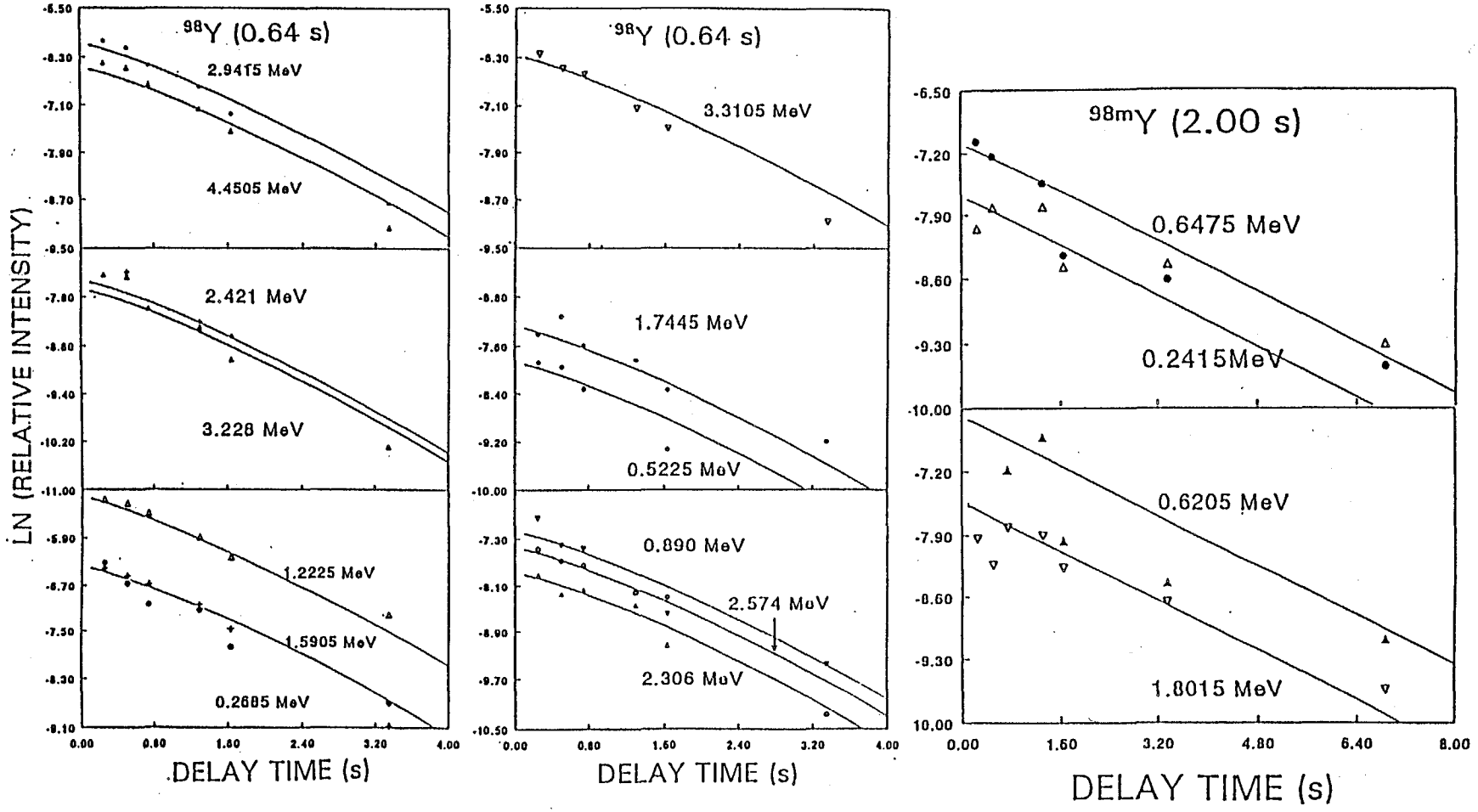

Figure G4. *Time evolution for selected gamma lines from $^{235}U(n_{th},ff)$ compared with ENDF/B-VI (normalized curves).*

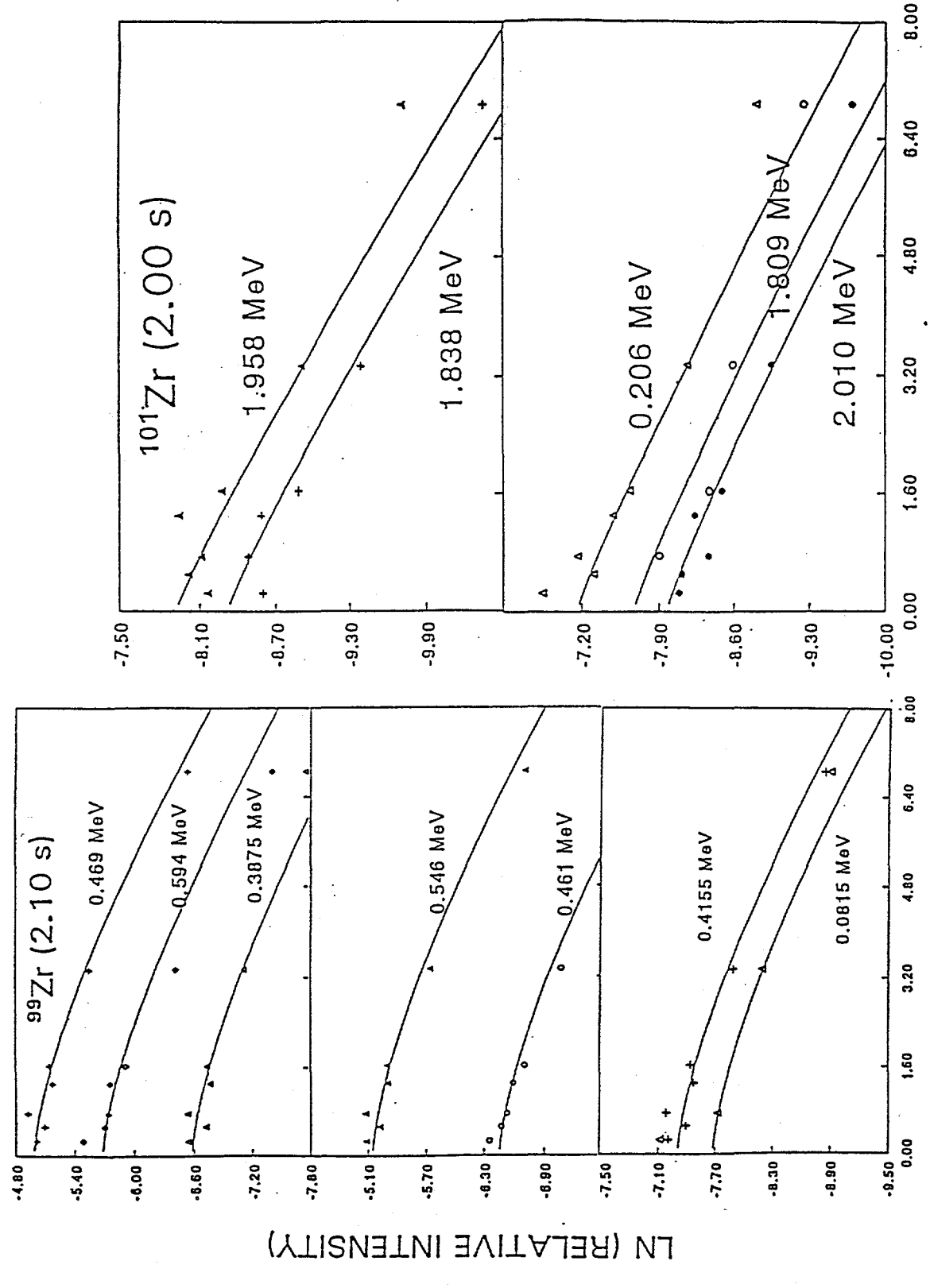

Figure G5. Time evolution for selected gamma lines from $^{235}U(n_{th},ff)$ compared with ENDF/B-VI (normalized curves).

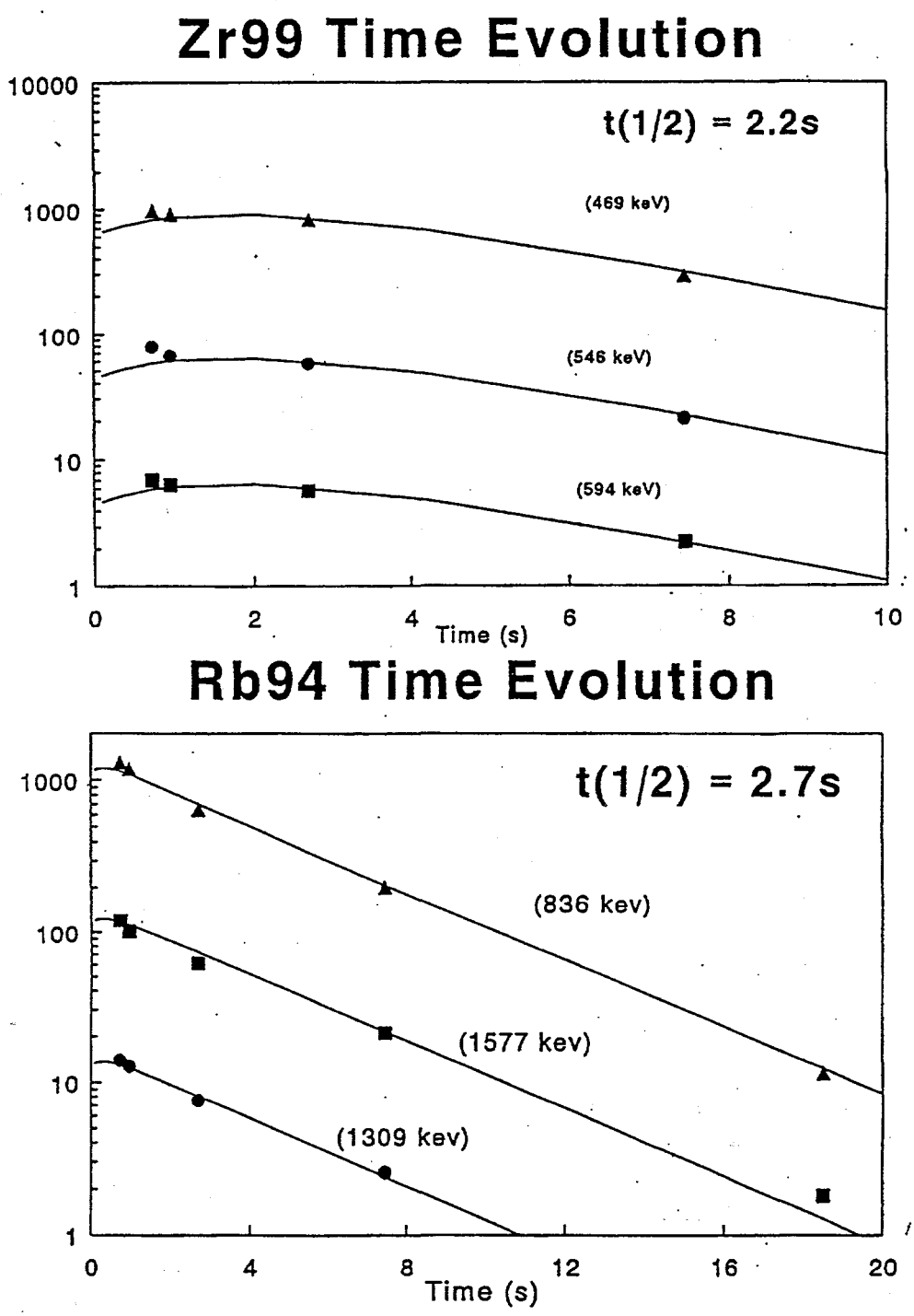

Figure G6. Time evolution of ^{99}Zr and ^{94}Rb from $^{238}U(n_{th},ff)$ compared with ENDF/B-VI (normalized curves).

H SPECTRAL DECOMPOSITION AND ANALYSIS CODES

i) HPGe Gamma Spectra

In the analysis of high-resolution gamma spectra measurements, an interactive graphics program (NDAA) was developed early in the project by M. Villani for background subtraction and fitting of resulting spectra with superposed Gaussian peaks. The widths of these Gaussians are fixed parameters, determined by an energy-dependent peak-width calibration function. The number of peaks is specified by the user, and an initial guess of their positions is made. The program then optimizes the peak areas and peak positions using a non-linear least-squares fitting procedure.

With the recent acquisition of our VAX/PC data analysis system, we are also now using the spectral analysis code GELIFIT2 to perform background subtraction and peak fitting. In this code the area of each peak is also determined using a gaussian-fitting routine but the code has some additional features to NDAA. It is used extensively in other laboratories.

Development of the spectral analysis code NDAA was part of the Ph.D. thesis of Marcel Villani.

ii) NaI(Tl) Gamma Spectra

The nearly continuous aggregate gamma spectra measured with NaI(Tl) result from the superposition of detector response functions for several thousand monoenergetic gamma lines from fission products which contribute to the spectrum. Knowing the response functions for monoenergetic gamma rays is, therefore, necessary in order to extract the true distribution of gamma energy from the measured spectra. With known response functions $R_i(E)$, each measured spectrum $S(E)$ can be expressed as a superposition of the response functions from the N contributors, i.e.,

$$S(E) = \sum_{i=1}^{N} a_i R_i(E)$$

The coefficients a_i represent a histogram of the true energy distribution if the response functions are normalized to unit area, viz:

$$\int_0^\infty R_i(E)\,dE = 1.$$

In practice, the response function cannot be known for every gamma line contributing to the spectrum but it is sufficient in determining $S(E)$ if the response function is known at energies

which are spaced apart by about one half the width of the full-energy peak. This means that typically about N = 100 response functions are sufficient to represent the gamma-ray energy distribution in our measured spectra.

Response functions were measured at 15 different energies in the range 0.081 - 6.13 MeV (Section B(i)) and an interpolation program (NGRC) was developed for constructing parameterized response functions at any desired intermediate gamma energy. In determining the response function parameters, each measured response function was divided into two parts, a tail region and peaks. The peaks consisted of the full energy peak, single and double escape peaks, and 0.511-MeV peak, which were all assumed to be Gaussian shaped. The tail was the remainder of the response function after subtracting the peak structure from it. Each response function was fitted in a least-squares fashion with a sum of Gaussians and a cubic polynomial to represent the background. From this fit the areas and widths of the full energy, 0.511-MeV, and escape peaks were obtained. The shape of the tail was obtained by subtracting the Gaussian functions used to fit the peak structure from the response function. A library consisting of fifteen such tails was generated and stored in separate computer files which were called when running program NGRC. With the energy dependence of the interpolation parameters established, these could be used to obtain any response function whose energy falls between two measured response functions.

In order to test this procedure, interpolated response functions were used to unfold a measured ^{152}Eu spectrum. Figure H1 shows the ^{152}Eu spectrum measured with both NaI(Tl) and HPGe detectors. The HPGe spectrum was used to assist in locating the centroid of the NaI(Tl) photopeaks and response functions were generated whose photopeaks coincided with the HPGe lines. Figure H2 shows the excellent agreement obtained between the measured and fitted ^{152}Eu spectrum.

The parameterized response functions, stored in a matrix file, were initially used as input files for the spectral decomposition code FERD-PC [Ref.15]. However, with this code the unfolding analysis was restricted to fewer than 80 response functions, each having only 100 energy bins. More recently, we developed our own decomposition program (CRSUP) which can handle up to 100 response functions, each having 1500 energy bins. Both NGRC and CRSUP were developed largely with the intent of maximizing the number of response functions that can be used to model a measured spectrum without having to reduce the number of energy bins contained in each of the response functions. In this way considerable structure could be retained in the parameterized response function. Figure H3 shows the excellent agreement obtained with CRSUP in fitting a gamma spectrum from ^{235}U(n_{th},ff) with 100 response functions.

Development of the response-function interpolation method and adaptation of the program FERD-PC for use with our computer system was part the MS thesis of Hung Nguyen. Development of the decomposition code CRSUP is part of his Ph.D. thesis.

iii) Beta Spectra

Monoenergetic electron sources with energies much above 1 MeV were not readily available to us. However, since electron response functions are relatively simple, essentially comprising a symmetric full-energy peak plus a low energy tail, measured low-energy response functions were used to generate trial response functions for the region 2.0 - 7.0 MeV. The validity of these trial functions was checked by using them to unfold measured beta spectra of known spectral shape. Such beta spectra, particularly if energetic, mimic the continuous spectra measured following fission, in which case a response function set which correctly describes these source spectra can also be applied to the decomposition of the aggregate beta spectra to yield their beta energy distribution.

The trial set of response functions shown in Fig. H4 was established based on the response function measurements below 1 MeV. This set has a gaussian peak with FWHM given by:

$$\Delta E = a + b\sqrt{E}$$

and a ramp function tail whose area relative to that of the peak remains constant. The set was tested by decomposing a measured beta spectrum from ^{24}Na using FERD-PC and comparing the result with the known shape of its beta energy distribution (Fig. H5). It was also tested by generating a beta spectrum from the known beta-energy distribution of ^{38}Cl and comparing the result with its measured spectrum. Good agreement was had in both cases once a small efficiency correction was applied at energies below 0.30 MeV, due mainly to the passage of low-energy betas through the disk scintillator.

The determination and testing of trial beta response functions is part of the Ph.D. thesis of Shengjie Li.

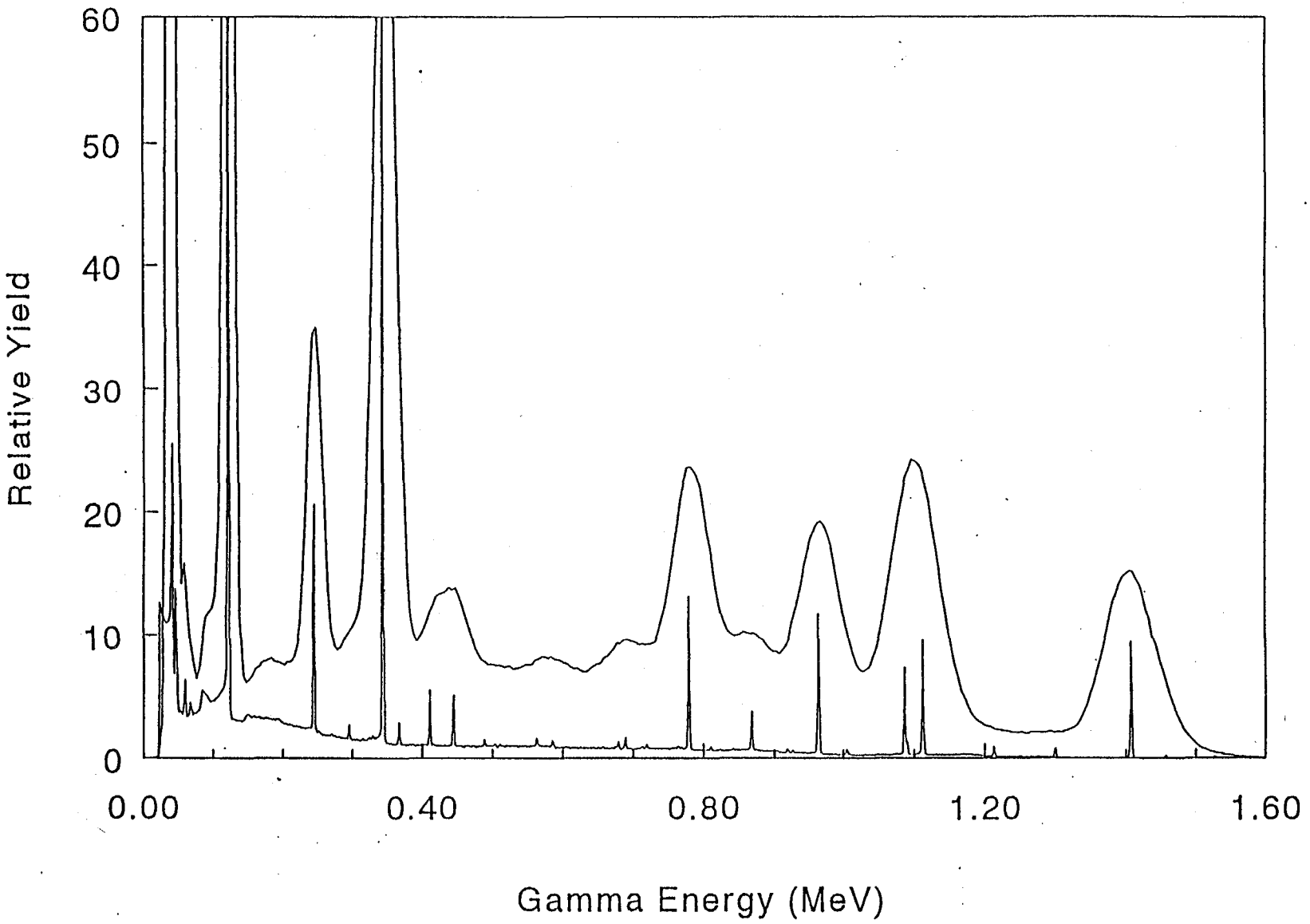

Figure H1. *Gamma-ray spectrum for ^{152}Eu measured using NaI(Tl) and HPGe spectrometers.*

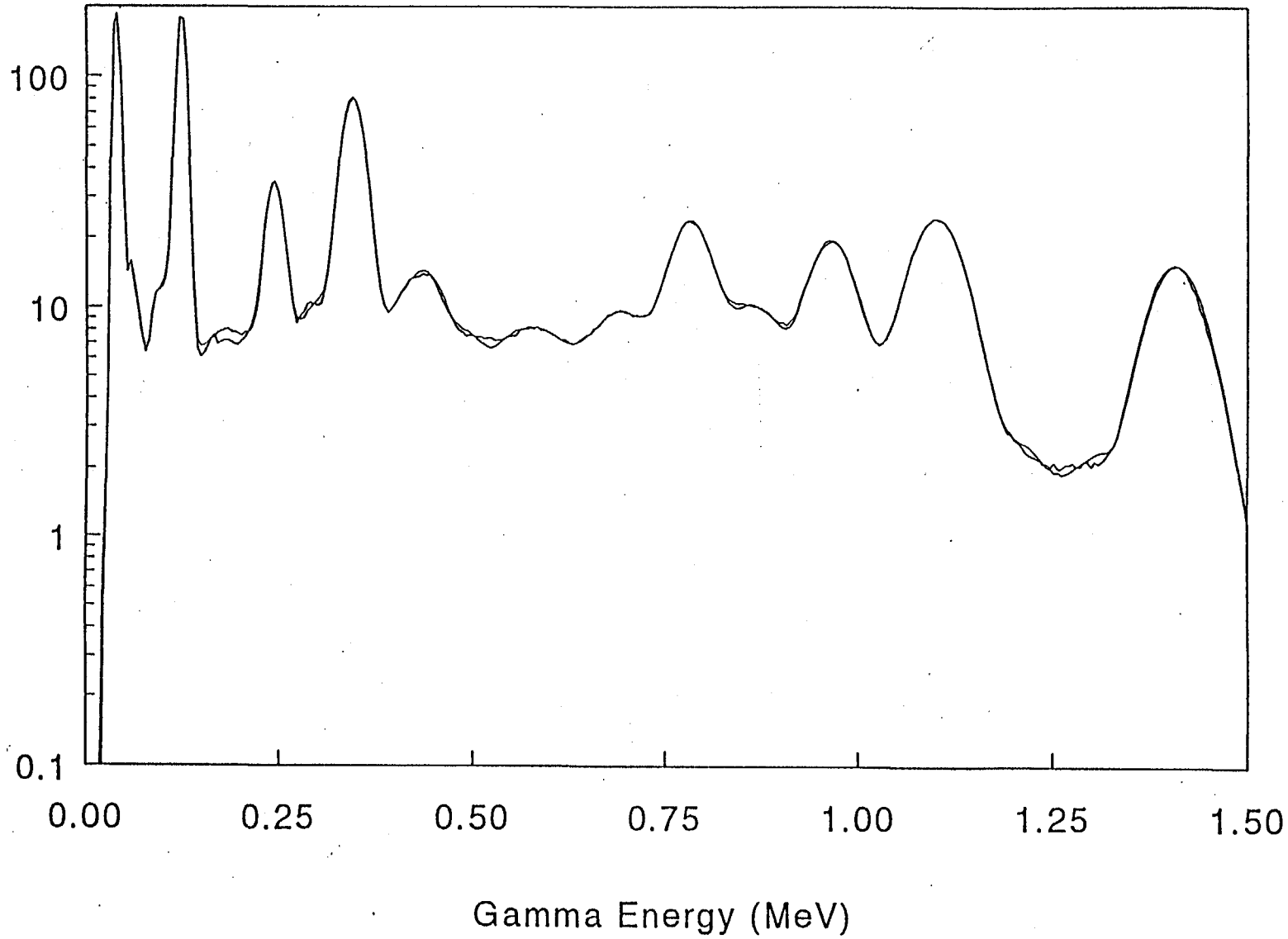

Figure H2. *Measured and fitted gamma-ray spectrum for ^{152}Eu.*

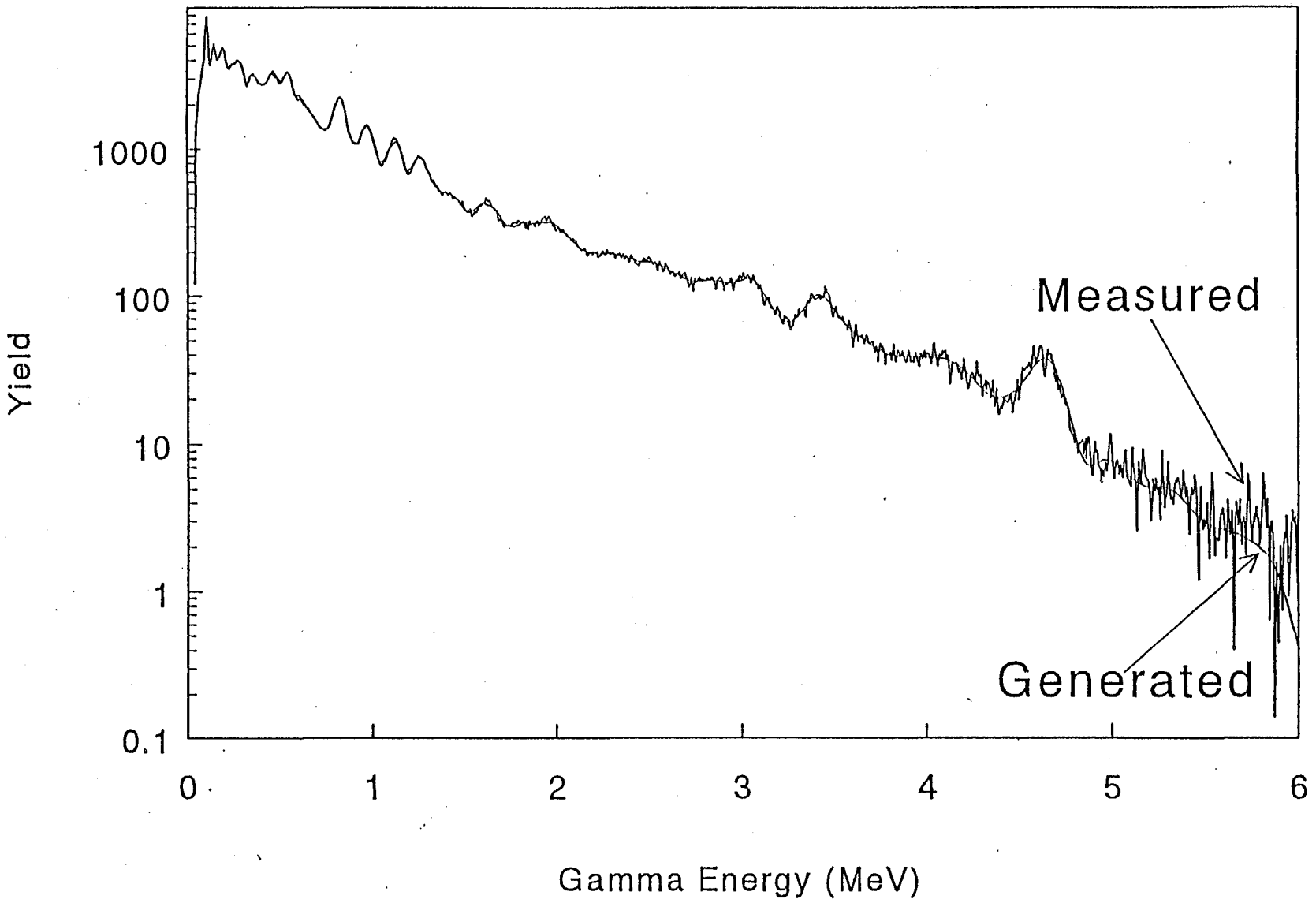

Figure H3. *A measured gamma-ray spectrum following $^{235}U(n_{th},ff)$ fitted with CRSUP.*

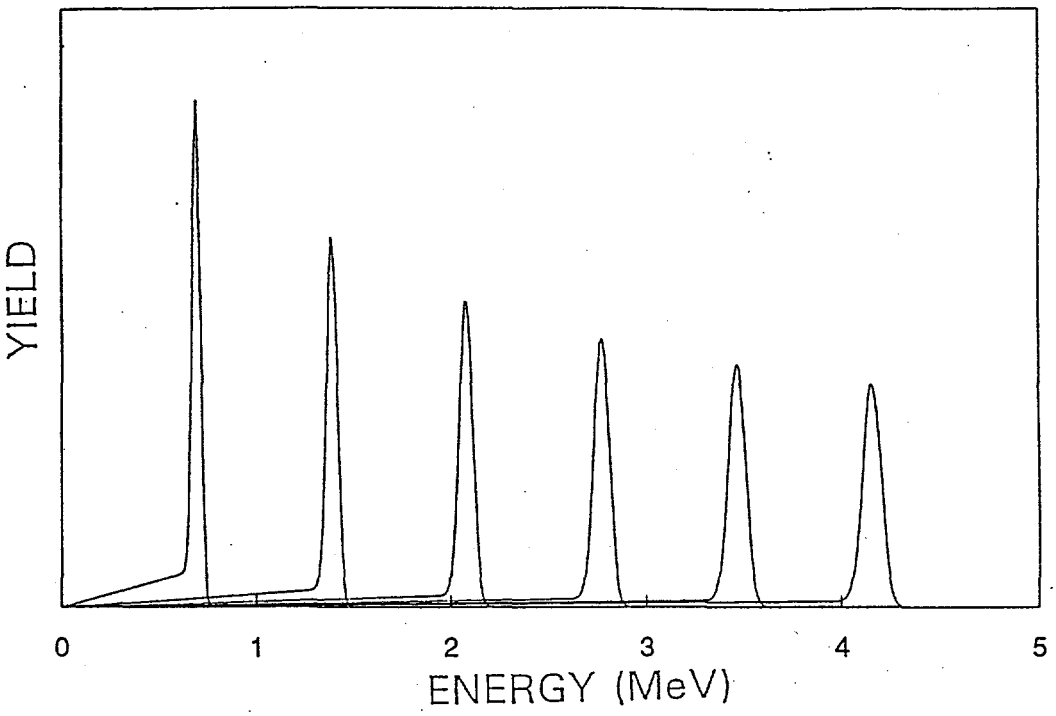

Figure H4. Trial set of beta response functions based on the response function measurements below 1 MeV.

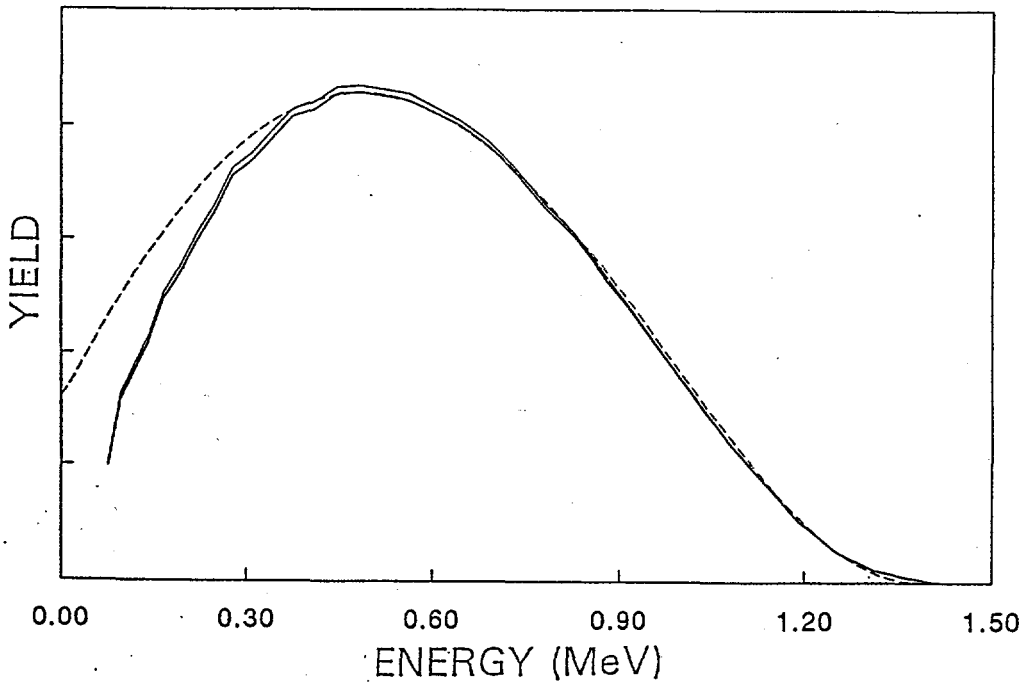

Figure H5. Decomposition of measured ^{24}Na beta spectrum compared with the beta energy distribution.

I. HELIUM JET FISSION FRAGMENT TRANSFER EFFICIENCY

The fission chamber and fission fragment transfer system were designed to transfer fission fragments to the beta or gamma spectrometer uniformly over the full mass distribution. Except for isotopes of the noble gases krypton and xenon, such uniformity of transfer was verified by measurements in which the activity transferred by helium jet was compared with that transferred by "rabbit" shuttle. In high-resolution gamma-ray measurements with the HPGe spectrometer, no lines associated with krypton and xenon isotopes were observed although, in an earlier study, the noble gases were shown to be efficiently transferred by the helium jet and could be retained for considerable time when deposited at the exit of the jet in activated charcoal chilled to liquid nitrogen temperatures. We attribute the loss of noble gases at the spectrometers to their non-adherence on the moving tape at the exit of the helium jet.

In measuring the transfer efficiency, fission fragments transported by the helium jet or rabbit shuttle were identified using x rays as signatures for the transferred elements. X-ray spectra measured with the He-jet and rabbit-shuttle systems are compared in Fig. I1 in which the peaks are labelled with the element at the position of its $K_{\alpha 1}$ line. The "2/5" and "3/5" mass regions are clearly evident and the overall agreement between the two transfer methods is seen to be very good. The relative transfer efficiency was deduced from the ratio of the normalized x-ray intensities for each identified element. In determining peak intensities careful account was taken of overlapping peaks, which distorted the background shape, and also of K_β lines from Z-1 neighbors which usually fell beneath the K_α lines of Z elements. Background subtraction was carried out by subjecting the spectra to a Gaussian-shaped filter, scaled to zero net area, which passed the peaks and rejected the background [Refs.23,24]. This yielded a background-free transform of the spectrum:

$$y_i' = \sum_{j=-r}^{+r} h_j y_{i+j}$$

In accounting for overlapping K_α and K_β lines, use was made of the known energies and relative intensities of the $K_{\alpha 1}$, $K_{\alpha 2}$, $K_{\beta 1}$ and $K_{\beta 2}$ lines for each element, together with the energy resolution of the detector, to form model spectra for each element identified in the measured spectrum. These model spectra, when appropriately normalized, were then sent through the same filter as above to produce their transforms, and the relative intensities then extracted for each spectrum by performing a weighted least-squares fit of the model spectra to the filtered measured spectra. A representative least squares fit of a filtered spectrum is shown in Fig. I2. and the helium-jet/rabbit-shuttle intensity ratios determined from this study are plotted in Fig. I3. With few exceptions, the masses are seen to be transferred with almost equal probability.

The presence of a characteristic x ray in the spectrum may arise from the transfer of two different elements, with one of them in an isomeric state. (Both xenon and krypton have known isomers, e.g., ^{143m}Xe, ^{135m}Xe, ^{91m}Kr and ^{85m}Kr.) For example, the precursor fission product, Z,

can be transferred and then beta decay to the daughter nucleus Z+1 in an excited state. This state will occasionally decay by internal conversion, thus ejecting an electron and giving rise to x rays associated with the Z+1 daughters. The presence of rubidium x-rays according to this interpretation would then suggest the transfer of krypton. However, if some of the Z+1 isotopes are transferred in long-lived isomeric states, which subsequently decay by internal conversion, these will also give rise to the same x ray. The presence of rubidium x rays in this case would suggest the transfer of rubidium isotopes in long-lived isomeric states. We interpret the diminished rubidium x rays and absence of krypton x rays in Fig. I3 as indicating that krypton atoms transferred in the helium jet do not adhere to the thin foam-plastic "catcher" at the exit of the helium jet. Likewise, the missing cesium x rays and diminished xenon x rays indicate that xenon atoms also do not adhere to the catcher.

Except for mainly the noble gases then, this study has established that our He-jet system transfers essentially the full mass distribution of fission fragments to the spectrometers with nearly equal probability. The noble gases account for only about 4% of the total activity at short delay times (< 1s) following fission, and the missing contributions can readily be excluded from the energy distributions calculated with ENDF/B-VI when comparing with our measured aggregate distributions without introducing large uncertainty.

This study of the helium jet fission-fragment transfer efficiency was part of the M.S. thesis of Paul Bennett.

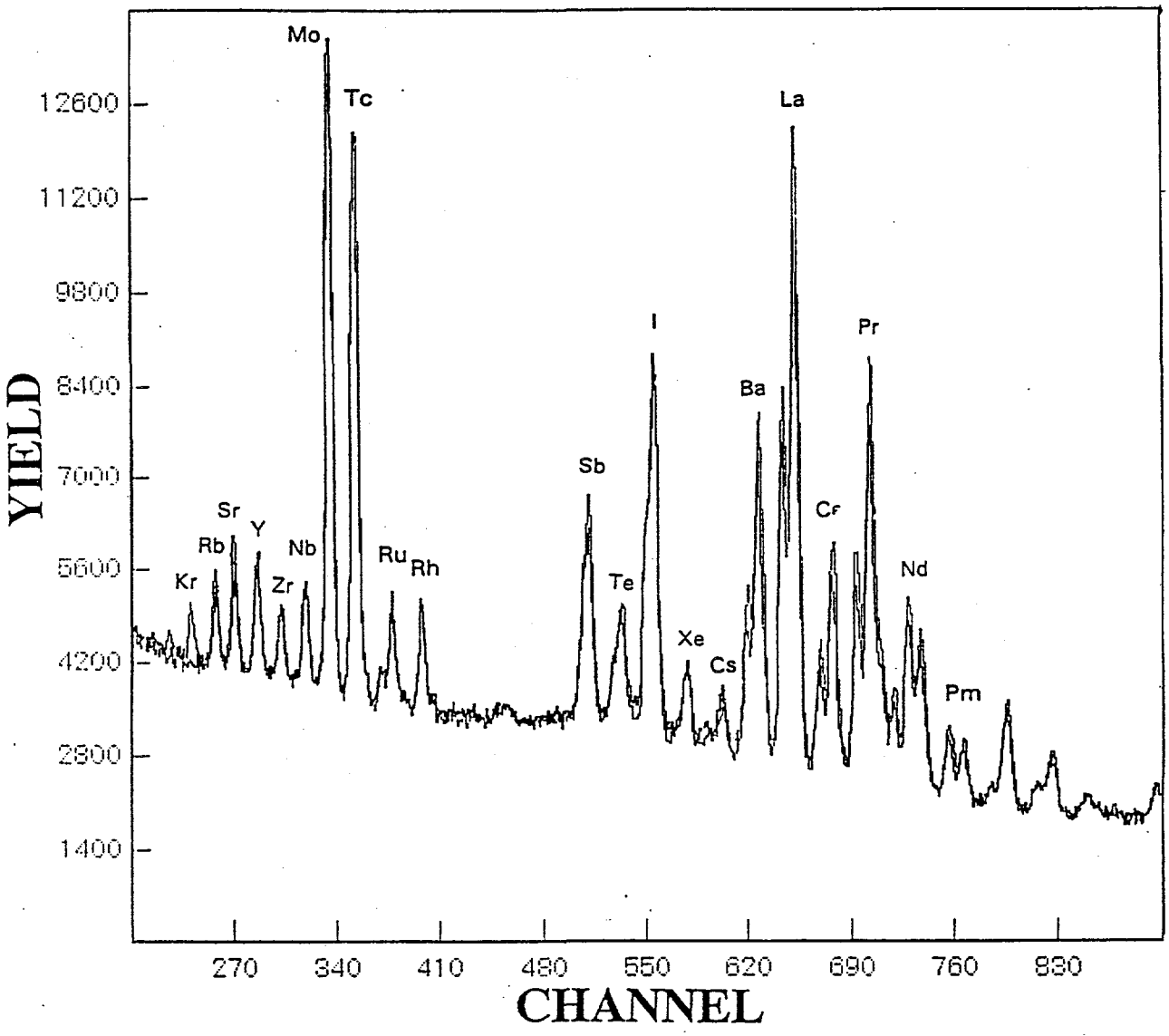

Figure 11. *Superimposed x-ray spectra from fission fragments transferred with the helium jet and with the rabbit shuttle system.*

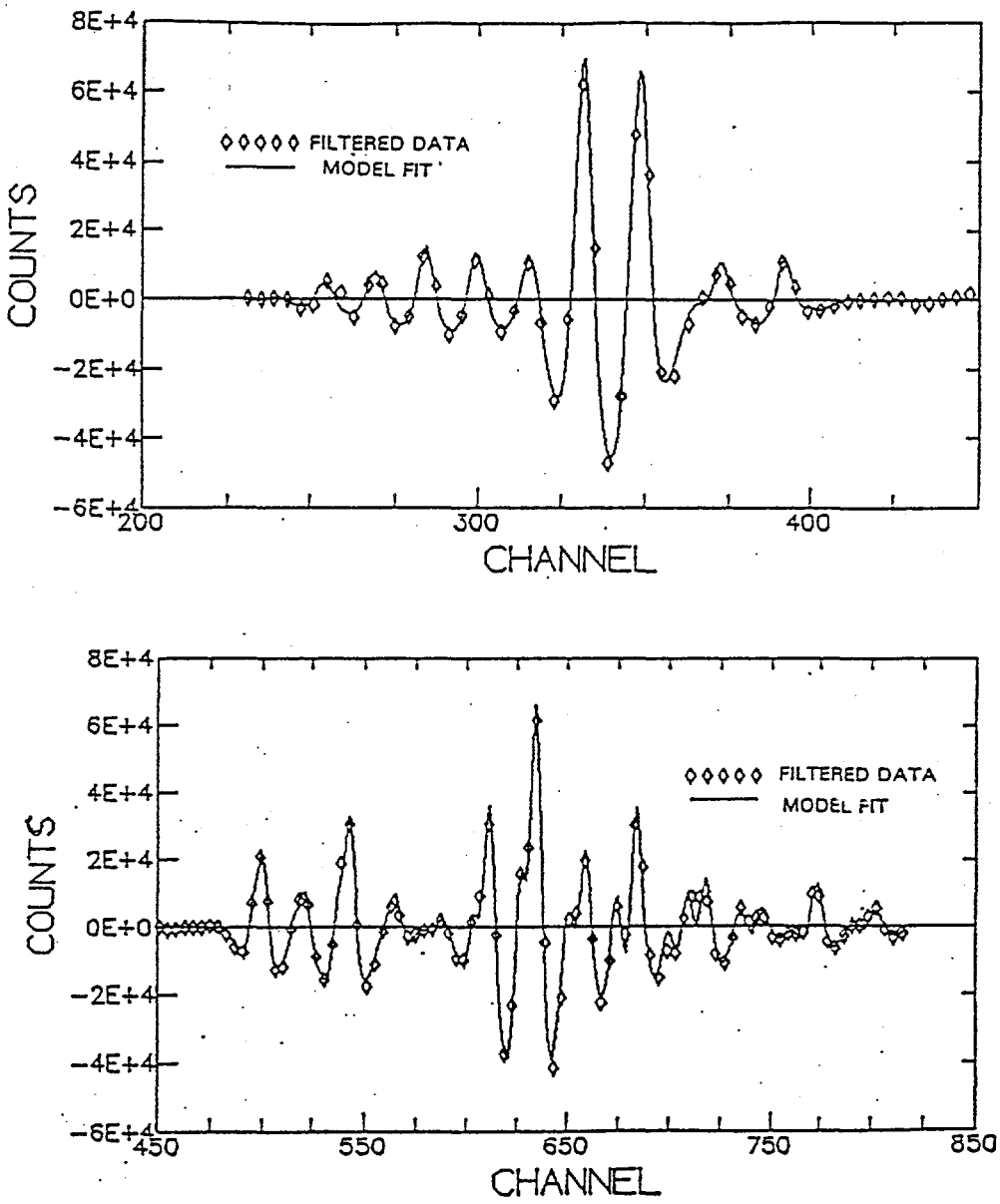

Figure I2. Comparison of filtered measured x-ray data and model fit in the "2/5 mass region" (above) and "3/5 mass region" (below).

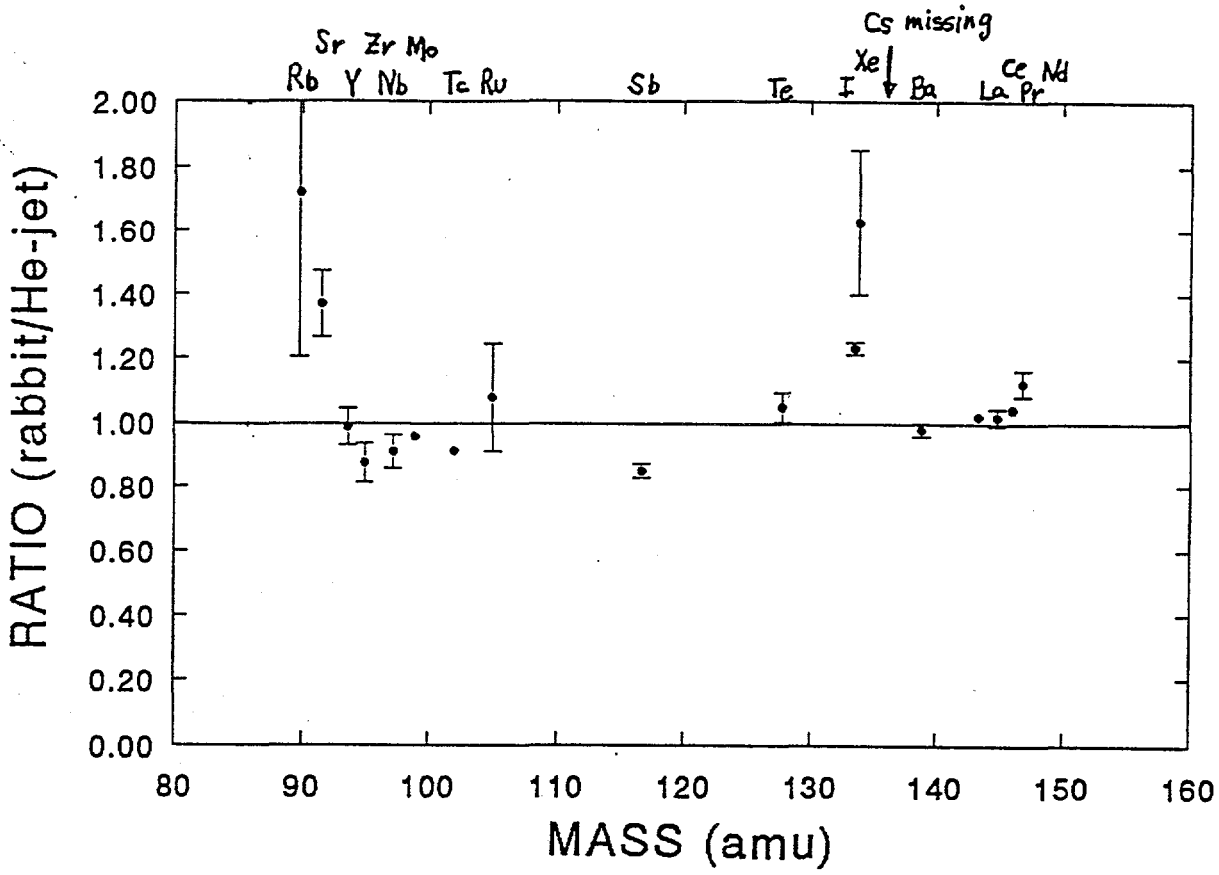

Figure I3. Ratio of x-ray intensities from fission fragments transferred with the rabbit shuttle system and with helium jet. The ratio includes a correction for the escape probability from a thick ^{235}U foil.

2. PROJECT RELATED PUBLICATIONS

A. REFEREED PAPERS

1. "Six-Group Decomposition of Composite Delayed Neutron Spectra from 235U Fission"; M.F. Villani, G.P. Couchell, M.H. Haghighi, D.J. Pullen, W.A. Schier and Q. Sharfuddin, Nucl. Sci. Eng. **111** (1992) 422.

2. "Energy Distributions of Gamma and of Beta Decay Heat as Function of Decay Time for ^{238}U(n,f)"; W.A. Schier, J.M. Campbell, G.P. Couchell, S. Li, H.V. Nguyen, D.J. Pullen, E.H. Seabury and S.V. Tipnis and T.R. England, Accepted for publication in Proceedings International Conference on Nuclear Data for Science and Technology, Gatlinburg, TN (1994).

3. "A Study of Gamma-Ray and Beta-Particle Decay Heat following Thermal Neutron Induced Fission of ^{235}U"; G.P. Couchell, J.M. Campbell, S. Li, H.V. Nguyen, D.J. Pullen, E.H. Seabury and S.V. Tipnis and T.R. England, Accepted for publication in Proceedings International Conference on Nuclear Data for Science and Technology, Gatlinburg, TN (1994).

4. " High Resolution Gamma-Ray Spectra for ^{235}U(n,ff)"; D.J. Pullen, J.M. Campbell, G.P. Couchell, S. Li, H.V. Nguyen, W.A. Schier, E.H. Seabury, S.V. Tipnis and T.R. England, Accepted for publication in Proceedings International Conference on Nuclear Data for Science and Technology, Gatlinburg, TN (1994).

5. "Programs in C for Parameterizing Measured 5"x5" NaI Gamma Response Functions and Unfolding of Continuous Gamma Spectra", H.V. Nguyen, J.M. Campbell, G.P.Couchell, S. Li, D.J. Pullen, W.A. Schier, E.H. Seabury and T.V. Tipnis, Computer Phys. Comm. (submitted).

6. "Beta Particle Spectrometer for Measuring Aggregate Beta Spectra Following Fission", W.A. Schier, J.M. Campbell, G.P. Couchell, S. Li, H.V. Nguyen, D.J. Pullen, E.H. Seabury and S.V. Tipnis, Nucl. Instr. Meth. (submitted).

7. "Relative Efficiency Measurements of Fission Fragment Transfer with a Helium Jet", P.R. Bennett, W.A. Schier, G.P. Couchell, E.S. Jacobs, D.J. Pullen and M.F. Villani, Nucl. Instr. Meth. (submitted).

B. PUBLISHED ABSTRACTS

1. "Use of a High Purity Germanium Detector in Aggregate Fission-Product Decay Heat Studies"; D.J. Pullen, J.M. Campbell, G.P. Couchell, H.V. Nguyen, W.A. Schier, S.V. Tipnis and M.F. Villani, Bull. Amer. Phys. Soc. II, **37** (1992) 1279.

2. "A NaI(Tl) Gamma-Ray Spectrometer for Aggregate Fission-Product Gamma Decay Heat Study"; G.P. Couchell, J.M. Campbell, H.V. Nguyen, D.J. Pullen, W.A. Schier, S.V. Tipnis and M.F. Villani, Bull. Amer. Phys. Soc. II, **37** (1992) 1292.

3. "Beta Spectrometer Design and Testing"; W.A. Schier, J.M. Campbell, G.P. Couchell, H.V. Nguyen, D.J. Pullen, S.V. Tipnis and M.F. Villani, Bull. Amer. Phys. Soc. II, **37** (1992) 1293.

4. "Helium Jet Fission Fragment Transfer Efficiency"; P.R. Bennett, J.M. Campbell, G.P. Couchell, H.V. Nguyen, D.J. Pullen, W.A. Schier, S.V. Tipnis and M.F. Villani, Bull. Amer. Phys. Soc. II, **37** (1992) 1293.

5. "Six-Group Decomposition of ^{238}U Delayed Neutron Composite Energy Spectra"; M.F. Villani, J.M. Campbell, G.P. Couchell, H.V. Nguyen, D.J. Pullen, W.A. Schier and S.V. Tipnis, Bull. Amer. Phys. Soc. II, **37** (1992) 1300.

6. "Comparison of Selected Gamma-Ray Yields Following Thermal Fission of ^{235}U with ENDF/B-VI Decay Data"; J.M. Campbell, G.P. Couchell, H.V. Nguyen, D.J. Pullen, W.A. Schier, S.V. Tipnis and M.F. Villani, Bull. Amer. Phys. Soc. II, **37** (1992) 1300.

7. "Response Functions for Beta-particle and Gamma-ray Spectrometers"; S. Li, J.M. Campbell, G.P. Couchell, H.V. Nguyen, D.J. Pullen, W.A. Schier and S.V. Tipnis, Bull. Amer. Phys. Soc. II, **38** (1993) 1652.

8. "Spectrum Analysis Programs for Decay Heat Studies of Aggregate Fission Products"; H.V. Nguyen, J.M. Campbell, G.P. Couchell, S. Li, D.J. Pullen, W.A. Schier, S.V. Tipnis and M.F. Villani, Bull. Amer. Phys. Soc. II, **38** (1993) 1652.

9. "Comparison of the Measured Time Evolution of Individual Fission-product Gamma-ray Lines with ENDF/B-VI Data"; S.V. Tipnis, J.M. Campbell, G.P. Couchell, S. Li, H.V. Nguyen, D.J. Pullen, W.A. Schier and T.R. England, Bull. Amer. Phys. Soc. II, **38** (1993) 1652.

10. "High Resolution Gamma-ray Spectra from Aggregate Fission-Products Measured in $^{235}U(n_{th},f)$"; J.M. Campbell, G.P. Couchell, S. Li, H.V. Nguyen, D.J. Pullen, W.A. Schier and S.V. Tipnis, Bull. Amer. Phys. Soc. II, **38** (1993) 1650.

11. "Study of Gamma-Ray Heat from Aggregate ^{235}U Fission Products"; G.P. Couchell, J.M. Campbell, S. Li, H.V. Nguyen, D.J. Pullen, W.A. Schier and S.V. Tipnis, Bull. Amer. Phys. Soc. II, **38** (1993) 1650.

12. "Study of Beta-Particle Decay Heat from Aggregate ^{235}U Fission Products"; W.A.Schier, J.M. Campbell, G.P. Couchell, S. Li, H.V. Nguyen, D.J. Pullen and S.V. Tipnis, Bull. Amer. Phys. Soc. II, **38** (1993) 1650.

13. "Gamma-Ray Study of Short-Lived Aggregate Fission Products $^{235}U(n,f)$"; W.A. Schier, J.M. Campbell, G.P. Couchell, S. Li, H.V. Nguyen, D.J. Pullen, E.H. Seabury and S.V. Tipnis and T.R. England, Bull. Am. Phys. Soc. **38** (1993) 1806.

14. "Experimental Arrangement for $^{238}U(n,f)$ Aggregate Beta-Particle and Gamma-Ray Decay Heat Study"; E.H. Seabury, J.M. Campbell, G.P. Couchell, S. Li, H.V. Nguyen, D.J. Pullen, W.A. Schier and S.V. Tipnis, Bull. Am. Phys. Soc. **39** (1994) 1271.

15. "Comparison of Halflives of Short-lived ^{235}U Fission Products with ENDF/B-VI"; S.V. Tipnis, J.M. Campbell, G.P. Couchell, S. Li, H.V. Nguyen, D.J. Pullen, W.A. Schier, E.H. Seabury and T.R. England, Bull. Am. Phys. Soc. **39** (1994) 1271.

16. "Time Evolution of Fission-Product Gamma Spectra using ENDF/B-VI"; J.M. Campbell, G.P. Couchell, S. Li, H.V. Nguyen, D.J. Pullen, W.A. Schier, E.H. Seabury, S.V. Tipnis and T.R. England, Bull. Am. Phys. Soc. **39** (1994) 1271.

17. "Interpolation of Response Functions Used in the Analysis of 5" x 5" NaI Gamma Ray Measurements"; H.V. Nguyen, J.M. Campbell, G.P. Couchell, S. Li, D.J. Pullen, W.A. Schier, E.H. Seabury and S.V. Tipnis, Bull. Am. Phys. Soc. **39** (1994) 1271.

18. "Aggregate Gamma Decay Heat Spectra Comparison following ^{238}U and ^{235}U Neutron Fission"; S. Li, J.M. Campbell, G.P. Couchell, H.V. Nguyen, D.J. Pullen, W.A. Schier, E.H. Seabury and S.V. Tipnis, Bull. Am. Phys. Soc. **39** (1994) 1271.

19. "Feasibility Study of Gamma-Ray Coincidence Measurements of $^{238}U(n,f)$ Fission Products"; E.H. Seabury, J.M. Campbell, P. Chowdhury, G.P. Couchell, S. Li, H.V. Nguyen, D.J. Pullen, W.A. Schier and S.V. Tipnis, (presented Oct. 7-8, 1994; to be published in Bull. Am. Phys. Soc. **40** (1995)).

20. "Beta Energy Spectra from U-238 Fission Products"; S. Li, J.M. Campbell, G.P. Couchell, H.V. Nguyen, D.J. Pullen, W.A. Schier, E.H. Seabury and S.V. Tipnis, (presented Oct. 7-8, 1994; to be published in Bull. Am. Phys. Soc. **40** (1995)).

21. "Comparison of the Measured Time Evolution of Individual Fission-Product Gamma-ray Lines with ENDF/B-VI Data"; J.M. Campbell, P. Chowdhury, G.P. Couchell, S. Li, H.V. Nguyen, D.J. Pullen, W.A. Schier, E.H. Seabury and S.V. Tipnis, (presented Oct. 7-8, 1994; to be published in Bull. Am. Phys. Soc. **40** (1995)).

22. "Unfolding of Gamma Spectra from Aggregate Fission Products using Response Functions"; H.V. Nguyen, J.M. Campbell, G.P. Couchell, D.J. Pullen, W.A. Schier, E.H. Seabury and S.V. Tipnis, (presented Oct. 7-8, 1994; to be published in Bull. Am. Phys. Soc. **40** (1995)).

23. "Comparision of Production Probabilities of ^{235}U Fission Products with ENDF/B-VI"; S.V. Tipnis, J.M. Campbell, G.P. Couchell, D.J. Pullen, W.A. Schier, E.H. Seabury and S.V. Tipnis, (presented Oct. 7-8, 1994; to be published in Bull. Am. Phys. Soc. **40** (1995)).

3. FACULTY ASSOCIATES AND STUDENTS

FACULTY RESEARCH ASSOCIATE

Dr. David Pullen

ADJUNCT FACULTY ASSOCIATE

Dr. Partha Chowdhury (1993-)

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Joann Campbell (Ph.D.)

Shengjie Li (Ph.D.)

Hung Nguyen (Ph.D.)

Edward Seabury (Ph.D.)

Sameer Tipnis (Ph.D.)

Degrees Conferred on Students Associated with Project

Joann Campbell, M.S.; Hung Nguyen, M.S.

4. ABSTRACTS OF PRESENTED PAPERS

The following are abstracts of three papers presented on this work at the International Conference on Nuclear Data for Science and Technology, Gatlinburg, Tennessee, 1994.

Energy Distributions of Gamma and of Beta Decay Heat as Function of Decay Time for $^{238}U(n,f)$*

W.A. SCHIER, J.M. CAMPBELL, G.P. COUCHELL, S. LI, H.V. NGUYEN, D.J. PULLEN, E.H. SEABURY AND S.V. TIPNIS,

University of Massachusetts Lowell, Dept. of Physics, Lowell, MA 01854, USA

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Separate aggregate gamma-ray and aggregate beta-particle decay heat measurements are underway on the UMASS Lowell 1-MW research reactor and we report here on these measurements and preliminary energy distributions derived from them. The helium jet/tape transport system transfers the fission fragments from the fission chamber in a fast neutron port to the gamma or the beta spectrometer outside. It allows measurements to be made as short as 0.2 s after fission. But the same system also allows one to extend measurements out to very long delay times, e.g. 50,000 s. Below 20 s, no previous aggregate decay heat measurements exist. This region is of particular interest in testing the ENDF/B-VI data base since in this time region 20-30% of the decay heat must be calculated from a statistical model. The gamma-ray spectrometer is a collimated, well-shielded 5"x5" NaI(Tl) detector run in beta-gamma coincidence. The beta spectrometer is a 3"x3" plastic scintillator that is gated by an optically isolated thin scintillator disk on its surface. The beta spectrometer has excellent gamma discrimination, sufficiently good energy resolution for decay heat measurements and a rather simple response function shape. Response functions to monoenergetic gamma rays have been measured for the gamma-ray spectrometer using both neutron capture sources and nuclear reactions. These were then decomposed into the full energy peak, single and double escape peaks, Compton band, etc. and a method for interpolating to other energies was established. The resulting set of response functions was tested by reproducing complex gamma spectra such as ^{152}Eu. Response functions for the beta spectrometer were measured for $E_\beta < 1$ MeV with internal transition (IT) sources such as ^{113m}Sn, ^{137}Cs and ^{207}Bi. Appropriate coincidence conditions must be applied in the ^{137}Cs and ^{207}Bi cases to suppress the continuous beta background and select the monoenergetic electron lines. For $E_\beta > 1$ MeV a different approach was applied since there is a paucity of suitable high energy IT sources. In this region a set of continuous beta spectra having known energy distributions were measured. Sets of trial response functions based on those measured below 1 MeV were applied to the full set of measured beta spectra to extract their energy distributions and compare to their theoretical shapes. Using such response functions, the program FERDO was employed to convert measured decay heat spectra into energy distributions. In the case of gamma rays, the resulting distributions must still be corrected for detection efficiency. Energy distributions and their average energies as a function of delay time can then be calculated and compared to ENDF/B-VI and other aggregate decay heat measurements.

* Supported by the U.S. Department of Energy

High-Resolution Gamma-Ray Spectra for $^{235}U(n_{th},ff)$*

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As part of a program involving separate measurements of the aggregate decay heats for beta and gamma-rays as a function of delay time following fission, high-resolution measurements of gamma-ray spectra following thermal neutron-induced fission of ^{235}U have been made with the UMASS Lowell 5.5-MV Van de Graaff accelerator. A helium-jet system provided rapid transfer of the fission fragments from the fission chamber to a low-background counting room, where spectra were measured in the energy range 0 - 6 MeV using a high purity germanium detector. The use of beta/gamma coincidence gating in the gamma-ray spectrometer reduced the overall background by about two orders of magnitude. Further improvement by a factor of 3 - 5 to the peak-to-background ratio was obtained using a NaI annulus for Compton suppression. By placing the HPGe detector nearer the front face of the annulus than in conventional geometries, good detection efficiency could still be had using the annulus. Gamma spectra were measured at 21 different delay time intervals following fission, within the time range 0.2s to 50,000s. These measurements provide information on the relative production probabilities of the fission products as well as their ratio of direct production in fission to production through beta-decay chains. Where the decay scheme is known, the ratio of metastable-to-ground state production can also be determined. The time evolution of the gamma line intensities enables the halflives of the precursor nuclides to additionally be determined. Nearly 300 gamma peaks have so far been analyzed for delay times <10s, corresponding to 56 different gamma lines (including several isomeric transitions) in 10 different fission products. No gamma lines are observed above 5 MeV energy, and the analysis is currently being extended to include the measurements at longer delay times. Relative production probabilities and ratios as well as halflives obtained from this study will be compared with values calculated with the CINDER10 code based on ENDF/B-VI fission-product data for ^{235}U fission.

* Supported by the U.S. Department of Energy

A Study of Gamma-Ray and Beta-Particle Decay Heat following Thermal Neutron Induced Fission of ^{235}U*

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Aggregate gamma-ray and aggregate beta-particle energy spectra have been measured for fission products resulting from thermal-induced fission of ^{235}U. Each of the two sets of spectra span an energy range of approximately 0.1 - 8.0 MeV and decay time ranges of 0.2s - 10,000s after fission. Preliminary energy distributions have been deduced from the measurements. The use of a helium-jet/tape transport system has facilitated studies of decay times well below 1 s, where no previous aggregate decay heat measurements for both beta and gamma radiation exist. Delay times below approximately 50s are of particular interest since summation calculations based on ENDF/B-VI fission product data fall well below results obtained from previous aggregate gamma-ray decay heat studies. The gamma-ray component of decay heat has been measured using a 5"x5" NaI(Tl) detector housed in a massive shield consisting of a combination of lead, brass and tungsten. A magnet in the nose of the shield deflected beta particles away from the detector. The NaI spectra were gated by a thin beta detector also viewing the particular section of transport tape of interest. This gating reduced background, including that due to betas that passed through the magnetic deflection system, by approximately two orders of magnitude. The beta spectrometer consists of a 3"x3" plastic scintillator gated by an optically isolated thin scintillator disk of somewhat smaller diameter. Gating by the disk discriminates against gamma rays entering the large scintillator and also against beta particles falling close to the outer wall. Response functions for both the gamma-ray and beta spectrometers were determined using a combination of radiaoctive sources and nuclear reactions. Because decay heat beta and gamma-ray spectra are continuous in nature, an interpolation code was written for generating response functions at any intermediate energy. The validity of the generated response functions was tested by using them to unfold complex gamma-ray and beta spectra of known energy distributions. Once the validity of both the beta and gamma sets of response functions was established, these were used as input files for the unfolding program FERDO employed to convert our measured decay heat spectra into energy distributions. The deduced energy distributions and their resulting average energies will be compared with those of previous studies as well as with summation calculations performed with CINDER10 using the ENDF/B-VI fission product data base. Sensitivity of our measurements to the absence of noble gas contributions will also be examined.

* Supported by the U.S. Department of Energy

The following are abstracts of papers presented on this work at various meetings of the American Physical Society.

1. Use of a High Purity Germanium Detector in Aggregate Fission-Product Decay Heat Studies.* D.J. PULLEN, J.M. CAMPBELL, G.P. COUCHELL, H.V. NGUYEN, W.A. SCHIER, S.V. TIPNIS, M. VILLANI, University of Massachusetts Lowell. --- Our study of gamma-ray decay heat from aggregate fission products will be complemented by high resolution spectral measurements for delay times 0.1 - 50,000 s using a high purity germanium detector of approximately 1.8-keV (FWHM) energy resolution. These measurements will facilitate selective comparisons of individual transitions with ENDF/B-VI as described in the previous abstract. The use of a helium jet transport system for rapid transfer of fission products to a low-background environment also allows beta-gamma coincidence to further suppress background counts, facilitating premium spectrum measurements. Shielding of the detector and means of reducing its sensitivity to chance-coincidence fission-product beta particles will be presented. Housing the germanium detector inside a NaI(Tl) annulus for Compton suppression is also considered, and use of this annulus in decay heat studies will by evaluated.

*Supported in part by the U.S. Department of Energy.

2. A NaI(Tl) Gamma-Ray Spectrometer for Aggregate Fission-Product Gamma Decay Heat Study.* G.P. COUCHELL, J.M. CAMPBELL, H.V. NGUYEN, D.J. PULLEN, W.A. SCHIER, S.V. TIPNIS, M.F. VILLANI, University of Massachusetts Lowell. — A 12.7-cm x 12.7-cm NaI(Tl) scintillation detector was recently acquired as one component of a project which will study both gamma-ray and beta decay heat from aggregate fission products as a function of delay time, 0.1 - 50,000 s. The use of a helium jet system for rapid transport of fission products to a low-background environment for study of their spectra also allows the use of beta-gamma coincidence to select a delay time interval of interest and to greatly suppress background counts. This coincidence also provides excellent sensitivity to high energy gammas by eliminating cosmic-ray background. A lead-brass shield with a tungsten two segment collimator also greatly reduces the background count rate and minimizes edge effects by selecting the central 7.6-cm diameter region of the crystal. Chance-coincidence beta particles are eliminated from gamma spectra by deflecting betas with a magnetic field between the collimator segments. Sample spectra illustrating the detector response, background and beta particle suppression, and sensitivity to high energy gamma rays will be presented. Also aggregate gamma-ray spectra following thermal neutron fission of ^{235}U will be shown.

*Supported in part by the U.S. Department of Energy.

3. Beta Spectrometer Design and Testing.* W.A. SCHIER, J.M. CAMPBELL, G.P. COUCHELL, H.V. NGUYEN, D.J. PULLEN, S.V. TIPNIS, M.F. VILLANI, University of Massachusetts Lowell. — A spectrometer for measuring beta energy spectra in decay heat studies has been constructed and tested. The spectrometer consists of a 6.4-cm diameter x 6.4-cm length BC408 plastic scintillator made light tight with 0.0013-cm aluminum foil gated by a 3.8-cm diameter x 0.025-cm thick scintillator disk positioned directly against the foil. The bare scintillator disk is viewed by two photomultipliers in an uncoupled manner. The gating essentially eliminates the gamma rays from mixed spectra since gammas rarely interact with the thin disk. The smaller disk diameter eliminates edge loss of secondary electrons, faithfully reproducing the allowed shapes of test beta sources even in the presence of severe gamma-ray backgrounds. Design features and comparisons of beta spectra from test sources and from aggregate fission products following the thermal neutron fission of ^{235}U will be presented.

*Supported in part by the U.S. Department of Energy.

4. Helium Jet Fission Fragment Transfer Efficiency.* P.R. BENNETT, J.M. CAMPBELL, G.P. COUCHELL, H.V. NGUYEN, D.J. PULLEN, W.A. SCHIER, S.V. TIPNIS, M.F. VILLANI, University of Massachusetts Lowell. — The helium jet has been shown to transfer fission fragments with essentially uniform relative efficiency over the full mass distribution[1] (with a few possible exceptions) using fore pump oil vapor as a nucleation agent in a small fission chamber. Relative efficiency is measured by comparing characteristic x rays from fission products captured in a catcher at the fission foil and at the exit of the helium jet. Here we attempt to improve on earlier efficiency measurements by matching irradiation-cooldown-detection cycles, minimizing self-absorption of x rays and suppressing background by deflecting the beta particles emitted by the fission products. Details of the experimental setup and measured relative transfer efficiency will be presented.

*Supported in part by the U.S. Department of Energy.

[1]G.P. Couchell et al., Proc. Int. Conf. on Nuclear Data for Science and Technology, Mito, Japan (1988) p. 415: publ. JAERI, 1988.

5. Six-Group Decomposition of ^{238}U Delayed Neutron Composite Energy Spectra.* M.F. VILLANI, J.M. CAMPBELL, G.P. COUCHELL, D.J. PULLEN, W.A. SCHIER, University of Massachusetts Lowell. — Delayed neutron composite energy spectra have been measured for six delay-time intervals following the fast fission of ^{238}U. The delay-time intervals span the range 0.17 to 10.2 seconds following fission and the measured spectra span neutron energies from 10 keV to 4 MeV. Six-group spectra have been deduced from the measured composite spectra utilizing the Keepin six-group model with parameters obtained from ENDF/B-VI. A standard least-squares method was found to produce unphysical six-group spectra that were overly oscillatory and often negative. Physically acceptable six-group solutions were obtained using a constrained least-squares technique. The deduced six-group spectra are compared to the corresponding ^{238}U six-group spectra constructed from the ENDF/B-VI data base as well as those deduced from measured composite spectra of ^{235}U and ^{239}Pu.

*Supported in part by the U.S. Department of Energy.

6. Comparison of Selective Gamma-Ray Yields following Thermal Fission of ^{235}U with the ENDF/B-VI Decay Data.* J.M. CAMPBELL, G.P. COUCHELL, H.V. NGUYEN, D.J. PULLEN, W.A. SCHIER, S.V. TIPNIS, M.F. VILLANI, University of Massachusetts Lowell and T.R. ENGLAND, Los Alamos National Laboratory. — Gamma-ray energy spectra of aggregate fission products resulting from thermal neutron fission of ^{235}U have been measured with a Ge(Li) detector over a delay time range of 0.17 - 85.5 s following fission. Numerous individual gamma transitions have been identified and the time evolutions of their intensities have been determined. The observed variations of intensities with time have been compared with those predicted by the ENDF/B-VI fission-product data base. These preliminary measurements serve as a feasibility study of the use of high resolution aggregate gamma-ray spectra for checking the consistency of the ENDF/B-VI summation calculations based on individual fission product yields and decay data.

*Supported in part by the U.S. Department of Energy.

7. Response Functions for Beta-Particle and Gamma-Ray Spectrometers.* S.LI, J.M.CAMPBELL, G.P.COUCHELL, H.V.NGUYEN, D.J.PULLEN, W.A.SCHIER, S.V.TIPNIS, University of Massachusetts Lowell.---Response functions that vary with incident particle energies have been measured for both our beta spectrometer (a large plastic scintillator) and NaI detector with its massive shield (consisting of a combination of lead, brass and tungsten). Three bare IT sources, Sn/In-113m, ^{137}Cs and ^{207}Bi were used for measuring beta response functions in the energy range 0 to 1 MeV. NaI response functions were determined from the sources, e.g. ^{198}Au, ^{24}Na, produced through neutron activation using our 1-MW swimming-pool reactor, and the ^{19}F(p,$\alpha\gamma$)^{16}O reaction on our 2-MeV Van de Graaff accelerator was used to produce the 6.13-MeV line from ^{16}O.

* Supported in part by the U.S. Department of Energy.

8. Spectrum Analysis Programs for Decay Heat Studies of Aggregate Fission Products.* H.V.NGUYEN, J.M.CAMPBELL, G.P.COUCHELL, S.LI, D.J.PULLEN, W.A.SCHIER, S.V.TIPNIS, M.F. VILLANI, University of Massachusetts Lowell.--- A multiple peak deconvolution routine was developed for the Ge(Li) and HPGe detectors used in the study of individual gamma rays from aggregate fission products. The routine fits a sum of Gaussians to a selected region of the spectrum by applying a nonlinear least squares fit. The width of each peak was calculated, using the calibrated resolution of the detector, and was then kept fixed in the fitting process. In addition an unfolding code, FERD[1], which was recently obtained from the Radiation Shielding Information Center at ORNL, will be used for the unfolding of the nearly continuous aggregate gamma-ray spectra obtained with our NaI(Tl) detector and for the continuous spectra measured with our beta-particle spectrometer. A subroutine for interpolating measured response functions to any intermediate energy has been written for the beta response functions and is being developed for the gamma-ray responses.

1) B. W. Rust, D. T. Ingersoll, and W. R. Burrus, *A User's Manual for the FERDO and FERD Unfolding Codes*, ORNL/TM-8720, 1983.

*Supported in part by the U.S. Department of Energy.

9. Comparison of the Measured Time Evolution of Individual Fission-Product Gamma-Ray Lines with ENDF/B-VI Data*. S.V. TIPNIS, J.M. CAMPBELL, G.P. COUCHELL, S. LI, H.V. NGUYEN, D.J. PULLEN and W.A. SCHIER, University of Massachusetts Lowell, and T.R. ENGLAND, Los Alamos National Laboratory. High resolution gamma-ray spectra from aggregate fission products from $^{235}U(n_{th}, f)$ have been measured for six delay time intervals over the range 0.17 - 85.5s using a Ge(Li) detector. Although the spectra are complex, many gamma lines can be identified and for the more prominent ones the time dependence of their yields has been determined. Since fission products can be formed directly in fission or as a product in a beta decay chain, this time evolution may not be simple exponential. Several non-exponential cases have been observed among the more than twenty cases examined. These have all been compared with the predicted time dependences based on summation calculations using individual fission product yields and decay data from the ENDF/B-VI fission product data base.

*Supported in part by the U.S. Department of Energy.

10. High Resolution Gamma-Ray Spectra from Aggregate Fission-Products Measured in $^{235}U(n_{th}, f)$*. J.M. CAMPBELL, G.P. COUCHELL, S. LI, H.V. NGUYEN, D.J. PULLEN, W.A. SCHIER, and S.V. TIPNIS, University of Massachusetts Lowell. A high purity germanium detector has been used to measure gamma-ray spectra from the aggregate of fission products produced in $^{235}U(n_{th}, f)$ for fifteen different delay-time intervals in the range 0.6 - 100,000s following fission. Random background contribution is reduced by more than an order of magnitude by utilizing beta-gamma coincidence techniques and a further improvement by a factor of 3 to 5 in the peak-to-background ratio is had when a NaI(Tl) annulus is used to suppress contributions from Compton gamma rays. Nearly 300 gamma-ray peaks have been measured in the time interval <1s following fission and in the gamma energy range 0 - 6 MeV. Principal fission-product contributors identified in the gamma spectra so far are ^{96}Sr, ^{97}Sr, ^{98}Sr, ^{97m}Y, ^{98m}Y, ^{98}Y, ^{99}Zr, ^{99}Nb, ^{143}Ba and ^{146}La.

*Supported in part by the U.S. Department of Energy.

11. Study of Gamma-Ray Decay Heat from Aggregate ^{235}U Fission Products.* G.P. COUCHELL, J.M. CAMPBELL, S. LI, H.V. NGUYEN, D.J. PULLEN, W.A. SCHIER and S.V. TIPNIS, University of Massachusetts Lowell. --- A 12.7-cm x 12.7-cm NaI(Tl) scintillation detector has been used to study the gamma-ray component of decay heat released by fission products resulting from the thermal neutron fission of ^{235}U. Moderated neutrons from the ^{7}Li(p,n)^{7}Be reaction were used to initiate fission in 93%-enriched uranium foils, and a helium jet system was used to rapidly transfer the products to a low-background counting area. Beta-gamma coincidence gating suppressed background counts and selected delay time intervals after fission. A lead-brass shield with a tungsten two-segment collimator also greatly reduced background counts and minimized edge effects by selecting the central region of the crystal. Measurements were performed at 16 time intervals spanning the delay-time range 0.2 - 15,440 s. These are the first reported aggregate gamma-ray decay heat spectra measured below a 1-s delay time. Sample spectra illustrating the excellent statistical quality of the measurements and particularly the sensitivity of the spectrometer to high-energy gamma rays (due to the beta coincidence gating) will be shown.

* Supported in part by the U.S. Department of Energy.

12. Study of Beta-Particle Decay Heat from Aggregate ^{235}U Fission Products.* W.A. SCHIER, J.M. CAMPBELL, G.P. COUCHELL, S. LI, H.V. NGUYEN, D.J. PULLEN and S.V. TIPNIS, University of Massachusetts Lowell. --- Aggregate beta spectra following the thermal neutron fission of ^{235}U have been measured at 17 time intervals spanning the delay time range 0.2 to 11,960 s. These are the first beta spectra measurements below a delay time of 2 s and the only corroboration below 20 s of previous measurements[1]. Fission fragments were transferred with a He-jet/tape-transport system. Thermalized neutrons from the ^{7}Li(p,n)^{7}Be reaction were incident on the 93%-enriched uranium foil lining the small hemispherical fission chamber. Our beta spectrometer, based on a plastic well scintillator, derives its gamma-ray insensitivity through coincidence with a very thin disk scintillator. The disk has a smaller diameter than the well-scintillator to minimize edge loss effects. Details of spectrometer design, gamma rejection, typical response functions and the evolution of beta spectra with delay time will be presented.

1) J.K. Dickens et al., Nucl. Sci. Eng. 74, 106(1980).

* Supported in part by the U.S. Department of Energy

13. Gamma-Ray Study of Short-Lived Aggregate Fission Products from $^{235}U(n,f)$.* W.A. SCHIER, J.M. CAMPBELL, G.P. COUCHELL, S. LI, H.V. NGUYEN, D.J. PULLEN, E.H. SEABURY and S.V. TIPNIS, University of Massachusetts Lowell and T.R. ENGLAND, Los Alamos National Laboratory. --- A high purity germanium detector was used to measure gamma-ray spectra following thermal neutron-induced fission of ^{235}U for eighteen delay-time intervals in the range 0.1-100,000 s following fission. A helium-jet system was used to rapidly transport fission products to a low-background counting area. The gamma-ray spectrometer employed beta-gamma coincidence as well as a NaI(Tl) annulus for background suppression. Nearly 300 gamma-ray peaks have been analyzed for delay-time intervals < 10 s after fission in the energy range 0-6 MeV. The time evolution of a peak provides information about the lifetime of the precursor nuclide as well as the ratio of its direct production in fission to its production through radioactive decay. Where decay schemes are known the measured gamma-ray intensities can be used to deduce relative yields and in some cases metastable-to-ground-state production probabilities. Results of lifetimes, yields, etc. of short-lived products will be compared with CINDER 10 calculations based on ENDF/B-IV fission-product data.

* Supported in part by the U.S. Department of Energy

14. Experimental Development of Aggregate Beta-Particle and Gamma-Ray Decay Heat Study of $^{238}U(n,f)$.* E.H.SEABURY, J.M.CAMPBELL, G.P.COUCHELL, S.LI, H.V.NGUYEN, D.J.PULLEN, W.A.SCHIER, S.V.TIPNIS, University of Massachusetts Lowell.---A helium-jet/tape transfer system has been developed at the UMASS-Lowell 1 MW swimming pool research reactor. Previously this system was at the Lowell Van De Graaff accelerator in order to study the decay heat of $^{235}U(n,f)$ and $^{239}Pu(n,f)$ for time intervals between 0.1s - 50,000s after fission. In order to study $^{238}U(n,f)$ decay heat, in particular for time intervals below 20s which have not yet been studied, it was necessary to move the system. Fast neutron fluence measurements of a reactor beam port, background radiation measurements of the containment building, details of the helium-jet and reactor, and preliminary gamma-ray spectra obtained with a NaI(Tl) detector will be presented.

*Supported in part by the U.S. Department of Energy

15. Comparison of Halflives of Short-lived ^{235}U Fission Products with ENDF/B-VI*. S.V. TIPNIS, J.M. CAMPBELL, G.P. COUCHELL, S. LI, H.V. NGUYEN, D.J. PULLEN, E.H. SEABURY and W.A. SCHIER, University of Massachusetts Lowell, and T.R. ENGLAND, Los Alamos National Laboratory. --- Gamma-ray spectra of aggregate fission products from $^{235}U(n_{th}, f)$ were measured by beta-gamma coincidence using a HPGe detector enclosed in a NaI(Tl) Compton suppression annulus. Fission products were transported from the fission chamber to a low background counting room with a helium-jet/tape transport system. Spectra were measured for seven delay time intervals over a range of 0.13 - 8.3s. Many individual gamma-ray lines were identified, and their lifetimes calculated assuming that they have a simple exponential time dependence. Comparisons are made with the CINDER10 calculations which are based on the ENDF/B-VI data base.

*Supported in part by the U.S. Department of Energy.

16. Time Evolution of Fission-Product Gamma Spectra using ENDF/B-VI.* J.M. CAMPBELL, G.P. COUCHELL, S. LI, H.V. NGUYEN, D.J. PULLEN, W.A. SCHIER, E.H. SEABURY, S.V. TIPNIS, University of Massachusetts Lowell, T.R. ENGLAND, Los Alamos National Lab. --- ENDF/B-VI (Evaluated Nuclear Data File) contains information on energies and relative intensities of fission product gamma rays. By combining the relative intensity of each gamma-ray line with the time-dependent activity of the corresponding fission product, an absolute fission-product gamma-ray spectrum can be determined at any time interval following fission for comparison with measurements. The time-dependent activities have been calculated using CINDER10, a code used in calculations of the production and decay of fission products. Nearly one-thousand coupled differential equations are involved in these calculations. Their method of solution and the resulting time-dependent gamma-ray spectra will be discussed.

* Supported in part by the U.S. Department of Energy

17. Interpolation of Response Functions Used in the Analysis of 5"x5" NaI Gamma-ray Measurements.* H.V. NGUYEN, J.M. CAMPBELL, G.P. COUCHELL, S.LI, D.J. PULLEN, W.A. SCHIER, E.H. SEABURY and S.V. TIPNIS, University of Massachusetts Lowell.--- A 5"x5" NaI detector has been used to measure gamma-ray decay heat resulting from the decay of aggregate fission products. In order to extract the true gamma-ray energy distribution from the measured spectra, the detector response functions for monoenergetic gamma rays spanning the energy range of the measurements must be determined. At present we have measured nine such response functions in the energy range 0.081-1.52 MeV. An interpolation scheme has been developed for estimating the response function at any other intermediate energy. A library of response functions generated in this way has been used to fit in a least-squares fashion the gamma-ray spectrum of ^{152}Eu measured by the same NaI detector. The fit is an excellent reproduction of both the photopeak and continuous regions of the entire measured spectrum.

*Supported in part by the U.S. Department of Energy.

18. Aggregate Gamma Decay Heat Spectra Comparison Following ^{238}U and ^{235}U Neutron Fission*. S. LI, J.M. CAMPBELL, G.P. COUCHELL, H.V. NGUYEN, D.J. PULLEN, W.A. SCHIER, E.H. SEABURY and S.V. TIPNIS, University of Massachusetts Lowell.--- Recently measured aggregate gamma-ray decay heat spectra following the fast neutron fission of ^{238}U will be compared with the aggregate gamma spectra from the thermal neutron fission of ^{235}U. Fission products were transported by the helium-jet/tape-transport system from the fission chamber to the detector. Both sets of spectra were obtained with a collimated 5" x 5" NaI(Tl) detector through beta-gamma coincidence. The ^{238}U fast neutron fission product measurements were made at eleven delay time intervals in the range 1 - 9,000 s. This set of measurements include the first aggregate gamma-ray measurements for ^{238}U neutron fission below 20 s delay time. Similar time intervals were chosen in the ^{238}U/^{235}U gamma spectra comparision.

*Supported in part by the U.S. Department of Energy.

19. Feasibility Study of Gamma-Ray Coincidence Measurements of $^{238}U(n,f)$ Fission Products.* E.H. SEABURY, J.M. CAMPBELL, P. CHOWDHURY, G.P. COUCHELL, S. LI, H.V. NGUYEN, D.J. PULLEN, W.A. SCHIER, S.V. TIPNIS, University of Massachusetts Lowell.---Some product nuclides of $^{238}U(n,f)$ emit gamma rays that have not been placed in energy level schemes. The goal of this study is to determine whether these gamma rays can be placed in existing level schemes, and if new transitions can be identified through the use of gamma-gamma coincidence measurements. A helium-jet transport system is already in place at the UMASS-Lowell 1 MW Research Reactor for the study of fission fragment beta and gamma decay. Two HP Ge detectors housed in Compton-anticoincidence shields were used to perform the measurements. In this feasibility study the system was used with an event-mode data-acquisition-and-analysis system. Details of the study and preliminary data will be presented.

*Supported in part by the U.S. Department of Energy

20. Beta Energy Spectra from U-238 Fission Products.* S. LI, J.M. CAMPBELL, G.P. COUCHELL, H.V. NGUYEN, D.J. PULLEN, W.A. SCHIER, E.H. SEABURY and S.V. TIPNIS, University of Massachusetts Lowell, and T.R. ENGLAND, Los Alamos National Laboratory.---We have recently measured the beta spectra from U-238 fission products at selected delay times as short as 1 s through 20 s, where few experimental results exist. A thin-disk-gated beta spectrometer was energy calibrated using beta and gamma sources. The low energy response for the beta spectrometer was established based on three measurements using bare IT sources, Sn-113m, Cs-137, and Bi-207, all having energies below 1 MeV. At higher energies trial response functions having similar shapes, but energy dependent widths, were tested by using them to unfold continuous beta spectra of known shape. By adjusting the widths excellent fits to the continuous spectra were obtained. These response functions were then used to unfold the U-238 fission-product beta spectra. The resulting beta energy distributions are remarkably similar to ones obtained earlier from U-235 fission products. Our U-238 beta energy distributions are somewhat less energetic than predictions based on ENDF/B-VI fission-product data.

*Supported in part by the U.S. Department of Energy.

21. Comparison of the Measured Time Evolution of Individual Fission-Product Gamma-Ray Lines with ENDF/B-VI Data.* J.M. CAMPBELL, P. CHOWDHURY, G.P. COUCHELL, S. LI, H.V. NGUYEN, D.J. PULLEN, W.A. SCHIER and S.V. TIPNIS, University of Massachusetts Lowell, and T.R. ENGLAND, Los Alamos National Laboratory. Gamma-ray energy spectra of aggregate fission products produced after fission of ^{238}U have been measured, extending our previous similar study of fission products resulting from thermal fission of ^{235}U. Spectra are measured using a high-purity germanium detector enclosed in a NaI(Tl) Compton suppression annulus. The rapid transfer of fission products from the fission chamber to a low-background counting room by means of a helium-jet/tape transport system leads to a marked reduction in background and allows measurement of spectra for delay times less than 1s. Beta-gamma coincidence leads to a further reduction in background by about two orders of magnitude. Spectra have been measured for 15 delay time intervals over a range of 0.6 - 25000s. In a preliminary analysis of spectra measured out to 50s, individual gamma transitions have been identified and the time evolution of intensities for a number of them have been determined. Comparisons are made with CINDER10 calculations which use ENDF/B-VI fission data.

*Supported in part by the U.S. Department of Energy.

22. Unfolding of Gamma Spectra from Aggregate Fission Products using Response Functions.* H.V. NGUYEN, J.M. CAMPBELL, G.P. COUCHELL, S.LI, D.J. PULLEN, W.A. SCHIER, E.H. SEABURY and S.V. TIPNIS, University of Massachusetts Lowell.--- A 5"x5" NaI detector has been used to measure gamma-ray spectra resulting from the decay of aggregate fission products. In order to extract the true gamma-ray energy distribution from the measured spectra, the detector response functions for monoenergetic gamma rays spanning the energy range of the measurements must be determined. At present we have measured 13 such response functions in the energy range 0.081-6.13 MeV. An interpolation scheme has been developed for estimating the response function at any other intermediate energy. In addition a C program has been written for obtaining the gamma-ray energy distribution. It assumes the measured spectrum consists of a superposition of a specified number of response functions placed at specified energies. The program then computes the area as a function of gamma energy of each response function in a least-squares fashion. It was possible to use up to 100 response functions in the representation of each spectrum, where each response function consists of 1500 bins.

*Supported in part by the U.S. Department of Energy.

23. Comparison of Production Probabilities of ^{235}U Fission Products with ENDF/B-VI*. S.V. TIPNIS, J.M. CAMPBELL, G.P. COUCHELL, S. LI, H.V. NGUYEN, D.J. PULLEN, E.H. SEABURY and W.A. SCHIER, University of Massachusetts Lowell, and T.R. ENGLAND, Los Alamos National Laboratory. --- Gamma-ray spectra of aggregate fission products from $^{235}U(n_{th}, f)$ were measured by beta-gamma coincidence using a HPGe detector enclosed in a NaI(Tl) Compton suppression annulus. Fission products were transported from the fission chamber to a low background counting room with a helium-jet/tape transport system. The spectra were measured over delay times ranging from 0.24 to 564s. Many individual gamma-ray lines were identified, and their relative intensities compared to the ones as predicted in the Nuclear Data Sheets. From the relative intensities, the production probabilities of the precursor isotopes were calculated and compared with the values listed in the ENDF/B-VI fission-product data base.

*Supported in part by the U.S. Department of Energy.

5. THESIS ABSTRACTS

Two MS degrees were conferred during this period for research based on this project. The thesis abstracts are presented below.

CALCULATION OF GAMMA ENERGY SPECTRA OF FISSION PRODUCTS FOLLOWING THE FISSION OF ^{235}U USING EVALUATED NUCLEAR DATA FILES

by

JOANN M. CAMPBELL

Gamma-ray energy spectra after thermal fission of ^{235}U are calculated for individual fission products using the Evaluated Nuclear Data Files (ENDF/B-VI). Delay times span the range of 0.1 to 100000 s after fission, while energies range from 0 to 6.0 MeV with this range divided into 1 keV energy bins. The activities for 891 fission products are found at 25 delay times using the program CINDER10, a well-established fission product and depletion code. These activities are then used to calculate a time-dependent gamma spectrum for 244 fission products using ENDF/B-VI spectral data. The 320 most intense nuclides at each time are found. The calculated data are compared to measurements performed at the University of Massachussetts Lowell 5.5 MV Van De Graaf accelerator, using a helium-jet/tape transfer system to quickly flush fission fragments from the fission chamber. The experimental time-evolutions of discrete gamma-ray lines of 14 nuclides are compared to those calculated by CINDER10. The measured intensity of six gamma-ray lines of ^{96}Sr are also compared to the spectral data for this nuclide contained in ENDF/B-VI.

INTERPOLATION OF RESPONSE FUNCTIONS USED IN THE ANALYSIS OF 5"x5" NaI GAMMA-RAY SPECTRUM MEASUREMENTS

by

HUNG V. NGUYEN

A 5"x5" NaI detector has been used to measure gamma-ray decay heat resulting from the decay of aggregate fission products. In order to extract the true gamma-ray energy distribution from the measured spectra, the detector response functions for monoenergetic gamma rays spanning the energy range of the measurements must be determined. Nine such response functions in the energy range 0.081 - 1.52 MeV have been measured and an interpolation scheme for estimating the response function at any other intermediate energy has been developed. In order to test the validity of the interpolation scheme, a program SPEC-FIT has been written which used a library of response functions generated by this procedure to fit, in a least-squares fashion, the gamma-ray spectrum of ^{152}Eu measured by the same NaI detector. The fit is an excellent reproduction of both the full-energy peak and continuous regions of the entire measured spectrum. The extraction of the energy distribution from the actual decay heat spectra will be performed by the program FERD-PC, which was obtained from the Radiation Shielding Information Center at ORNL. The implementation of this program has been successfully tested using a "measured" spectrum that was artificially generated by performing a superposition of a set of analytical response functions weighted by a linear distribution function. The analytical response function used consisted of a Gaussian and a tail. For future work, it is intended to carry out the response function interpolation scheme to the energy range beyond 1.5 MeV.

HIGH RESOLUTION GAMMA-RAY SPECTRA FOR $^{235}U(n_{th},ff)$[a]

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ABSTRACT

High-resolution gamma ray spectra from thermal fission of ^{235}U have been measured for 21 delay time intervals in the range 0.2s to nearly 100,000s. More than 170 gamma transitions have so far been been studied out to 1000s in 27 different fission products. Relative line intensities and time evolutions for the gamma lines provide information on relative production probabilities for fission products, direct -vs.- beta chain production ratios, isomeric to ground-state production ratios, as well as precursor halflives, for comparison with ENDF/B-VI.

I. INTRODUCTION AND METHOD

The present high-resolution gamma ray measurements are part of a general study under way at Lowell, in collaboration with Los Alamos, of separate aggregate beta- and gamma decay heats measured for $^{235}U(n_{th},ff)$ as a function of time following fission. Measurements are carried out with the UML Van de Graaff accelerator and a helium jet system[1] provides rapid transfer of the fission fragments to a low background counting area. Here they are sprayed onto a moving tape having adjustable speed which then transports them to a gamma-ray spectrometer.

The spectrometer, shown in Fig. 1, consists of a high purity germanium detector with a NaI(Tl) annulus for Compton suppression. Incorporation also of beta/gamma coincidence gating in the spectrometer is found to reduce the overall background in the gamma spectra by about a factor 10^2, at the same time sharply defining the delay-time interval for a spectral measurement. In order to maintain satisfactory count rates, the HPGe detector is located close to the tape carrying the fission products and this requires a long cryostat pipe to bring the detector to the front face of the annulus rather than the center. Although this reduces the suppression efficiency of the annulus, the peak-to-background ratio is nevertheless improved by further factors of 3 to 5.

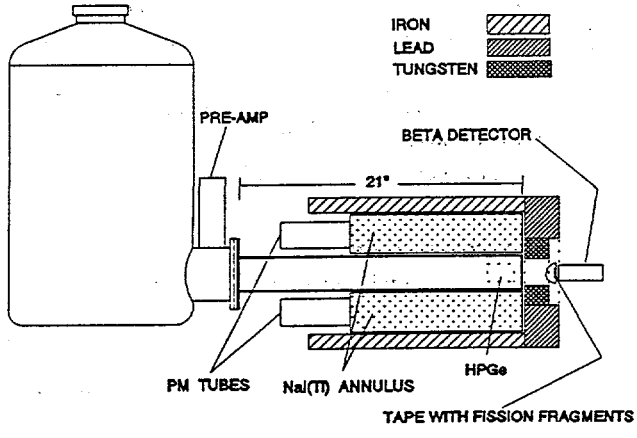

Figure 1. HPGe spectrometer with NaI(Tl) annulus.

[a]Supported by the U.S. Department of Energy

This reduced background not only enables measurements to be extended to weaker gamma lines but also extends the time period over which an individual peak can be followed, which aids considerably in the identification of the precursor nuclides.

II. RESULTS

Gamma spectra were measured between 0 - 6 MeV for 21 time intervals in the range 0.2s - 100,000s. Figure 2 shows the energy region 0 - 1.5 MeV for one such spectrum, corresponding to a time interval 0.58 - 0.90 s, with a number of prominent gamma lines identified by their precursor. Although the spectrum is rich in gamma lines it is nevertheless quite selective in the nuclides which are predominant contributors, since only those whose half lives approximately match the interval tend to predominate, (see caption to Fig. 2). This greatly simplifies the observed spectra.

Analysis of these spectra has so far been carried out for delay times ranging from 0.2s to nearly 1000s. After correcting for detector efficiency and relatively normalizing between time intervals, intensities have been determined for over 700 gamma peaks, from which a total of 170 different gamma transitions have been identified in 27 fission products. Semi-log plots of intensities as a function of time are shown for several representative cases in Fig. 3. The lines represent the gamma activities calculated at various delay times with the LANL code CINDER, which uses spectral information from the ENDF/B-VI data file. They are normalized here to provide best fits to the measured relative intensities. The fits are generally very satisfactory and the figures demonstrate the sensitivity of the measurements to short-lived fission products (e.g., ^{98}Y: $T_{1/2}$ = 0.64s, and ^{97}Sr: $T_{1/2}$ = 0.42s) as well as to the direct-vs.-beta chain production, as depicted by the curvatures which are apparent in many of the plots (e.g., ^{99}Zr and ^{145}La).

Measured relative gamma line intensities are compared with published NDS[2] values for two typical cases (^{98}Y and ^{145}La) in Tables I and II. The generally good agreement indicates the measured lines are not appreciably intermixed with lines from other precursors.

Relative production probabilities can also be determined from the measured spectra with the aid of known gamma activities[2]. Preliminary values determined in the present work are compared in Table III with the relative

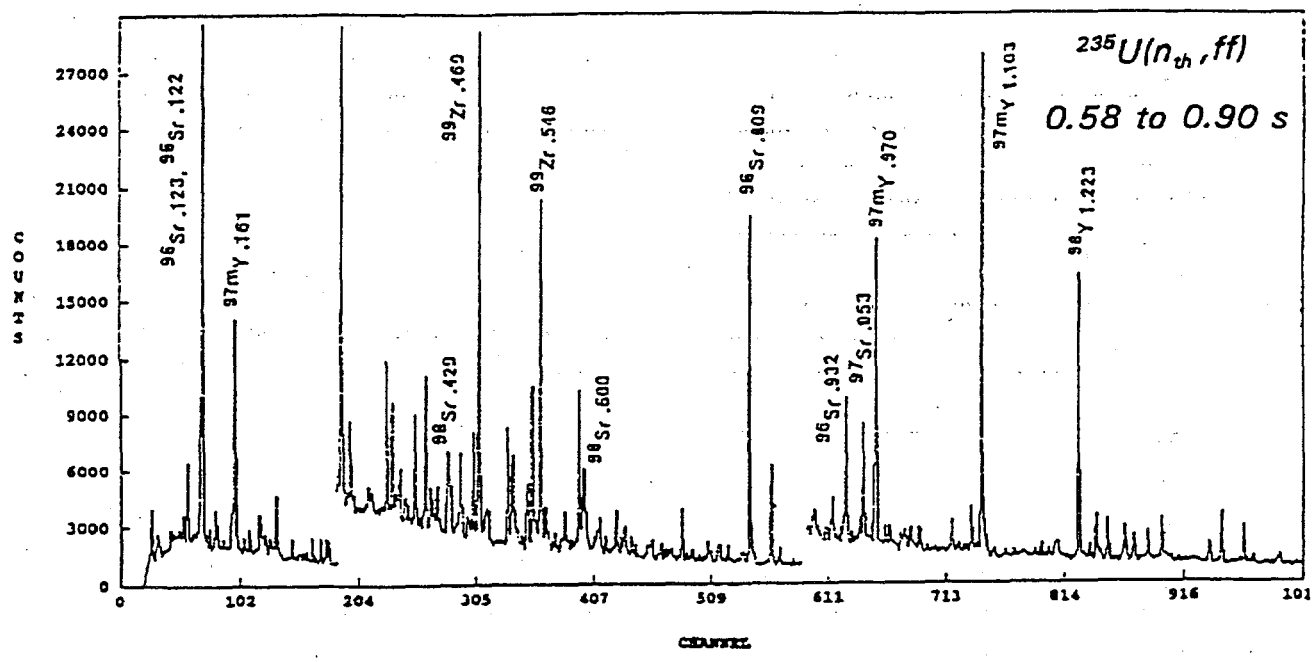

Figure 2. First 1.5 MeV of gamma ray spectrum measured in the time interval 0.58 - 0.90s following ^{235}U fission. Precursors (halflives) identified with the most prominent lines are ^{96}Sr (1.06s), ^{97}Sr (0.42s), ^{97m}Y (1.23), ^{98}Sr (0.64s), ^{98}Y (0.64s) and ^{99}Zr (2.1s).

TABLE I. Present measurement vs. NDS relative gamma line intensities following beta decay of ^{98}Y ($T_{1/2}$ = 0.64s).

Level (keV)	Gamma (keV)	RELATIVE INTENSITIES NDS (ref.2)	Present
4451	3228	1.4	1.1
	4450	3.4	3.3
4164	2306	0.7	0.7
	2421	1.6	1.4
	2574	0.8	1.2
	2941	5.8	4.7
	3310	2.4	4.0
1859	269	2.6	2.7
	636	0.5	-
1744	522	0.7	0.7
	890	0.4	1.5
	1744	1.4	1.6
1437	214	1.5	-

TABLE II. Present measurement vs. NDS relative gamma line intensities following beta decay of ^{145}La ($T_{1/2}$ = 24.8s).

Level (keV)	Gamma (keV)	RELATIVE INTENSITIES NDS (ref.2)	Present
2377	2377	3.22	-
2360	2359	3.83	3.72
2156	2156	3.22	3.72
1890	1820	1.84	1.59
1002	932	1.72	-
1021	787	2.83	2.89
672	672	3.10	2.89
447	447	0.96	1.18
356	356	1.38	1.07
234	170	0.61	0.71

production probabilities from ENDF/B-VI based on individual precursor data. The uncertainties in these preliminary measurements are estimated at no more than 15% and the assigned ENDF uncertainties (%) are listed in parentheses. Analysis of these data still continues.

A comparison of isomeric to ground state production ratios in fission can be made where different gamma transitions are involved. Table IV shows such comparisons for $^{97m}Y/^{97}Y$ and $^{98m}Y/^{98}Y$. The ^{97}Y ratio is in fair agreement with ENDF/B-VI whereas there is substantial disagreement in the case of ^{98}Y.

TABLE III. Relative production probabilites for $^{235}U(n_{th},ff)$. Values in parentheses are the percentage uncertainties assigned in the *pre* ENDF/B-VI evaluation.

Nuclide	ENDF/B-VI	Present
^{91}Rb	2.68 (1.4)	1.78
^{94}Rb	0.79 (2.8)	0.81
^{95}Rb	0.37 (4.0)	0.40
^{94}Sr	3.07 (1.4)	4.35
^{96}Sr	1.17 (6.0)	1.60
^{97}Sr	0.83 (2.8)	0.64
^{98}Sr	0.39 (6.0)	0.44
^{96m}Y	0.97 (45)	0.69
^{97}Y	1.45 (23)	4.56
^{97m}Y	0.88 (-)	1.38
^{98}Y	0.53 (23)	2.17
^{98m}Y	0.63 (32)	0.37
^{99}Y	1.00 (6.0)	0.90
^{100}Y	0.15 (64)	0.15
^{99}Zr	2.69 (2.8)	3.06
^{140}Cs	2.96 (2.8)	2.02
^{145}La	1.85 (6.0)	4.02

TABLE IV. Isomeric/Ground-state production ratios.

Nuclide	ENDF/B-VI	Present
$^{97m}Y/^{97}Y$	0.60	0.94
$^{98m}Y/^{98}Y$	1.73	0.06

III. CONCLUSIONS

The wealth of information obtained from high-resolution aggregate gamma-ray measurements following ^{235}U fission and the numerous comparisons which can be made with ENDF/B-VI data prove the value of such studies in testing ENDF. Analysis of the present measurements is currently being extended to delay times out to nearly 100,000s and to the longer-lived fission products. Similar composite high-resolution gamma spectral measurements are also being carried out for ^{238}U and ^{239}Pu fission.

REFERENCES

1. Walter A. Schier et al., these Proceedings.

2. "Nuclear Data Sheets", Vols. 35 - 64, Natl. Nucl. Data Center, Publ. Academic Press Inc., NY.

ENERGY DISTRIBUTIONS OF GAMMA AND OF BETA DECAY HEAT AS FUNCTION OF DECAY TIME FOR $^{238}U(n,f)$[a]

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ABSTRACT

Separate aggregate gamma-ray and aggregate beta-particle measurements following fast neutron fission of ^{238}U were made as a function of delay time over the range 1-4400s. Using sets of response function based on measured monoenergetic gamma and beta sources, the program FERDO was employed to convert these spectra to energy distributions for comparison with ENDF/B-VI.

I. INTRODUCTION

Separate aggregate gamma-ray and aggregate beta-particle decay heat measurements following $^{238}U(n,f)$ were made at the UMASS Lowell 1-MW reactor and energy distributions extracted from these spectra are presented. Measurements at delay times below 20 s are of particular interest in this study so a helium-jet/tape-transport system (Fig. 1) is employed for rapid transfer (0.2 s). The uniformity of the full mass distribution has been verified with x-ray measurements. Only the inert gases are found not to stick to the tape and will require additional measurements in the future but these account for only 4-20% of the decay heat in this delay time range. The reactor core is the source of fast neutrons producing sufficient fission rates in the depleted ^{238}U foil lining the small (17mm radius) hemispherical chamber. Fission of any residual ^{235}U in the foil is suppressed with the cadmium wrapping. Delay time is selected by varying the tape speed or position of the spectrometer along the tape.

[a] Supported by the U.S. Department of Energy

II. GAMMA DECAY-HEAT MEASUREMENTS

The gamma-ray spectrometer used in these decay heat measurements is a collimated, well-shielded 5"x5" NaI(Tl) detector. A thin (1mm) plastic scintillator detecting mainly beta particles is placed along the tape and gated in coincidence with this spectrometer for precise selection of a delay-time interval and for background suppression. Details of the spectrometer and its response functions are given in these proceedings in the ^{235}U decay-heat paper with the same authorship[1].

The 0-3 MeV portion of measured gamma spectra are shown in Fig. 2 to display the time evolution of the structure. Fig. 3 shows the energy distributions unfolded with FERDO[2] from four cases compared with ENDF/B-VI[3].

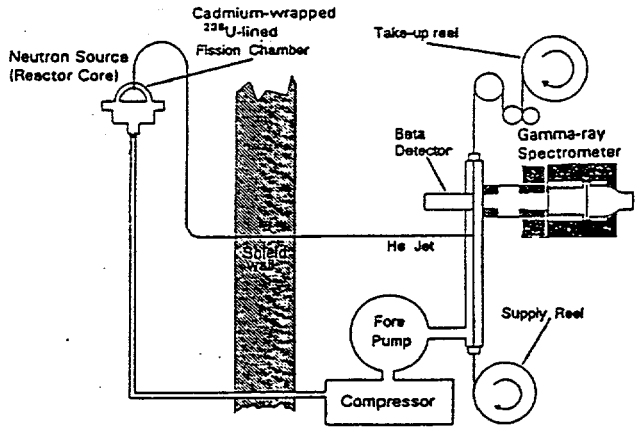

Figure 1. He-jet/tape-transport system with spectrometer.

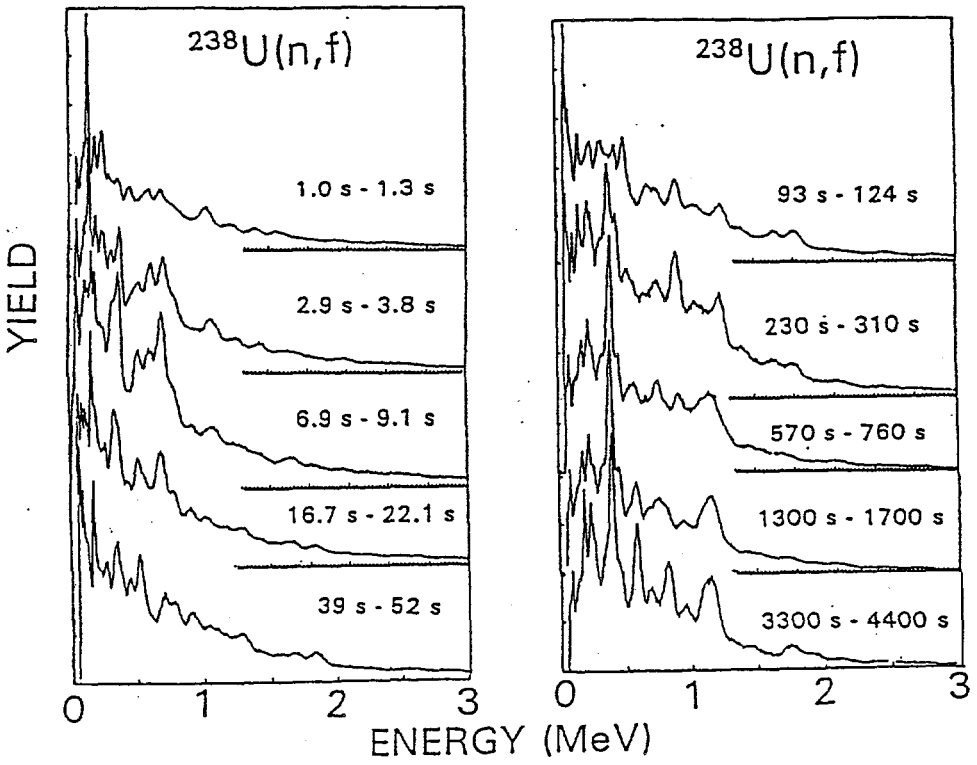

Figure 2. The 0-3 MeV portions of measured aggregate gamma-ray spectra following fast neutron fission of ^{238}U.

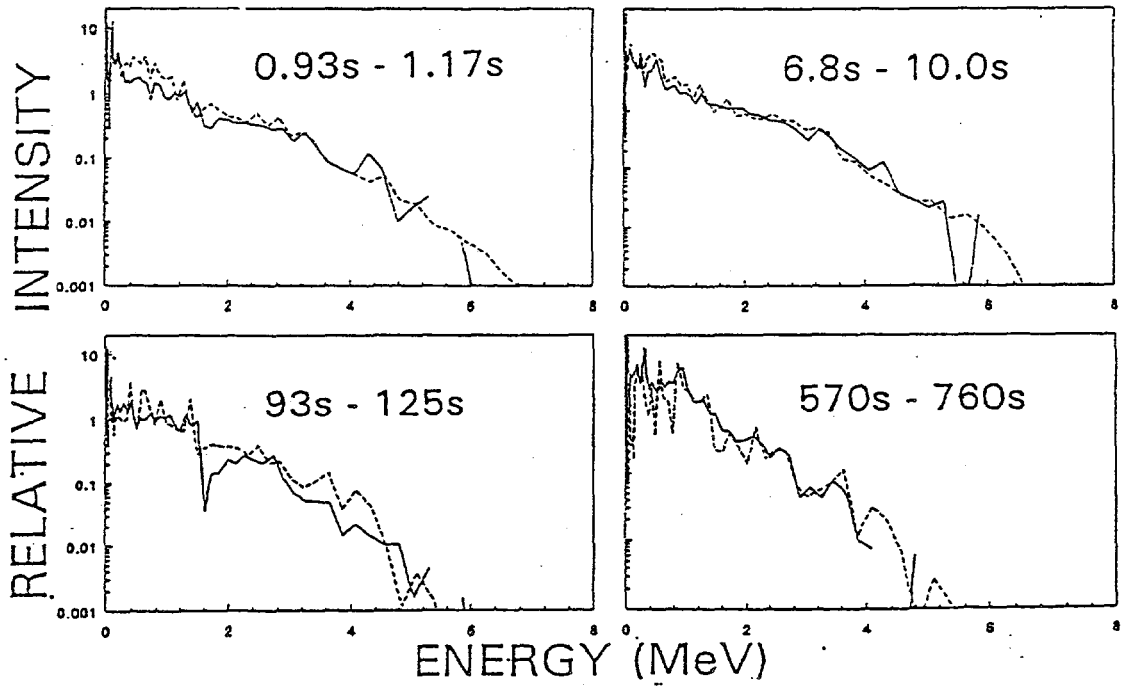

Figure 3. Our aggregate gamma energy distributions (solid line) at selected delay times following $^{238}U(n,f)$ compared with ENDF/B-VI (dashed line).

III. BETA DECAY-HEAT MEASUREMENTS

The beta spectrometer shown in Fig. 4 consists of a 3"x3" plastic scintillator optically isolated by an aluminum foil from a thin scintillator disk (0.04"x1.5") on its surface. Beta particles passing through the disk act as trigger to the large cylinder scintillator whereas gamma rays seldom interact with the thin disk scintillator. This high degree of gamma insensitivity is essential since a similar number of gamma rays accompany the betas.

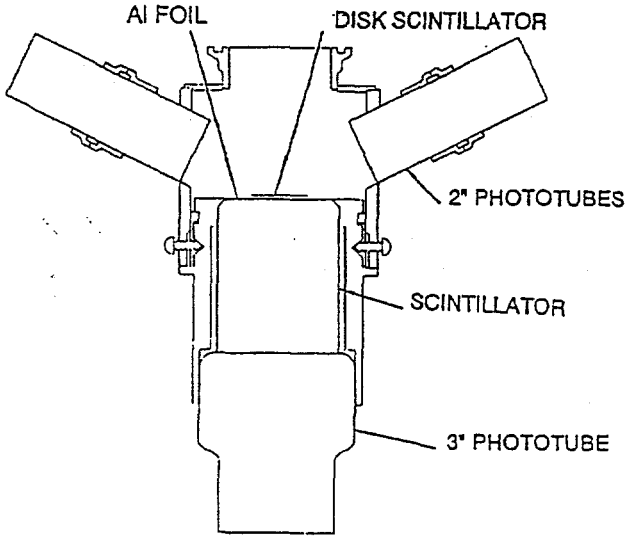

Figure 4. Diagram of the beta spectrometer.

For $E_\beta \leq 1$MeV, bare IT sources (^{113m}Sn, ^{137}Cs and ^{208}Bi) are used to measure response functions. In the case of ^{137}Cs shown in Fig. 5 the response function's low energy tail is extracted from the beta spectrum by performing an x-ray coincidence for this 2.6min. metastable state. The response function fit is also shown.

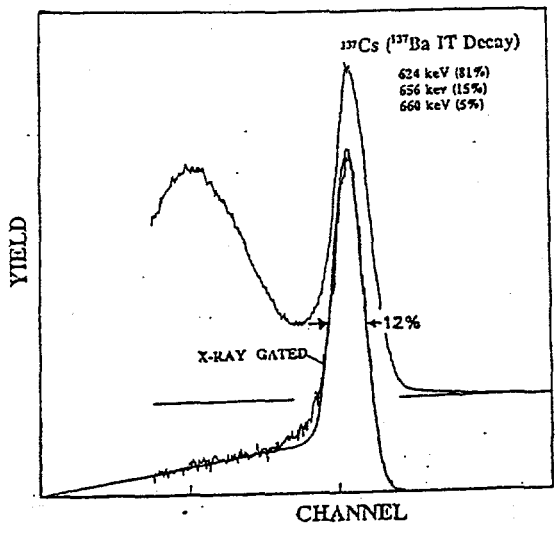

Figure 5. Beta response function from bare ^{137}Cs.

A trial set of beta response functions based on measured ($E_\beta \leq 1$MeV) ones was generated (Fig. 6). To test this set

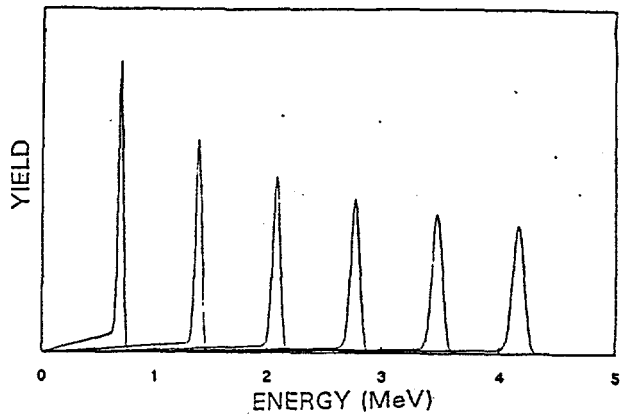

Figure 6. Trial set of beta response functions.

above 1MeV, beta spectra with known shapes (^{24}Na, ^{38}Cl, etc.) were measured and analyzed with the FERDO unfolding program. The ^{24}Na beta energy distribution is compared to the resolution-broadened theoretical shape in Fig. 7. Good agreement is observed in such comparisons except at low energies where the beta detection efficiency falls off due to the betas traversing the disk scintillator.

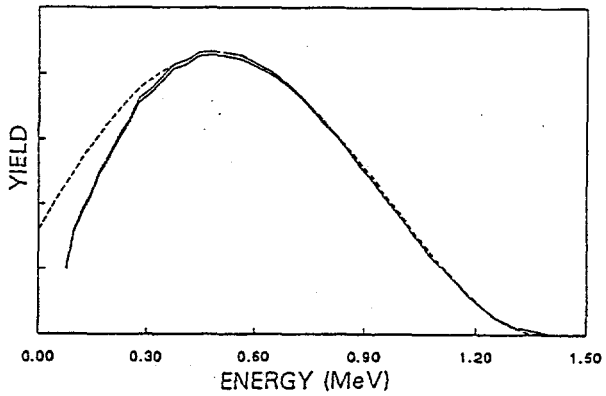

Figure 7. FERDO output compared to theoretical shape of ^{24}Na beta energy distribution.

Aggregate ^{238}U(n,f) beta spectra were measured between 1-1000s and converted to energy distributions using FERDO with this trial set of beta response functions. Four representative energy distributions are shown in Fig. 8 at delay times of approximately 1, 10, 100 and 1000s. The beta energy distributions of Dickens et al.[4] for ^{235}U(n^{th},f) are represented by dashed curve at three similar delay times. ENDF/B-VI predicts that the beta spectra for the ^{235}U and ^{238}U should be nearly identical and this comparison bears out that prediction. But comparing directly to ENDF/B-VI (dot-dashed curves) our spectra are generally softer and have somewhat different shapes.

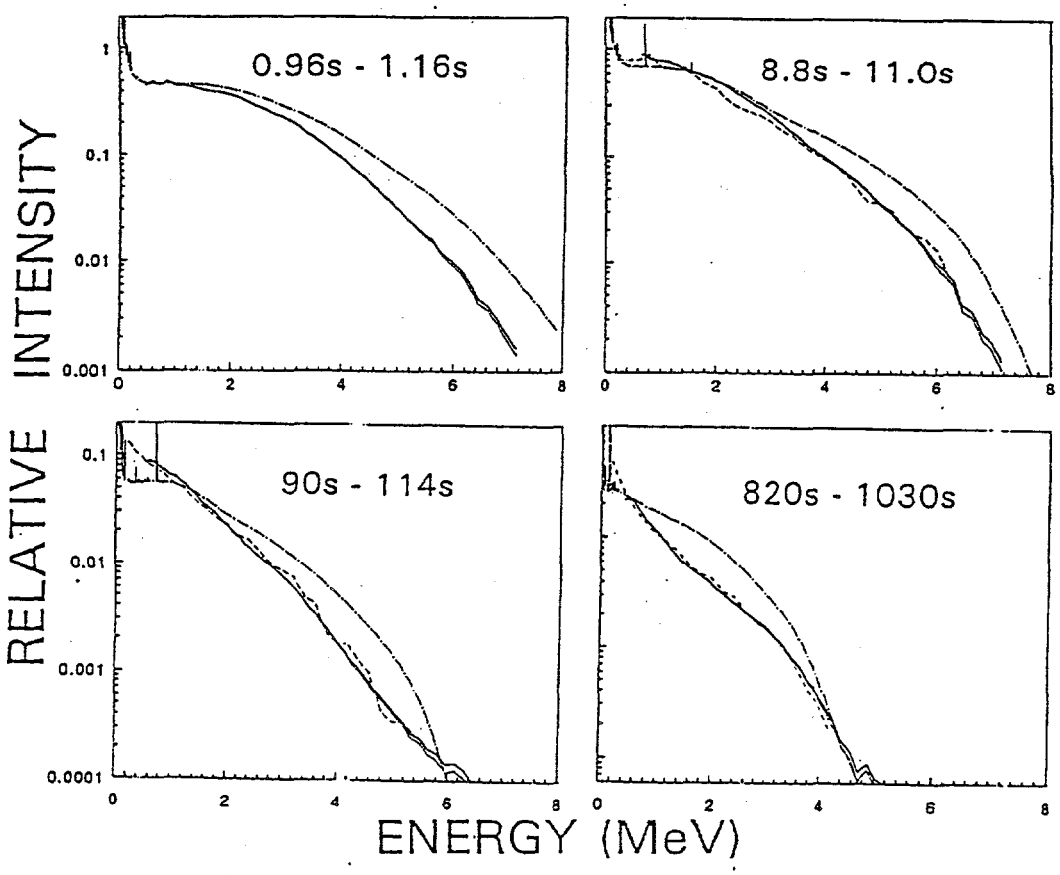

Figure 8. *Our aggregate beta energy distributions (solid) following $^{238}U(n,f)$. The dashed curves represent beta distributions following $^{235}U(n_{th},f)$ at similar delay times[Ref. 4]. The ENDF/B-VI are the dash-dot curves.*

IV. CONCLUSIONS

Separate aggregate gamma-ray and beta-particle energy distributions were measured following fast neutron fission of ^{238}U as a function of delay time over the range 1-4400s. These are the first measurements of this type to study both beta and gamma aggregate spectra for delay times shorter than 20s after fission. Comparisons of gamma-ray spectra made at four selected delay times with summation calculations using the ENDF/B-VI data base show good agreement in each case. Similar comparisons of our measured beta energy distributions with summation calculations are only in fair agreement. In particular at short delay times the measured beta spectra are considerably less energetic than the ENDF/B-VI predictions. However, the presently measured ^{238}U beta distributions are in excellent agreement with similar ^{235}U beta distributions measured earlier at ORNL. Such agreement between ^{235}U and ^{238}U spectra is predicted by ENDF/B-VI summation calculations.

REFERENCES

1. G.P. Couchell et al., paper presented at this conference (1994).

2. B.W. Rust, D.T. Ingersoll and W.R. Burus, *A User's Manual for the FERDO and FERD Unfolding Codes,* ORNL/TM-8720 (1983).

3. T.R. England, R. Wilczynski and N.L. Whittemore, *CINDER-7: An Interim Users Report,* Los Alamos National Laboratory report LA-5885-MS (April 1975).

4. J.K. Dickens, J.F.Emery, T.A. Love, J.W. McConnell, K.J. Northcutt, R.W. Peelle and H. Weaver, *Fission-Product Energy Release for Times Following Thermal-Neutron Fission of ^{235}U between 2 and 1400 Seconds,* ORNL/NUREG-14 (1977).

A STUDY OF GAMMA-RAY AND BETA-PARTICLE DECAY HEAT FOLLOWING THERMAL NEUTRON INDUCED FISSION OF ^{235}U[a]

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ABSTRACT

Aggregate gamma-ray and aggregate beta-particle energy spectra have been measured for fission products resulting from thermal neutron-induced fission of ^{235}U for decay times ranging from approximately 0.2s to 12,000s after fission. Preliminary energy distributions have been deduced from the measurements and these have been compared with summation calculations performed with CINDER using the ENDF/B-VI fission product data base.

I. INTRODUCTION

Aggregate fission-product gamma-ray and beta-particle energy spectra have been measured following thermal-neutron-induced fission of ^{235}U. Neutrons from the $^{7}Li(p,n)^{7}Be$ reaction produced at the UMASS Lowell 5-MV Van de Graaff accelerator were thermalized and used to initiate fission in a highly enriched ^{235}U foil lining the inner surface of a hemispherical fission chamber. A helium-jet/tape-transport system was used to rapidly transfer fission products from the chamber to a low-background counting area (see paper by W. Schier et al.[1] at this meeting). Delay time after fission was selected by varying the tape speed or the position of the spectrometer relative to the He-jet spray point.

[a]Supported by the U.S. Department of Energy

These are the first reported measurements of both the beta and gamma spectra of aggregate ^{235}U fission products to extend to delay times well below 1s. Special emphasis was placed on measurements at short delay times where summation calculations based on individual fission product nuclei display their largest uncertainties[2].

II. GAMMA-RAY ENERGY SPECTRA

Gamma-ray spectra were measured using a 5"x5" NaI(Tl) detector housed in a massive shield consisting of a combination of lead, brass and tungsten as depicted in Fig. 1. The collimator restricts gamma rays to the central 3" and its magnet deflects beta-particles so only the gamma component of the tape activity is seen by the detector. A thin beta detector is located on the opposite side of the

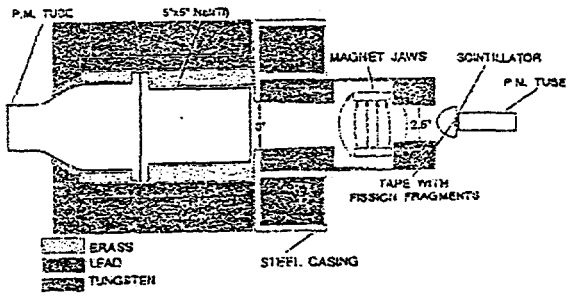

Figure 1. Gamma-ray spectrometer with beta-gating.

transport tape as shown, and is used to gate the NaI spectrum.

Although the NaI detector is well shielded and views only a limited portion of the tape, it is the beta detector which is essentially in contact with the tape which defines the delay time interval. Equally important, the coincidence reduces background, including that due to betas that pass through the magnetic deflection field by about two orders of magnitude.

Spectrometer response functions were determined at 15 gamma-ray energies spanning the region 0.05-6.13MeV. Algorithms were written to interpolate these measured response functions to any intermediate energy. Some sample calculated response functions are shown in Fig. 2.

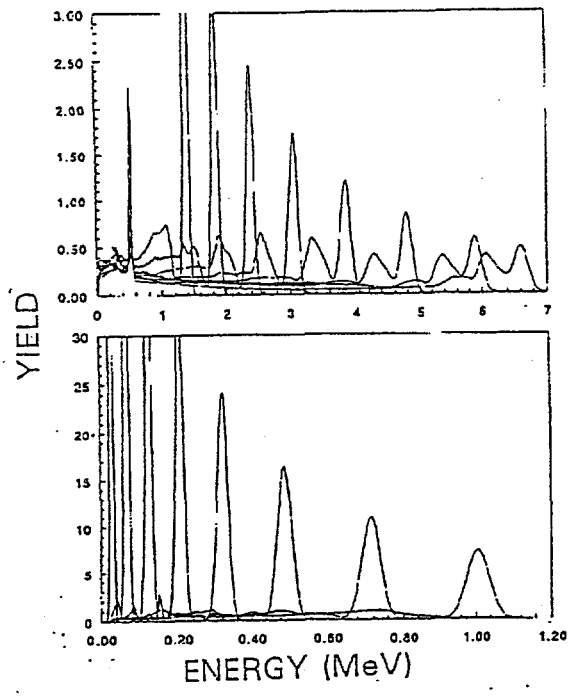

Figure 2. Gamma-ray response functions.

To test the validity of the measured response functions as well as the interpolation scheme, the measured ^{152}Eu spectrum was fitted as shown in Fig. 3 using the known energies of the ^{152}Eu lines. The agreement is excellent.

Aggregate gamma-ray spectra of ^{235}U fission products have been measured at 19 time intervals spanning the delay time range 0.14s to 15,440s. A number of the spectra are

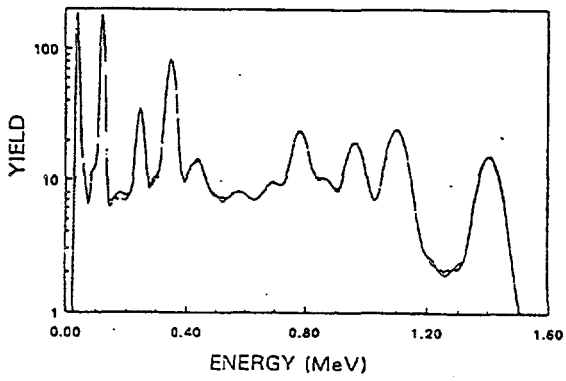

Figure 3. Measured and response-function fitted ^{152}Eu spectra superimposed.

shown in Fig 4 to demonstrate their statistical quality and the time evolution of their structure. Several of these spectra have been unfolded in a preliminary fashion using FERD-PC with a library of 79 response functions to span the energy region 0.10-8.0 MeV. The resulting energy distributions are shown in Fig. 5 along with summation calculations with code CINDER[4] using the ENDF/B-VI fission product data base. In general the agreement is good.

III. BETA-PARTICLE ENERGY SPECTRA

The beta spectrometer used in this study is described in the paper by W. Schier et al.[1] Aggregate beta spectra following thermal neutron fission of ^{235}U have been measured at 19 time intervals spanning the delay time range 0.25s to 11,960s. These are the first aggregate beta spectra measurements below a delay time of 2s. The spectra are being currently analyzed with the response function unfolding program FERD-PC to extract the true beta-particle energy distributions. A preliminary analysis of a few spectra using 70 response functions to span the region E_β=0.2-8.0MeV is shown in Fig. 6. Also shown on the figure are results from an earlier study by Dickens et al.[5](ORNL) and also summation calculations performed with CINDER using the ENDF/B-VI fission production data base.

The present measurements are in fair agreement with the ORNL data but show considerable discrepancy with ENDF at higher beta energies for the two shorter delay times. Subsequent refinements in the spectrometer gating electronics incorporated into our recent study[1] of ^{238}U have yielded beta spectra in excellent agreement with those of

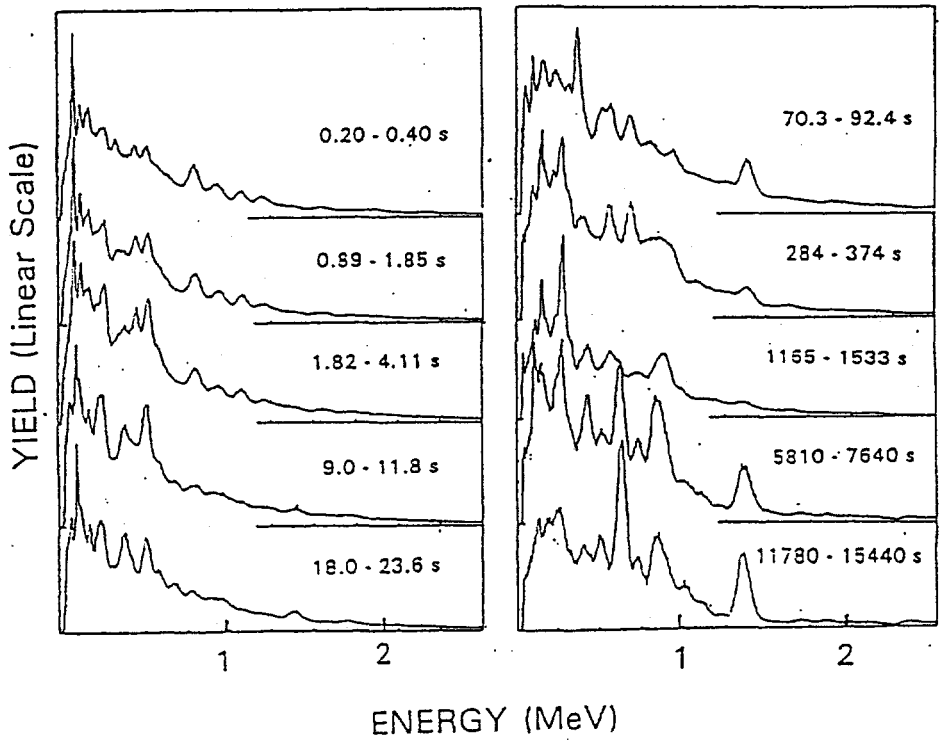

Figure 4. The 0-2.5MeV portions of measured aggregate gamma-ray spectra following thermal neutrons fission of ^{235}U.

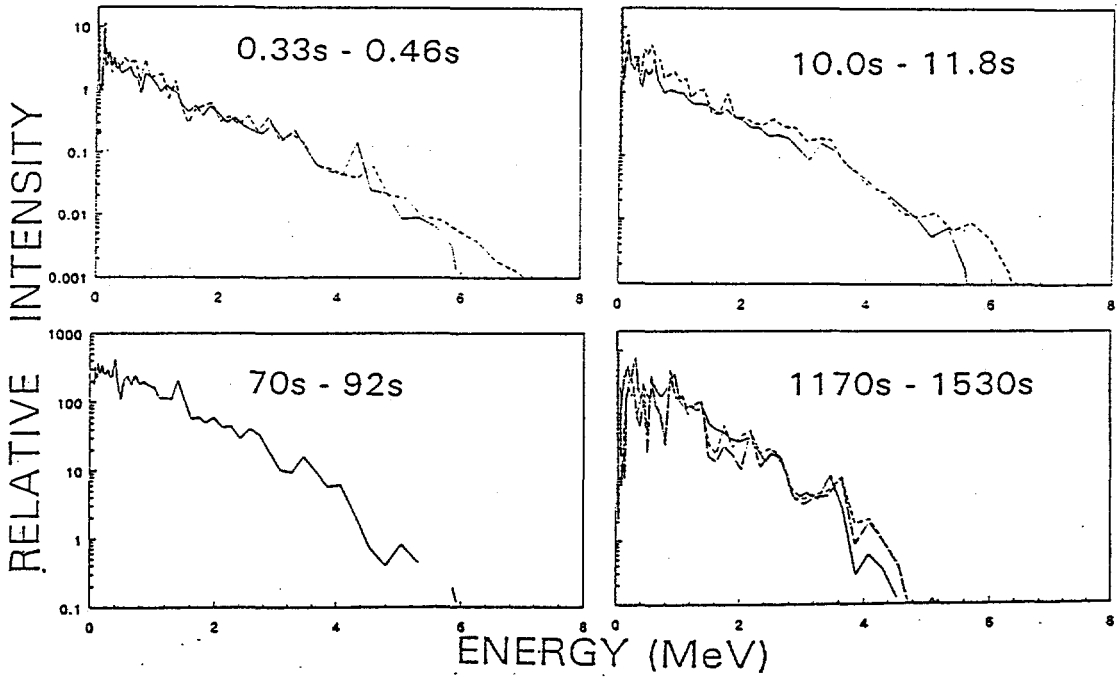

Figure 5. Our gamma-ray energy distributions (solid lines) at selected delay times following $^{235}U(n_{th},f)$ compared with ENDF/B-VI (dashed lines).

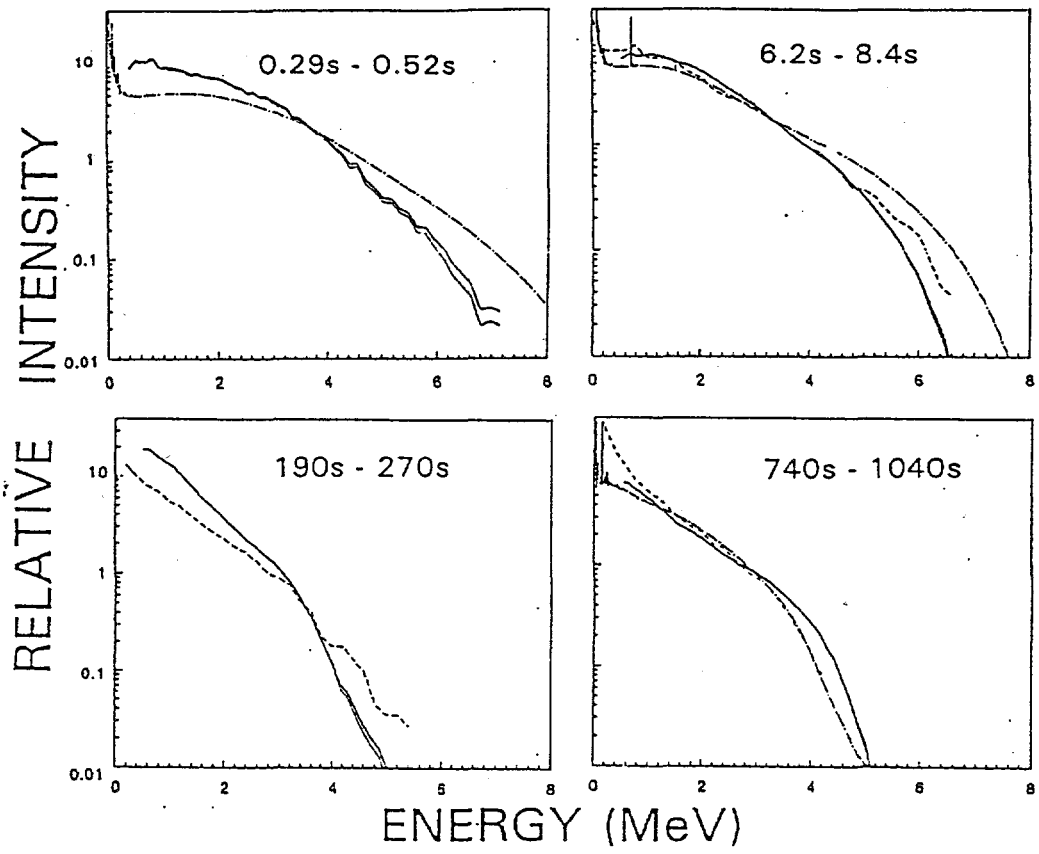

Figure 6. Our beta energy distributions (solid) following $^{235}U(n_{th},f)$. The dashed curves represent earlier ORNL studies[5] at similar delay times. ENDF/B-VI based summation results are the dash-dot curves.

ORNL. We plan to repeat the present ^{235}U beta measurements in anticipation of equally good agreement with the ORNL data to provide confidence in measurements at short delay times where no data exists.

IV. CONCLUSIONS

Both gamma-ray and beta-particle energy spectra have been measured from the aggregate fission products of ^{235}U as a function of time after fission. This is the first study to determine both beta and gamma spectra at times less than 1s after fission, where summation calculations based on individual fission-product evaluations such as ENDF have their largest uncertainties. Preliminary results show good agreement between the measured gamma-ray spectra and those obtained from summation calculations using the ENDF/B-VI fission-product data base. The beta spectra show considerably more disagreement for short times after fission, especially at higher beta energies.

REFERENCES

1. W.A. Schier et al., paper at this Conference.

2. G. Rudstam and T. England, *Test of Pre-ENDF/B-VI Decay Data and Fission Yields*, LA-11909-MS(1990).

3. B.W. Rust, D.T. Ingersoll and W.R. Burrus, *A User's Manual for the FERDO and FERD Unfolding Codes*, ORNL/TM-8720 (1983).

4. T.R. England, R. Wilczynski and N.L. Whittemore, *CINDER-7: An Interim Users Report*, Los Alamos National Laboratory report LA-5885-MS (April 1975).

5. J.K. Dickens, J.F. Emery, T.A. Love, J.W. McConnell, K.J. Northcutt, R.W. Peele and H. Weaver, *Fission-Product Energy Release for Times Following Thermal-Neutron Fission of ^{235}U between 2 and 1400 Seconds*, ORNL/NUREG-14(1977).

References

1. J.K. Dickens, *Fission Product Decay Heat for Thermal Reactors*, Proc. Int. Conf. Nuclear Cross Sections for Technology, Knoxville, TN (1979) 25-33.

2. G. Rudstam and T. England, *Test of Pre-ENDF/B-VI Decay Data and Fission Yields*, LA-11909-MS (1990).

3. K. Takahashi and M. Yameda, Prog. Theor. Phys. **41**, 1470 (1969).

4. T. Yoshida and J. Katakura, Nucl. Sci. Eng. **93**, 193 (1986).

5. J. Katakura and T. England, *Augmentation of ENDF/B Fission Product Gamma-Ray Spectra by Calculated Spectra,* to be published by Los Alamos National Laboratory.

6. G.P. Couchell, J.M. Campbell, S. Li, H.V. Nguyen, D.J. Pullen, E.H. Seabury and S.V. Tipnis and T.R. England, *A Study of Gamma-Ray and Beta-Particle Decay Heat Following Thermal Neutron Induced Fission of ^{235}U*, Proc. Int.l Conf. on Nucl. Data for Science and Technology, Gatlinburg, TN (1994), to be published.

7. W.A. Schier, J.M. Campbell, G.P. Couchell, S. Li, H.V. Nguyen, D.J. Pullen, E.H. Seabury and S.V. Tipnis and T.R. England, *Energy Distributions of Gamma and of Beta Decay Heat as Function of Decay Time for ^{238}U*, Proc. Int.l Conf. on Nucl. Data for Science and Technology, Gatlinburg, TN (1994), to be published.

8. D.J. Pullen, J.M. Campbell, G.P. Couchell, S. Li, H.V. Nguyen, W.A. Schier, E.H. Seabury, S.V. Tipnis and T.R. England, *High Resolution Gamma-Ray Spectra for $^{235}U(n_{th},ff)$*, Proc. Int.l Conf. on Nucl. Data for Science and Technology, Gatlinburg, TN (1994), to be published.

9. K. Takahashi and M. Yamada, Prog. Theor. Phys. **41** (1969) 1470; S. Koyama, K. Takahashi and M. Yamada, Prog. Theor. Phys. **44** (1970) 633; K. Takahashi, Prog. Theor. Phys. **45** (1971) 1466.

10. G. Rudstam and T. England, *Test of Pre-ENDF/B-VI Decay Data and Fission Yields*, LANL Report LA-11909-MS (1990).

11. J. Katakura and T. England, *Augmentation of ENDF/B Fission Product Gamma-Ray Spectra by Calculated Spectra*, LANl Report LA-12125-MS (1991).

12. T.R. England, W.B. Wilson and M.G. Whittemore, *CINDER-7: An interim Users Report*, LANL report LA-5885-MS (1975)

13. T.R. England, unpublished

14. J. Campbell, M.S. thesis, University of Massachusetts Lowell (1993)

15. B.W. Rust, D.T. Ingersoll and W.R. Burrus, *A User's Manual for the FERDO and FERD Unfolding Codes*, ORNL/TM-8720, 1983.

16. J.K. Dickens, J.F. Emery, T.A. Love, J.W. McConnell, K.J. Northcutt, R.W. Peele and H. Weaver, *Fission-Product Energy Release for Times Following Fission of ^{235}U Between 2 and 1400 Seconds*, ORNL Report ORNL/NUREG-14 (1977).

17. M. Akiyama, K. Furuta, T. Ida, K. Sakata and S. An, J. At. Energy Soc. Japan, **24** (1982) 709.

18. M. Akiyama and S.An, *Measurement of Fission-Product Decay Heat for Fast Reactors*, Proc. Int. Conf. on Nucl Data for Sci. and Technology, Antwerp (1982) 237.

19. M. Akiyama and J. Katakura, *Measured Data of Delayed Gamma-Ray Spectra from Fissions of ^{232}Th, ^{233}U, ^{235}U and ^{239}Pu by Fast Neutrons, Tabular Data*, Japan Atomic Energy Research Inst. report JAERI-M-88-252 (1988).

20. G. Rudstam, P.I. Johansson, O. Tengblad, P. Aagaard and J. Erikson, *Beta and Gamma Spectra of Short-Lived Fission Products*, Atomic Data and Nuclear Data Tables **45** (1990) 239.

21. *Nuclear Data Sheets*, Vols. 35 - 64, Natl. Nucl. Data Center, Publ. Academic Press Inc., NY.

22. A.C. Wahl, *Nuclear Charge Distribution and Delayed-Neutron Yields for Thermal Neutron-Induced Fission of ^{235}U, ^{233}U, ^{239}Pu and ^{241}Pu and for Fast Neutron-Induced Fission of ^{238}U*, Proc. Spec. Mtg. on Data for Decay Heat Predictions, Studsvik, Sweden, (1987) 9-19.

23. P.J. Statham, Anal. Chem. **49** (1977) 2149

24. F. Janssens and J.P. Francois, Anal. Chem. **63** (1991) 320