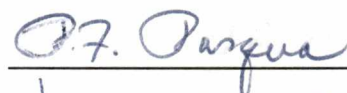
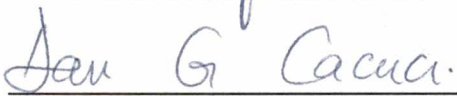
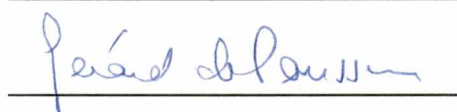


To the Graduate Council:

I am submitting herewith a thesis written by Roger Lawrence Macklin entitled "High Resolution Measurement of the ^{238}U Neutron Capture Yield for Incident Neutron Energies from 1 keV to 100 keV." I have examined the final copy of this thesis for form and content and recommend that it be accepted in partial fulfillment of the requirements for the degree of Master of Science, with a major in Nuclear Engineering.


R. B. Perez, Major Professor

We have read this thesis
and recommend its acceptance:

Accepted for the Council:




Vice Provost
and Dean of The Graduate School

HIGH RESOLUTION MEASUREMENT OF THE ^{238}U NEUTRON
CAPTURE YIELD FOR INCIDENT NEUTRON ENERGIES
FROM 1 keV TO 100 keV

A Thesis
Presented for the
Master of Science
Degree
The University of Tennessee, Knoxville

Roger Lawrence Macklin

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ABSTRACT

The purpose of this work is the precise determination of the ^{238}U capture yield (i.e., the probability of neutron absorption) as a function of neutron energy with the highest available neutron energy resolution. The motivation for the undertaking of such a complex and lengthy task arises from the central role played by the ^{238}U neutron capture process in the calculation of the neutron balance of both thermal reactors and fast breeder reactors. Requirements from the reactor design community for improved understanding and representation of the ^{238}U neutron capture led the Nuclear Energy Agency Nuclear Data Committee (NEANDC) to set up the ^{238}U Task Force to evaluate the available ^{238}U neutron capture data. This NEANDC task force recommended that a new high energy resolution measurement of the ^{238}U capture yield should be performed. With this added incentive, a cooperative program was initiated between the Oak Ridge National Laboratory and the University of Tennessee Nuclear Engineering Department to perform the present measurement using the Oak Ridge Electron Linear Accelerator (ORELA) facility.

The pulsed beam of neutrons from the ORELA facility is collimated on a sample of ^{238}U . The neutron capture rate in the sample is measured, as a function of neutron time-of-flight (TOF), by detecting the gamma rays from the $^{238}\text{U}(n,\gamma)^{239}\text{U}$ reaction with a large gamma-ray detector surrounding the ^{238}U sample. At each energy, the capture yield is proportional to the observed capture rate divided by the measured intensity of the neutron beam. The constant of proportionality (the normalization constant) is obtained as the ratio of

theoretical to experimentally measured areas under small ^{238}U resonances where the resonance parameters have been determined from high resolution ^{238}U transmission measurements. The cross section for the reaction $^{238}\text{U}(n,\gamma)^{239}\text{U}$ can be derived from the measured capture yield if one applies appropriate corrections for multiple scattering and resonance self-shielding.

Use of fast electronics and the 150-meter flight-path station at ORELA resulted in the world's highest energy resolution measurement of the ^{238}U neutron capture yield. This feature of the present work will allow the extension of the resolved resonance region in ^{238}U from its current limit of 4 keV up to 20 keV. In addition, some 200 new ^{238}U neutron resonances in the energy range from 250 eV to 10 keV have been discovered which had not been detected in previous measurements. These results are truly of historic importance in regard to both reactor design and the wealth of information they provide for the understanding of the structure of the ($^{238}\text{U} + n$) compound nucleus.

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CHAPTER I

INTRODUCTION

The first nuclear reactors depended on ^{235}U fission neutrons inducing further fission reactions. For slow or thermalized neutrons with energies below about 5 electron volts (eV), conditions were favorable to this chain reaction, even in natural uranium. As the fission neutrons start near a million eV, ways to get them moderated to low energy without being captured had to be devised. As most of the neutron capture in natural uranium (which is 99.3% ^{238}U) occurs at narrow resonances, the resonance self-shielding effect could be exploited by using relatively thick rods or plates of fuel. The use of low mass nuclei for moderators gave an average energy reduction per neutron scattering greater than the width of most resonances, so a neutron being slowed down in a reactor did not have more than one chance to be captured at a particular resonance.

Modern power reactors use the protons in water for neutron moderation but need slightly enriched uranium ($\approx 2\%$ ^{235}U) to compensate for their non-resonant capture. The nuclear data base used to design reactors (ENDF/B) has included resonance parameters for ^{238}U up to 4 keV based on available experimental techniques. Smoothed or average cross sections from there to about 10 keV have not been adequate to predict the changes in reactivity as the effective broadening of resonances by thermal motion increases with temperature. This Doppler broadening is adequately understood theoretically as a function of temperature but requires individual resonance parameters in the 4- to 10-keV energy range. Requests for such resonance parameter data have

been given the highest priority by representatives of the United States, the United Kingdom and the USSR.

Though the ^{238}U capture cross section is known to $\pm 5\%$ between 30 and 500 keV,¹ the uncertainty is as large as $\pm 10\%$ between 1 and 30 keV. In particular,

1. The spread in the differential data is greater than expected from the assigned errors, especially below 30 keV.
2. The differential data do not meet the accuracy of $\pm 3\%$ or less required by the designers² of liquid-metal fast-breeder reactors (LMFBRs) and high-temperature gas-cooled reactors (HTGRs).
3. Integral measurements in fast reactors imply values of effective ^{238}U capture cross sections which are $\approx 10\%$ lower than the values that are calculated from the best estimates based on differential data.

These inconsistencies in the ^{238}U capture cross section, which were discussed at the Antwerp Conference on Nuclear Data for Science and Technology in September 1982, led to the formation of the ^{238}U Task Force for the Nuclear Energy Agency Nuclear Data committee (NEANDC).

The ^{238}U Task Force³ was set up to deal with two problems: (1) disagreement among the neutron widths of the resolved resonances above 1.4 keV (derived from transmission measurements at several laboratories), and (2) the ^{238}U capture cross section in the resolved and unresolved resonance regions. Among the most relevant conclusions arrived at by the NEANDC Task Force were that most of the capture cross section measurements in the lower energy range (≈ 1 to 30 keV) were old⁴⁻⁸ and that they disagreed among themselves by more than

expected from their respective error assignments.⁹⁻¹¹ To cope with these longstanding discrepancies, the ^{238}U Task Force³ issued the following recommendations:

1. Reanalysis of the old ^{238}U capture data would be impossible, hence new measurements are required.
2. A high resolution capture cross section measurement (≈ 150 m in flight path) is urgently needed.
3. Previous inconsistencies in the normalization of the capture cross section in the 1- to 30-keV neutron-energy range must be resolved.
4. Extension of the resolved resonance energy range to higher energies. This would allow a consistent analysis of high resolution transmission and capture data in the energy region between 1 and ≈ 10 keV.

In response to these requests, a measurement of the ^{238}U capture yield was performed at the 150-meter flight-path station of the Oak Ridge Electron Linear Accelerator (ORELA) facility.¹² The present experiment has utilized ORELA as a powerful pulsed neutron source which, combined with the long (150-m) flight path, allows achievement of adequate energy resolution up to at least 20 keV. To get good statistical accuracy with low backgrounds, the large liquid scintillator tank (ORELAST) with an internal septum was used as a detector for capture gamma rays from the $^{238}\text{U}(n,\gamma)^{239}\text{U}$ reaction. The septum in the tank provides isolation of the signals from the left and right halves of the capture gamma-ray detector. Electronically requiring a time coincidence between the two halves drastically reduces the background from

single ionizing events, yet it still leaves the response to capture gamma-ray cascades adequately high.

The content of this thesis is organized as follows. Chapter II describes the principle of the measurement and gives detailed information on the experimental arrangement such as the detectors and electronic set-up. The methodology used for the analysis of the experimental data and the uncertainty analysis is described in Chapter III. The capture yield results and comparisons with other measurements are given in Chapter IV. The theoretical interpretation of the data is discussed in Chapter V. Chapter VI presents the conclusions derived from the present work and discusses future work in the area of ^{238}U capture yield measurements.

CHAPTER II

DESCRIPTION OF THE EXPERIMENT

2.0 INTRODUCTION

This chapter describes the experimental setup, methods, and instrumentation used for the determination of the ^{238}U capture yield.

2.1 PRINCIPLE OF THE MEASUREMENT

A pulsed beam of neutrons is collimated on a sample of ^{238}U . The neutron capture rate in the sample is measured, as a function of neutron time-of-flight (TOF), by detecting the gamma rays from the $^{238}\text{U}(n,\gamma)^{239}\text{U}$ reaction with a large gamma-ray detector surrounding the ^{238}U sample. At each energy, the capture yield is proportional to the observed capture rate divided by the measured intensity of the neutron beam. The constant of proportionality (the normalization constant) is obtained as the ratio of theoretical to experimentally measured areas under small ^{238}U resonances where the resonance parameters have been determined from high resolution ^{238}U transmission measurements.¹³ The cross section for the reaction $^{238}\text{U}(n,\gamma)^{239}\text{U}$ can be derived from the calculated capture yield if one applies appropriate corrections for multiple scattering and resonance self-shielding.

2.2 EXPERIMENTAL ARRANGEMENT

The Oak Ridge Electron Linear Accelerator (ORELA)¹⁴ was used to produce a pulsed source of neutrons. Pulses of approximately 140 MeV electrons of 12-A peak intensity impinge on a water-cooled tantalum

target where they produce intense bremsstrahlung gamma rays which, in turn, generate neutrons by the (γ, n) reaction. Some of these neutrons are slowed down to the energies of interest by water surrounding the target assembly. This water is canned in a 2.4-mm-thick beryllium housing. The target-moderator assembly¹⁵ is shown in Figure 2-1.

The capture gamma-ray detector was positioned in the 150-m station of flight path 6 of ORELA. This flight path is normal to the surface of the moderator. The ^{238}U sample was placed at the center of the capture gamma-ray detector at a nominal distance of 151.96 m from the neutron source. The incident neutron detector was located between the ^{238}U sample and the neutron source at a nominal distance of 150.86 m from the latter.

The neutron collimator system of flight path 6 has been described in detail by Silver et al.¹⁶ and is shown in Figure 2-2. The collimators are made of copper and lead to avoid the energy degradation caused by neutron scattering in lighter materials such as boron, lithium, or paraffin. The neutron beam is collimated to a 7.63-cm-diameter umbra at the sample position with an 8.16-cm-diameter penumbra. A shadow bar, rectangular in cross section and comprised of 61 cm of copper followed by 16 cm of lead, was positioned 2.5 cm from the ORELA target to shield the ^{238}U sample from the bremsstrahlung radiation generated by the electrons impinging upon the tantalum target. This bar considerably reduces the "gamma flash" due to the gamma rays and unmoderated neutrons arriving at the detector a few microseconds after the electron burst hits the target.

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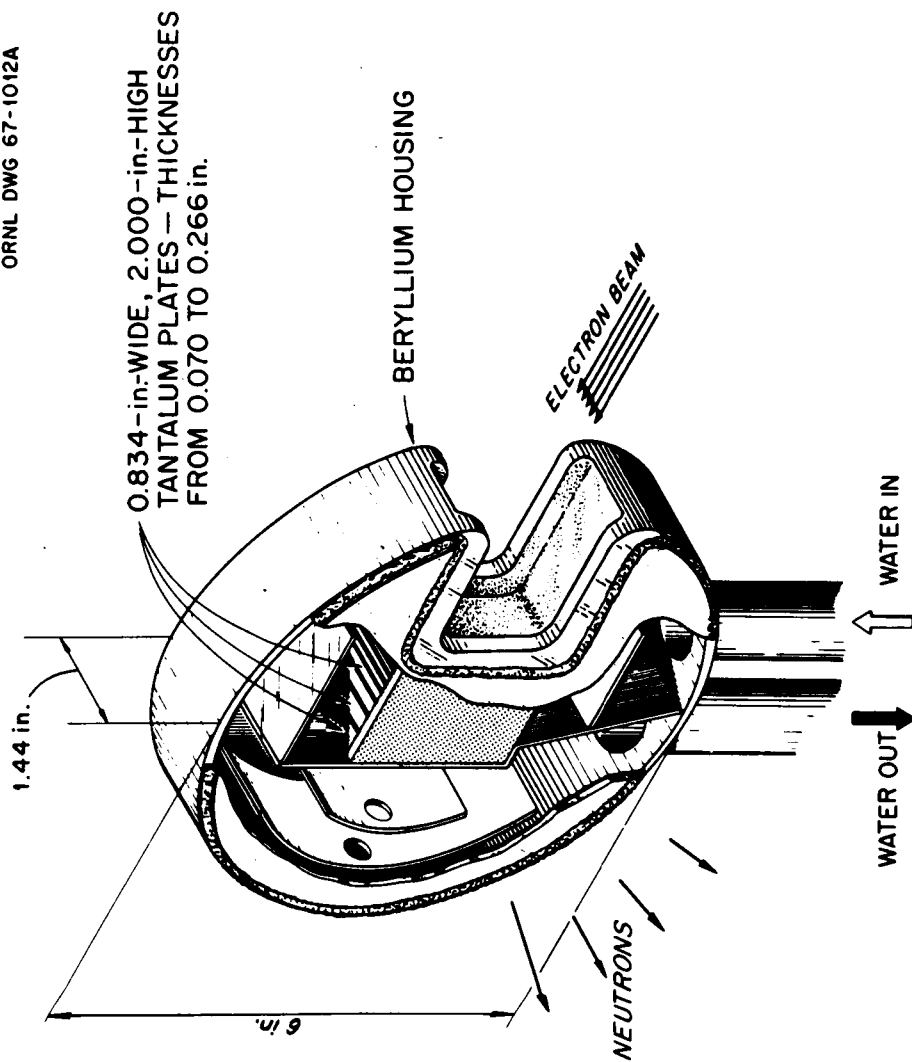


Figure 2-1. The High Power (50 kW) Water-Cooled Tantalum Neutron Target and Moderator Assembly for ORELA.

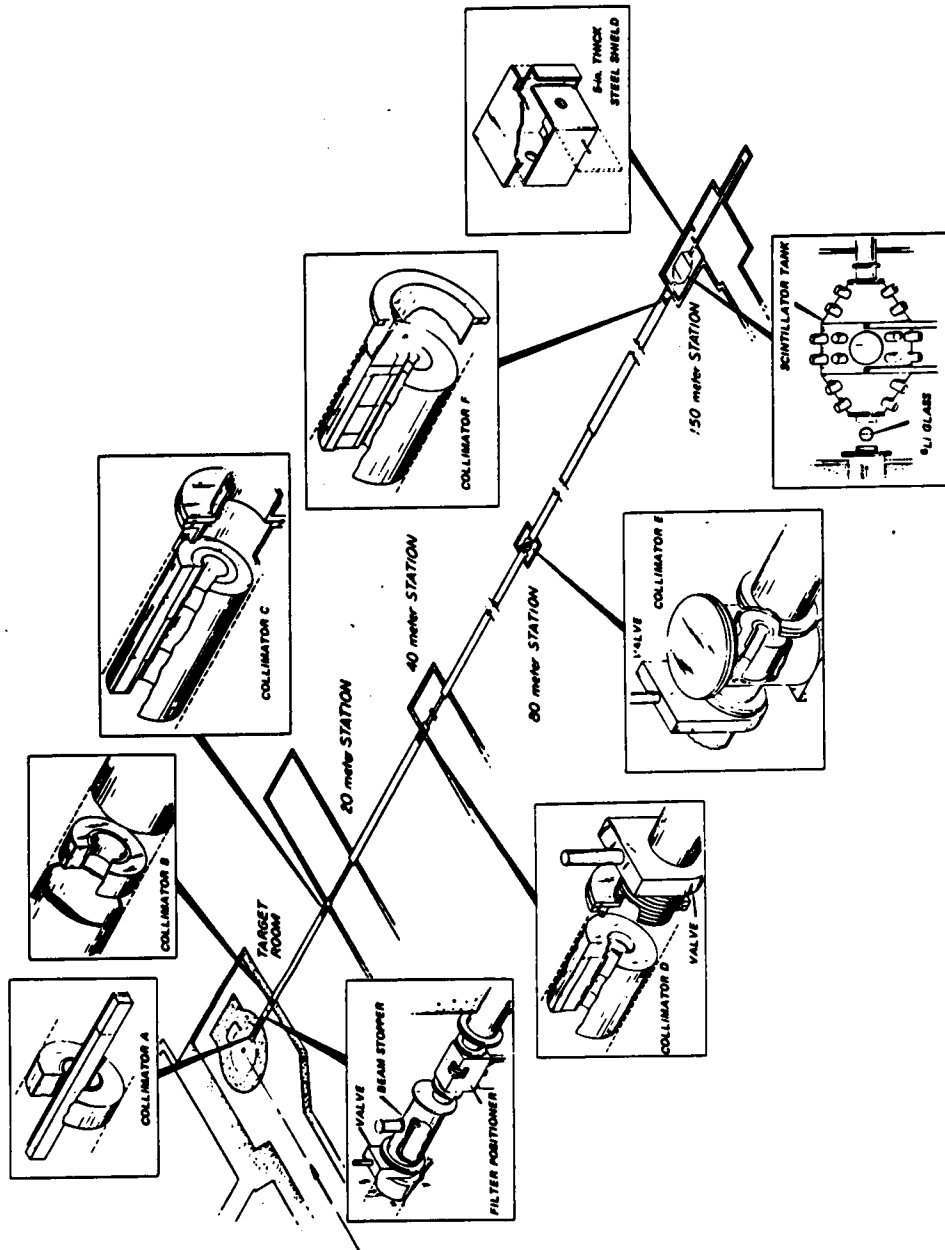


Figure 2-2. The Collimation System for the 150-meter Station of Flight Path 6 at the ORELA Facility, Showing in Particular the Beam Stopper, the Filter Positioner, and the Copper and Lead Collimators.

2.3 THE DETECTOR

The ORELA liquid scintillator tank (ORELAST)¹⁶ is located on a flight path of 150 meters and serves as the detector for capture gamma rays in this measurement of the ^{238}U capture yield. A sketch of this detector is shown in Figure 2-3. The tank contains approximately 3000 liters of solution, consisting of three parts commercial liquid scintillator¹⁷ (NE-224) to one part trimethylborate. The function of the trimethylborate additive is to reduce background counts arising from the 2.22-MeV capture gamma ray in hydrogen. Thus, scattered neutrons which are thermalized in the solution can be captured by boron rather than hydrogen. Approximately 96% of the neutrons captured in boron result in a 0.478-MeV gamma ray.

The surface of the ORELAST tank is studded with thirty-two RCA Model 4522 photomultiplier tubes which are positioned to cover equal surface areas of the liquid scintillator. The gain for each photomultiplier tube is individually adjusted to provide optimum overall pulse-height resolution of the detector. The ORELA neutron beam passes through an aluminum tube which traverses the ORELAST tank and in which the ^{238}U sample is centered. A liner composed of lithium hydride (152.6-cm length and 2.5-cm thickness) is placed inside the through-tube which serves to further reduce the background caused by neutrons scattering off the ^{238}U sample and decreases the probability of fast neutrons scattered by the scintillator which might return to the ^{238}U sample.

The ORELAST tank is optically divided into two equal halves by 0.05-mm-thick aluminized-mylar sheets in the vertical plane which

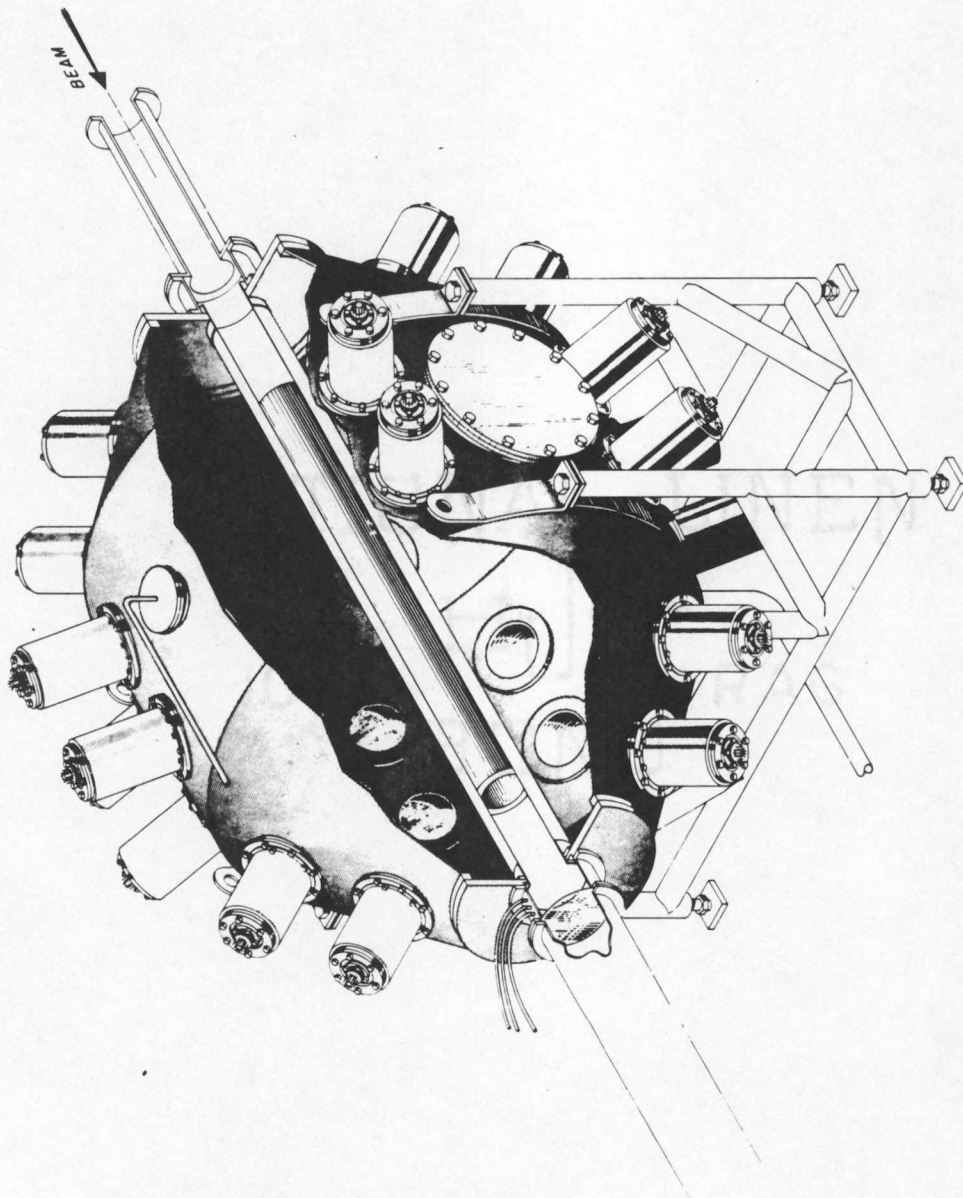


Figure 2-3. Sketch of the ORELA Liquid Scintillator Tank (OREFAST). This tank is viewed by thirty-two 12.7-cm-diameter RCA Model 4522 photomultiplier tubes. It was separated into two equal sides with an aluminized mylar septum in the vertical plane containing the beam axis. This septum is not shown in the figure.

contains the neutron-beam axis. Requiring coincidence between the signals from the two halves of the tank provides an approximately eight times higher signal-to-background ratio than could be obtained using "singles" mode. This takes advantage of the high multiplicity of gamma rays from neutron capture in the ^{238}U sample at the center of the tank, whereas the background is in a large measure composed of single gamma rays which have a lower probability of being seen in both halves of the tank.

The ^{238}U samples used were one of 0.635-mm nominal thickness (65.47 grams in weight and 8.26-cm mean diameter), and a second of 2.54-mm nominal thickness (473.70 grams in weight and 11.11-cm mean diameter). A particular sample is placed in a light aluminum holder and positioned at the center of the ORELAST tank inside the lithium-hydride liner. Both ends of the tank are sealed with mylar windows, and argon gas is passed continuously through the through-tube (because argon scatters fewer neutrons than air).

The neutron flux is measured by a ^6Li glass scintillator¹⁸ positioned just "upstream" of the ORELAST tank. This detector is a disk of 83 mm diameter and 1 mm thickness (Nuclear Enterprises Co., type NE-912) mounted in a light-tight housing which is viewed by two RCA Model 4522 photomultiplier tubes.

2.4 THE ELECTRONICS

A simplified block diagram of the electronics system is shown in Figure 2-4. The fast signals from the anodes of 16 photomultiplier tubes are summed and then amplified for each side of the tank. The outputs from these modules are coupled to separate constant fraction

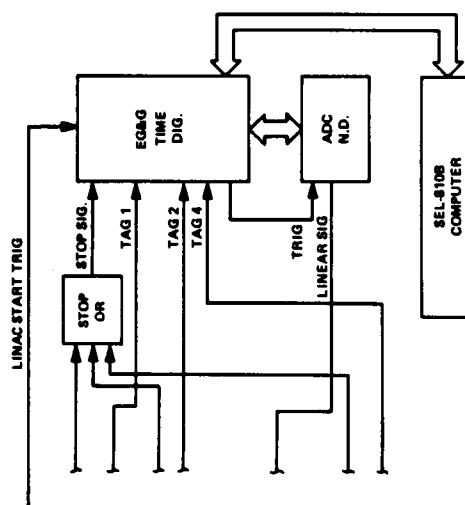
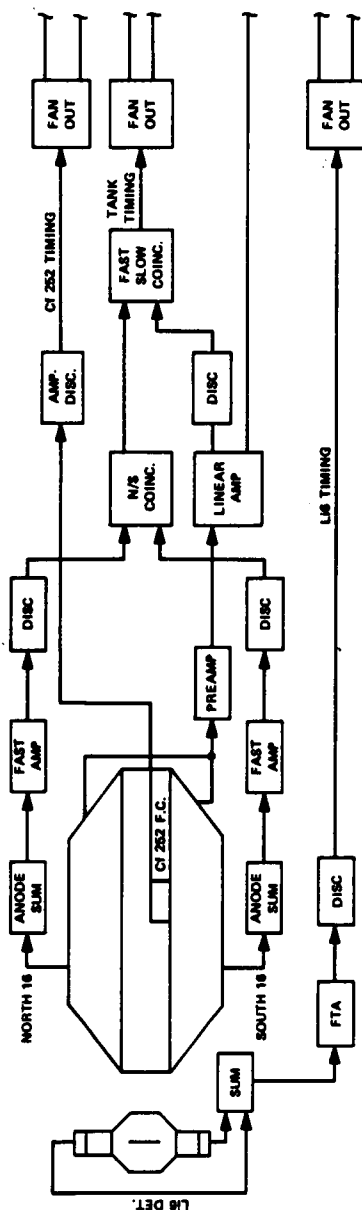


Figure 2-4. Simplified Block Diagram of the Electronic Circuitry Used in the Measurements of the ²³⁸U Capture Yield. The time-of-flight spectra were accumulated on a fast random access disk linked to the SEL Model 810-B computer and later unloaded on magnetic tape and analyzed on a DEC Model VAX 11/785 computer by batch process.

discriminators which are then fed to a coincidence circuit which produces an output only when both sides of the tank detect the capture gamma rays.

The linear signal from the tank comes from the dynodes of the photomultiplier tubes, all thirty-two of which are summed together. This signal is processed by an electronic preamplifier and a Tennelec model TC-200 linear amplifier. The output from this amplifier is fed to the data-acquisition system in the data-processing lab and to a local cross-over discriminator. This discriminator determines the gamma-ray pulse-height energy range over which the measurement is conducted (in this case, from 2.4 MeV to 10.4 MeV).

Signals from the "fast" coincidence circuit and the "slow" cross-over discriminator are coupled to a coincidence circuit. The "fast" signal determines the timing resolution, and the "slow" signal determines the gamma-ray energy range. The output from this coincidence circuit is presented to the data-acquisition system as a timing pulse.

The timing signal from the ${}^6\text{Li}$ glass neutron-flux detector is derived in a similar manner. Two RCA model 4522 photomultiplier tubes, one at either side of the lithium glass scintillator and well clear of the neutron beam, view the light output induced by the alpha particle and triton of the ${}^6\text{Li}(n,\alpha)\text{T}$ reaction. Anode signals are processed by fast sum and discriminator circuits. The discriminator bias window is set just below and just above the peak from the ${}^6\text{Li}(n,\alpha)\text{T}$ reaction. (Both the alpha particle and the tritium nucleus produce light output since both are charged particles.)

A small ^{252}Cf fission chamber with a fission count rate of 65 per second is also placed in the lithium hydride through-tube liner slightly "downstream" of the ^{238}U sample but below the neutron beam. The fission fragment signal from the amplifier-discriminator circuit is used to monitor continuously the tank timing and gain. The full-width-at-half-maximum (FWHM) of the time spectrum between the ^{252}Cf and the scintillator tank is approximately 6 nanoseconds.

The timing and pulse-height information are processed simultaneously in the data-acquisition lab. The neutron time-of-flight is measured using an EG&G model TDC-100 multistop time digitizer. The start trigger for this device is derived from the linear accelerator (LINAC) gamma flash. Pulses from the scintillator tank, the neutron flux monitor, and the ^{252}Cf circuits are coupled to the TDC stop input.

Scintillator tank pulse-height information is derived from a Nuclear Data model 560 analog-digital converter (ADC). The binary information from the ADC and TDC are combined in the time digitizer and fed to a Systems Engineering Laboratories (SEL) model 810-B digital computer which stores the information (on disk).

The separate detector signals are presented to "tag" inputs in the TDC, allowing the computer to identify each signal.

2.5 THE DATA ACQUISITION SYSTEM

Binary information representing neutron time-of-flight, gamma-ray pulse height, and identification tags are combined to form a 30-bit word for each event which is read by the data-acquisition computer.

The ORELA data-acquisition computer system¹⁹ consists of three SEL model 810-B digital computers used for on-line data acquisition, a peripheral equipment controller (also a SEL 810-B), and a DEC model VAX 11/785 digital computer used for immediate data analysis. Each data-acquisition computer can simultaneously record data from four experimental systems. Data to the computers are handled by a priority multiplexing scheme with continuous input, and storage is accomplished using a fixed-head disk.

The data space available for the three SEL 810-B computers is 350K, 750K, and 1000K sixteen-bit words.

Programs for the data-acquisition computer are stored in core memory when active. The data-acquisition programs and system-monitor program are organized into the ORELA Data-Acquisition Monitor System.²⁰ The experimenter's data-acquisition program is commonly called a "crunch routine." Program constants that determine time-channel width, pulse-height conversion gain, and multiparameter storage are entered to the program by a card reader or teletype. Input data for the multiplexer are passed through the "crunch" table for sorting before updating a specific address on the disk.

2.6 MULTIPARAMETER TOF-PULSE-HEIGHT SYSTEM

For each sample filter configuration, five TOF spectra were simultaneously recorded into five pulse-height bins. Table 2-1 shows the pulse-height ranges (in MeV) used for this purpose, and Table 2-2 gives the time-of-flight program which applies to each pulse-height bin, for an accelerator repetition rate of 800 pulses per second. The

pulse-height spectrum was calibrated by measuring the gamma-ray pulse-height spectrum of a standardized ^{60}Co source.

Table 2-1. Bias Levels for the TOF-Pulse-Height Multiparameter System

TOF Section	Pulse-height Range (MeV)
1	0.00 - 1.86
2	1.86 - 2.79
3	2.79 - 4.65
4	4.65 - 6.51
5	6.51 - 8.33

Table 2-2. Time-of Flight Program for Measurement of ^{238}U Capture Yield (for Accelerator Repetition Rate of 800 Pulses per Second)

Number of Channels	Channel Width (nsec)	Time-of-Flight (μsec)	Energy (keV)
1000	32	0 - 32	2500 - 136.409
25000	4	32 - 132	136.409 - 7.168
10000	8	132 - 212	7.168 - 2.743
10000	16	212 - 372	2.743 - 0.883
10000	32	372 - 692	0.883 - 0.254

Figure 2-5 shows pulse-height data for the gamma-ray spectra of a ^{252}Cf source.

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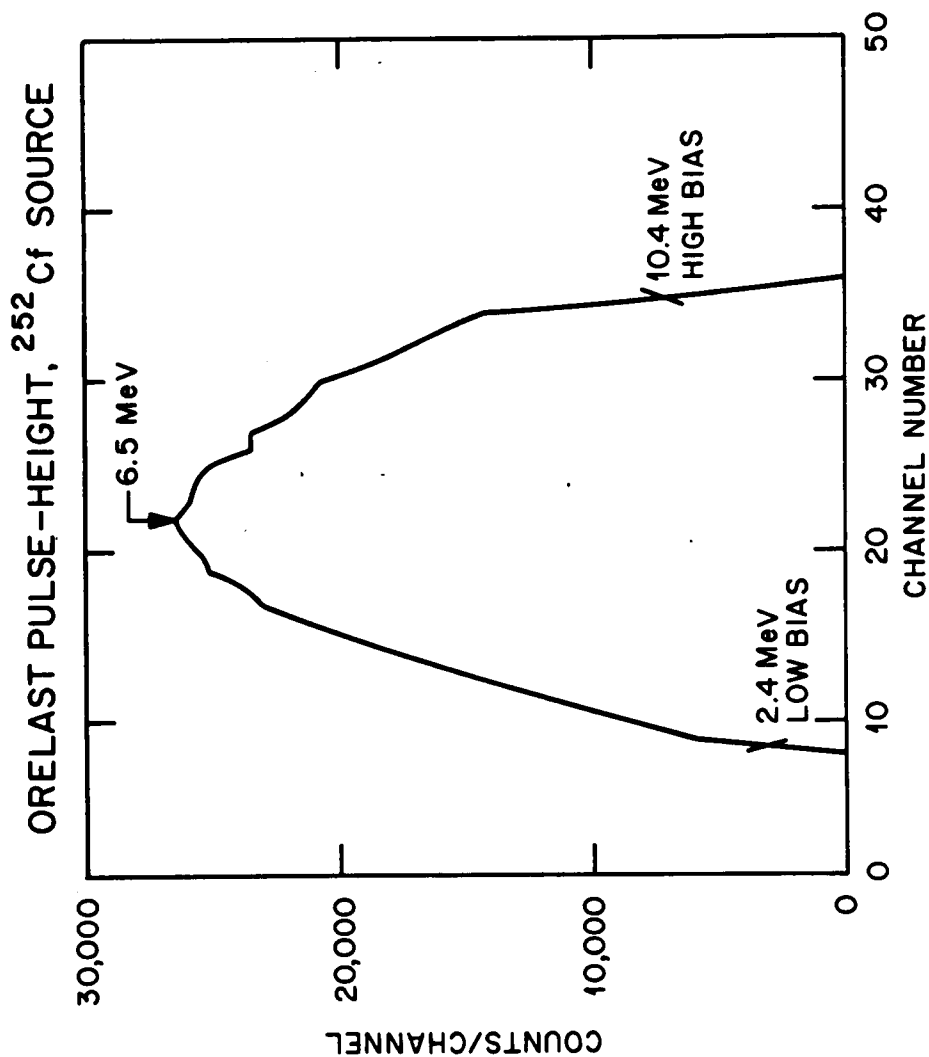


Figure 2-5. Californium-252 Source Pulse-Height Spectrum Taken with the ORELAST Detector, Showing Count Rate Versus Channel Number. The low bias and high bias energies, in MeV, were determined by comparison with the known gamma-ray energies from a standardized ^{60}Co source.

CHAPTER III

ANALYSIS OF THE DATA

3.0 INTRODUCTION

The purpose of the analysis of the data is to obtain the capture yield, $Y_c(E)$ (i.e. the probability of neutron absorption), as a function of neutron energy for each of the ^{238}U samples used in this measurement. The capture yield is defined as a function of the neutron energy, E , by the relation

$$E^{1/2} Y_c(E) = a_n \left[\frac{C(E)}{S(E)} \right] R(E) (b \cdot \text{eV}^{1/2}) \quad (3.1)$$

where

$C(E)$ = background subtracted count rate from the ORELAST detector,

$S(E)$ = background corrected count rate from the lithium glass neutron flux monitor,

$R(E)$ = a factor which accounts for the deviations of the $^6\text{Li}(n,\alpha)$ cross section from $\langle\langle 1/v \rangle\rangle$ behavior,

a_n = normalization constant.

Five measurements of the gamma-ray TOF spectrum were performed for each of the two ^{238}U samples. Measurements were done with and without aluminum filters in the neutron beam, at LINAC repetition rates of 800 and 400 pulses per second, to determine the energy-dependent background which arises from neutron-pulse overlap and room return. The fifth measurement was performed using a polyethylene filter in the beam to determine the energy independent ("constant")

component of the background. A ^{10}B filter was used in all runs to reduce low-energy (< 250 eV) neutrons from the "tail" of the previous pulse (pulse "overlap"). The successive steps in the data analysis to obtain the capture yield are discussed in detail below.

3.1 DEAD-TIME CORRECTION

All time-of-flight spectra were taken simultaneously using the same digital time-interval counter. Thus the dead-time correction is the same for all TOF spectra for any given time-of-flight channel. However, this dead-time correction does not cancel in the ratio of the counts from the gamma-ray detector to those from the neutron-flux monitor (see Eq. 3-1) because the energy corresponding to a given time-of-flight channel is not the same for the two detectors. A fixed dead-time of 32.768 microseconds (larger than the dead-time of any of the electronic components) was artificially imposed on the equipment. This arrangement allowed a straightforward calculation of the dead-time correction which was typically of the order of 1 to 2% and never exceeded 12%.

3.2 ENERGY CALIBRATION AND EXPERIMENTAL RESOLUTION

The conversion of measured time-of-flight, τ , to neutron energy is performed by the relation (neglecting relativistic effects)

$$E = \mu^2 \frac{L^2}{(\tau + \tau_0)^2} \text{ (eV)} \quad (3-2)$$

where

- μ = $72.300 \text{ (eV)}^{1/2}(\mu \text{ sec})/\text{m}$,
 L = neutron flight path (m), and
 τ_0 = initial delay of the time-of-flight "clock" due to delays in the electronics chain.

Rather than measuring L and τ_0 , these two parameters were obtained by solving Eq. 3-1 for pairs of resonances of known energies in the uranium and aluminum nuclei. The resulting energy scales were then realigned with the precise energy calibration of Olsen et al.¹³ Table 3-1 gives the values for L and τ_0 , for each sample thickness, derived from this realignment.

Table 3-1. Flight Path Distance and Initial Clock Delay for the ^{238}U Capture Yield Data as Derived from Alignment with Energy Scale of Olsen et al. (Reference 13)

Sample (atoms/barn)	Flight Path Distance, L (meters)	Initial Clock Delay, τ_0 (microseconds)
0.01236	151.939	-2.256
0.00309	151.947	-2.255

The experimental resolution broadening is well described²¹ by a Gaussian convolution of width given by the expression

$$\Delta^2 = \Delta_D^2 + \Delta_M^2 + \Delta_t^2, \quad (3-3)$$

where Δ_D is the Doppler broadening width, Δ_M represents the broadening effect of the moderator, and Δ_t is the width associated with the timing resolution. These quantities are given by the relations²²⁻²⁵

$$\Delta_D^2 = \frac{4kTE}{A} , \quad (3-4)$$

$$\Delta_M^2 = \frac{2}{3} \left(\frac{s^2 + \ell^2}{L^2} \right) E^2 , \quad (3-5)$$

and

$$\Delta_t^2 = \frac{2}{3} \left(\frac{(\tau_b^2 + \tau_c^2)}{\mu^2 L^2} \right) E^3 , \quad (3-6)$$

where

- k = Boltzmann constant = 8.61781×10^{-5} (eV)(°K),
- T = effective sample temperature = 300°K,
- A = target mass number = 238,
- s = "equivalent" moderator thickness (FWHM) = 2.16 cm,
- ℓ = "equivalent" length corresponding to the neutron slowing-down time in the moderator (cm) = 2.63 cm,
- τ_b = pulse width of the neutron burst (nanoseconds) (typically on the order of 14 nsec), and
- τ_c = channel width (μ sec) (variable, from a minimum of 4 nanoseconds to a maximum of 32 nanoseconds).

With the above parameters one obtains

$$\Delta_D = 2.08 \times 10^{-2} E^{1/2} \text{ (eV)} , \quad (3-7)$$

$$\Delta_M = 1.83 \times 10^{-4} E \text{ (eV)} , \quad (3-8)$$

and

$$\Delta_t = 7.43 \times 10^{-8} \left(196 + \tau^2 \right)^{1/2} E^{3/2} \text{ (eV)} , \quad (3-9)$$

with τ being the channel width in nanoseconds.

Figure 3-1 shows plots of the resolution width versus neutron energy for the present measurement (at a flight-path distance of 150 m). Note that below 6 keV the Doppler broadening provides the largest contribution to the experimental resolution broadening, whereas above 10 keV the timing resolution and the moderator broadening become more important in determining the overall resolution. However, for a flight path of 40 meters (as was used in the 1973 ORELA ^{238}U capture measurement of de Saussure et al.), as shown in Figure 3-2, the resolution width due to the moderator is the dominant contribution below 6 keV. Table 3-2 shows the overall resolution width, Δ (eV), (defined in Eq. 3-3) for these two flight-path distances as well as the partial resolution widths. Note that at neutron energies around 10 keV, the overall resolution width for an experiment with $L = 40$ m is three times the corresponding resolution width achievable at $L = 150$ m, and about half the average level spacing for s-wave resonances in the ($^{238}\text{U} + n$) compound nucleus.

3.3 BACKGROUND SUBTRACTION

The time-of-flight (TOF) spectra obtained from the capture gamma-ray detector are contaminated by two background components--an energy-dependent component and a "constant" background contribution. The energy-dependent background arises from several sources:

1. Beam neutrons generated in previous neutron bursts (pulse overlap),
2. Room return neutrons which reach the ^{238}U sample,

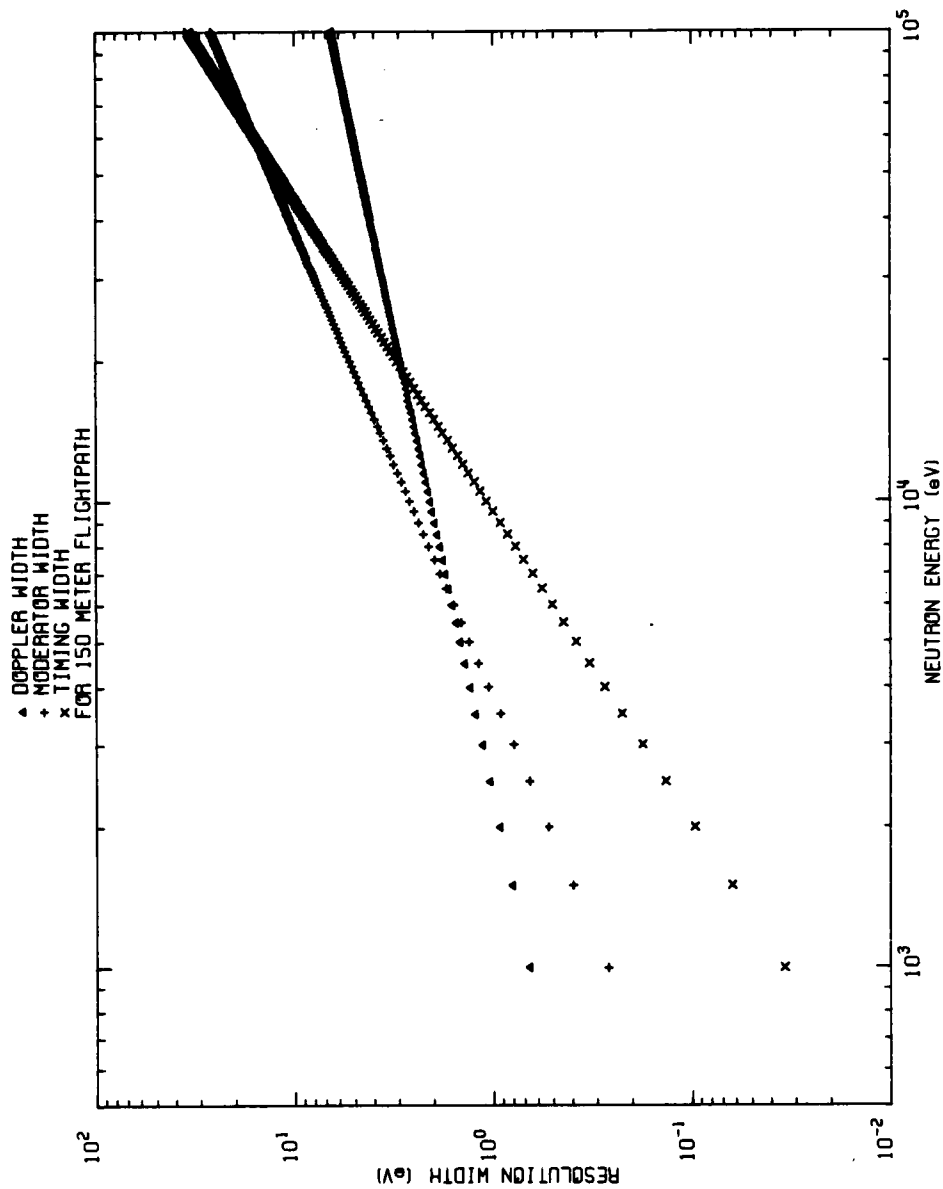


Figure 3-1. Plots of the Calculated Doppler Width, Moderator Width, and Timing Width Versus Neutron Energy for the 150-meter Flight-Path Station.

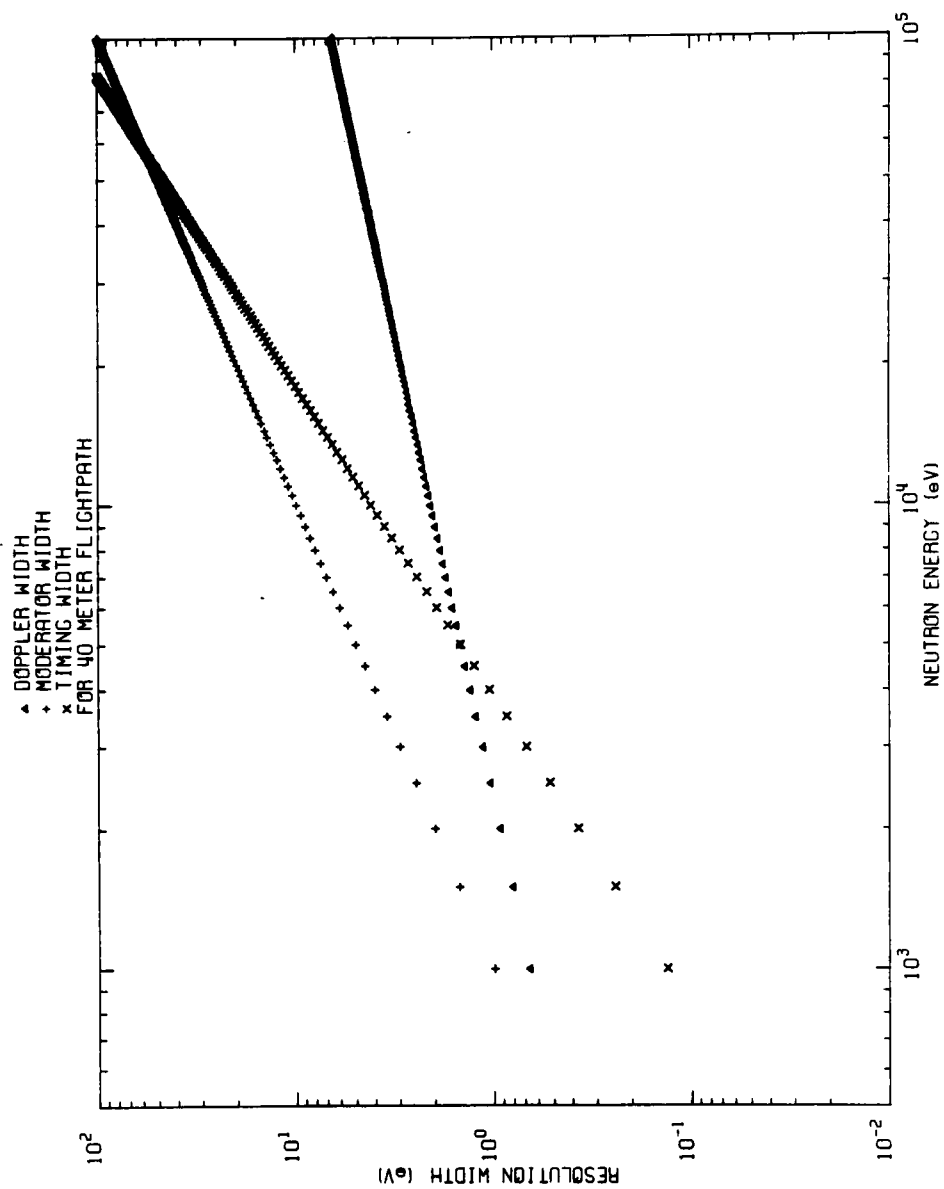


Figure 3-2. Plots of the Calculated Doppler Width, Moderator Width, and Timing Width Versus Neutron Energy for the 40-meter Flight-Path Station.

3. Beam neutrons producing prompt gamma-rays upon capture in materials around the detector.

Table 3-2. Calculated Resolution Widths (eV) for Neutron Flight-Path Lengths of 40 Meters and 150 meters

Partial resolution widths at neutron energy of	1 keV	10 keV	100 keV
Doppler width (eV)			
40 m, 150 m	.660	2.08	6.6
Moderator width (eV)			
40 m	1.00	10.0	100.
150 m	.264	2.64	26.4
Timing width (eV)			
40 m	.094	2.9	94.
150 m	.034	1.08	34.2
Overall resolution width (eV)			
40 m	1.2	10.6	137.4
150 m	.71	3.5	43.7

The first two energy-dependent background sources have the common feature of arising from neutrons generated more than one pulse before detection. Their TOF spectrum, which is proportional to the energy-dependent background, can be obtained by running the pulsed neutron source at half the repetition rate, f_H , of the foreground run. Neutrons taking longer than $2/f_H$, which would also contribute to the background, fall in an energy range such that they are absorbed by the ^{10}B filter (199 g in weight and with areal density of 0.05577 atoms/barn). Hence, for each experimental configuration, two runs are performed: a "high frequency" run at a repetition rate of 800 pulses per second and a "low frequency" run at 400 pulses per second.²⁶ The

third energy-dependent background contribution cannot be measured directly, but it can be minimized by the use of good neutron beam collimation.

The constant background component is produced by two sources: (1) cosmic rays, and (2) long-lived radioactivities in the detector, surrounding materials, and the ^{238}U samples.

The constant background component consisting of muons (generated in the atmosphere by cosmic rays) is considerably reduced through the orientation of the internal aluminized-mylar septum in the ORELAST detector. These muons have predominantly vertical tracks so that the vertical septum is clearly superior to a horizontal septum for rejecting this background component.

To determine the constant background contribution, TOF spectra were measured for each experimental configuration, with a polyethylene plug (four contiguous 2.54-mm-thick pieces) inserted in the neutron beam. Below 100 keV, the polyethylene effectively removes the beam neutrons but is relatively transparent to the gamma-ray background, which includes gamma rays produced by the accelerator ("gamma flash"), and gamma rays created in the water-cooled neutron moderator by the nuclear reaction $\text{H} + \text{n} \rightarrow \text{D} + \gamma$.

3.3.1 Treatment of the Energy-Dependent Background

Figure 3-3 shows TOF spectra for a high-frequency run (upper curve) and a low-frequency run (lower curve). Note that because of the different time-of-flight intervals spanned in these runs, their respective channel structures are also different. The count rate in the low-frequency run, for time-of-flight values above $t_D = \frac{1}{f_H}$,

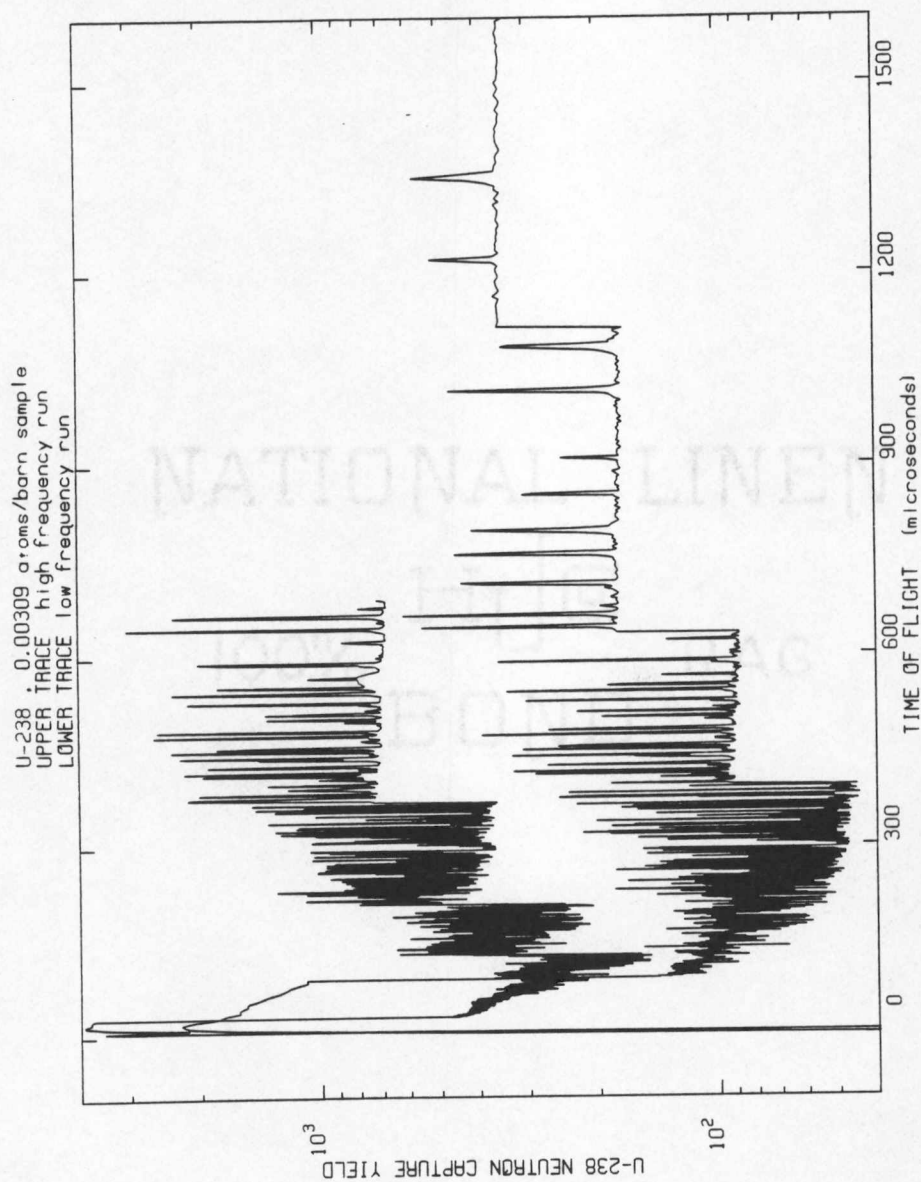


Figure 3-3. Uranium-238 Capture Yield TOF Spectra Versus Neutron Time-of-Flight for the 0.00309 atoms/barn sample. The upper trace is the TOF data for the high-frequency run, and the lower trace is the TOF data for the low-frequency run. The data shown have been averaged over 50-channel intervals. The discontinuities seen in the TOF spectra arise from changes in the channel time-width ("crunch factor").

contains information on the energy-dependent background appearing in the high-frequency run. Calling $E(t_F)$ the neutron energy corresponding to the time-of-flight, t_F , in the high-frequency run, the associated energy background will appear in the low-frequency run at the neutron energy,

$$E^* = E(t_F + t_D) \quad , \quad (3-10)$$

where

$$(t_D = \frac{1}{f_H}) \quad .$$

A program, SHIFT, was written to convert the channel structure of the low-frequency run, in the time interval $0 \leq t \leq t_D$, to the channel structure of the high-frequency run. By introducing a delay of t_D (μsec) in the SHIFT program, the TOF spectrum of the low-frequency run, in the time interval $t_D < t < 2t_D$, was displaced to the time interval, $0 \leq t \leq t_D$, and given the channel structure of the high-frequency run. Figure 3-4 shows the high-frequency run (upper curve) together with the tail of the low-frequency run displaced by the above procedure (lower curve). Note the presence of two broad peaks in the background. The first peak appears at $102.6 \mu\text{sec}$ and the second peak at $560 \mu\text{sec}$. This TOF value corresponds to energies of $E^*(102.6 + t_D) = 66.0 \text{ eV}$ and $E^*(560 + t_D) = 37.2 \text{ eV}$ in the tail of the low-frequency run. Hence, these background peaks must be generated by the 36.7 (eV) and 62.02 (eV) ^{238}U resonances. Indeed the neutrons from the previous ORELA pulse, at the above resonance energies, will contribute counts appearing in the high-frequency runs as broad peaks at energies around $E(202.6) = 11 \text{ keV}$ and $E(560) = 396 \text{ eV}$. The next two lower large ^{238}U

resonances at 22 eV and 6.7 eV do not contribute because the former is outside the time range covered by the high-frequency run and the latter corresponds to neutrons which are absorbed by the ^{10}B filter. This feature of the background, illustrated in Figure 3-4, aids in the fine-tuning of the background subtraction procedure.

Calling HF_i the counts in the i^{th} channel of the high-frequency run, one can write

$$\text{HF}_i = A_i - \left[B_i + B_{o_i} \right] , \quad (3-11)$$

where

- A_i = foreground counts,
- B_i = energy-dependent background counts, and
- B_{o_i} = constant background counts.

Similarly, calling AT_i the counts in the i^{th} channel for the low-frequency run in the time interval $0 < t < t_D$, and BT_i the counts in the i^{th} channel for the displaced tail of the low-frequency run, one obtains

$$\text{AT}_i = A'_i + B'_{o_i} \quad (3-12)$$

and

$$\text{BT}_i = B'_i + B'_{o_i} , \quad (3-13)$$

with

- A'_i = foreground counts (low-frequency run),
- B'_{o_i} = constant background (low-frequency run), and

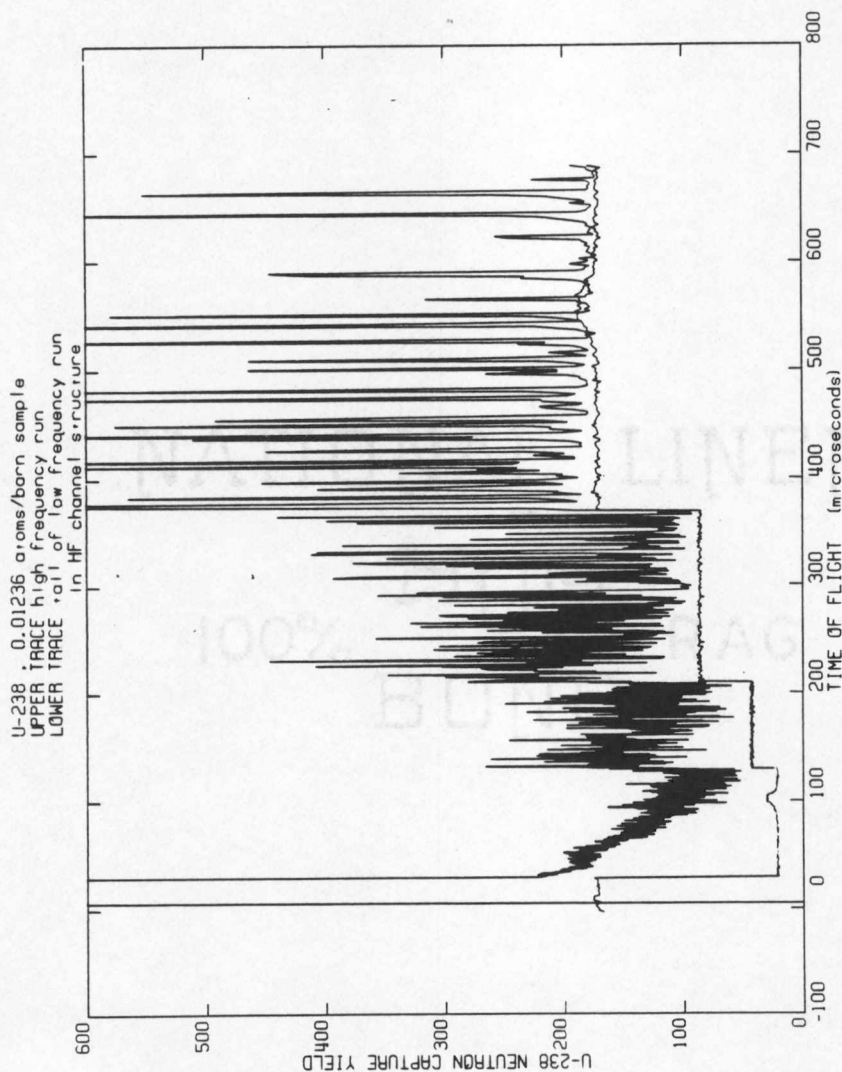


Figure 3-4. Uranium-238 Capture Yield TOF Spectra Versus Neutron Time-of-Flight for the 0.01236 atoms/barn Sample. The upper trace is the TOF data for the high-frequency run. The lower trace is the overlap portion ("tail") of the TOF data for the low-frequency run, which has been placed in the same channel time-width structure ("crunch") as the high-frequency run using the SHIFT program. The data shown have been averaged over 50-channel intervals. The discontinuities seen in the TOF spectra arise from changes in the channel time-width ("crunch factor").

B'_i = energy-dependent background (low-frequency run).

Next the TOF spectrum, TT_i is defined as

$$TT_i = AT_i + BT_i, \quad (3-14)$$

which from Eqs. (3-11) and (3-12) becomes

$$TT_i = A'_i + B'_i + 2B'_{o_i}. \quad (3-15)$$

Comparing Eqs. 3-13 and 3-14 (and in view of the beam dependence of the foreground and energy-dependent background), one must have the proportionality relation

$$A_i + B_i = a_0(A'_i + B'_i), \quad (3-16)$$

where the internormalization ratio is given by the relation

$$a_0 = X/X' \quad (3-17)$$

with

$$X = \sum_{i=I_1}^{I_2} (A_i + B_i) \quad (3-18)$$

and

$$X' = \sum_{i=I_1}^{I_2} (A'_i + B'_i), \quad (3-19)$$

where I_1 and I_2 define a region in the TOF spectrum. However, because of the presence of the yet unknown backgrounds, B_{o_i} and B'_{o_i} , the parameter a_0 cannot be calculated. Rather one has to use the ratio of the known quantities

$$a = \frac{\sum_{i=I_1}^{I_2} HF_i}{\sum_{i=I_1}^{I_2} TT_i} = \frac{a_0 + R}{1 + R'} \quad (3-20)$$

with

$$R = \frac{1}{X_1} \sum_{i=I_1}^{I_2} B_{oi} \quad \text{and} \quad R' = \frac{2}{X_1} \sum_{i=I_1}^{I_2} B'_{oi} \quad (3-21)$$

From the analysis of the runs with the polyethylene plug in the beam, one concludes that the number of constant background counts per channel is a small fraction of the contributions to the count rate arising from the foreground and energy-dependent background. Hence, one can assume that R' is much less than unity, leading to the following approximation to Eq. 3-19:

$$a = a_0 + \Delta_a \quad (3-22)$$

with

$$\Delta_a = R(1 - R') - a_0 R' \quad (3-23)$$

The energy-dependent background corrected TOF spectrum, EBC_i , is then obtained as

$$EBC_i = HF_i - aBT_i \quad (3-24)$$

which from Eqs. 3-3, 3-5, and 3-8 becomes

$$EBC_i = A_i + B_{oi} - aB'_{oi} - \Delta aB'_i \quad (3-25)$$

which shows that there still remains a fraction of the energy dependent background in the TOF spectrum.

3.3.2 Treatment of the Constant Background Component

Let P_i be the counts in the i^{th} channel for the TOF spectrum measurement with the polyethylene plug in the beam which, except for a constant, represents the constant background in the capture detector TOF spectra. To calculate the internormalization constant, b , one selects several regions between resonances in the EBC_i TOF spectrum, such that the foreground component, A_i , can be neglected. Hence, one can rewrite Eq. 3-14 as

$$(EBC_i)_0 = B_{oi} - (aB'_{oi} + \Delta a B'_i) \quad (3-26)$$

The internormalization constant is then given by

$$b = \frac{1}{P} \sum_{i=I_2}^{I_3} (EBC_i)_0 = b_0 - \frac{\Delta a}{P} \sum_{i=I_2}^{I_3} (B'_{oi} + B'_i) \quad (3-27)$$

with

$$b_0 = \frac{1}{P} \sum_{i=I_2}^{I_3} (B_{oi} - a_o B'_{oi}) \quad (3-28)$$

and

$$P = \sum_{i=I_2}^{I_3} P_i \quad (3-29)$$

Note that b_0 would be the correct normalization constant if the correction factor, Δa , would vanish. Because of the relative

smallness of the constant background, the values of P_i fluctuate considerably even though long background runs were taken. Hence, an average of the background, $\langle P_I \rangle$, was taken every 1000 TOF channels, so that the constant background subtracted TOF spectrum was obtained by the relation

$$CBS_i = EBC_i - b\langle P_I \rangle \quad (3-30)$$

with

$$\langle P_I \rangle = \frac{1}{1000} \sum_{i \in I} P_i \quad , \quad (3-31)$$

where I represents a 1000-channel region in the TOF spectrum. Insertion of Eqs. 3-24 and 3-26 into 3-29 yields

$$CBS_i = A_i + \Delta a \left\{ \left[\frac{\langle P_I \rangle}{P} \sum_{i=I_2}^{I_3} B'_{oi} - B'_{oi} \right] + \left[\frac{\langle P_I \rangle}{P} \sum_{i=I_2}^{I_3} B'_i - B'_i \right] \right\} \quad (3-32)$$

where one uses the relation

$$B_{oi} - a_0 B'_{oi} = b_0 \langle P_I \rangle \quad . \quad (3-33)$$

Then Eq. 3-32 can be rewritten as

$$CBS_i = A_i + \Delta a \left\{ \left[\frac{\langle P_I \rangle}{\langle P \rangle} \langle B'_{oi} \rangle - B'_{oi} \right] + \left[\frac{\langle P_I \rangle}{\langle P \rangle} \langle B'_i \rangle - B'_i \right] \right\} \quad (3-34)$$

where one introduces the averages

$$\langle P \rangle = \frac{1}{I_3 - I_2} P \quad , \quad (3-35)$$

$$\langle B'_{o_i} \rangle = \frac{1}{I_3 - I_2} \sum_{i=I_2}^{I_3} B'_{o_i} , \quad (3-36)$$

and

$$\langle B'_i \rangle = \frac{1}{I_3 - I_2} \sum_{i=I_2}^{I_3} B'_i . \quad (3-37)$$

In view of the smoothness of the backgrounds over limited channel regions (certainly true for the constant background), one can assume that

$$\langle P \rangle \approx \langle P_I \rangle , \quad B'_{o_i} \approx \langle B'_{o_i} \rangle , \quad \text{and} \quad B'_i \approx \langle B'_i \rangle .$$

Hence to a very good approximation Eq. 3-34 becomes the desired result

$$CBS_i = A_i . \quad (3-38)$$

As an illustration of the background subtraction methodology, Figure 3-5 shows the TOF spectra of a high-frequency run, the energy-dependent background component, and the background subtracted final result for the 2.54-mm nominal thickness ^{238}U sample, where each has been averaged over 500 TOF channels for clarity.

3.4 DETERMINATION OF THE NEUTRON BEAM SPECTRUM

The shape of the neutron beam energy spectrum, $C(E)$, is obtained from the TOF spectrum, M_i , of the neutrons detected by the ^6Li glass flux monitor. Several factors may affect this TOF spectrum:

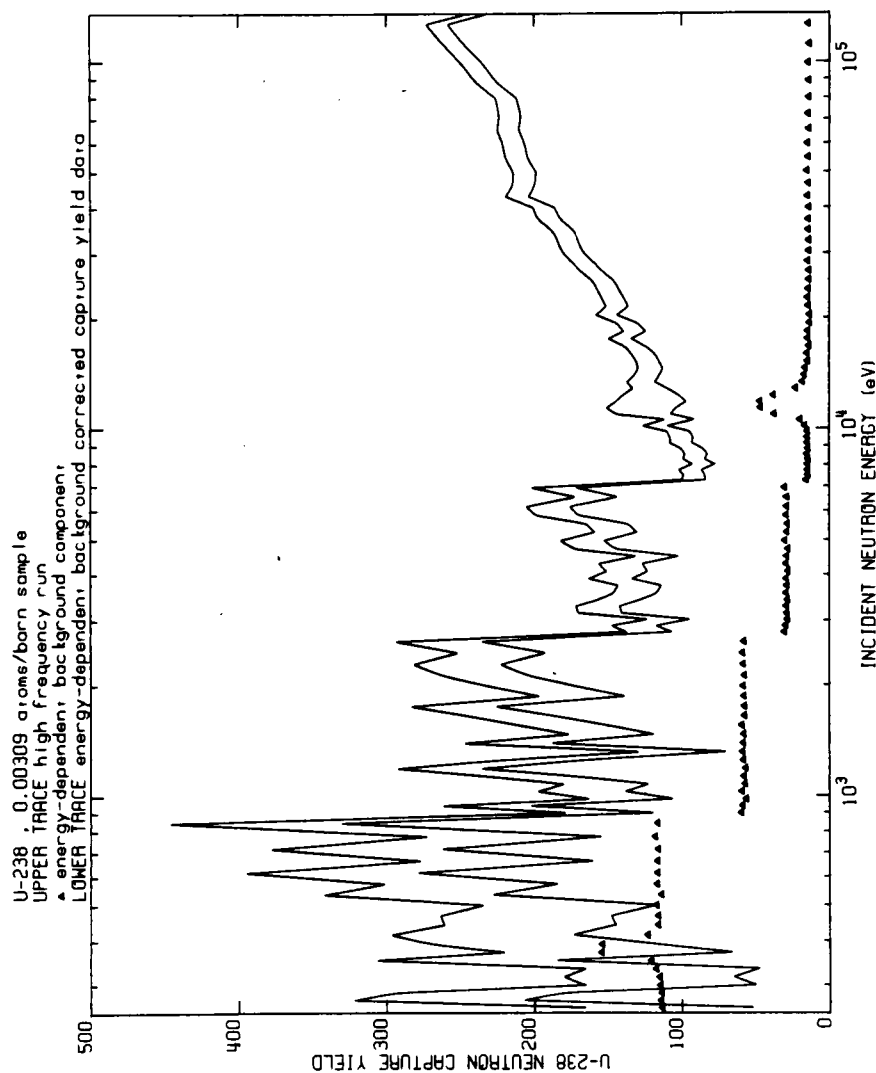


Figure 3-5. Uranium-238 Capture Yield TOF Spectra Versus Neutron Energy for the 0.00309 atoms/barn sample. The upper trace is the TOF data for the high-frequency run. The lower trace corresponds to the high-frequency run TOF data after subtraction of the energy-dependent background which is also shown. Note the broad overlap peaks at 396 eV and 11 keV. The data shown have been averaged over 50-channel intervals. The discontinuities seen in the TOF spectra arise from changes in the channel time-width ("crunch factor").

1. Shape distortion arising from changing conditions at the ORELA target-moderator assembly,
2. Background contamination,
3. Multiple neutron scattering within the ^6Li glass, and
4. Effects of oxygen, silicon, and cerium constituents of the glass.

Shape distortion at the target appears because of changes in the moderator environment (such as steam bubble generation at high-power operation of the ORELA accelerator). To minimize these effects, each sample was assigned its own monitor spectrum so that the same accelerator conditions prevailed for the gamma-ray and monitor-spectra runs. However, a posteriori checks of the ^6Li glass TOF spectra for the two samples showed them to be fairly consistent in shape, as illustrated in Figure 3-6. The background subtraction was performed following the technique described in Section 3.3, with the exception that the internormalization constant, b , for the constant background was obtained from the count rate at the bottom of the 5.903-keV aluminum resonance dip. The effects of the cerium and silicon resonances at 1.3 keV and 56.0 keV, respectively, were eliminated by graphically interpolating through the corresponding peaks in the monitor TOF spectrum. The effect of the oxygen nuclei in the glass is to enhance the detector efficiency via multiple neutron scattering. However, it has been shown¹⁸ that for a 0.5-mm-thick ^6Li glass, the multiple-scattering correction is fairly flat in the energy region between 1 keV and 100 keV, except for the effects of the cerium and silicon resonances, as shown in Figure 3-7. Hence, in the energy region of interest, the multiple scattering effects do not distort the spectrum

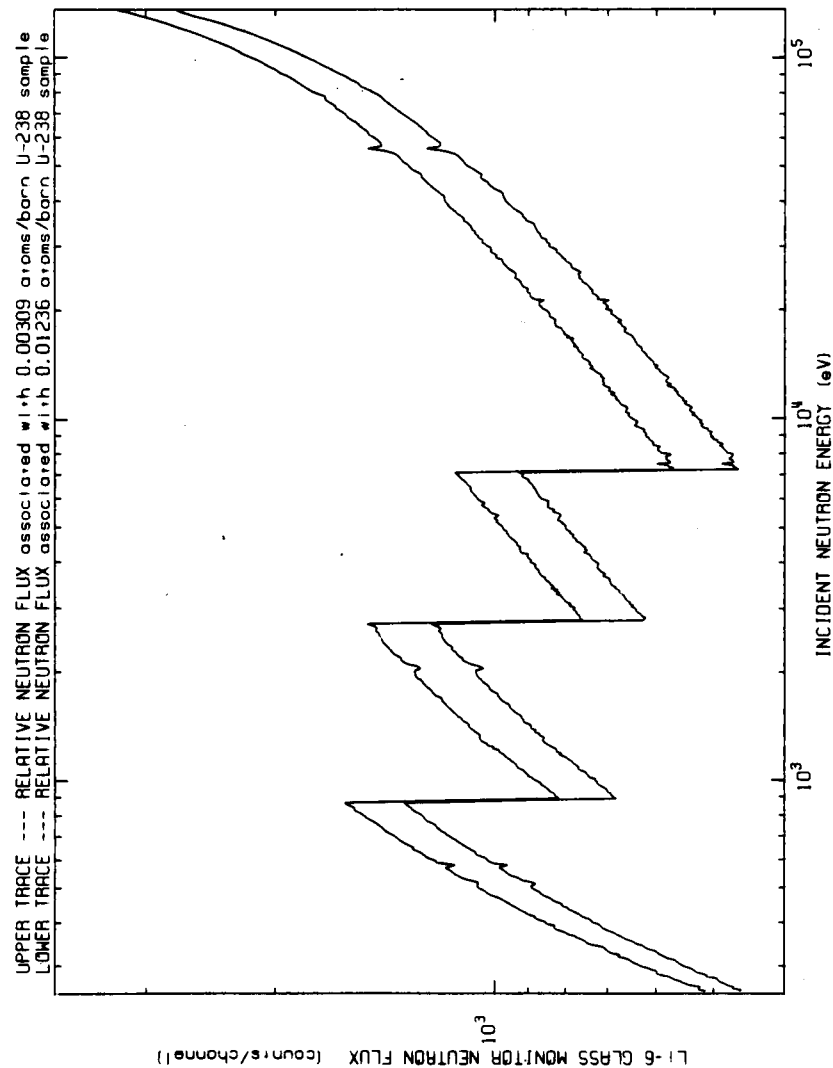


Figure 3-6. Lithium-6 Glass Monitor Relative Neutron Flux Versus Incident Neutron Energy. The upper trace is the relative neutron flux associated with the 0.00309 atoms/barn sample. The lower trace is the relative neutron flux associated with the 0.01236 atoms/barn sample. The effect of the ^{28}Si resonance at 56.0 keV is clearly seen in the data. The data have been averaged over 100-channel intervals. The discontinuities seen in the TOF spectra arise from changes in the channel time-width ("crunch factor").

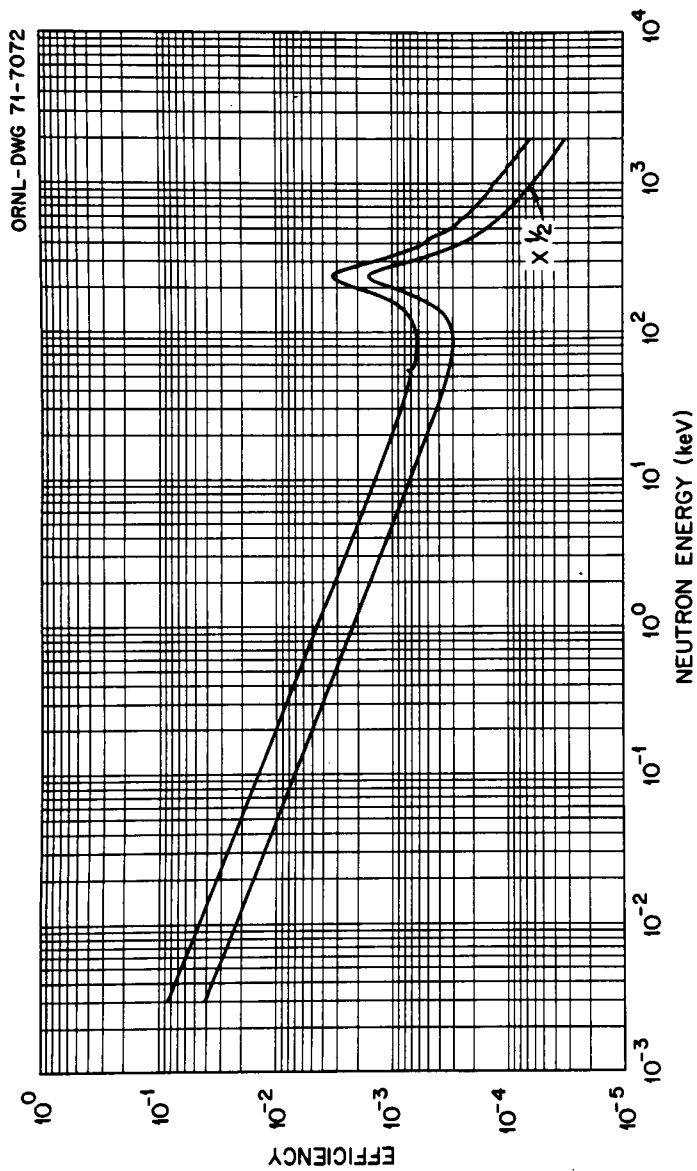


Figure 3-7. Transmitted Neutron Efficiency of a 0.05-cm ^6Li Glass Scintillator With and Without Self-Absorption, Scattering, and Resonance Structure Effects. (From Reference 27.)

shape and can be included in the normalization factor k_0 of Eq. 3-1. Previous studies at ORELA²⁷ showed that the transmitted neutron efficiency (see Figure 3-7) very closely follows the $1/v$ law, hence the factor $R(E)$ in Eq. 3-1 was made unity. (The slope in the graph of Figure 3-7 is 0.5).

3.5 NORMALIZATION OF THE CAPTURE YIELD DATA

To determine the normalization constant, a_n , in Eq. 3.1, a technique proposed by Moxon²⁸ was utilized. This technique is based on the idea that for small s-wave resonances (i.e. with $g\Gamma_n$ small), self-shielding and multiple scattering effects can be neglected. (See Appendix for further elaboration of this idea.) Hence, for these resonances, sample thickness effects are negligible and the measured capture yield can be described quite accurately by the Breit-Wigner single-level approximation formula

$$\sigma_{n,\gamma}(E) = \frac{\pi}{k^2} \frac{g_J \Gamma_n \Gamma_\gamma}{(E - E_0)^2 + (\frac{1}{2}\Gamma)^2} \quad (3-39)$$

where

- g_J = spin statistical factor = $\frac{(2J + 1)}{2(2I + 1)}$,
- I = spin of the target nucleus,
- J = total angular momentum,
- Γ_n = neutron width (eV),
- Γ_γ = capture width (eV),
- E_0 = resonance energy (eV),
- Γ_T = total width (eV) = $\Gamma_\gamma + \Gamma_n + \Gamma_n'$, (eV), with

Γ_n , - inelastic neutron width (which has the value zero below the first inelastic channel at 45 keV);

and where the quantity, k , is given by²⁹

$$k = k_0 E^{1/2} \text{ (cm}^{-1}\text{)} \quad (3-40)$$

with

$$k_0 = 2.196771 \left(\frac{(\text{AWR})}{(\text{AWR} + 1)} \right) 10^9 \quad (3-41)$$

For the ^{238}U nucleus the Atomic Weight Ratio (AWR in Eq. 3-41 above) is 236.006. Insertion of Eq. 3-40 into Eq. 3-39 yields

$$E^{1/2} \sigma_{n,\gamma}(E) = \frac{c_0}{E^{1/2}} \frac{g_J \Gamma_n \Gamma_\gamma}{[(E - E_0)^2 + 1/4\Gamma^2]} \quad (3-42)$$

with $c_0 = 1/k_0^2 \text{ (cm}^2\text{)}$.

Taking A_{TH} as the theoretical area under the Breit-Wigner resonance,

$$A_{\text{TH}} = \int_{E_1}^{E_2} dE E^{1/2} \sigma_{n,\gamma}(E) \quad (\text{b} \cdot \text{eV}^{3/2}) \quad (3-43)$$

and A_{EXP} as the experimental area under the measured capture yield data

$$A_{\text{EXP}} = \int_{E_1}^{E_2} dE E^{1/2} Y_c(E) \quad (\text{b} \cdot \text{eV}^{3/2}) \quad (3-44)$$

the normalization constant, a_n , is given by the ratio

$$a_n = \frac{A_{\text{TH}}}{A_{\text{EXP}}} \quad (3-45)$$

The integral in Eq. 3-43 can be calculated readily; using the change of variable

$$x = \frac{E - E_0}{1/2\Gamma_T} \quad (3-46)$$

and neglecting the energy dependence of Γ_T and Γ_n , yields

$$A_{TH} = \left(\frac{c}{E^{1/2}} \right) \left(\frac{g_J \Gamma_n \Gamma_\gamma}{\Gamma_T} \right) F(x_2, x_1) \quad (3-47)$$

with

$$F(x_2, x_1) = \frac{1}{\pi} [\tan^{-1}(x_2) - \tan^{-1}(x_1)] \quad , \quad (3-48)$$

$$x_2 = \frac{E_2 - E_0}{1/2\Gamma_T} \quad , \quad x_1 = \frac{E_1 - E_0}{1/2\Gamma_T} \quad (3-49)$$

and

$$c = 2\pi c_0 = 4.12509 \times 10^6 \quad (\text{b} \cdot \text{eV}) \quad (3-50)$$

The values of $g_J \Gamma_n$, were obtained from the resonance data of Olsen et al.³⁰ The ENDF/B-V recommended value of 0.0235 eV was used for the capture width, Γ_γ , for all the small s-wave resonances used in the calculation of the normalization constants.

Previous work at ORNL concerning the measurement of gamma-ray intensities³¹ showed that for both Breit-Wigner and Gaussian line shapes which are contaminated with background noise, a nearly optimal estimation of the area under the curve is obtained using the limits of integration

$$E_1 = E_0 - \frac{1}{2} \Delta E(1/5) \quad (3-51)$$

and

$$E_2 = E_0 + \frac{1}{2} \Delta E(1/5) \quad , \quad (3-52)$$

where $\Delta E(1/5)$ is the full width (eV) at one-fifth (1/5) of the maximum amplitude.

Tables 3-3 and 3-4 show the normalization constant calculations for the TOF spectra for the 0.01236 atoms/barn and 0.00309 atoms/barn ^{238}U samples, respectively. The average normalization constants, a_n , given in Tables 3-3 and 3-4 are the simple arithmetic means, and the given uncertainties are the standard deviations of the mean value for each sample thickness. (It is emphasized that the TOF spectrum for each sample has been taken as the simple sum of the two pulse-height groups with the best signal-to-background ratios, which is then divided by the corresponding neutron-flux spectrum prior to normalization. This procedure was followed to improve the statistical quality of the normalized capture yield data.)

Table 3-3. Normalization Constants for the
0.01236 atoms/barn ^{238}U Sample

E_0 (eV)	$g\Gamma_n$ (meV)	a_n
624.20	.82	80.31
677.75	.66	71.16
756.24	.55	77.92
808.16	.44	70.56
846.65	.87	78.43
891.23	.77	79.68
977.37	.77	75.07
1062.68	.96	84.51
Average normalization constant $a_n =$		77.2 ± 1.7

Table 3-4. Normalization Constant for the
0.00309 atoms/barns ^{238}U Sample

E_0 (eV)	$g\Gamma_n$ (meV)	a_n
624.20	.82	203.98
677.75	.66	227.36
756.24	.55	215.33
808.16	.44	266.18
846.65	.87	210.28
891.23	.77	257.88
977.37	.77	211.29
1062.68	.96	239.25
Average normalization constant $a_n =$		228.9 ± 8.2

3.6 UNCERTAINTIES IN THE MEASUREMENTS

Two types of uncertainties must be considered: the statistical counting uncertainty, which is uncorrelated between time-of-flight channels, and the fully correlated systematic uncertainties.

The statistical uncertainty for each sample was obtained by systematically following the propagation of the standard deviation of Poisson distributed statistical samples, at each step in the data reduction process. The relative error in the i^{th} channel of the capture yield TOF spectrum is then given by the relation

$$\frac{\delta Y_c(i)}{Y_c(i)} = \frac{[HF(i) + a BT(i)]^{1/2}}{CBS(i)}, \quad (3-53)$$

where all the symbols in Eq. 3-53 have been previously defined in Section 3.3.2.

The main sources of systematic uncertainties are the background subtraction uncertainties in the TOF spectra for the ^{238}U counts in

the ORELAST and the neutron monitor counts, and normalization uncertainties. To estimate the uncertainties associated with the background subtraction procedure, the sensitivity coefficients, S_i , below were computed for each channel, i , in the TOF spectra.

$$S_i = \frac{a}{Y_c(i)} \frac{\Delta Y_c(i)}{\Delta a} \quad , \quad (3-54)$$

where

$\Delta a/a$ = percent change in the background subtraction constant,

$\Delta Y_c(i)/Y_c(i)$ = percent change in the capture yield, and

$\Delta Y_c(i)$ = change in the capture yield upon a change Δa in the background subtraction constant.

The sensitivity coefficients were computed for the energy-dependent and constant background subtractions for both the ORELAST and ${}^6\text{Li}$ glass neutron monitor TOF spectra. These calculations were carried out by changing the energy-dependent background subtraction constant (or the constant background subtraction constant) by 5% and proceeding through all following steps in the data reduction process with no other changes.

The estimated systematic uncertainties are given as the root mean square (RMS) combination of two sources of uncertainty:

1. Energy-dependent uncertainties arising from background subtraction and neutron beam spectrum determination, and
2. Normalization uncertainties.

The energy-dependent uncertainties were obtained by the convolution of the sensitivity coefficients with the estimated uncertainties in the

constant, a , for each of the background subtraction procedures. These uncertainties were then combined with a 3% normalization uncertainty estimated from the calculation of the normalization constant, a_n . (See Tables 3-3 and 3-4). Following this procedure, the systematic uncertainties in the capture yield data were estimated, on the average, to be 6% up to 10 keV and 5% from 10 keV up to 100 keV.

CHAPTER IV

RESULTS

4.0 COMPARISON OF CAPTURE YIELD RESULTS AS A FUNCTION OF PULSE-HEIGHT DISTRIBUTION

The time-of-flight pulse-height multiparameter system described in Section 2.6 was used to observe whether appreciable change in pulse-height spectrum or gamma-ray multiplicity occurred as a function of time-of-flight. Such changes could be produced either by electronic bias shifts or by an energy dependence of the average properties of the gamma-ray cascade, following neutron capture. Both effects could change the measured value of the capture yield.

Figure 4-1 shows the pulse-height spectrum measured at the neutron time-of-flight corresponding to the 518.3-eV ^{238}U resonance. The large spike seen in the figure is due to various background contributions (such as from the electronics, from radioactivity in the sample, etc.). This background contribution is seen in Figure 4-2, which shows a pulse-height spectrum around 500 eV where there are no large resonances in the ($^{238}\text{U} + n$) compound nucleus. Examination of the background subtracted pulse-height spectrum in Figure 4-3 shows that pulse-height bins 3 and 4 (see Table 2-1) exhibit the best signal-to-background ratio. Consequently, the time-of-flight spectra for these two sections were separately analyzed and then combined (prior to normalization) to improve the statistical accuracy, for each of the two ^{238}U samples.

Capture yields for each of the two selected pulse-height bin time-of-flight spectra are shown in Figures 4-4 and 4-5 for both

ORELAST PULSE-HEIGHT (INCLUDING BACKGROUND)
AT 518 eV INCIDENT NEUTRON ENERGY

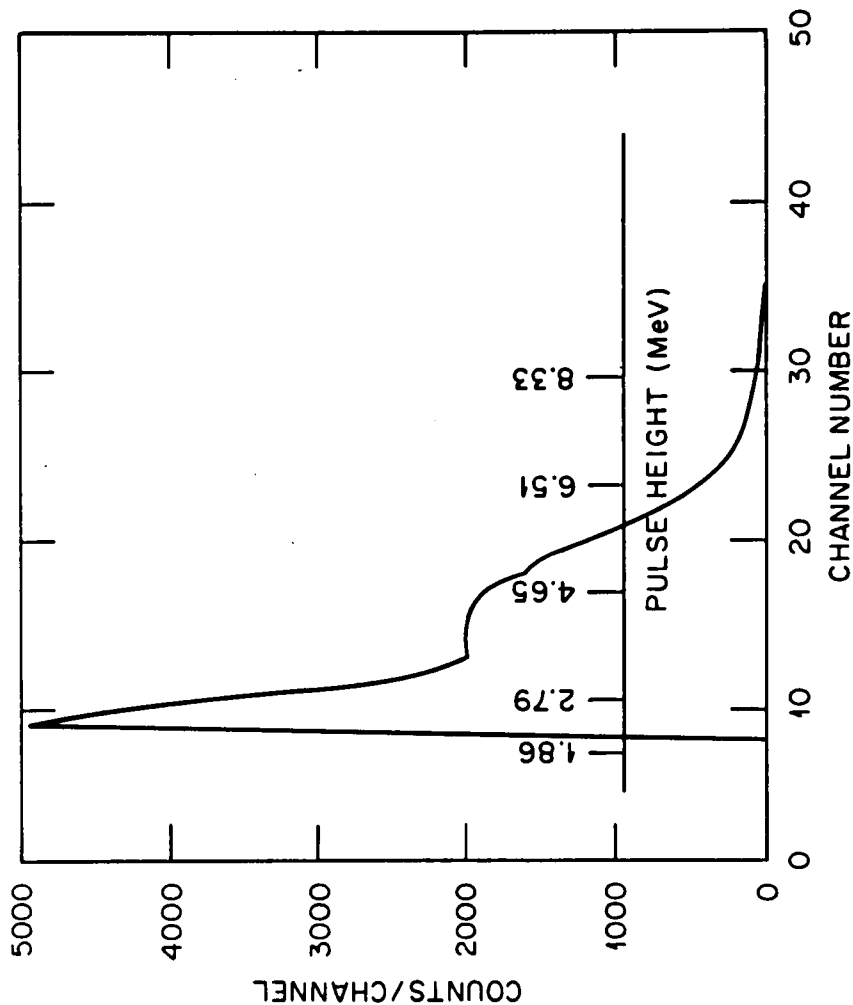


Figure 4-1. ORELAST Pulse-Height Spectrum (Including Background) at TOF Corresponding to 518 eV, Showing Count Rate Versus Channel Number. The large spike is due to background contributions. The second horizontal scale shows the pulse-height ("bias") group boundaries.

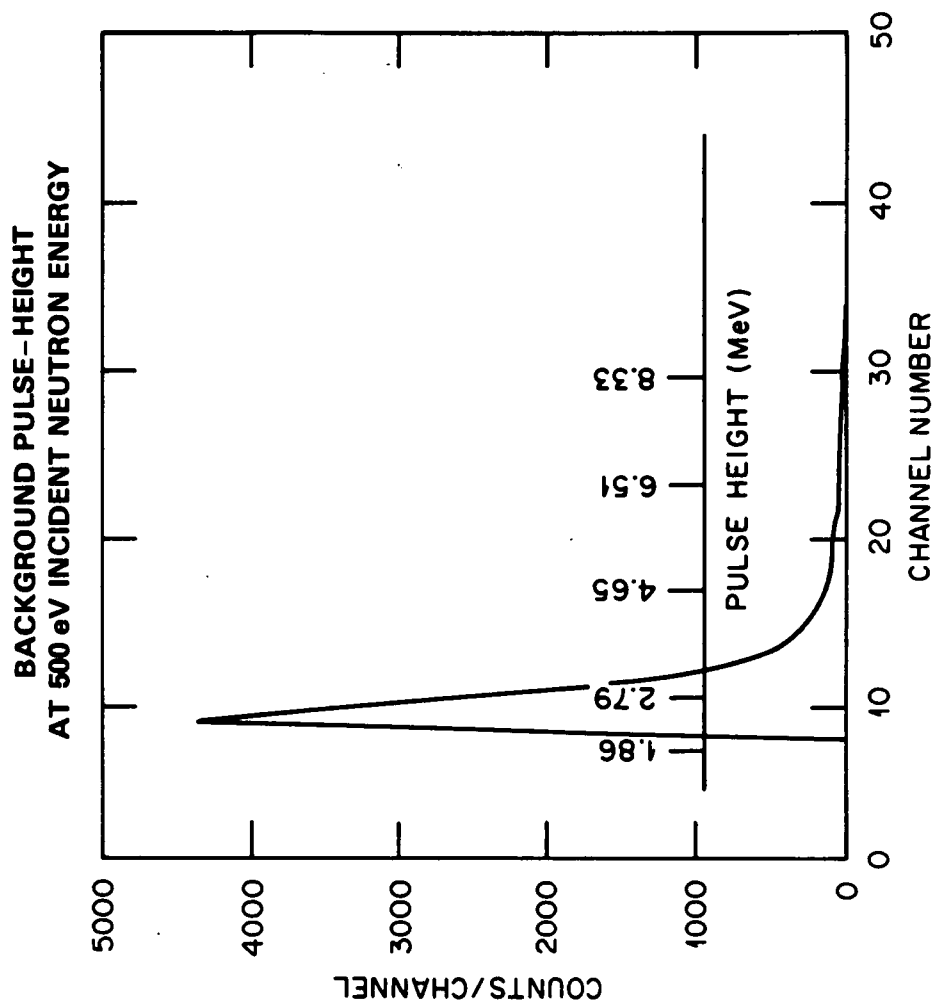


Figure 4-2. ORELAST Pulse-Height Spectrum at TOF Corresponding to 500 eV, Showing Count Rate Versus Channel Number for the Background Contribution. The second horizontal scale shows the pulse-height ("bias") group boundaries.

**ORELAST BACKGROUND-SUBTRACTED PULSE-HEIGHT
AT 518 eV INCIDENT NEUTRON ENERGY**

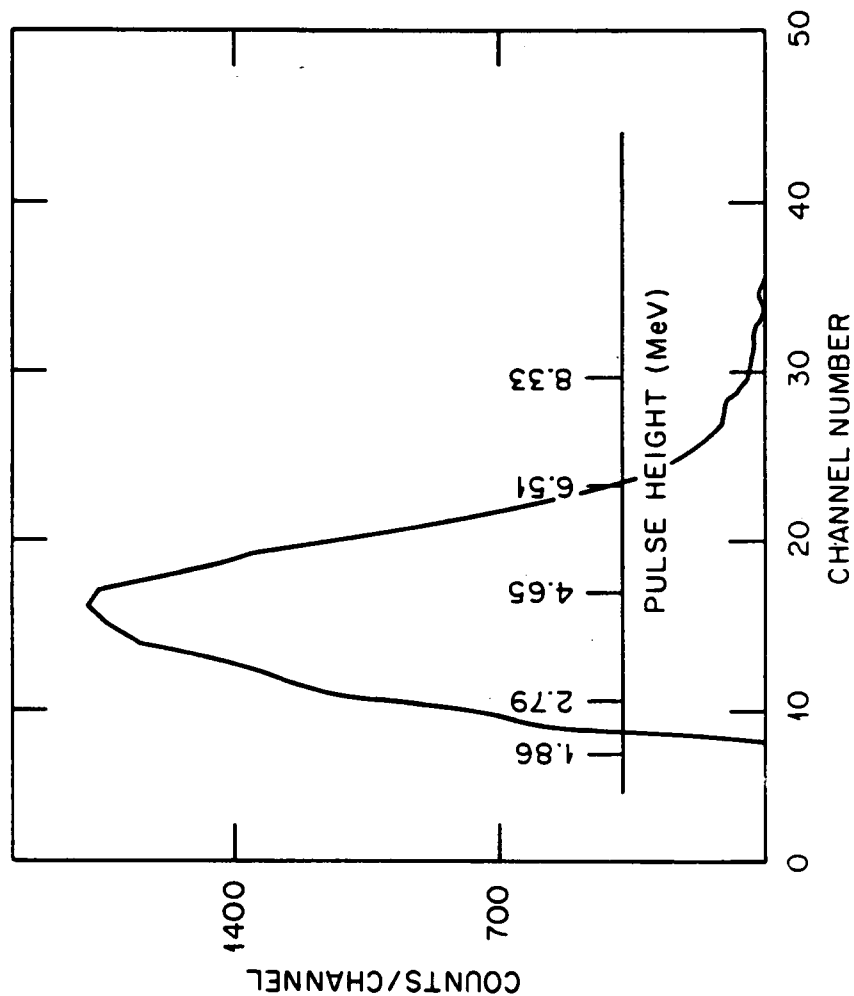


Figure 4-3. ORELAST Pulse-Height Spectrum at TOF Corresponding to 518 eV, Where the Background Contribution Has Been Subtracted, Showing Count Rate Versus Channel Number. The second horizontal scale shows the pulse-height ("bias") group boundaries.

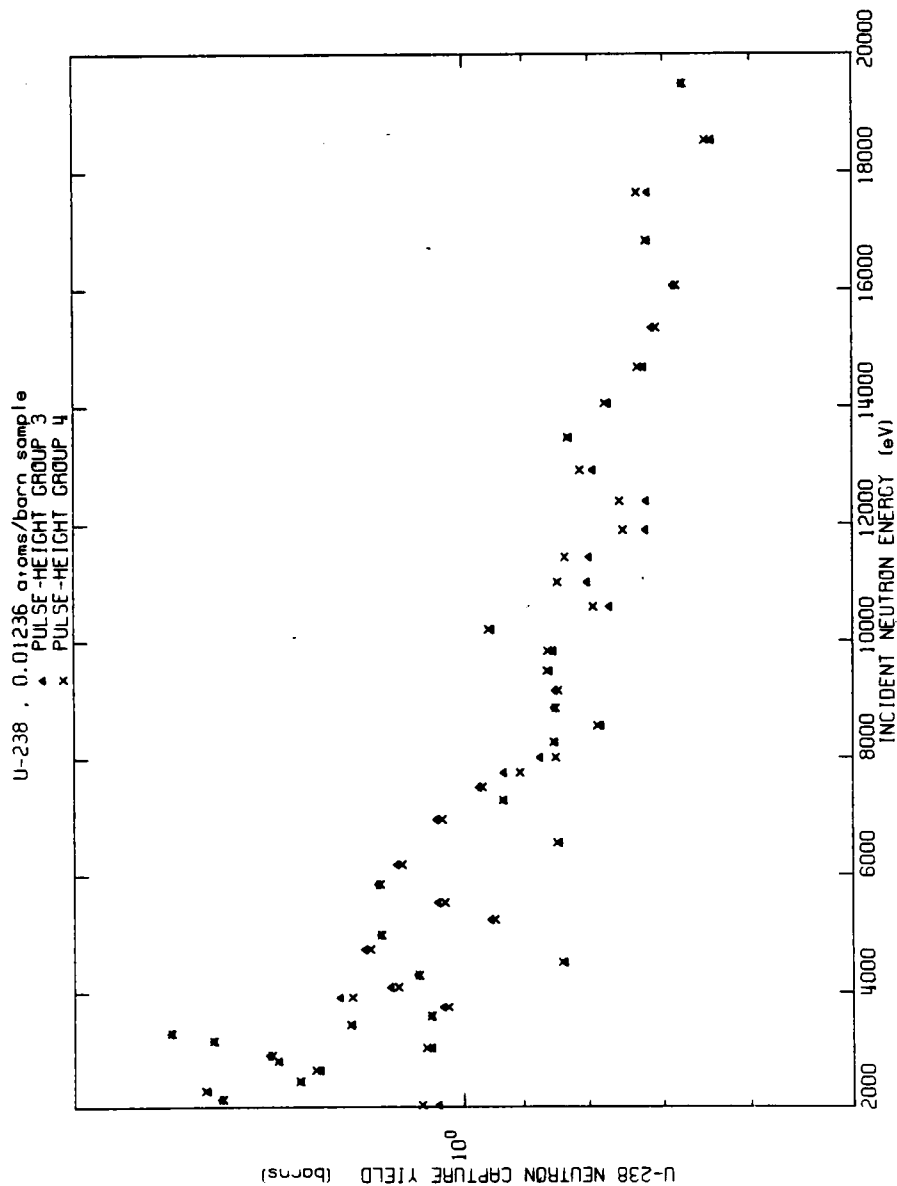


Figure 4-4. Comparison of the Capture Yield Data for the Two Selected Pulse-Height Groups for the 0.01236 atoms/barn Sample in the Energy Range from 2 keV to 20 keV. The data shown have been averaged over 500-channel intervals.

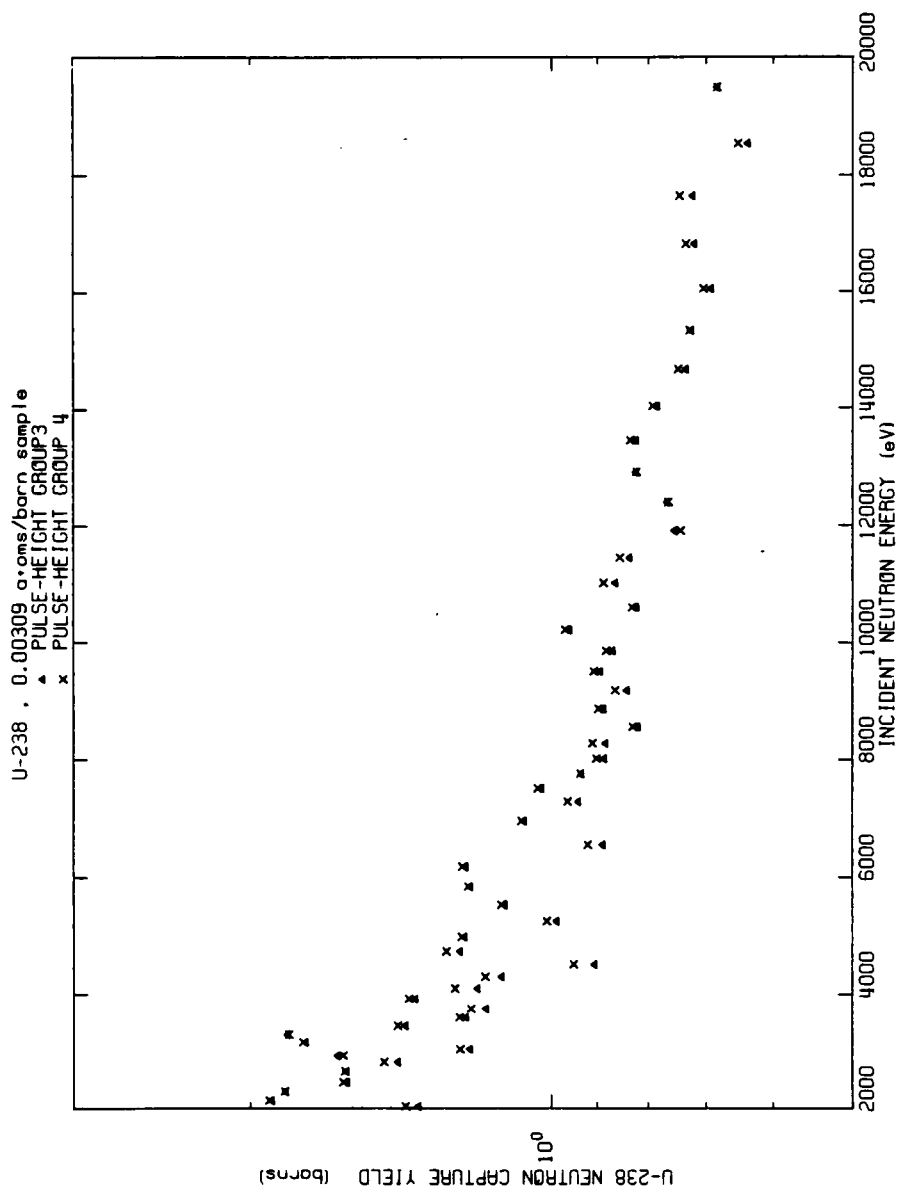


Figure 4-5. Comparison of the Capture Yield Data for the Two Selected Pulse-Height Groups for the 0.00309 atoms/barn Sample in the Energy Range from 2 keV to 20 keV. The data shown have been averaged over 500-channel intervals.

samples in the neutron energy range between 2 keV and 20 keV. Figures 4-6 and 4-7 show a similar comparison in the energy range above 20 keV. Note that whereas there is satisfactory agreement between the data for the two pulse-height bins below 50 keV, there are systematic differences above 50 keV. However, in both samples, the observed differences above 50 keV are in magnitude and not in shape. This leads to the belief that these differences may arise primarily from background subtraction problems in this particular energy region. In fact, it appears very difficult to detect the effect of tank-coincidence bias changes and energy-dependent gamma-ray cascades on the measurement. These effects are probably overshadowed by the uncertainties in the background subtraction which depend strongly on the signal-to-background ratio which is different for each pulse-height bin.

4.1 SAMPLE THICKNESS EFFECTS

The capture data obtained from the interaction of a beam of neutrons with a ^{238}U sample of finite thickness cannot be interpreted directly as capture cross section because of self-shielding and multiple scattering effects within the sample. The self-shielding effect arises from the attenuation of the neutron beam as it propagates through the sample, thus resulting in a non-uniform distribution of neutrons as a function of sample depth. The depression of the neutron flux then decreases the number of capture events in the sample. The multiple scattering effect arises from neutrons which, after undergoing one or several scattering events, remain in the sample and are finally captured at an energy lower than their initial energy. The

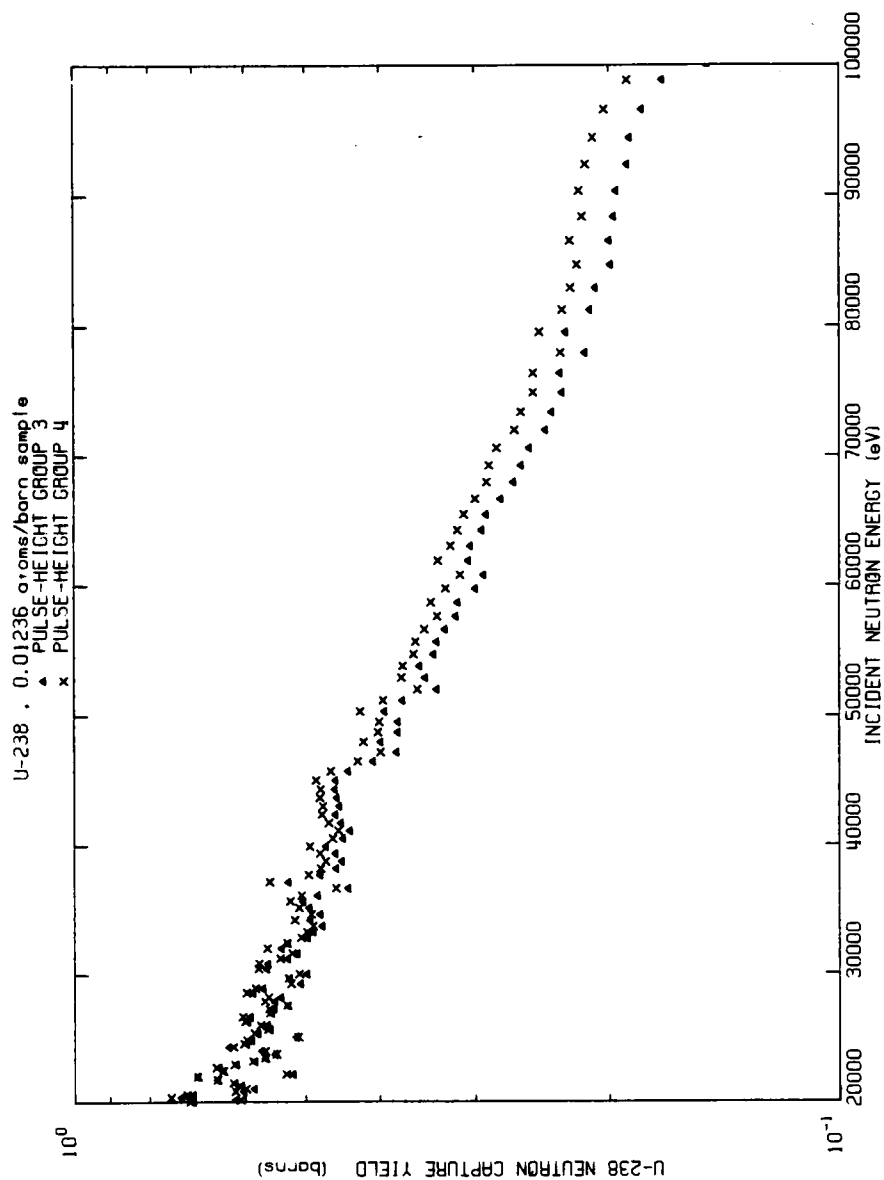


Figure 4-6. Comparison of the Capture Yield Data for the Two Selected Pulse-Height Groups for the 0.01236 atoms/barn Sample in the Energy Range from 20 keV to 100 keV. The data shown have been averaged over 100-channel intervals.

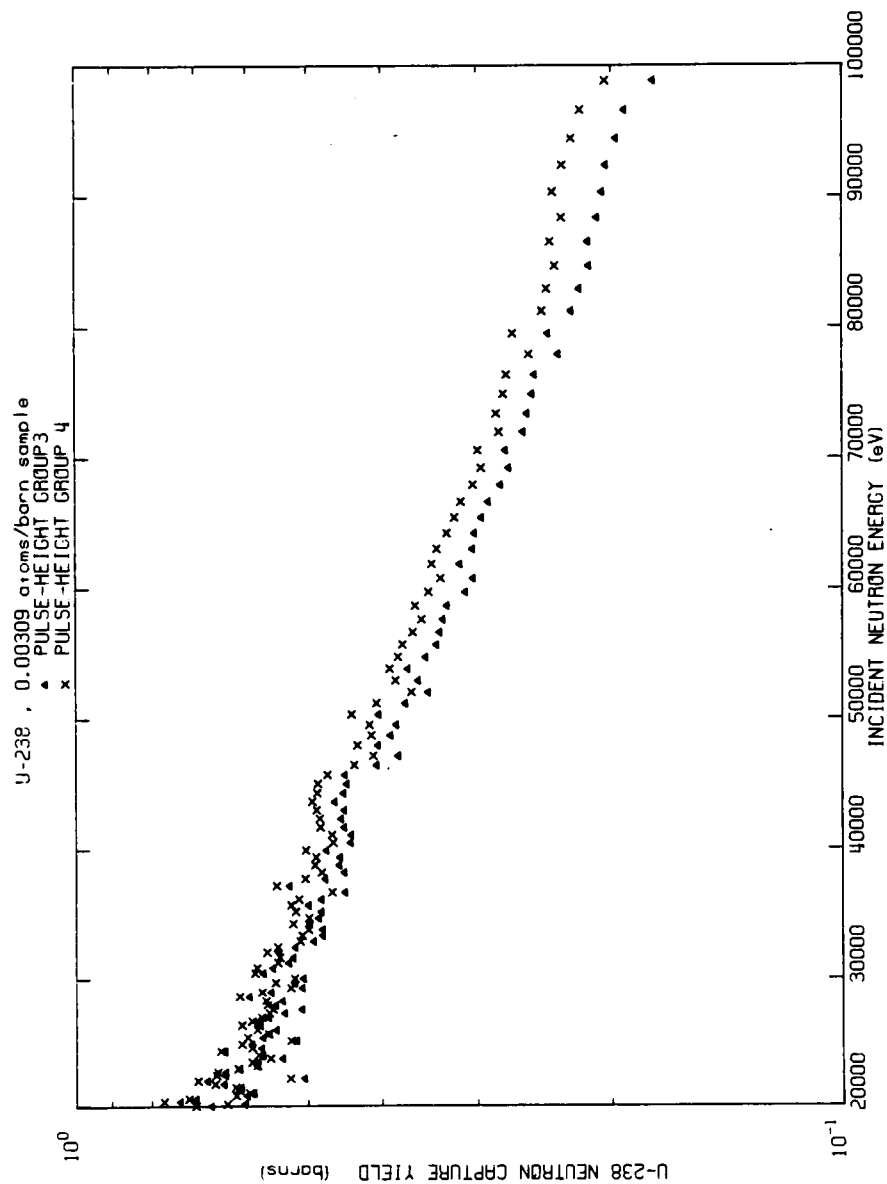


Figure 4-7. Comparison of the Capture Yield Data for the Two Selected Pulse-Height Groups for the 0.00309 atoms/barn Sample in the Energy Range from 20 keV to 100 keV. The data shown have been averaged over 100-channel intervals.

effect of these off-energy capture events is to increase the measured capture rate. Hence the self-shielding effect decreases the rate of capture events, whereas the multiple scattering effect acts in the opposite way. The relative magnitude of these competing effects is a function of the resonance parameters of the particular resonance.

The capture yield is related to the neutron capture cross section $\sigma_{n,\gamma}(E)$, by the relation

$$Y_c(E) = \sigma_{n\gamma}(E) S_s(E,N) M_s(E,N) \quad (4-1)$$

where

$$S_s(E,N) = \text{self-shielding factor} = [1 - \exp\{-N\sigma_T(E)\}]/N\sigma_T(E) \quad (4-2)$$

and

$$M_s(E,N) = \text{multiple scattering factor} = \left[1 - \frac{\sigma_{n\gamma}(E)}{\sigma_T(E)} P_c(N,E) \right]^{-1} \quad (4-3)$$

with

N = sample thickness (atoms/barn),

$\sigma_T(E)$ = total neutron cross section, and

$P_c(N,E)$ = mean collision probability ($P_c(0,E) = 0$). (4-4)

Note that it is only for infinitely thin samples (i.e. $N \rightarrow 0$), that $S_s(0,E)$ and $M_s(0,E)$ become unity and there is equivalence between capture cross section and capture yield.

Figure 4-8 shows a Monte Carlo calculation⁴ of the self-shielding and multiple scattering effects on neutron capture in the neutron-energy mentioned, the self-shielding process "chops off" the resonance

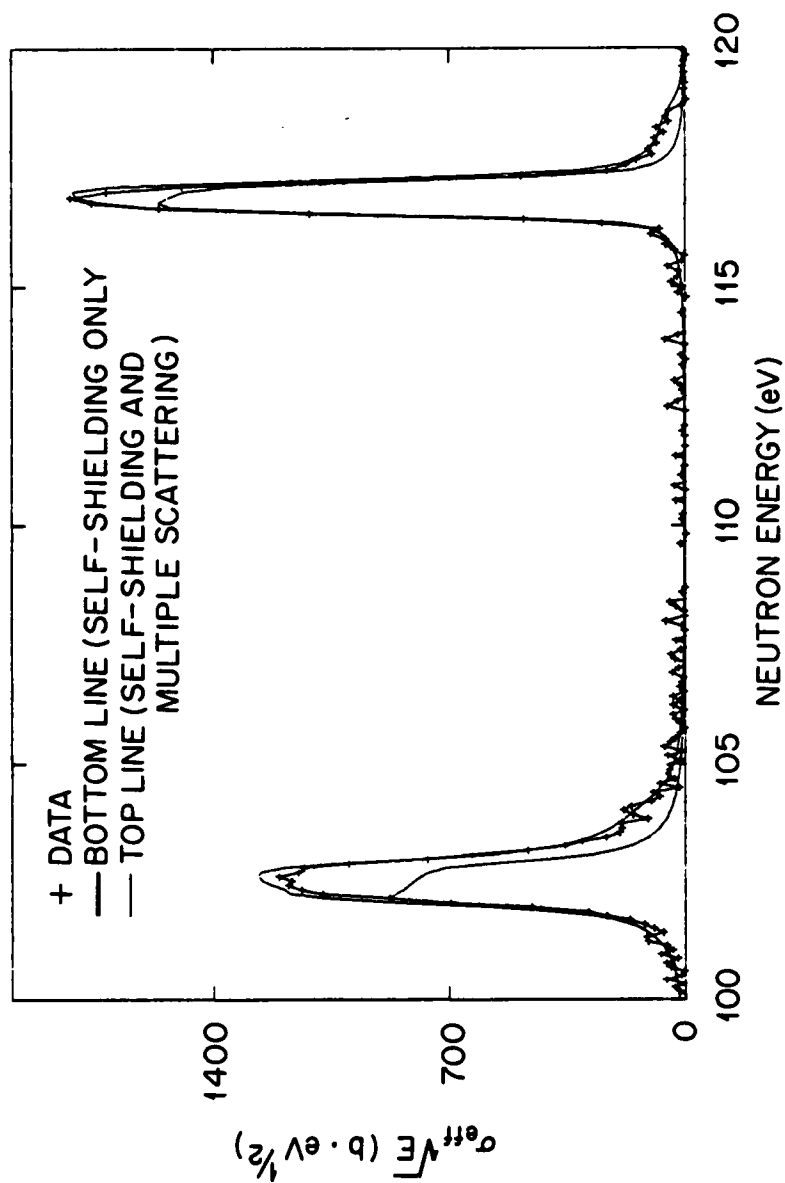


Figure 4-8. Monte Carlo Calculations of the Self-Shielding and Multiple Scattering Effects for a 0.0028 atoms/barn Sample Thickness. (From Reference 4.) Note that the relative contributions of self-shielding and multiple scattering effects depend on the shape and strength of the resonances.

peaks, and that the relative magnitudes of the sample thickness effects indeed depend on the resonance parameters.

Capture data for the two ^{238}U samples used in the present measurements are shown in Figure 4-9 for neutron energies in the resolved resonance region. These results clearly show substantial sample thickness effects in the large resonances. Figure 4-10 displays capture yield data for the two samples in the neutron-energy range 25 to 100 keV, where the data in each case have been averaged over 250 time-of-flight channels. Note that in the region from 25 up to 50 keV, there appears to be a considerable cancellation between the self-shielding and multiple scattering effects for the two sample thicknesses.

4.2 RESULTS AND DISCUSSION

Our capture yield data averaged over decimal energy intervals between 0.5 keV and 100 keV are shown in Table 4-1 for the 0.01236 atoms/barn and 0.00309 atoms/barn samples. Comparison of our data with the results from previous measurements is hindered by the fact that most researchers report their results as cross-section data--without specifying in any detail the self-shielding and multiple-scattering corrections which were applied to their capture yield data (to derive the reported cross-section data). Thus, we shall compare the present capture yield results with two other ^{238}U capture yield measurements performed at the Oak Ridge Electron Linear Accelerator facility. These are the 1973 measurement of de Saussure et al.⁴ and the 1982 measurement of Yang.³² The comparison of experimental conditions and data-treatment methods, displayed in Table 4-2, shows that

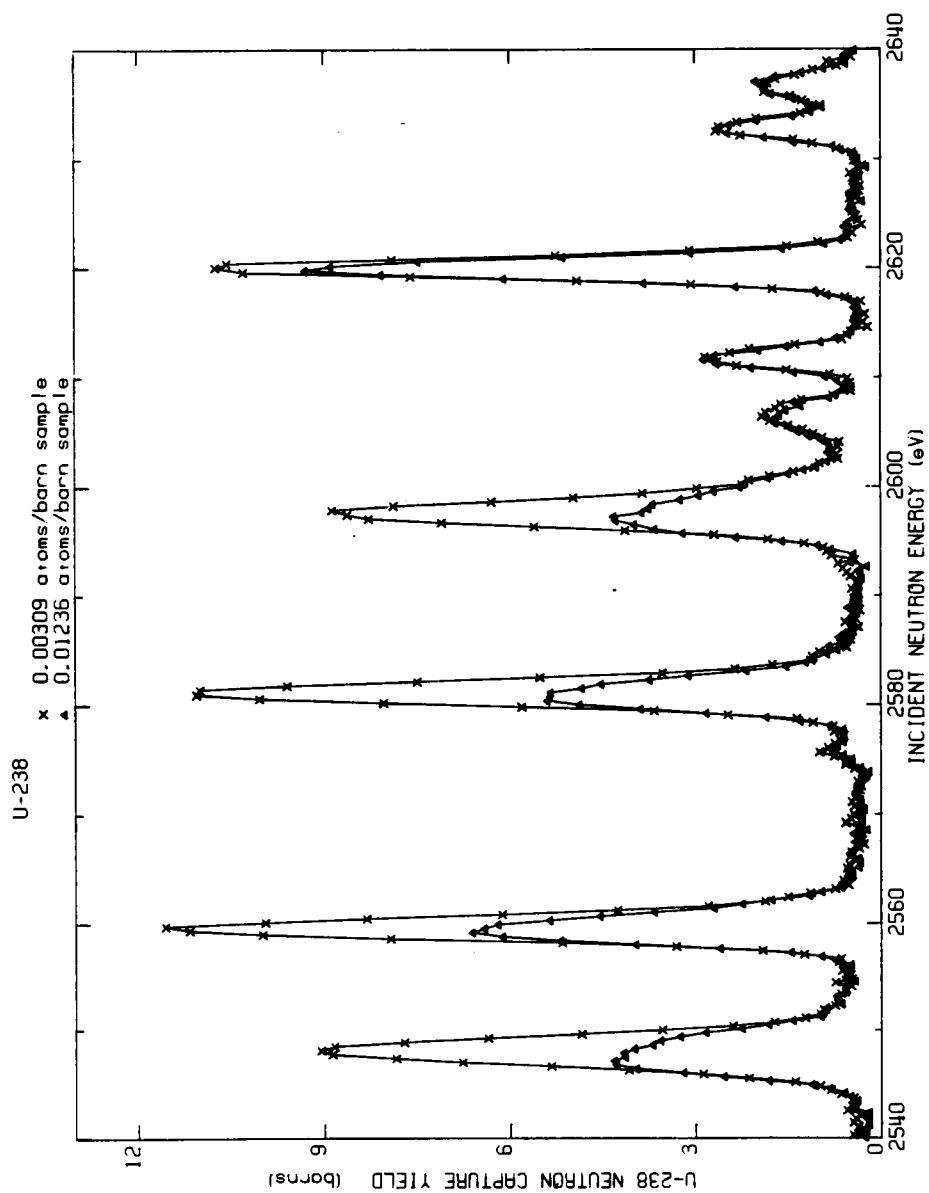


Figure 4-9. Capture Yield Data for the 0.00309 atoms/barn and 0.01236 atoms/barn Samples in the Neutron Energy Range from 2540 eV to 2640 eV. The lines shown merely connect the data symbols for each sample for clarity. Note the appearance of substantial sample thickness effects on the larger resonances.

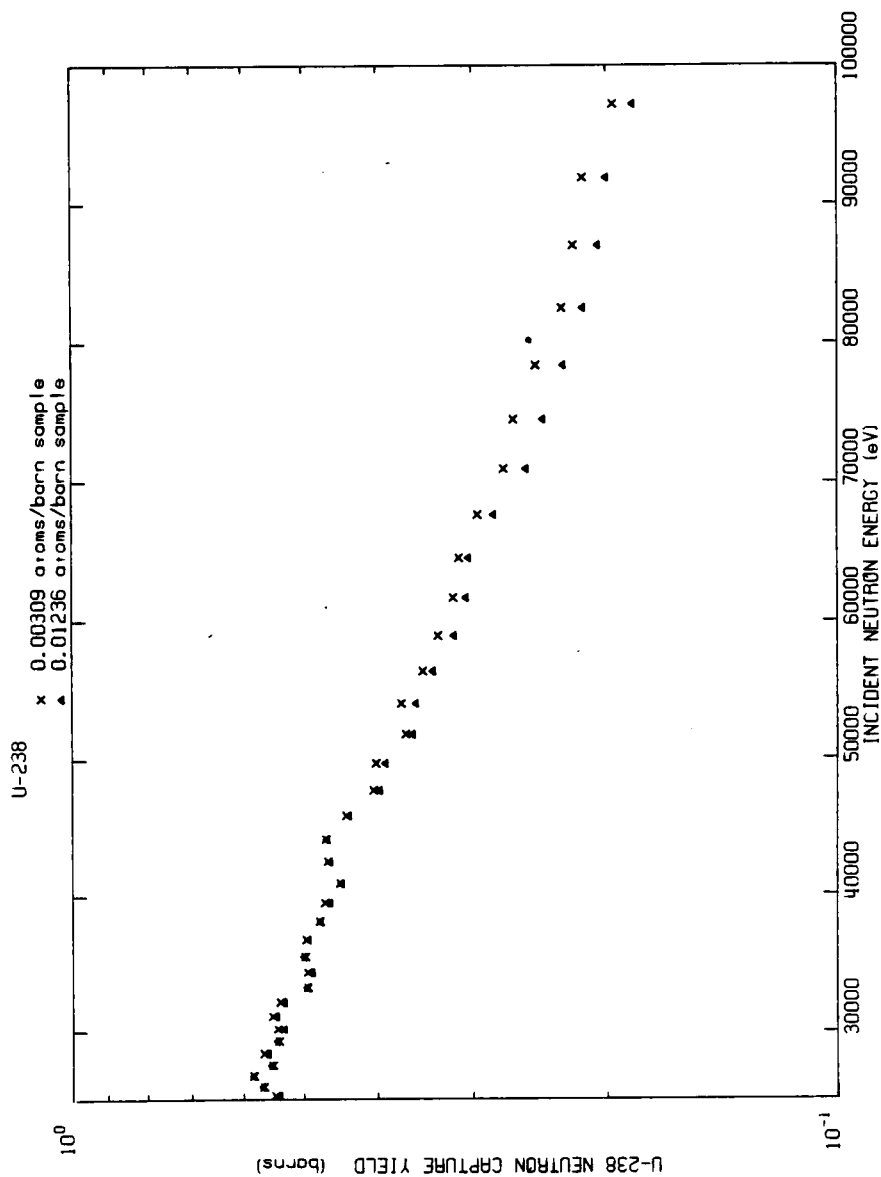


Figure 4-10. Capture Yield Data for the 0.00309 atoms/barn and 0.01236 atoms/barn Samples in the Neutron Energy Range from 25 keV to 100 keV. The data have been averaged over 250-channel intervals. Note the cancellation between self-shielding and multiple scattering effects in the energy region from 25 keV to 50 keV, and the dominance of the multiple scattering effect above 50 keV.

Table 4-1. Capture Yield Over Decimal Energy Intervals from 0.5 keV up to 100 keV for the 0.01236 atoms/barns and 0.00309 atoms/barn Samples. Also shown are capture yield data of de Saussure et al. and Yang.

ΔE (keV)	Y ₁ (150) (0.01236 atoms/barn)	Y ₂ (150) (0.00309 atoms/barn)	Y ₁ (40) ^a (0.00283 atoms/barn)	Y ₂ (40) ^b 0.00283 atoms/barn)
.5 - .6	2.24±.13	4.10±.25	4.00±.18	3.55±.23
.6 - .7	2.19±.13	3.33±.20	3.20±.14	2.85±.20
.7 - .8	1.91±.11	2.23±.13	2.04±.13	1.71±.11
.8 - .9	1.98±.12	2.93±.18	2.83±.13	2.54±.18
.9 - 1	2.23±.13	3.50±.21	3.41±.15	3.04±.23
1 - 2	1.55±.09	2.06±.12	1.93±.09	1.84±.13
2 - 3	1.37±.08	1.63±.10	1.53±.08	1.42±.09
3 - 4	1.28±.08	1.45±.09	1.32±.08	1.20±.08
4 - 5	1.08±.06	1.15±.07	1.05±.07	1.05±.06
5 - 6	1.07±.06	1.13±.07	1.05±.06	1.02±.06
6 - 7	1.01±.06	1.08±.06	1.03±.06	.900±.05
7 - 8	.943±.06	.968±.06	.889±.05	.821±.05
8 - 9	.805±.05	.841±.05	.797±.04	.722±.05
9 - 10	.862±.05	.881±.05	.799±.04	.735±.05
10 - 20	.741±.04	.755±.05	.715±.03	
20 - 30	.584±.04	.586±.04	.572±.03	
30 - 40	.499±.03	.503±.03	.505±.03	
40 - 50	.434±.03	.439±.03	.435±.02	
50 - 60	.348±.02	.360±.02	.352±.02	
60 - 70	.293±.02	.304±.02	.306±.02	
70 - 80	.241±.01	.262±.02	.268±.02	
80 - 90	.211±.01	.226±.01	.242±.02	
90 - 100	.191±.01	.204±.01	.216±.02	

^a40-m flight-path measurement of de Saussure et al. (1973).

^b40-m flight-path measurement of Yang (1982).

Table 4-2. Comparison of Experimental Conditions and Data Treatment Methods for Three ORELA Measurements of the ^{238}U Capture Yield

EXPERIMENT	de Saussure et al. (1973)	Yang (1982)	This work (1987)
FLIGHT - PATH	40 m	40 m	150 m
GAMMA-RAY DETECTOR	ORELAST	Four C_6F_6 liquid scintillators viewing the ^{238}U sample ^a	ORELAST
NEUTRON MONITOR	$^{10}\text{BF}_3$ ionization chamber	$^{10}\text{BF}_3$ ionization chamber	^6Li -glass monitor
BACKGROUND SUBTRACTION	Resonance filters	Resonance filters	Neutron overlap method
NORMALIZATION	Saturated resonance technique	Saturated resonance technique	Small resonance method
SAMPLE AREAL DENSITIES (atoms/barn)	0.000140, 0.000396, and 0.00283	0.000396, 0.00062, and 0.00283	0.01236 and 0.00309

^aThe Maier-Leibnitz pulse-height weighting technique was used to obtain a response proportional to the ^{238}U binding energy and independent of the gamma-ray cascade multiplicity. (See Macklin et al.³³)

there are substantial differences in the methodologies among the three measurements. These differences are deemed sufficient to remove any question of "inbreeding."

The 1973 and 1987 measurements used the ORELAST tank in coincidence but had different neutron-beam monitors. The background subtraction methods and normalization techniques were also quite different. The 1982 measurement by Yang, although using data treatment methods similar to the 1973 experiment of de Saussure et al., measured the neutron capture gamma rays by viewing the ^{238}U sample with four non-hydrogenous C_6F_6 liquid scintillator detectors. In addition, Yang's measurement used the Maier-Leibnitz pulse-height weighting technique.³³

The 1973 experiment and the 1982 measurement used two of the same ^{238}U samples, but the present work used one similar but slightly different sample of 0.00309 atoms/barn. However, Monte Carlo calculations³² of sample-thickness effects versus sample thickness show that self-shielding and multiple-scattering effects differ by less than 1% between the 0.00283 and 0.00309 atoms/barn samples. Hence, for the purpose of the present capture yield data comparison, one can safely assume that the three measurements, although differing in methodology, can be associated with the same sample thickness.

With the exception of the region above 70 keV, our data appear somewhat lower than the 1973 results but remain within the bounds of the given uncertainties for both measurements. The comparison with the 1982 data shows the present results to be systematically higher than those of Yang, with discrepancies above 10% in some of the energy intervals. There is no reason to believe that the entirety of these

substantial differences could arise from the different gamma-ray detection techniques used in the two experiments. Rather one suspects that these discrepancies originate in the background subtraction procedure.

In the 1982 measurement the background below 3.5 keV was estimated from the count rate between well-resolved resonances, where the capture cross section is negligible. Afterwards this background was extrapolated by a polynomial fit to the background evaluated at the 5.9 keV and 35.2 keV aluminum resonances. This background subtraction procedure was dictated by the restricted neutron-energy range in Yang's experiment. The 1973 experiment was performed over a larger neutron-energy range which allowed the use of the sulfur resonances at 96 keV and 110 keV, and a direct (no extrapolation) determination of the background in the energy range above 3.5 keV range was then possible. The present measurement, which relied on the neutron pulse overlap method, also provided a direct determination of the background. It is suspected that the discrepancies observed above 3 keV (between the 1973 and 1987 measurements as compared to the measurement of Yang) are due to the fact that too much background was subtracted in the 1982 experiment by Yang. Even small differences in background between experiments may lead to substantial discrepancies between measurements when the results are averaged over relatively large energy intervals.

The high energy resolution achieved in the present work is illustrated in Figures 4-11 and 4-12, which show examples of the capture yield data around 15 keV and 23 keV, respectively, for the 0.01236 atoms/barn sample. The energy resolution of these data will allow the possibility of extending the resolved resonance energy region for ^{238}U

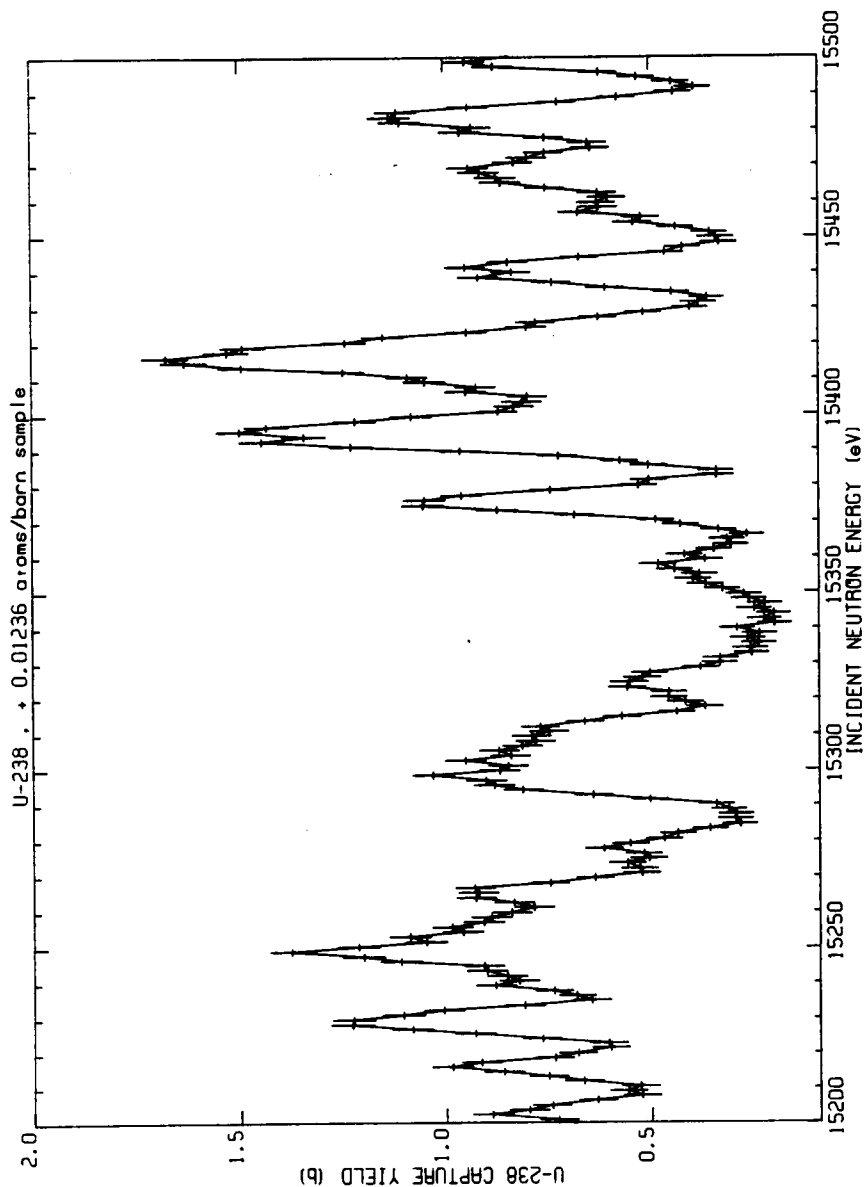


Figure 4-11. Capture Yield Data in the Neutron Energy Range from 15.2 keV to 15.5 keV for the 0.01236 atoms/barn Sample. Note the high resolution achieved by the present measurement which is a factor of three better than any previous measurement in this energy region. The vertical bars indicate statistical uncertainties only.

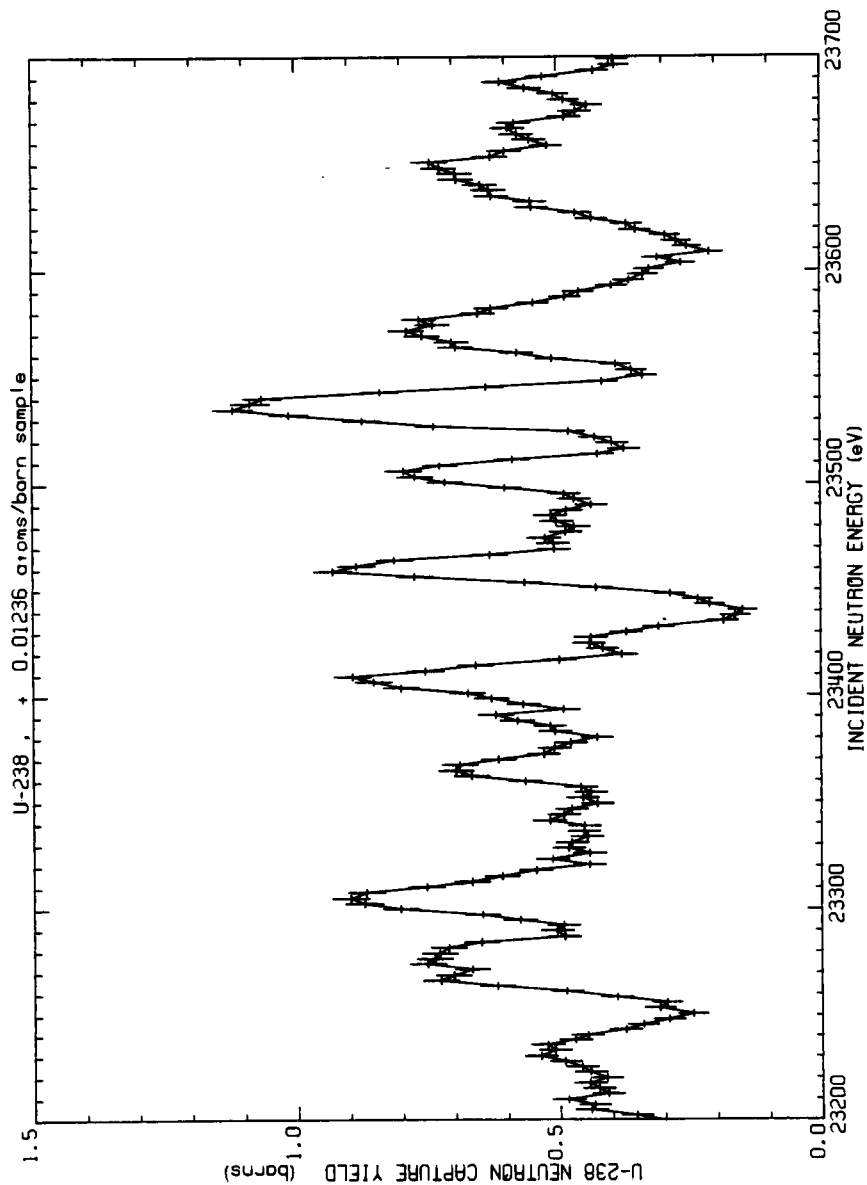


Figure 4-12. Capture Yield Data in the Neutron Energy Range from 23.2 keV to 23.7 keV for the 0.01236 atoms/barn sample. Note the high resolution achieved by the present measurement which is a factor of three better than any previous measurement in this energy region. The vertical bars indicate statistical uncertainties only.

beyond its current 4-keV limit. Above 20 keV the present data exhibit a substantial amount of fine structure in the capture yield. This is illustrated in Figures 4-13 and 4-14 which show capture yield data for the two ^{238}U samples in the energy regions covering 25 to 50 keV and 50 to 90 keV, respectively. These data, which have been averaged over fifty time-of-flight channels to eliminate statistical fluctuations, show a remarkable amount of fine structure which appears extremely well reproduced in both samples. The existence of this fine structure in the ^{238}U neutron capture yield is of importance for the calculation of self-shielded neutron group cross sections and, particularly in the 4- to 10-keV range, for the determination of the Doppler coefficient of reactivity. Figures 4-15 and 4-16 show the ^{238}U neutron capture cross section between 10 keV and 100 keV as prescribed by the ENDF/B-V evaluation,³⁴ with the capture yield data for each of the two samples in the present work superimposed. Evidently the ENDF/B-V evaluation is not providing the amount of fine structure needed for reactor design in this energy region.

As previously noted, it is not quite consistent to compare the present capture data with previous ^{238}U neutron capture measurements because of the lack of detailed information on the sample thickness corrections. Nevertheless, above 10 keV the self-shielding and multiple scattering corrections for a 0.00309-atoms/barn sample are on the average about 4% so that, in principle, one can provide a qualitative comparison between capture cross-section data and capture yield data. This is illustrated in Figure 4-17 which shows a comparison of various sets of ^{238}U neutron capture data (from the evaluation by Davey³⁵) with the present capture yield data for the 0.00309-atoms/barn sample.

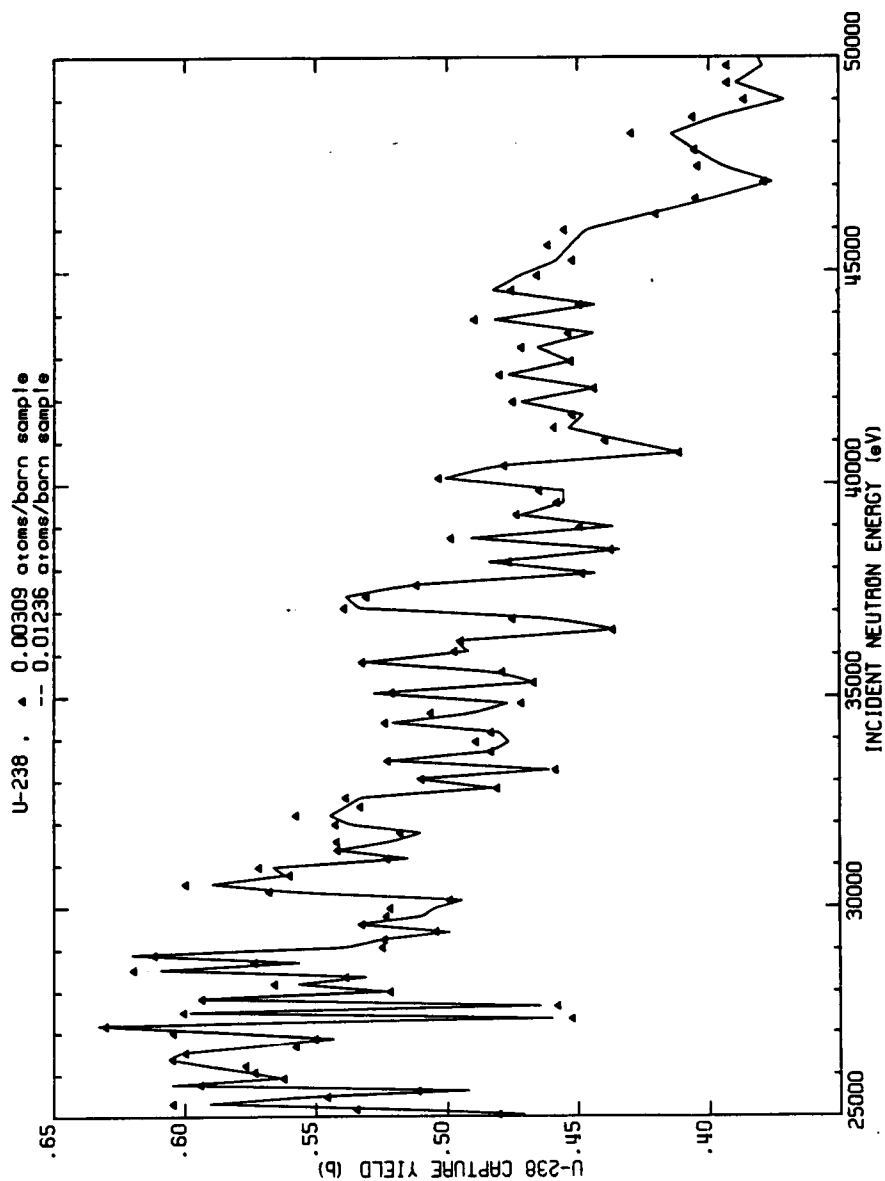


Figure 4-13. Capture Yield Data for the 0.00309 atoms/barn and 0.01236 atoms/barn Samples in the Neutron Energy Range from 25 keV to 50 keV. The data have been averaged over 50-channel intervals. Note the good reproducibility of the observed fine structure between the two samples.

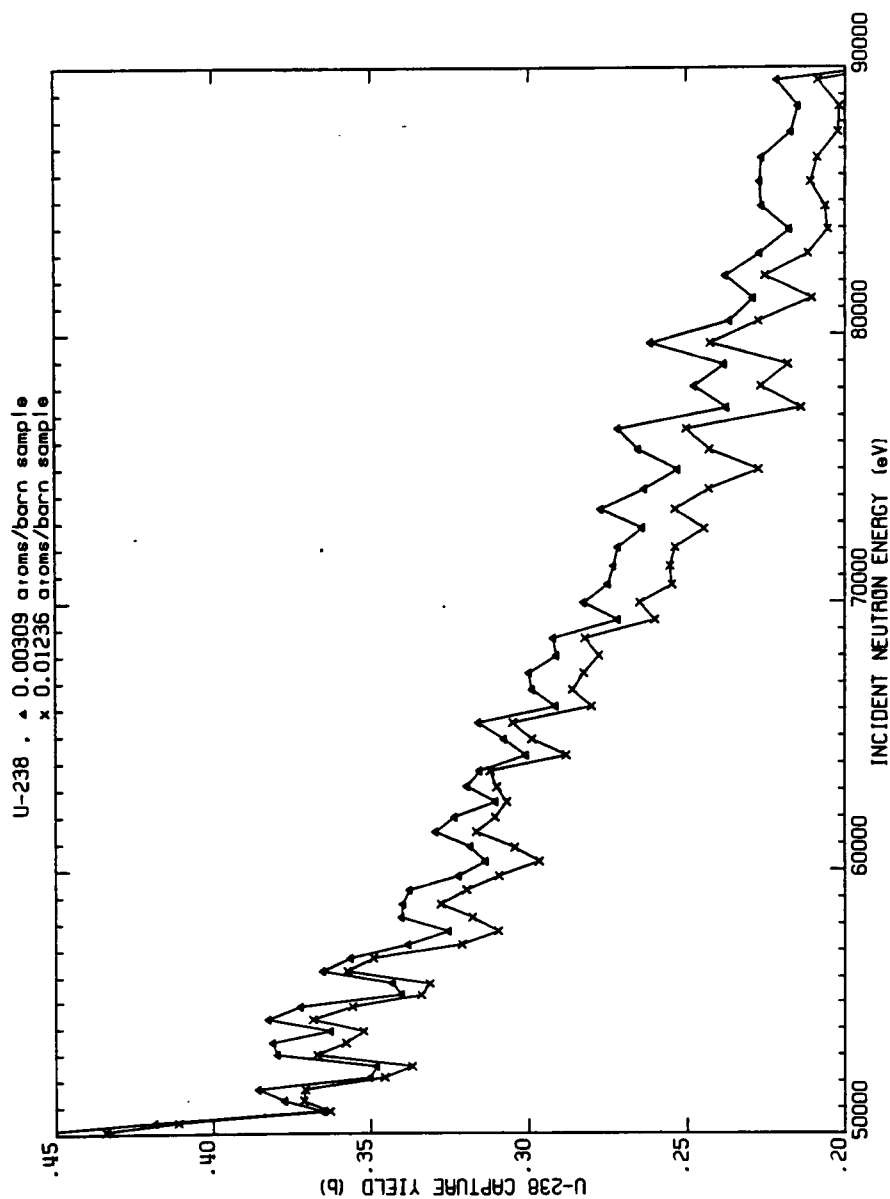


Figure 4-14. Capture Yield Data for the 0.00309 atoms/barn and 0.01236 atoms/barn Samples in the Neutron Energy Range from 50 keV to 90 keV. The data have been averaged over 50-channel intervals. Note the good reproducibility of the observed fine structure between the two samples.

CURVE - U-238, 0.01236 atoms/barn SAMPLE
 LINE - ENDF/B-V EVALUATION

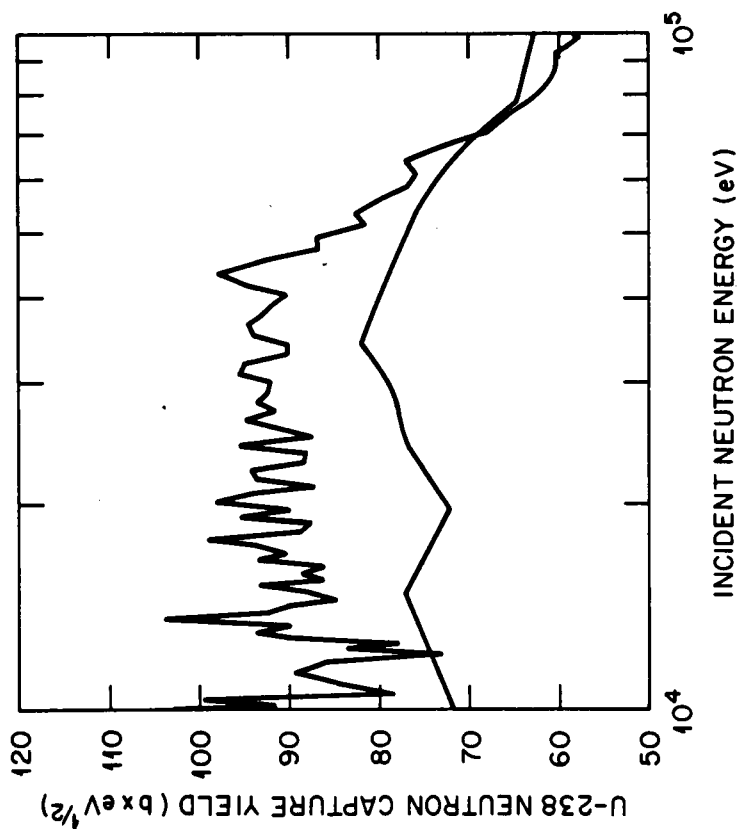


Figure 4-15. Comparison of the Fine Structure Observed in the Capture Yield Data for the 0.00309 atoms/barn Sample with the Structure Introduced in the ^{238}U Capture Cross Section by the ENDF/B-V Evaluation (Reference 34) in the Neutron Energy Range from 10 keV to 100 keV. The data have been averaged over 250-channel intervals. Because of the lack of sample thickness corrections in the capture yield data, the present comparison is only intended to be of a qualitative nature.

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CURVE - U-238, 0.00309 atoms/barn SAMPLE

LINE - ENDF/B-V EVALUATION

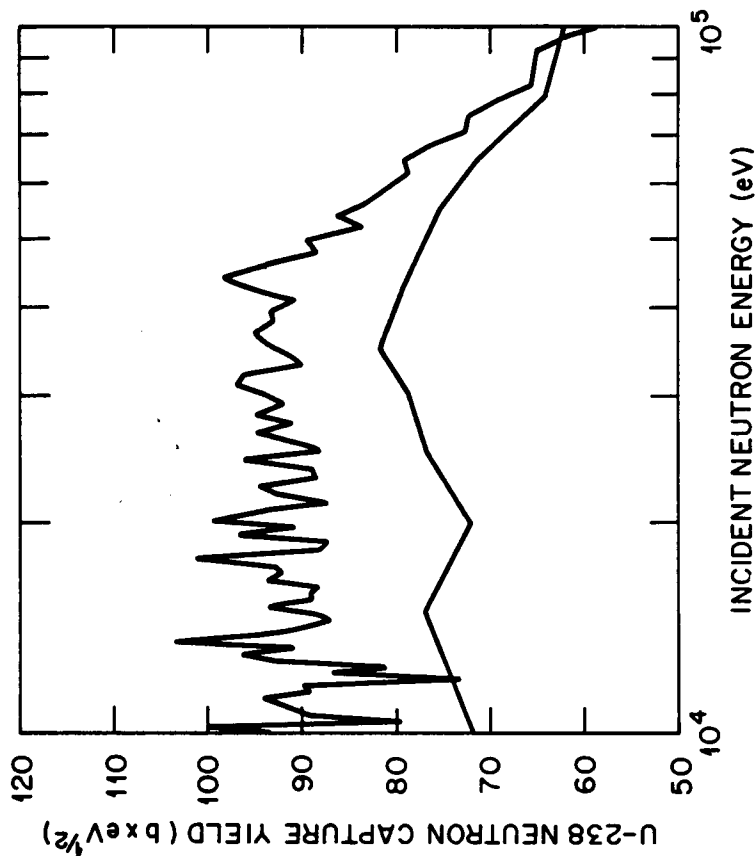


Figure 4-16. Comparison of the Fine Structure Observed in the Capture Yield Data for the 0.01236 atoms/barn Sample with the Structure Introduced in the ^{238}U Capture Cross Section by the ENDF/B-V Evaluation (Reference 34) in the Neutron Energy Range from 10 keV to 100 keV. The data have been averaged over 250-channel intervals. Because of the lack of sample thickness corrections in the capture yield data, the present comparison is only intended to be of a qualitative nature.

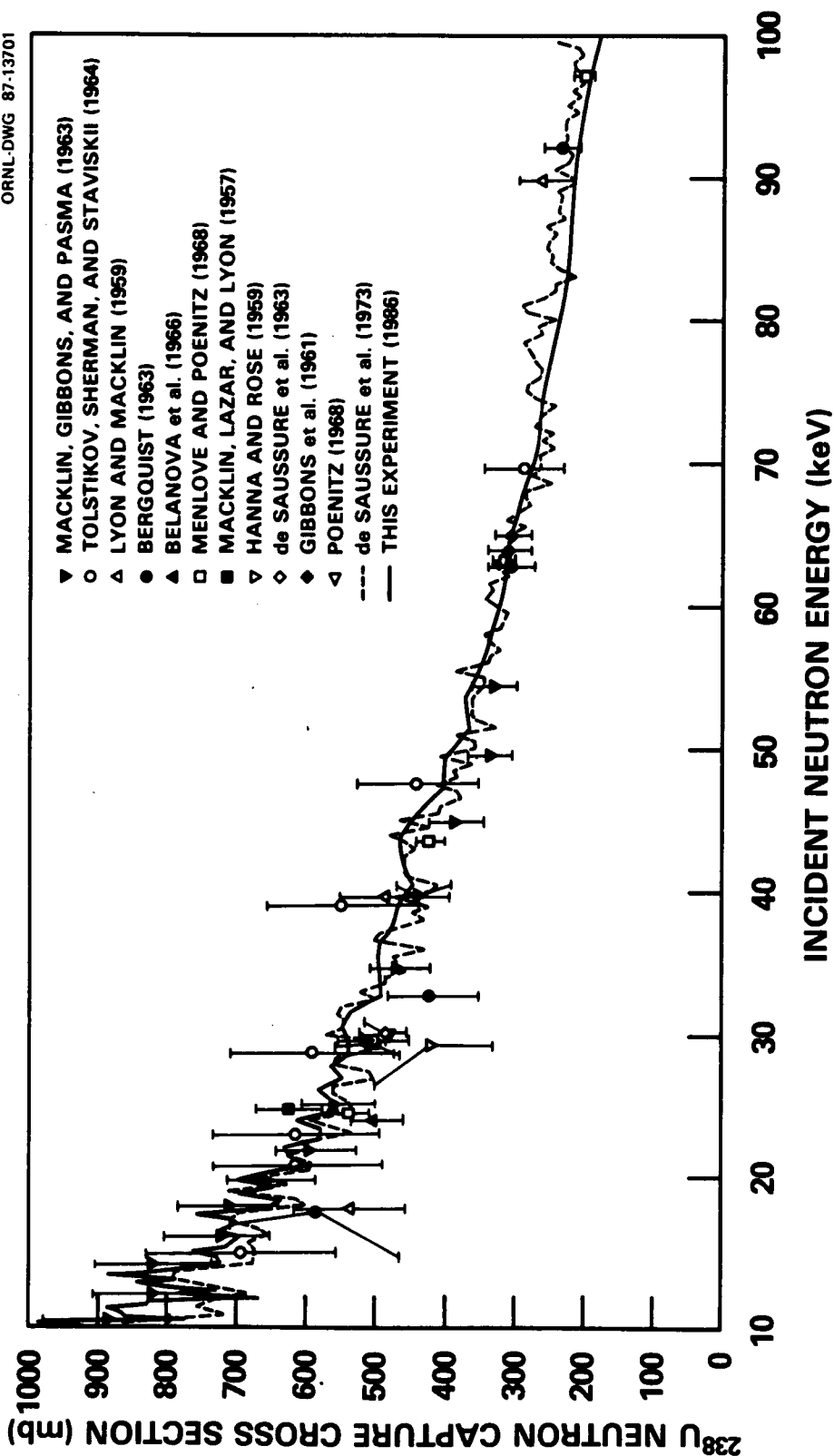


Figure 4-17. Comparison of Various ^{238}U Neutron Capture Cross Section Measurements from Davey's Evaluation (Reference 35) with the Present Capture Yield Data for the 0.00309 atoms/barn Sample in the Neutron Energy Range from 10 keV to 100 keV. The references for the previous data sets shown in the figure are given in Reference 35. The present data have been averaged over 250-channel intervals.

There is a generally good agreement in both shape and magnitude between the various sets of data and the present capture yield data, within the limits of the self-shielding and multiple scattering corrections. A similar comparison is shown in Figure 4-18 for the 0.01236-atoms/barn sample. Note that, in this instance, the capture data for this sample show self-shielding effects above 70 keV. In the energy region between 10 keV and 70 keV, there is an apparent cancellation of self-shielding and multiple scattering effects in the present capture data for both samples, which explains the observed generally good agreement between capture cross section and capture yield data sets.

Perhaps the most exciting result of the present work has been the discovery of many small resonances of the ($^{238}\text{U} + n$) compound nucleus which have not been detected in previous capture or transmission measurements. In addition, multiple splittings of what appeared to be single resonances in previous measurements have been observed. Table 4-3 presents a listing of approximate energies for some 200 resonances which were not seen in the high resolution transmission measurement of Olsen et al.^{13,30} at 150 m in the energy range from 250 eV to 10 keV.

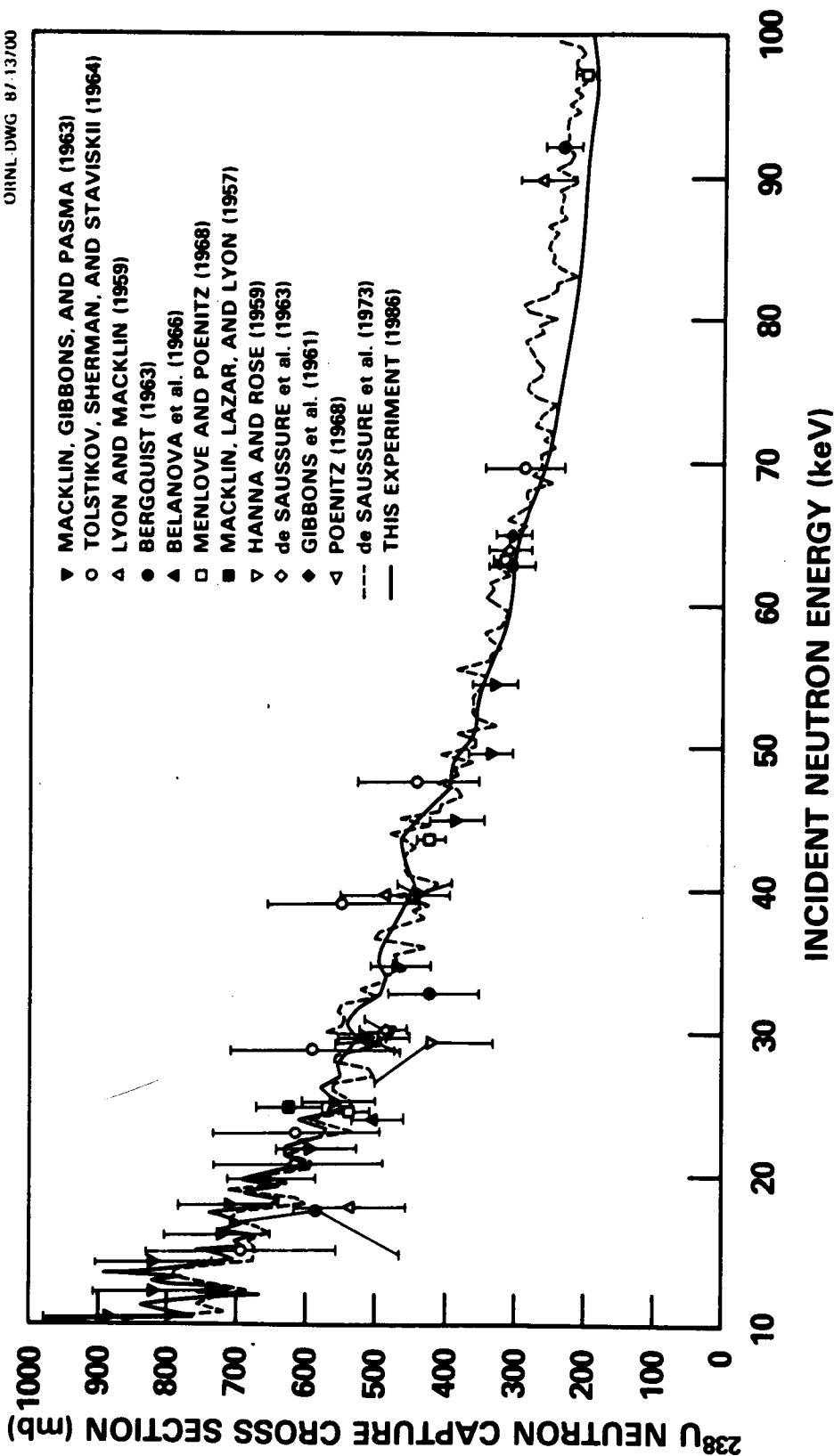


Figure 4-18. Comparison of Various ^{238}U Neutron Capture Cross Section Measurements from Davey's Evaluation (Reference 35) with the Present Capture Yield Data for the 0.01236 atoms/barn Sample in the Neutron Energy Range from 10 keV to 100 keV. The references for the previous data sets shown in the figure are given in Reference 35. The present data have been averaged over 250-channel intervals.

Table 4-3. Preliminary List of Resonances (250 eV to 10 keV) Not Detected in Previous ^{238}U Transmission Measurement¹³ at 150 Meters

Energy (eV)	Energy (eV)	Energy (eV)	Energy (eV)	Energy (eV)	Energy (eV)
734.9	2825.3	4497.6	5703.7	7515.1	8973.2
770.6	2846.4	4529.9	5727.8	7530.1	8978.2
772.3	3048.5	4551.6	5907.6	7583.7	8994.2
800.9	3157.6	4573.5	5910.8	7631.8	9056.0
823.3	3174.0	4584.2	6099.9	7706.4	9060.7
834.2	3210.2	4602.7	6102.4	7707.8	9111.8
907.2	3214.9	4677.1	6334.0	7714.9	9121.8
982.0	3233.0	4716.0	6369.5	7740.0	9123.8
984.2	3257.8	4775.9	6466.0	7745.9	9183.0
1278.5	3413.6	4812.5	6530.1	7793.3	9193.9
1456.4	3447.4	4839.6	6557.9	7795.3	9195.3
1492.2	3471.1	4870.8	6559.8	7820.0	9204.2
1562.6	3506.7	4961.1	6601.0	7865.1	9207.5
1577.6	3542.1	5063.8	6603.7	7892.4	9224.8
1614.1	3547.2	5067.1	6617.8	7917.3 ^a	9228.0
1766.6	3584.0	5095.9	6621.5	8018.6	9291.8
1985.4	3603.5	5110.3	6672.6	8059.3	9311.4
2035.2	3746.5	5128.7	6717.6	8066.7	9339.3
2038.4	3791.1	5150.1	6719.4	8078.0	9344.5
2063.6	3809.1	5152.2	6730.0	8122.7	9516.8
2126.0	3949.5	5177.7	6732.0	8168.7	9540.5
2140.5	3962.4	5180.4	6754.0	8171.6	9567.7
2179.0	3974.8	5327.5	6768.2	8233.3	9716.7
2192.5	3979.5	5329.2	6775.9	8268.0	9721.0
2197.3	3993.2	5330.8	6778.3	8295.9	9783.3
2219.9	4052.3	5441.1	6941.4	8303.1	9784.7
2304.9	4100.9	5443.8	6989.6	8342.2	9810.9
2434.9	4104.9	5567.6	6993.0	8343.8	9813.7
2439.4	4158.1	5569.2	7055.4 ^a	8427.7	9821.6
2452.3	4182.9	5570.5	7076.4	8439.2	9876.3
2468.0	4202.5	5597.6	7110.1 ^a	8442.2	9878.5
2509.5	4236.3	5598.9	7226.2	8520.6	9888.0
2534.5	4245.0	5601.7	7332.8	8621.3	9890.3
2575.8	4270.4	5632.3	7352.4	8643.9	9920.0 ^a
2647.4	4319.5	5635.3	7382.6	8680.1	9938.5 ^a
2654.4	4341.1	5658.2	7387.0	8697.4	9959.9
2688.1	4413.3	5664.1	7469.0	8717.7 ^a	9979.3
2720.5	4426.8	5688.8	7477.0	8796.2	
2732.8	4439.8	5690.6	7490.1	8798.5	
2779.1	4441.7	5702.1	7512.2	8811.1	

^aPossible doublet or multiple.

CHAPTER V

THEORETICAL INTERPRETATION OF THE CAPTURE YIELD DATA

The measured capture yield data are interpreted using the Breit-Wigner resonance formalism which gives the expression below for the capture cross section:

$$\sigma_{n\gamma}^J(E) = \frac{C_0}{E} \sum_{\lambda} \frac{g_J \Gamma_{n\lambda} \Gamma_{\gamma\lambda}}{(E - E_{\lambda})^2 + \left[\frac{1}{2} \Gamma_{T\lambda} \right]^2} \quad (5-1)$$

where the summation goes over the number of resonance peaks, and

- g_J = spin statistical factor = $\frac{2J + 1}{2(2I + 1)}$
- I = spin of the target nucleus
- J = total angular momentum
- $\Gamma_{n\lambda}$ = neutron width (eV)
- $\Gamma_{\gamma\lambda}$ = radiation width (eV)
- E_{λ} = resonance energy (eV)
- $\Gamma_{T\lambda}$ = total width = $\Gamma_{\gamma\lambda} + \Gamma_{n\lambda} + \Gamma_{n\lambda}'$, (eV)
- $\Gamma_{n\lambda}'$ = inelastic neutron width (which has the value zero below the first inelastic channel at 45 keV).

The Breit-Wigner formalism above describes the capture cross section for nuclei where the level-level interference is small (i.e. the ratio of the total level width to the average level spacing is less than unity). This is indeed the case for the ($^{238}\text{U} + n$) compound nucleus.³⁴ However, for Eq. 5-1 to describe the measured capture yields, one has to correct for sample thickness effects and

experimental resolution effects. To this end the least-squares fitting code OLSFIT³⁶ is used. In this program, the capture yield data are divided into regions containing a maximum of sixteen resonances, and the associated neutron energy file is binned into channels of equal energy width. The iteration procedure starts from "educated guesses" for the resonance parameters. The calculated cross section is then corrected for self-shielding and multiple scattering effects, and convoluted with a Gaussian resolution function, giving a theoretical capture yield $Y_{C_{Th}}(\Gamma_n, \Gamma_\gamma, E_\lambda, E)$. Afterwards, the OLSFIT code performs a calculation of the optimal parameter set $(\Gamma_n, \Gamma_\gamma, E_\lambda)$ which minimizes the functional

$$F = \sum_i W_i \left[Y_{C_{Exp}}(E_i) - Y_{C_{Th}}(\Gamma_n, \Gamma_\gamma, E_\lambda, E_i) \right]^2 \quad (5-2)$$

where the summation goes over the energy points in the chosen energy region, and the quantities, W_i , are weight factors, usually taken as the inverse of the variance of the capture yield data. The capture yield data for the neutron energy range from 250 eV to 1 keV have been analyzed using the OLSFIT program. For the resonances seen in the transmission measurement of Olsen et al.,³⁰ their resonance parameters were used as the initial guesses. For resonances not seen in the transmission data, "educated" guesses were utilized, although in some instances parameters were available from the Mughabghab compilation.³⁷

Samples of the excellent theoretical fits to the capture yield data are given in Figures 5-1 and 5-2. Shown in these figures are comparisons between theory and experiment in the neutron energy range

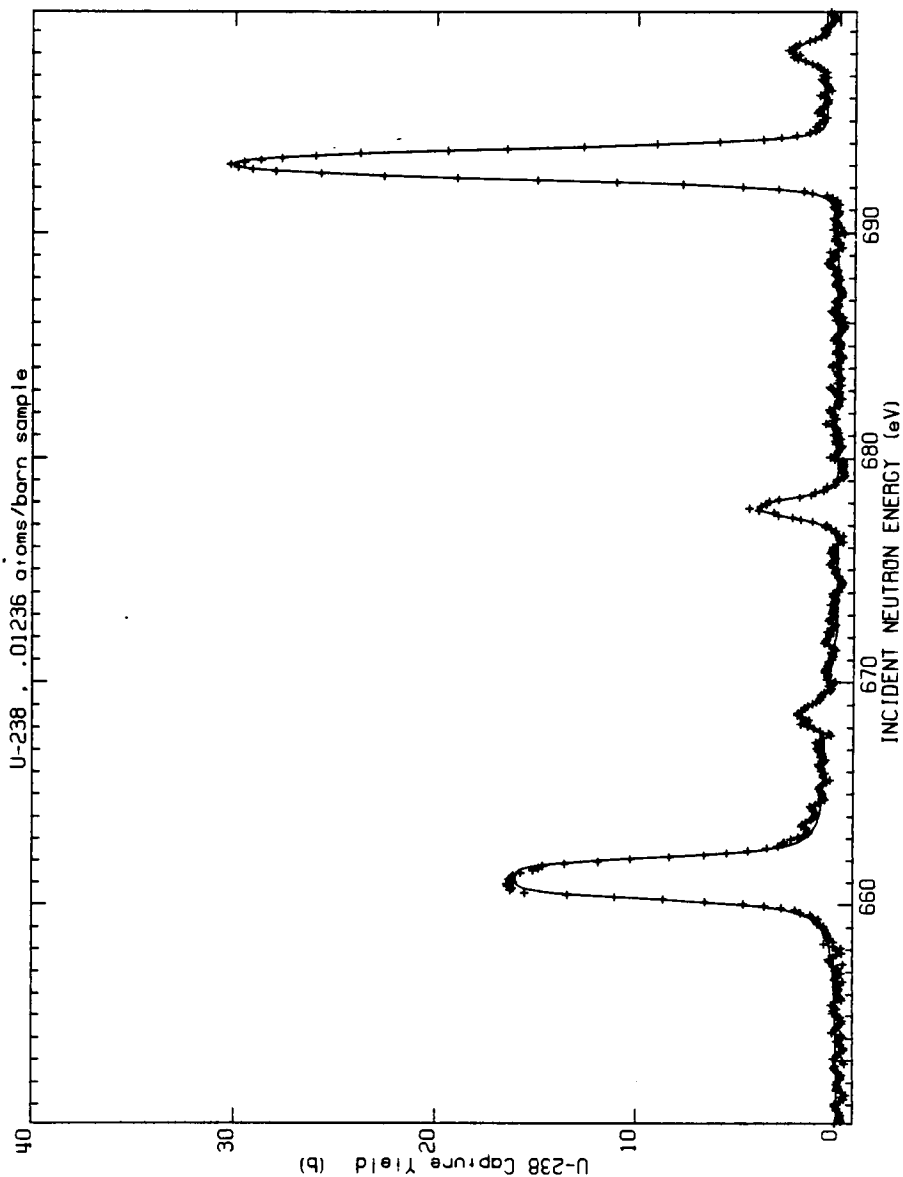


Figure 5-1. Comparison of the Measured Capture Yield Data with the Results of the Resonance Code OLSFIT in the Neutron Energy Range from 650 eV to 700 eV for the 0.01236 atoms/barn Sample. The crosses mark the data and the curve is the least-squares fit.

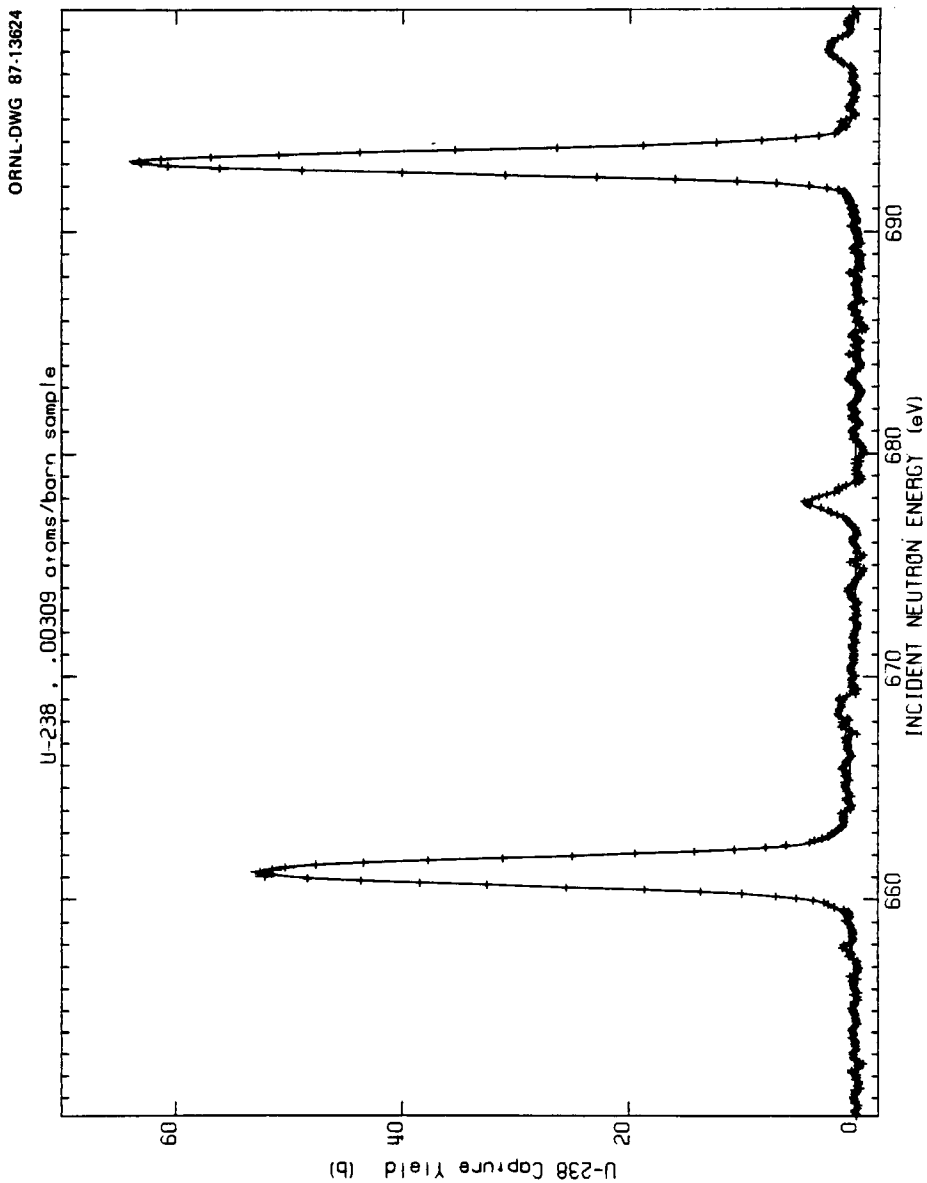


Figure 5-2. Comparison of the Measured Capture Yield Data with the Results of the Resonance Code OLSFIT in the Neutron Energy Range from 650 eV to 700 eV for the 0.00309 atoms/barn Sample. The crosses mark the data and the curve is the least-squares fit.

between 650 eV and 700 eV for the 0.01236 atoms/barn sample and the 0.00309 atoms/barn sample, respectively.

Figures 5-3 and 5-4 show the capture yield data in the neutron-energy region from 2108 eV up to 2208 eV, as well as the theoretical fit obtained from the resonance parameters of Olsen et al.,³⁰ for the 0.01236-atoms/barn and 0.00309-atoms/barn samples, respectively. Notice that the small capture resonances not seen in the transmission measurement are well reproduced in the capture yield data for both ^{238}U samples. Figure 5-5 shows the excellent theoretical fit to the 0.00309-atoms/barn sample data after including all the detected capture resonances.

The theoretical interpretation of capture data in terms of resonance parameters is substantially more difficult than the resonance parameterization of transmission data because of the sample thickness effects in the former. In dealing with capture yield data, the sample thickness effects must be convoluted with the resonance formalism to fit the experimental points. As these sample effects are themselves functions of the resonance parameters and their calculation is made on the basis of simplified models of the multiple scattering processes within the sample, one finds that in many instances the resonance parameters derived from transmission measurements must be iterated on to obtain the theoretical fits to capture data shown here. For this reason, it was not possible to find a set of parameters consistently fitting both the transmission and capture data. Clearly one needs to engage in a substantial, systematic effort combining transmission and

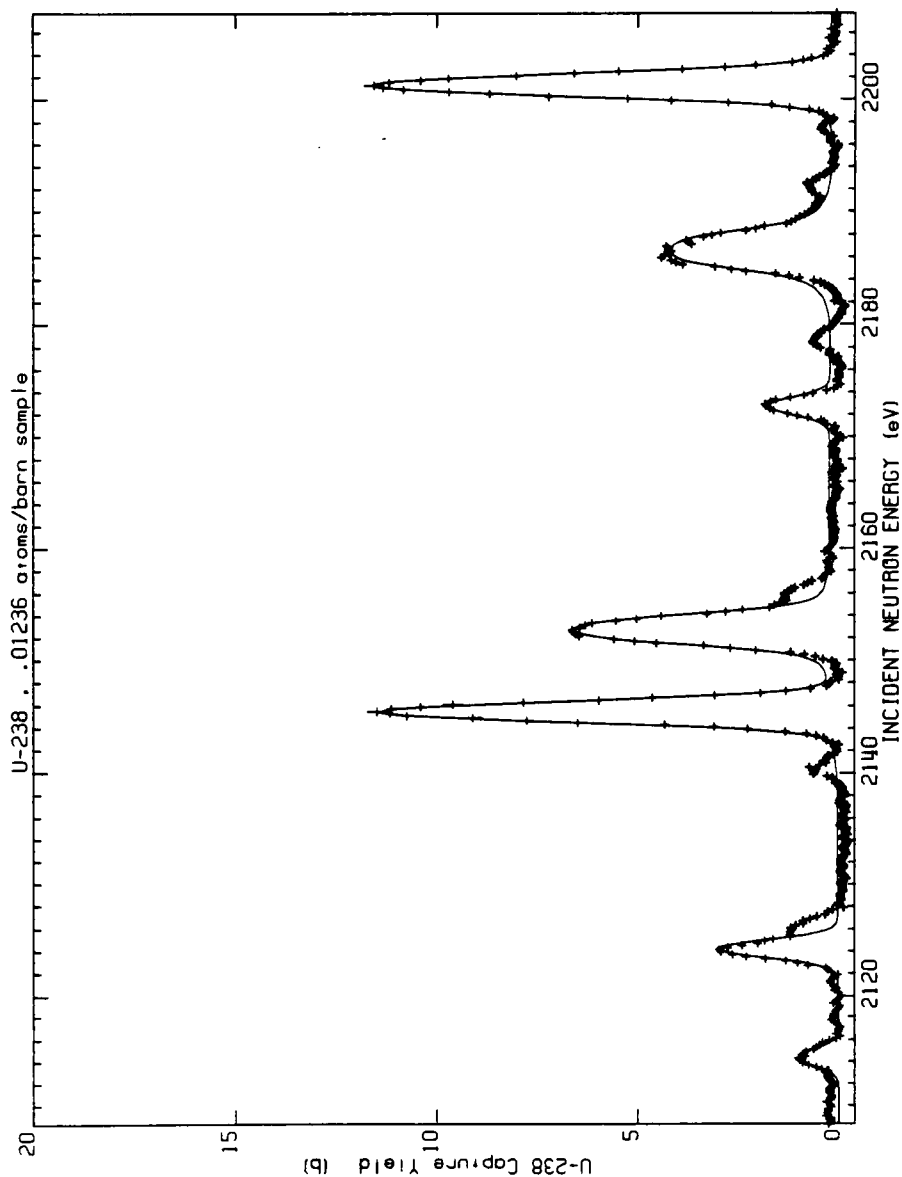


Figure 5-3. Comparison of the Measured Capture Yield Data with the Results of the Resonance Code OLSFIT in the Neutron Energy Range from 2108 eV to 2208 eV for the 0.01236 atoms/barn Sample. The resonance parameters of Reference 30 were used as input parameters. The crosses mark the data and the curve is the least-squares fit. Note that the data exhibit small resonances not seen in the transmission measurement of Reference 13.

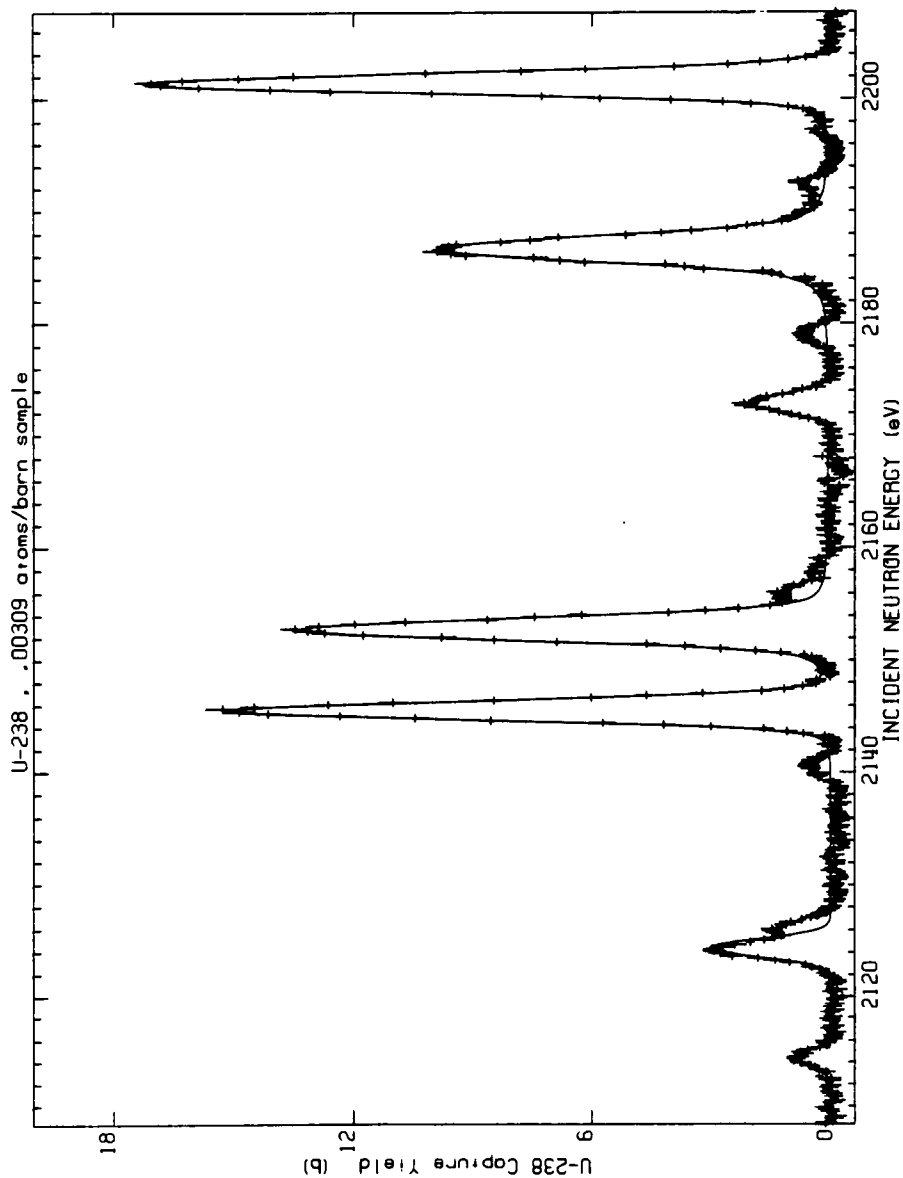


Figure 5-4. Comparison of the Measured Capture Yield Data with the Results of the Resonance Code OLSFIT in the Neutron Energy Range from 2108 eV to 2208 eV for the 0.00309 atoms/barn Sample. The resonance parameters of Reference 30 were used as input parameters. The crosses mark the data and the curve is the least-squares fit. Note that the data exhibit small resonances not seen in the transmission measurement of Reference 13.

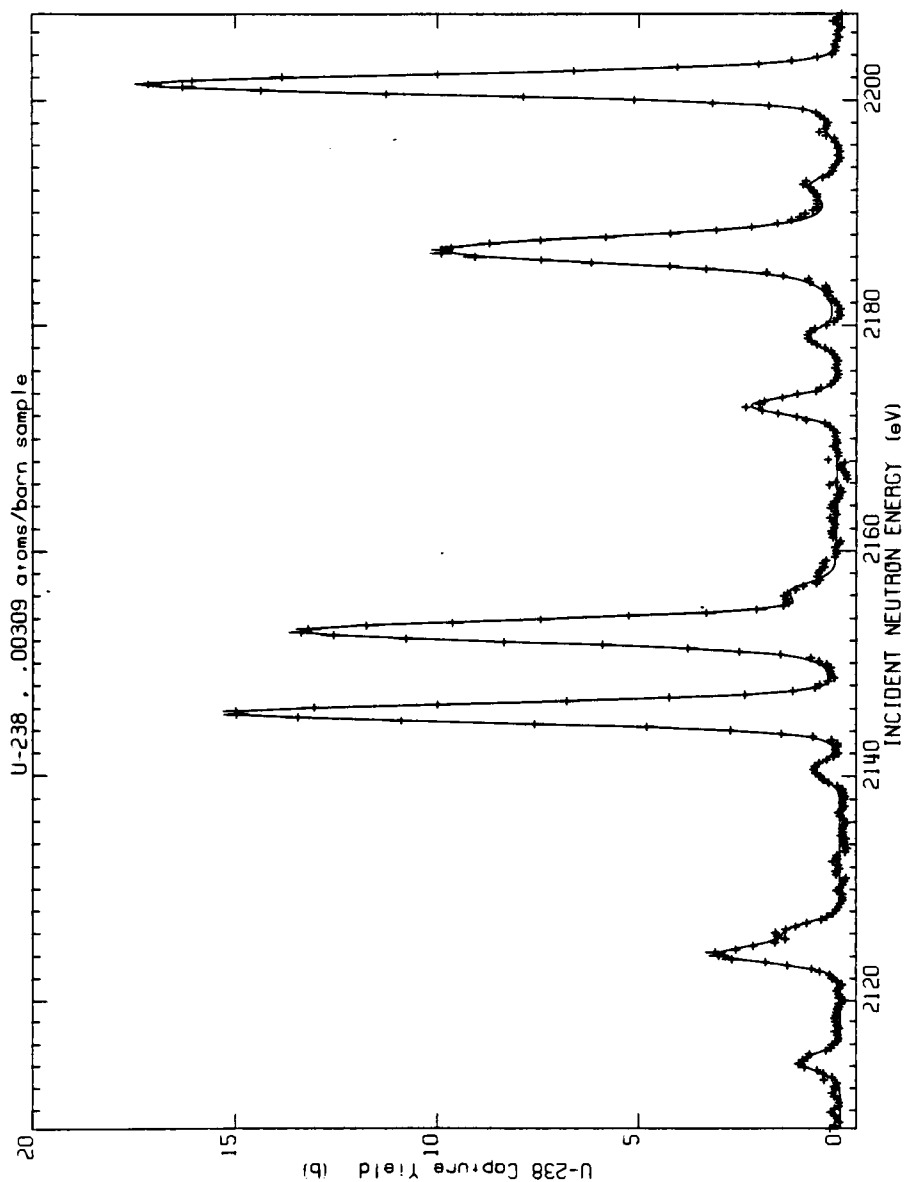


Figure 5-5. Comparison of the Measured Capture Yield Data with the Results of the Resonance Code OLSFIT in the Neutron Energy Range from 2108 eV to 2208 eV for the 0.00309 atoms/barn Sample. The crosses mark the data and the curve is the least-squares fit which, in this case, included the newly discovered small resonances in the theoretical resonance analysis.

capture data obtained from several ^{238}U sample thicknesses, with intercomparison of resonance codes, to ascertain that sample thickness effects are correctly applied. Such a program is outside the scope of this work.

CHAPTER VI

CONCLUSIONS AND RECOMMENDATIONS FOR FUTURE WORK

6.0 CONCLUSIONS

The world's highest energy resolution measurement of the ^{238}U neutron capture yield has been performed using the Oak Ridge Electron Linear Accelerator (ORELA) as the pulsed neutron source.

A powerful technique has been developed for the estimation of the energy-dependent background, which is based on the measurement of the neutron pulse overlap, by performing measurements of the time-of-flight (TOF) spectra at different ORELA pulse rates.

A careful analysis has been performed of the pulse-height distribution of the gamma-ray detector (ORELAST) counts, with the purpose of ascertaining the effect of the energy dependence of the gamma-ray cascades and of possible electronic bias shifts at the ORELAST on the TOF spectra. To this end, the pulse-height spectrum was divided into five bins, and each one of the TOF spectra was separately analyzed. It was found that the different signal-to-background ratios exhibited by the TOF spectra in each pulse-height bin were sufficient to explain the discrepancies among the TOF spectra corresponding to the various pulse-height bins. Hence, any effect of gamma-ray cascade changes with neutron energy, resonance spin and parity, or of electronic bias shifts are probably masked by the uncertainties associated with the background subtraction process.

No attempt has been made to correct the present capture yield data for sample thickness effects. Such correction (necessary to

derive capture cross section) would require a consistent set of resonance parameters, which are not as yet available, for both the resolved and unresolved resonance energy regions. In the past, sample thickness corrections have been performed with inconsistent parameter sets and approximate calculational algorithms which introduced additional uncertainties to the data. These uncertainties resulted in a situation where the discrepancies among various sets of ^{238}U neutron capture measurements were larger than their quoted experimental uncertainties.⁹ In view of this, it was concluded that the neutron cross-section technology community would be better served by providing high energy resolution capture yield data for two largely different sample thicknesses. These data can then be used as a testing ground for resonance parameter sets and calculational algorithms now in use for reactor design. In this regard, the present data have been released to (and are currently in use by) both the ^{238}U International Task Force and the National Nuclear Data Center at Brookhaven National Laboratory.

Some 200 small resonances of the ($^{238}\text{U} + n$) compound nucleus have been discovered which have not been detected in transmission measurements, in the energy range from 250 eV to 10 keV. These results will allow the development of improved resonance parameter sets, particularly when combined with high resolution ^{238}U transmission measurements currently in progress at ORELA.

The high energy resolution achieved in this measurement frequently resulted in doublet and triplet splittings of what appeared to be single resonances in previous measurements. This resolution should

allow extension of the resolved resonance energy region in ^{238}U from the present 4 keV limit up to 15 or 20 keV incident neutron energy.

A substantial amount of fine structure has been detected above 20 keV which is very well reproduced in the capture yield measurements of two largely different sample thicknesses of ^{238}U . Addition of this fine structure to future generations of the ENDF/B evaluations will allow improved calculation of self-shielded group cross sections and Doppler coefficients of reactivity, which are important parameters for the design of both thermal and fast fission reactors, as well as possible "blanketed" fusion reactors of the future.

6.1 RECOMMENDATIONS FOR FUTURE WORK

Although the present ^{238}U capture yield measurement was performed using one of the world's most advanced pulsed neutron sources, in combination with a large liquid scintillator tank as the capture gamma-ray detector, it is very likely that advancements in gamma-ray spectroscopy techniques will allow improved measurements in the future. With the benefit of experimental experience and hindsight, however, there is also room for improvement in some of the data-taking and data analysis procedures used in the methodology of the present work. In particular:

1. The boundaries of the pulse-height bins could have been more carefully chosen to improve the signal-to-background ratios of the TOF spectra. (Among the five pulse-height groups used in the present experiment, the TOF spectra of only two groups exhibited sufficiently high signal-to-background ratios to be included in the final results. Implementation of such improved pulse-height

bin choices would not have decreased the accelerator operating time necessary to achieve the same number of ^{238}U capture events for each sample thickness; however, more detailed information on the energy distribution of capture gamma-ray cascades would have resulted.)

2. The calculation of constants for the background subtraction procedure could be made easier and more accurate by utilizing minimization of the cross-correlation function between the foreground and the shape of the energy-dependent background obtained from the low frequency TOF spectrum.
3. By recording the TOF spectra for the single capture events in each side of the ORELAST detector, one could check on the sensitivity of the detector efficiency to the energy dependence of the capture gamma-ray cascades.

It is also suggested that capture yield measurements should be performed with a larger variety of ^{238}U sample thicknesses. This information would be useful in the development and validation of numerical codes for the calculation of sample thickness effects.

The present high-resolution ^{238}U capture yield data should be parameterized in terms of the resolved resonance R-matrix formalism up to 15 keV, and perhaps up to 20 keV, by performing simultaneous resonance parameter fits to high resolution transmission data. Clearly such calculations would involve a concerted effort by several groups and are beyond the scope of this work.

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APPENDIX

SAMPLE SIZE CORRECTIONS FOR WEAK CAPTURE RESONANCES IN ^{238}U
IN THE NORMALIZATION REGION (600 eV - 1100 eV)

SAMPLE SIZE CORRECTIONS FOR WEAK CAPTURE RESONANCES IN ^{238}U
IN THE NORMALIZATION REGION (600 eV - 1100 eV)

This appendix elaborates on the neglect of sample size corrections in the calculation of the normalization constant (see Section 3.5). For weak capture resonances (i.e., $g\Gamma_n \ll \Gamma_\gamma$), the capture probability is proportional to the neutron width. Hence the probability of capturing those neutrons which reached the resonance energy after several collisions in the sample is indeed very small. Thus, it appears justifiable to neglect multiple scattering corrections in weak capture resonances. The self-shielding factor, S_S , (see Eq. 4-2) is large even for small resonances, due to the potential cross section contribution, σ_p , to the total neutron cross section, $\sigma_T(E)$. However, the potential cross section is constant over a large neutron energy range, and its contribution to the resonance self-shielding can be included in the normalization constant.

By writing

$$\sigma_T(E) = \sigma_p + \sigma_c(E) \quad (\text{A-1})$$

where

$\sigma_c(E)$ = capture cross section (eV),

the expression for the self-shielding factor can be rewritten in the form

$$\frac{S_S}{S_{S_p}} = \left(1 + \frac{\langle \sigma_c \rangle}{\sigma_p} \right)^{-1} + \frac{e^{-N\sigma_p}}{S_{S_p}} \left[\frac{\langle \sigma_c \rangle / \sigma_p}{1 + \frac{\langle \sigma_c \rangle}{\sigma_p}} \right] \quad (\text{A-2})$$

where

N = sample areal density (atoms/barn)

and the self-shielding factor for potential scattering is given by

$$S_{S_p} = \frac{\left(1 - e^{-N\sigma_p} \right)}{N\sigma_p} \quad (\text{A-3})$$

In Eq. A-2, one must also define

$\langle \sigma_c \rangle$ = average capture cross section over the ^{238}U resonance

and make the approximation (for $N\sigma_c \ll 1$) that

$$e^{-N\langle \sigma_c \rangle} = 1 - N\langle \sigma_c \rangle \quad (\text{A-4})$$

For a ^{238}U capture resonance with $g\Gamma_n = 1.06$ meV and $\Gamma_\gamma = 23.6$ meV, the value $\langle \sigma_c \rangle$, calculated as half the peak cross section is equal to 3.605 barns. With a value of $\sigma_p = 9.38$ barns for the ^{238}U nucleus, one obtains the following values for the ratio of the overall self-shielding factor, S_S , to the potential scattering self-shielding factor, S_{S_p} :

1. Thick sample ($N = 0.01236$ atoms/barn)

$$\frac{S_S}{S_{S_p}} = 0.984$$

and

2. Thin sample ($N = 0.00309$ atoms/barn)

$$\frac{S_S}{S_{S_P}} = 0.996 \quad .$$

Thus the energy-dependent part of the self-shielding factor is less than 2% for the thick sample and only a fraction of 1% for the thin sample. In fact, these corrections are upper bounds, as the range of neutron widths for the particular set of resonances used in the normalization procedure (see Tables 3-3 and 3-4) was from 0.44 to 0.96 meV.

VITA

Roger Lawrence Macklin was born in Oak Ridge, Tennessee, on May 19, 1949, and attended Cedar Hill Elementary School in that city. His family spent one year in England, during which time he was enrolled at John Mason High School in Abingdon, Berkshire. Upon returning to the United States, he attended Jefferson Junior High School in Oak Ridge, Tennessee, and Oak Ridge High School. He participated in a National Science Foundation summer institute in physics at Cornell University, Ithaca, New York, in 1966 and received the Bell Telephone Science Award in 1967. He was graduated from Oak Ridge High School in June 1967.

He enrolled at Yale University, New Haven, Connecticut, in September 1967, majoring in physics. He was subject to the Vietnam-era military draft lottery, chose to reject his student deferment status, and received conscientious objector classification from the local draft board upon his return to Tennessee. Although he did not pass the physical examination required for alternate service, he performed volunteer work at the Oak Ridge Regional Mental Health Center while attending the University of Tennessee in Knoxville on a part-time basis. To satisfy his curiosity, he took advantage of the situation to pursue a wide-range program with exposure to topics of interest in the humanities and social sciences, as well as the natural sciences. He received the Bachelor of Arts degree in physics from the University of Tennessee in March 1980.

He accepted a graduate research assistantship with the University of Tennessee Nuclear Engineering Department in 1984, performing research at the Oak Ridge National Laboratory in collaboration with staff members at the Oak Ridge Electron Linear Accelerator. He represented the University of Tennessee in the 1986 American Nuclear Society Student Design Competition, Graduate Division, for which his team achieved the second-place national ranking. Mr. Macklin was awarded the Master of Science degree in Nuclear Engineering in August 1987.