Battelle

Pacific Northwest Laboratories
Richland, Washington 99352

AEC Research and Development Report

COMPARISON OF ALTERNATE
NONDESTRUCTIVE TECHNIQUES
FOR THE DETERMINATION
OF ^{238}U REACTION RATE RATIOS

January 1972

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately-owned rights.

PACIFIC NORTHWEST LABORATORY
operated by
BATTELLE
for the
U.S. ATOMIC ENERGY COMMISSION
Under Contract AT(45-1)-1830

3 3679 00062 1070

BNWL-1637

~~Special Distribution~~

COMPARISON OF ALTERNATE NONDESTRUCTIVE
TECHNIQUES FOR THE DETERMINATION
OF ^{238}U REACTION RATE RATIOS

By

Dale Robert Oden, Jr.

Reactor Physics Department

January 1972

BATTELLE
PACIFIC NORTHWEST LABORATORIES
RICHLAND, WASHINGTON 99352

TABLE OF CONTENTS

LIST OF FIGURES	iv
LIST OF TABLES	v
ACKNOWLEDGEMENTS	vi
ABSTRACT	vii
INTRODUCTION	1
BACKGROUND	4
DESCRIPTION OF EXPERIMENTS	10
A. Experimental Facilities	10
1. Plutonium Recycle Critical Facility (PRCF).	10
2. Thermal Test Reactor (TTR)	11
B. Lattice and Fuel Specifications	15
C. Foil Fabrication	15
D. Foil Arrangements	16
1. Lattice	16
2. Thermal Column	20
E. Foil Irradiations	20
DETERMINATION OF ²³⁹ Np ACTIVITY	23
A. NaI-NaI Coincidence Technique	23
1. Introduction	23
2. Description of Counting System	29
3. Data Accumulation, Reduction and Analysis	33
B. NaI Singles Technique	39
1. Introduction	39
2. Description of Counting System	40

3. Data Accumulation, Reduction and Analysis	41
C. Ge(Li) Singles Technique	47
1. Introduction	47
2. Description of Counting System	48
3. Data Accumulation, Reduction and Analysis	50
SYSTEMATIC ERRORS AND CORRECTIONS	58
A. Perturbations Due to the Presence of Cadmium in the Lattice - The Thermal Activation Technique	58
B. The Use of Fuel Spacers Inside the Cadmium Box	64
C. The Use of Uranium Metal Rather Than Oxide Foils	65
D. Thermal Flux Peaking and Dipping in Lattice and Thermal Column Foils	66
RESULTS AND CONCLUSIONS	68
COMPARATIVE EVALUATION OF THE THREE COUNTING TECHNIQUES	74
1. NaI Singles Counting	75
2. NaI-NaI Coincidence Counting	76
3. Ge(Li) Singles Counting	77
REFERENCES	79
APPENDICES	
APPENDIX A CORRECTIONS FOR RANDOM COINCIDENCES	84
APPENDIX B CORRECTIONS FOR COINCIDENCE LOSSES	88
APPENDIX C MEASUREMENT OF COINCIDENCE RESOLVING TIME	91
APPENDIX D DESCRIPTION OF CALCULATIONS AND MODELS USED	94

LIST OF FIGURES

1	Schematic Diagram of the Thermal Test Reactor	13
2	Foil Arrangements in the Fuel Rod	18
3	Decay Scheme of ${}_{93}\text{Np}^{239}$	25
4	γ - and X-Ray Spectrum from an Irradiated Depleted Uranium Foil	26
5	Schematic Diagram of the Coincidence Counting System	30
6	NaI Singles and Coincidence Spectra for Irradiated Depleted and Enriched Uranium Foils	35
7	Schematic Diagram of the Ge(Li) Counting System	49
8	Typical γ -Ray Spectra for Irradiation Highly Depleted (Lower Spectrum) and Highly Enriched (Upper Spectrum) Uranium Foils [20 cc Ge(Li) Detector]	51
9	γ - and X-Ray Spectrum from an Irradiated Depleted Uranium Foil from 0 to 200 keV	52
10	Comparison of γ - and X-Ray Spectra from an Irradiated Depleted Uranium Foil Using 20 cc (Upper Spectrum) and 0.4 cc (Lower Spectrum) Ge(Li) Detectors	55

LIST OF TABLES

1	Fuel and Clad Specifications	15
2	Summary of Foils Irradiated	21
3	Energies of Photopeaks in Figure 4	27
4	Typical Counting Cycle for NaI Coincidence and Singles Counting	42
5	Source and Energy of Photopeaks in Figure 8	54
6	Quantities Used in the Determination of ρ^{28} by the Thermal Activation Technique	64
7	Summary of ^{239}Np Specific Activities of Depleted Uranium Foils for Three Counting Techniques	69
8	Summary of Measured and Calculated Values for the ^{238}U Cadmium Ratio and ρ^{28}	71

ACKNOWLEDGEMENTS

I wish to thank Dr. Gene L. Woodruff who directed this work and Dr. Loren C. Schmid who suggested and supervised this research project.

The assistance of Mr. J. E. Choate in setting up, calibrating, and maintaining the operation of the NaI singles and coincidence spectrometer was invaluable. I am also indebted to Mr. D. L. Haggard for providing the same functions for the Ge(Li) spectrometer used in this research.

I would also like to thank Dr. A. J. Haverfield, both for allowing me to use his Ge(Li) spectrometers and for his helpful discussions and suggestions concerning their use.

ABSTRACT

The generally large disagreement which exists between measured and calculated values of ρ^{28} (the ratio of episcadmium to subcadmium neutron capture in ^{238}U) as well as that between measured values using different experimental techniques, suggests the possible presence of systematic experimental errors in these measurements. One possible source of systematic experimental error in these measurements lies in the counting technique for the determination of the activation product (^{239}U , ^{239}Np , or ^{239}Pu) activity. Unless chemical separations are performed, this activity must be determined in the presence of an interfering activity due to fission product disintegrations. The success of a given counting technique will then depend upon its ability to properly segregate the activation product activity from that due to fission products.

In order to determine if systematic biases due to the mode of counting are present in these measurements, three counting techniques for the determination of ^{239}Np disintegration rates were compared in the measurement of ^{238}U reaction rate ratios in a water moderated lattice of UO_2 rods. Depleted uranium foils, irradiated bare and cadmium covered in the fuel rod, were counted alternately on a NaI singles spectrometer, a NaI-NaI coincidence system, and via a new technique employing a 20 cc Ge(Li) singles spectrometer.

Although the three counting methods yielded the same cadmium ratio results within experimental uncertainties ($\pm 1\%$) (indicating the absence of a systematic bias due to the mode of counting in the present experiment) comparisons on the basis of several other criteria indicated the NaI-NaI

coincidence method to be presently the best overall. However, these experiments showed the promise that the relatively simple Ge(Li) singles spectrometry method could, with a little development, be demonstrated to be as good or better than the NaI-NaI coincidence system on an overall basis. The efficiency and energy resolution of the 20 cc Ge(Li) detector used in these experiments were found to be adequate for this application and it was recommended that neither be increased at the expense of a reduction in the other.

In addition, values of ρ^{28} obtained from the direct measurement of the ^{238}U cadmium ratio were compared to those obtained from the thermal activation technique to determine the perturbation due to the presence of cadmium in the lattice. Measured values were also compared to calculated values from two analytical models. The effect of fuel spacers inside the cadmium box, used in the fuel rod for the prevention of epithermal neutron streaming from the moderator through the cadmium buttons and into the foils, was also measured.

COMPARISON OF ALTERNATE NONDESTRUCTIVE
TECHNIQUES FOR THE DETERMINATION
OF ^{238}U REACTION RATE RATIOS

Dale Robert Oden, Jr.

INTRODUCTION

The nature of resonance neutron capture in heterogeneous assemblies presents a considerable obstacle to the accurate measurement of ^{238}U reaction rates. Although many different experimental techniques have been developed, and variations upon variations have been applied, there is as yet no conclusive evidence as to which technique is entirely satisfactory. The generally large disagreement which has been observed⁽¹⁻⁴⁾ between measured and calculated values of ρ^{28} (the ratio of epithermal to subcadmium neutron capture in ^{238}U) as well as that between measured values using different experimental techniques,^(2,3,5,6) have suggested the presence of systematic experimental errors as well as biases in the analytical models. As a result measurements of ^{238}U reaction rates in UO_2 fueled, light water moderated lattices still have more doubts and uncertainties associated with them than do any of the other standard neutronic parameters.

One possible source of systematic experimental error in these measurements lies in the counting technique for the determination of the activation product activity (^{239}U , ^{239}Np or ^{239}Pu). Unless chemical separations are performed, this activity must be determined in the presence of an interfering activity due to fission product disintegrations. Several counting techniques which have been applied include:

1. The determination of the 23.5 minute half-life activity of ^{239}U via
 - β -ray detection⁽⁷⁾
 - γ -ray detection⁽⁸⁻¹⁰⁾

2. The determination of the 2.35 day half-life activity of ^{239}Np via
 - β -ray detection⁽¹¹⁾
 - γ -ray detection⁽¹²⁻¹⁴⁾
 - β - γ delayed coincidence detection⁽¹⁵⁾
 - γ - γ coincidence detection⁽¹⁶⁾
 - γ -X-ray coincidence detection^(12,17-20)
3. The determination of the 2.44×10^4 year half-life activity of ^{239}Pu via
 - α particle detection⁽²¹⁾

It is the purpose of this present work to compare a few of these counting techniques to determine if systematic biases due to the mode of counting are present, and to recommend which methods are best suited for a particular situation. The methods utilized in this study were restricted to nondestructive techniques employing the detection of γ and X-rays resulting from the decay of ^{239}Np to ^{239}Pu . These methods seem to be the most popular in the literature due to their relative ease of application.

The three counting techniques chosen to be compared are:

1. Detection of the ^{239}Np 106 keV γ -ray* via NaI singles spectrometry.
2. Detection of coincidences between ^{239}Np γ -rays and ^{239}Pu X-rays via NaI-NaI coincidence spectrometry.

* Note that although the 106 keV γ -ray results from an energy transition between two excited states of a plutonium nucleus (see Figure 3) it is referred to as a neptunium γ -ray. Identification of a γ -ray with the parent (Np) rather than the daughter (Pu) nucleus is a standard practice but it does sometimes lead to confusion. No such ambiguity exists when referring to X-rays since only atomic transitions are involved here with the initial and final nucleus being one and the same.

3. Detection of the ^{239}Np 106 keV γ -ray and ^{239}Pu 103 keV X-ray via Ge(Li) singles spectrometry.

The latter method is a new approach and to the author's knowledge has never been successfully attempted previously.

Two other possible sources of systematic experimental error were also investigated. The first, dealing with possible perturbations caused by the presence of cadmium in the lattice, was studied by comparing results obtained by direct ^{238}U cadmium ratio measurements to those obtained via the "thermal activation technique" which does not require a cadmium covered ^{238}U activation in the lattice. The second possible source of systematic error studied involved the use of fuel pellets inside the cadmium box in the lattice irradiation.

BACKGROUND

Radiative neutron capture in ^{238}U plays a basic role in determining the physics characteristics of both thermal and fast reactors. Its influence on core design derives primarily through its effect on lattice reactivity (e.g., critical mass) and neutron economy (e.g., ^{239}Pu production rate). Definitions of several quantities which can be measured to aid in quantifying these effects are given in this section. Some considerations associated with making ^{238}U activation measurements are also indicated.

Thermal neutron capture in ^{238}U represents 5 to 15% of the total thermal neutron captures in typical slightly enriched uranium fueled, light water moderated, thermal power reactor lattices. The resonance neutron capture rate in ^{238}U , however, represents essentially all of the epithermal capture in these lattices and is typically one to four times the thermal capture rate in ^{238}U . Most of these nonfission captures during slowing down occur in the large ^{238}U resonance capture peaks at neutron energies greater than 6 eV. Since most of the power reactor lattices of interest today have a characteristically low moderator to fuel volume fraction, and hence a relatively high proportion of epithermal capture and fission events, an accurate accounting of these events is necessary for reliable prediction of lattice physics characteristics.

In thermal production and fast breeder type reactors the importance of ^{238}U capture is best illustrated through the conversion and breeding ratios respectively. These ratios basically relate fuel produced (^{239}Pu from neutron capture in ^{238}U) to fuel destroyed (^{235}U or ^{239}Pu through fission and capture). In thermal power reactors, capture rates in ^{238}U are then directly applicable to core design through determination of the

conversion ratio. Since the attainable fuel burnup is sensitive to the initial conversion ratio, an accurate knowledge of this ratio is imperative in predicting fuel economy.

Accurate measurements of ^{238}U capture rates in reactors are, therefore, necessary to provide data with which to check theoretical models for predicting these quantities. In addition to testing the ability of a theoretical model to predict the correct spectrum, these measurements can also provide a check on the accuracy of input cross sections and resonance parameters.

Measurements of ^{238}U reaction rates in lattices are usually performed on a relative rather than absolute basis due to the comparative simplicity with which the former measurements can be made. What is usually quoted as a result of these measurements is then a ratio of reaction rates. Several forms of these measured reaction rate ratios have been quoted in the literature. Definitions of each of the most common forms is given below with the symbol or abbreviation commonly associated with each.

$$\begin{aligned} C^{28} &= {}^{238}\text{U} \text{ cadmium ratio} \\ &= \frac{\text{Total capture in } {}^{238}\text{U}}{\text{Epicadmium capture in } {}^{238}\text{U}} \end{aligned}$$

$$\begin{aligned} \rho_c^{28} &= \frac{1}{C^{28} - 1} \\ &= \frac{\text{Epicadmium capture in } {}^{238}\text{U}}{\text{Subcadmium capture in } {}^{238}\text{U}} \end{aligned}$$

$$C^* = \text{MCR} = \text{Modified conversion ratio}$$

$$= \frac{\text{Total capture in } {}^{238}\text{U}}{\text{Total fissions in } {}^{235}\text{U}} = \left[\frac{\hat{\Sigma}_c^{28}}{\hat{\Sigma}_f^{25}} \right] \cdot \left[\frac{1 + \rho_c^{28}}{1 + \rho_f^{25}} \right]$$

where $\rho_f^{25} \equiv \frac{1}{C_{25}-1}$ and $C_{25} \equiv {}^{235}\text{U}$ fission cadmium ratio.

RMCR = Relative modified conversion ratio

$$\begin{aligned} & \frac{\text{Total } {}^{238}\text{U} \text{ capture in lattice}}{\text{Total } {}^{235}\text{U} \text{ fission in lattice}} \\ &= \frac{\text{Total } {}^{238}\text{U} \text{ capture in thermal spectrum}}{\text{Total } {}^{235}\text{U} \text{ fission in thermal spectrum}} \end{aligned}$$

$$\text{Note that } C^* = \text{RMCR} \left[\frac{\hat{\Sigma}_a^{28}}{\hat{\Sigma}_a^{25}} \right]_{\text{thermal}}$$

C = ICR = Initial conversion ratio

$$\begin{aligned} &= \frac{\text{Total } {}^{238}\text{U} \text{ capture in lattice}}{\text{Total } {}^{235}\text{U} \text{ capture in lattice}} \\ &= \frac{\text{MCR}}{(1+\alpha^{25})_{\text{lattice}}} = \text{RMCR} \left[\frac{\hat{\Sigma}_a^{28}}{\hat{\Sigma}_a^{25}} \right]_{\text{thermal}} \cdot \left[\frac{(1+\alpha^{25})_{\text{thermal}}}{(1+\alpha^{25})_{\text{lattice}}} \right] \end{aligned}$$

P = Resonance escape probability

$$= \frac{1}{1+\rho_c^{28} fF}$$

In the preceding

α^{25} = Capture to fission ratio of ^{235}U

$\hat{\Sigma}_a^{28}$ = Average macroscopic absorption cross section of ^{238}U
in thermal spectrum

$\hat{\Sigma}_a^{25}$ = Average macroscopic absorption cross section of ^{235}U
in thermal spectrum

f = Thermal utilization $\equiv \frac{\text{Total sub-cadmium absorption in the fuel}}{\text{Total sub-cadmium absorption in the cell}}$

$F = \hat{\Sigma}_c^{28} / (\hat{\Sigma}_c^{28} + \hat{\Sigma}_a^{25}) \equiv \frac{\text{Sub-cadmium capture rate in } ^{238}\text{U}}{\text{Total sub-cadmium absorption in the fuel}}$

$\bar{\Sigma}_c^{28}$ = Average thermal macroscopic radiative capture cross section for
 ^{238}U in the lattice fuel

$\bar{\Sigma}_a^{25}$ = Average thermal macroscopic absorption cross section for
 ^{235}U in the lattice fuel

$\bar{\Sigma}_f^{25}$ = Average thermal macroscopic fission cross section for ^{235}U
in the lattice fuel.

The ultimate product of neutron capture in ^{238}U is ^{239}Pu through the chain

Either the ^{239}U , ^{239}Np or ^{239}Pu activity may be measured to determine the capture rate in ^{238}U . Since activated ^{238}U foils must be counted within a few hours after irradiation if the ^{239}U β or γ activity is to be measured one must have quick access to foils after the irradiation. If access is

not available until a few hours after the irradiation, the ^{239}Np β or γ activity can be measured after about four hours to allow decay of essentially all of the ^{239}U to ^{239}Np . In addition, β and α counting techniques will in general require chemical separations to remove sources of interfering activities and to minimize particle attenuation in the foils.

In low power reactors counting statistics requirements may dictate near saturation activities. For ^{239}Np this would require irradiations on the order of a week, but for ^{239}U this would require a few hours. Which activity is to be measured then depends not only on the counting system available but also on the reactor in which the irradiations are made, and in some cases the availability of a lab to perform the required chemical separations.

A central problem in the nondestructive determination of ^{238}U capture rates from irradiated uranium foils is the difficulty of measuring the activity resulting from neutron capture in ^{238}U in the presence of both ^{235}U and ^{238}U fission product activities. Corrections for these fission product activities can be minimized to a certain extent in thermal reactor applications by utilizing uranium foils highly depleted in ^{235}U . Since typically 3 to 10% of the fissions in thermal reactors occur in ^{238}U , minimizing the ^{235}U content of a foil will not completely eliminate the need for a fission product activity correction. This will, however, minimize the correction to that necessary to account for fission product activity from fission in ^{238}U . Since the ^{238}U capture to fission ratio in these low enriched lattices is relatively large (5 to 10) a fission product correction <5% can be achieved for most counting techniques.

In the case of fast reactors one faces a more difficult problem since in a fast spectrum the fission cross section of ^{238}U may be larger than the capture cross section. Thus, in order to avoid large fission product activity corrections, one must concentrate on optimizing foil counting techniques to determine ^{238}U activation products in the presence of fission products or employ chemical separation techniques before counting.

DESCRIPTION OF EXPERIMENTS

The determination of ^{238}U activation rates requires the irradiation of foils in the medium of interest. The two experimental reactors used for foil irradiations in the present experiments are described in this section. Details of the lattice and fuel rods (the medium of interest in the present experiments) are also given. The fabrication of foils, their arrangement in the fuel rod and thermal column, and their irradiation are also discussed.

A. EXPERIMENTAL FACILITIES

1. Plutonium Recycle Critical Facility (PRCF)

The PRCF was originally built to measure physics and loading parameters for plutonium experiments in conjunction with the USAEC Plutonium Utilization Program. The facility has the capability of utilizing either H_2O or D_2O as a moderator and reflector.

The concrete reactor cell is located below grade. The 10-foot wide by 13-foot long by 32-foot deep cell houses the reactor tank, fuel element transfer facilities, moderator and cooling systems, and unirradiated fuel element storage racks. The reactor cell walls shield the surrounding facilities during irradiated fuel element transfer and reactor operation. A removable block, 3 ft $\times$ 3 ft $\times$ 4 ft thick, provides personnel access to the cell.

The reactor loadings tested in the PRCF are contained in a cylindrical aluminum tank, 6 ft in diameter and 9 ft high. This reactor tank is located in the reactor cell with the top of the tank about 17 ft below grade level. A grating at the minus 17-ft level provides access to the top of the reactor tank and one at the minus 26-ft level provides a working area

around the reactor vessel. The reactor itself is unshielded, with the cover blocks, cell walls and surrounding earth providing radiation shielding for the surrounding areas.

The tank provides a container to hold a variety of reactor systems. For a given system the fuel elements are spaced in the moderator by exchangeable lattice templates. Fine reactivity control and reactor scram are accomplished normally by six control and safety devices. Additional reactivity compensation is accomplished by adjustment of the moderator level. The reactor operates at powers up to 10 kW.

Following the completion of an experimental program, the reactor structure including the fuel, lattice templates, control and safety devices, and flux monitoring chambers are removed from the reactor tank and the structure for the next experiment is installed. In most new loadings many of the existing components, with exception of the lattice templates and fuel elements, can be utilized. This flexible arrangement provides a facility which can test a wide range of water moderated reactor systems.

Another feature of the facility provides for irradiated fuel elements to be transferred into the reactor cell from a water-filled loadout canal through an air-to-water fuel transfer lock. These elements can be cooled both during transfer and while positioned in the reactor. Irradiated fuel measurements have been successfully completed using this system.

2. Thermal Test Reactor (TTR)

The Thermal Test Reactor (TTR) is a light-water moderated graphite-reflected zero-power test reactor designed for a low critical mass, with an additional feature of having both internal and external thermal

columns. The reactor is used primarily as a neutron source. The low critical mass and enhanced thermal flux in the internal column are of advantage in nuclear material purity tests and foil standardization.

The fuel core or active lattice region of the TTR (Figure 1) is contained in a vertical cylindrical annulus made of aluminum. The annulus contains 20 aluminum fuel tubes. The fuel tubes are 2 in. in inside diameter and are equally spaced in a 15 in. diameter circle. Both the tubes and the annulus contain light water.

The annulus is 18 in. in height and outside diameter and has a 12 in. inside diameter. An internal thermal column fits inside the annulus and is 12 in. in diameter. A test hole, which is variable in size, but usually 2 in. in diameter, runs the full length of the reactor at the center of the internal thermal column.

The annulus fits horizontally in the center of a graphite reflector and vertically such that there are 25-1/8 in. and 18-11/16 in. of reflector at the bottom and top of the tank, respectively. The reflector has a 63-3/4 in. square base and is 62-15/16 in. high.

The fuel elements consist of discs of an uranium-aluminum alloy mounted on an aluminum tube. The discs are 1.96 in. in diameter, 0.075 in. thick, and have a 0.60 in. diameter hole in the center. Each disc is clad with 10 mils of aluminum and then coated with a thin layer of paraffin to protect the fuel element from corrosion by the water in which they are immersed. The uranium-aluminum alloy contains 25% uranium which is enriched to approximately 93% ^{235}U . About 57 of these uranium-aluminum discs, mounted on an aluminum tube 1/2 in. in diameter and 18 in. long, comprise a

FIGURE 1. Schematic Diagram of the Thermal Test Reactor

fuel assembly. Aluminum spacers, 5/8 in. OD $\times$ 0.2 in. thick and cut on a bias, separate the discs. A total of 1139 discs form a critical loading of 2.54 kg.

The reactor is controlled by two control rods, four safety rods, and four safety sheets which use cadmium to absorb neutrons. The safety rods and control rods penetrate the core annulus at a diameter of 16-5/8 in. and are equally spaced in the annulus. For the safety and control rods, 0.020-in. cadmium is wrapped around an aluminum rod 0.375 in. in diameter and the assembly is inserted in a 0.460 OD stainless steel tube. The cadmium portions of the control and safety rods are 18 in. long. The safety rods, when in the annulus, are within 4 in. of the bottom of the core and the control rods are aligned with the core. The absorbing material of the safety sheets is cadmium, 3 in. wide by 18 in. long by 0.030 in. thick. These cadmium sheets are supported between sheets of steel slightly larger in size.

The sheets are positioned at the periphery of the annulus between the annulus and the reflector. The sheets are arranged at angles of 54°, 108°, 54° and 144° from each other.

The external thermal column is a 46-in. cube and adjoins one side of the graphite reflector. This external thermal column provides a source of neutrons whose distribution with respect to energy is a Maxwellian with a mean energy which is related to the physical temperature of the external thermal column. The external column is enclosed in insulation and contains calrod heaters at its edges to provide the capability of raising the temperature of the graphite from room temperature to approximately 900°K.

B. LATTICE AND FUEL SPECIFICATIONS

Measurements were made in the PRCF in an 0.750 in. pitch square lattice consisting of aluminum clad UO_2 rods in a light water moderator. Fuel and clad specifications are tabulated below.

TABLE 1. Fuel and Clad Specifications

Fuel Form	UO_2 Pellets (Vibratory Compacted)
Fuel Enrichment	2.35 ± 0.05 wt% ^{235}U
Fuel Pellet Density	9.20 g/cc (84% Theoretical Density)
Fuel Pellet Diameter	0.4405 in.
Fuel Clad Material	Aluminum (6061)
Fuel Clad OD	0.500 in. (0.030 in. wall)

The active fuel length in the critical water reflected core consisting of 461 fuel rods was 36 inches.

C. FOIL FABRICATION

Three types of foils were used in the present experiments. Depleted uranium foils provided the bare and cadmium covered ^{238}U activation rate data. Highly enriched uranium-aluminum foils provided data for a fission product activity correction applied to the depleted uranium foil activity. Dysprosium-aluminum foils yielded activation data necessary for the determination of ρ^{28} by the "thermal activation technique".

The foils used in these experiments were all specially cut to closely match the 0.4405 in. fuel pellet diameter. This is a necessary condition

to obtain good experimental reaction rate data especially in the case of ^{238}U , since the epithermal absorption rate in ^{238}U is a strong function of radius near the fuel rod surface.*

The 20-fold depleted uranium (355 wppm ^{235}U) metal foils were all cut from a single piece of material which had been rolled to a nominal thickness of 0.003 inches. Highly enriched U(93.5)-Al foils (7 wt% U in Al) were cut from existing 0.500 in. diameter X 0.005 in. thick foils. Dysprosium-aluminum foils (4.18 wt% Dy in Al) were cut from a single sheet of material of 0.005 in. nominal thickness. All foils used in the experiment were chosen so that they were free from surface defects and were of uniform thickness and free from burrs. Foils appeared to be uniformly flat in that no crowning or dishing of the foils was detectable.

Each foil was weighed with an accuracy of about $\pm 0.1\%$ on a precision balance. Foils of each type were selected to be closely matched by weight for each irradiation. The 20-fold depleted uranium foils were weighed both prior to and after irradiation and counting. Weights of these foils were found to decrease by 0.2 to 0.5% due to the formation and loss of an oxide coating but the resulting corrections to the ratios of foil weights or activities were found to be less than 0.1% in all cases.

D. FOIL ARRANGEMENTS

1. Lattice

Two packets of foils were used in each lattice and thermal column irradiation. The first packet provided the bare or cadmium covered

* Measurements of the radial distribution of the ^{238}U absorption rate in different types of fuel rods have been reported in the literature. The interested reader is referred to References 8 and 22-26.

activation data, while the second packet served as monitor foils to allow normalization of foil activities between irradiations.

The foil packet consisted of a 20-fold depleted uranium metal foil, a highly enriched uranium-aluminum foil, a dysprosium-aluminum foil and aluminum catcher foils. The function of the depleted uranium foil was to sample the ^{238}U activation rate in the fuel rod. The highly enriched uranium-aluminum foil was necessary to allow corrections for fission product activity in the irradiated depleted uranium foil. The dysprosium-aluminum foil provided activation data necessary in the determination of ρ^{28} by the "thermal activation technique". The aluminum catcher foils prevent the contamination of each detector foil by an adjacent detector foil or fuel pellet.

The foil arrangement used in the lattice irradiations is shown in Figure 2. All lattice irradiations were made in a separable rod in the center of the PRCF core. The foil packet was positioned axially in the separable rod so as to be directly at the core center. The monitor foil packet was arranged as shown for the "bare foil irradiation" in Figure 2 and positioned axially 5 in. below the core midplane in the separable rod in all lattice irradiations.

The separable rod consisted of three segments held together by two layers of 0.001 in. thick mylar tape applied to each of the two joints. These tape sections were centered axially 1.4 in. above the detector foil packet and 1.3 in. below the monitor foil packet and were considered to have no perturbing effects on the experimental results. This is a reasonable assumption since only a small amount of water moderator was

BARE FOIL IRRADIATION

CADIUM COVERED FOIL IRRADIATION

FIGURE 2. Foil Arrangements in the Fuel Rod

displaced by the chemically similar mylar and any resulting perturbation should be much smaller in the region of the foil packets.

The foil assemblies were placed between specially selected 0.125 in. fuel pellets. These pellets adjacent to the foil packet were selected to be flat and free from surface defects and chipped edges so as to leave no gaps between the foil packet and fuel pellets. A compressive force was applied when assembling the separable fuel rod to insure that no gaps existed in the fuel pellet-foil assembly.

The purpose of the 0.125 in. fuel pellets inside the cadmium "box" was to eliminate the possibility of streaming of epithermal neutrons from the moderator through the cadmium buttons and into the foil packet without having had a possible interaction in the fuel. Justification for the use of these pellets inside the cadmium box will be given in a later section where experimental results obtained with and without these pellets in the cadmium "box" are compared.

An important feature of the design of the separable fuel rod was the allowance for cadmium covers which did not displace moderator in the lattice cell. In the bare foil irradiations the foil packet, sandwiched between two 0.125 in. fuel pellets, was surrounded radially by a 0.020 in. thick by 1.25 in. long aluminum sleeve. This sleeve was centered on the fuel packet and displaced the inner 2/3 of the aluminum cladding. In the cadmium covered foil irradiations the aluminum sleeve was replaced by a cadmium sleeve of the same dimensions and 0.020 in. thick cadmium buttons were inserted between the 0.125 in. fuel pellets and adjacent pellets as shown in Figure 2. Thus the need for corrections for systematic errors

introduced when cadmium displaces moderator in the vicinity of the foils was eliminated by the separable rod design. Corrections for the effect of moderator displacement are difficult to evaluate and become most important for tight pitched lattices. It is therefore desirable to eliminate the need for these corrections, whenever possible, through the design of the experiment.

2. Thermal Column

Foils of each type were irradiated in the TTR thermal column on a graphite rotator. The rotator was positioned just outside the core reflector in the center of the thermal column. The purpose of these irradiations was to provide activation data in a standard reference spectrum for the thermal activation technique. Although the spectrum was not purely thermal at this position counting rates of both bare and cadmium covered foils were sufficient to provide statistically precise cadmium ratio data for each foil type, thus allowing a precise determination of the thermal activity from the bare and cadmium covered activities.

As in the lattice irradiations two sets of foils were used in each thermal column irradiation. One set, positioned four inches from the other on the rotator, served as a monitor set to provide normalization between bare foil activities in the bare and cadmium covered foil irradiations. Each foil was irradiated in an individual, 0.020 in. thick, cadmium cover so as to be consistent with lattice cadmium cover thickness.

E. FOIL IRRADIATIONS

The types of foils, where they were irradiated, and for what purpose, are summarized in Table 2. In addition to the foils listed, a bare

monitor foil of each type was included in each irradiation to allow normalization between the bare and cadmium covered foil irradiations.

TABLE 2. Summary of Foils Irradiated

<u>Foil Type</u>	<u>Facility</u>	<u>Bare/Cd Covered</u>	<u>Purpose*</u>
Depleted U	Lattice	Bare	C^{28} and T.A.
		Cd Covered	C^{28}
	Thermal Column	Bare	T.A.
		Cd Covered	T.A.
U(93.5)-Al	Lattice	Bare	} Fission product activity corrections for above depleted uranium foils.
		Cd Covered	
	Thermal Column	Bare	
		Cd Covered	
Dysprosium-aluminum	Lattice	Bare	T.A.
		Cd Covered	T.A.
	Thermal Column	Bare	T.A.
		Cd Covered	T.A.

* C^{28} = direct ^{238}U cadmium ratio measurement.

T.A. = ρ^{28} via the thermal activation technique.

Three irradiations were made in the PRCF lattice and two in the TTR thermal column. Bare foil irradiations were made simultaneously in both reactors. A waiting time of two hours after the irradiations was necessary before foils could be collected so that personnel exposure would be minimized. Cadmium covered foil irradiations were also made simultaneously in both reactors one week after the bare foil irradiations were made and under the same reactor conditions. A third irradiation was made in the

PRCF lattice with the 0.020 in. cadmium button directly adjacent to the foil packet to determine the effect of the 0.125 in. fuel spacers in the cadmium "box".

All irradiations in the PRCF were made at a power level of 200 watts for one hour and irradiations in the TTR were made at a power level of 100 watts for one hour. These irradiation times and power levels were found to be adequate to obtain good counting statistics on all three foil counting systems employed.

DETERMINATION OF ^{239}Np ACTIVITY

The three counting techniques utilized in the determination of the ^{239}Np activities from the irradiated depleted uranium foils are discussed in this section. The NaI-NaI coincidence technique is introduced first since it requires the most thorough knowledge of the ^{239}Np decay scheme. With this knowledge as background information the introduction of the NaI singles and Ge(Li) singles techniques is greatly simplified.

The three counting systems are described along with methods used for data accumulation, reduction and analyses in each case. Typical spectra from each system are shown. The ^{238}U cadmium ratios and ρ^{28} values obtained from each counting technique are given in a later section.

A. NaI-NaI COINCIDENCE TECHNIQUE

1. Introduction

The principle of the coincidence spectrometry method is the measurement of the number of coincident events between two types of radiation emitted "simultaneously" in time from a given radioactive source. Two possible combinations of radiation would be a β -ray and a γ -ray or two γ -rays. The number of coincident events is measured utilizing two radiation detectors and a coincidence circuit which determines the simultaneity of two events within a resolving time τ_c . The coincidence circuit gives an output pulse whenever two input pulses (one from each detector) are "simultaneous" within the resolving time, but gives no output pulse when only one of the input pulses is present. The main advantage of coincidence spectrometry over single detector spectrometry in the present application is

its ability to selectively detect radiation from a particular isotope (^{239}Np) in the presence of interfering radiation from many other sources (fission products).

The feasibility of applying coincidence spectrometry to the determination of ^{239}Np specific activities is demonstrated in the decay scheme of ^{239}Np shown in Figure 3 adapted from Reference 27. In the decay of ^{239}Np , 32% of the β decays populate the 392 keV energy level of ^{239}Pu which deexcites primarily to the 286 keV energy level by emission of a 106 keV γ -ray in coincidence with 278, 228 or 210 keV γ -rays from the 286 keV energy level. In addition to deexcitation by γ ray emission there is a high probability in some cases for transitions to occur by internal conversion of orbital elections. This type of transition is evidenced in γ -ray spectra by the presence of photopeaks caused by characteristic X-rays when the orbital elections readjust to fill the vacancy left by the converted electron.

The energies and relative intensities of ^{239}Np γ -rays and ^{239}Pu X-rays are relatively well known.⁽²⁷⁻³³⁾ A γ and X-ray spectrum from an irradiated depleted uranium foil used in the present experiment is shown in Figure 4. This spectrum was taken with a high resolution (~ 800 eV at 100 keV) Ge(Li) detector at 108 hours decay time and gives an idea of the relative intensities of the various γ and X-rays in the decay scheme. Energies of the photopeaks identified are indicated in Table 3.

DECAY SCHEME FOR $^{239}_{93}\text{Np}$

FIGURE 3. Decay Scheme for $^{239}_{93}\text{Np}$

FIGURE 4. γ and X-ray Spectrum from an Irradiated Depleted Uranium Foil

TABLE 3. Energies of Photopeaks in Figure 4.

^{239}Np γ -ray Energies, keV		U and Pu X-ray Energies, keV	
γ_1	44.65	<u>U</u>	
γ_2	49.40	$U_{k\alpha 1}$	98.44
γ_3	57.26	$U_{k\alpha 2}$	94.66
γ_4	61.46	$U_{k\beta 1}$	111.30
γ_5	67.86	$U_{k\beta 2}$	114.56
γ_6	88.06	$U_{k\beta 3}$	110.43
γ_7	106.14	$U_{k\beta 4}$	114.33
γ_8	106.47	$U_{k\beta 5}$	111.98
γ_9	166.4	<u>Pu</u>	
γ_{10}	181.7	$Pu_{k\alpha 1}$	103.76
γ_{11}	209.8	$Pu_{k\alpha 2}$	99.55
γ_{12}	226.4	$Pu_{k\beta 1}$	117.26
γ_{13}	228.2	$Pu_{k\beta 2}$	120.70
γ_{14}	254.4	$Pu_{k\beta 3}$	116.27
γ_{15}	272.9	$Pu_{k\beta 4}$	120.44
γ_{16}	277.6	$Pu_{k\beta 5}$	117.85
γ_{17}	285.5		
γ_{18}	315.9		
γ_{19}	334.3		

In the case of ^{239}Np decay the 278, 228 and 210 keV γ transitions are highly internally converted while the 106 keV transition is not.^(28,29)

It is therefore possible to observe coincidences between the 106 keV γ -ray and the 99-121 keV ^{239}Pu X-rays resulting from the internal conversion of the 278, 228 and 210 keV lines instead of coincidences between the 106 keV γ -ray and 278, 228 and 210 keV γ -rays.⁽¹⁷⁾ An advantage of this method is that it allows the use of a symmetrical system yielding a higher counting efficiency in that the higher intensity and lower energy 104 keV ^{239}Pu X-rays are employed instead of the 278, 228 and 210 keV γ -rays themselves.

In addition this energy discrimination reduces the relative contribution to the total coincidence rate resulting from true but undesirable coincidences with the many fission product γ -rays present in the other portions of the spectrum. For these reasons this method was used in the present experiments.

A few interfering effects observed in coincidence spectrometry worth noting are pointed out in Reference 34, and enumerated here:

1. The coincidence spectra have the usual single and multiple Compton back scattering components found with singles spectrometers but in addition the Compton scattering from detector to detector produces false coincidences. For example, γ -rays of energy E_γ which are back scattered from a Compton process in detector 2 will result in a prominent peak near $E_{\text{bsc}} = E_\gamma - \hat{E}_{\text{comp}}$ in detector 1, where $\hat{E}_{\text{comp}}$ is the energy of the Compton edge* for an incident photon of energy E_γ . Similarly a peak at $\hat{E}_{\text{comp}}$ will appear in the coincidence spectrum as a result of a Compton scattering in detector 1 with the scattered photon being detected in detector 2. Antiscattering shields⁽³⁴⁾ are normally used to reduce this effect which is important for higher energy (>200 keV) γ -rays.
2. In singles spectrometry the escape of an X-ray following a photo-electric process produces an escape peak, but in coincidence work the

* The Compton edge corresponds to the maximum energy that an incident photon can impart to an electron in the scintillation crystal by Compton scattering and is given by⁽³⁵⁾

$$\hat{E}_{\text{comp}} = E_\gamma / (1 + \frac{mc^2}{2E_\gamma})$$

escaping X-ray reaching the other detector produces a coincidence count. This process results in the appearance of a strong 28 keV line (iodine X-rays) in coincidence with each low energy γ -ray in the coincidence spectrum. These false lines can be reduced with the same type of shield⁽³⁴⁾ used for Compton scattered photons.

3. Pair production will also produce a false coincidence line due to the production of annihilation radiation. This effect is most easily reduced by placing the detectors at 90° rather than at 180° from one another.
4. Random sum lines in the coincidence spectrum are expected as in singles spectrometry but a further difficulty, the coincident random sum line, appears whenever coincident and noncoincident radiation are both present. The random sum line of one coincident radiation and a noncoincident one is also coincident.

Due to the technique used in the present experiment and the primarily low energy γ and X-rays in the decay scheme of ^{239}Np none of the above interfering effects were found to provide any difficulties. The coincidence spectra shown in Figure 6 (see Page 35) do however show the effect indicated in (2) due to the escape of the 28 keV iodine X-ray.

2. Description of Counting System

A schematic diagram of the coincidence counting system is shown in Figure 5. The conditions shown in the diagram are those actually used during coincidence counting. In channel 1 the pulse height signals from the photomultiplier pass through a preamplifier and amplifier and are applied to the analog to digital converter at input "A" of the 512 channel analyzer. The signal from the photomultiplier in channel 2, after passing

FIGURE 5. Schematic Diagram of Coincidence Counting System

through a preamplifier and amplifier, is applied to the input of a single channel analyzer. If the pulse is of sufficient magnitude to pass between the window settings of the single channel analyzer an output pulse is sent to the coincidence gate of the multichannel analyzer through a coincidence driver. The function of the coincidence driver is to modify output pulses from the single channel analyzer and pulse height discriminator to be compatible with input pulse requirements of the coincidence gate of the 512 channel analyzer. If the gating pulse comes within a specified time after the pulse to the analyzer the analyzing cycle begins and the input pulse from channel 1 is recorded in the proper channel of the analyzer. If the gating pulse does not have the proper time relationship with the input pulse the analyzing cycle does not commence and the input pulse is cancelled. Thus, a pulse is analyzed by the analyzer only if an event occurred in the first detector simultaneously with one in the second detector which fulfilled the pulse height requirement of the single channel analyzer.

With this arrangement the single channel analyzer, which determines the pulse height and hence the energy range of pulses that gate the multichannel analyzer, can be easily calibrated. For the calibration only pulses from detector number 2 are used. The pulses, in addition to going to the single channel analyzer as usual, are also fed directly into the multichannel analyzer at input B as indicated by the dashed line in Figure 5. Thus, the multichannel analyzer analyzes all the pulses which enter the single channel analyzer. A 256 channel spectrum of these pulses is accumulated and retained in the memory. The multichannel analyzer gate is then switched on so that subsequent input pulses are analyzed only

if they fulfill the window requirement of the single channel analyzer. This mode of operation is possible because the delay time of the gate is the same as the time for a pulse to be analyzed by the single channel analyzer. These pulses are accumulated only in the channel positions corresponding to the pulse heights of the accepted signal and at points corresponding to counts already stored in the 256 channels in the memory. By using the overlap function of the 512 channel analyzer the position of the window of the single channel analyzer is displayed relative to the complete spectrum. By changing the window level or width the region of pulses is moved over the spectrum and positioned on the energy region that is to gate the coincidence circuit. In this manner the window level of the single channel analyzer can be set in a very short time using an irradiated depleted uranium foil or a ^{243}Am source. After the window is adjusted input B is switched off and input A is switched on so the multichannel analyzer can accumulate the coincidence data.

In general the best results using coincidence counting are obtained by using large solid angles (detectors close together), efficient counters, short resolving times, and a source of not too high specific activity. A large solid angle was obtained by placing 2 in. diameter by 1/4 in. thick NaI crystals $\sim 1/2$ in. apart. A 1/8 in. thick aluminum plate, with a 1/2 in. diameter hole centered on the detector axis, was fixed between the two detectors. This allowed accurate positioning of the foils which were mounted on 3/32 in. thick plexiglass plates which fit snugly into a receptacle mounted on the aluminum plate. The aluminum plate also reduced the amount of radiation back scattered from one crystal that would reach the other crystal. Coincidence background was minimized by completely enclosing the pair of detectors (and source) in a 2 in. thick lead cave.

Since the efficiency of the detectors is fixed and the resolving time is set by the electronics of the system an optimum source strength (discussed in Appendix A) for this system was achieved by careful selection of irradiation conditions. This allowed the random coincidence rate to be kept small compared to the true coincidence rate and yet allowed a sufficient number of coincidences to be recorded in a reasonable time.

3. Data Accumulation, Reduction and Analysis

The coincidence activity of each depleted uranium foil was measured a total of six times during the period 20-200 hours after irradiation. The observed coincidence activities were found to decay with the half life of ^{239}Np (2.35 days)⁽²⁷⁾ during this period of time indicating that there were no gross contaminants present in the foils. The high internal consistency observed between the ^{239}Np activity determinations at different decay times testifies as to the reproducibility of the counting geometry. A 10 minute coincidence count on the depleted uranium foils was found to yield adequate (<1%) counting statistics during the major portion of this period.

A ^{243}Am source was counted periodically to check for counting system stability to gain shift and zero drift as well as to monitor the position and width of the "gating pulse" window. The ^{243}Am is an excellent monitor for ^{239}Np counting due to its mode of decay, i.e.,

The short half life of the daughter ^{239}Np relative to the parent ^{243}Am results in the two being in secular equilibrium, thus yielding a constant activity source of ^{239}Np .

In addition, standard sources (^{241}Am , ^{243}Am and ^{203}Hg) were used in each leg of the coincidence circuit for energy calibration and system stability checks. Overall, the long-term stability of the counting equipment was excellent in that no corrections to foil activities were judged to be necessary for these effects. Counting was performed in a temperature controlled room with equipment power supplied from a well regulated and isolated line.

Referring to Figure 5 the coincidence data consisted of the spectra of pulses N_1 , from channel 1, which were detected as occurring simultaneously with "gating pulses" N_2 , which lie within the energy limits set by the single channel analyzer and pulse height discriminator in channel 2. The spectra N_1 were recorded over the energy range 0-512 keV. The energy window in channel 2 was centered on the 105 keV composite peak to accept pulses whose energies were in the range 90-122 keV. This window position and width, as recommended by previous experimenters^(12,17,18) was found to yield a good signal to noise ratio while still being relatively stable to slight gain and zero shifts. The number of "gating pulses" N_2 , passing through the window was recorded with the scaler shown in Figure 5. The number of input pulses from channel 1 was determined via integration of the NaI singles spectra taken just prior to or immediately after the coincidence spectra were taken.

Typical coincidence spectra for both depleted and enriched uranium foils are shown in Figure 6. Singles spectra are also shown in the figure for purposes of comparison. All spectra shown were taken at ~25 hours cooling time.

FIGURE 6. NaI Singles and Coincidence Spectra for Irradiated Depleted and Enriched Uranium Foils.

In both the coincidence and singles spectra for the depleted uranium foil there are three principle energy groups. The 105 keV peak is a composite of the 106 keV ^{239}Np γ -ray and Pu and U X-rays. The second major peak in the spectrum appears at ~ 228 keV and results primarily from 210 and 228 keV ^{239}Np γ -rays. The third peak at ~ 278 keV is due to the ^{239}Np γ -ray at that energy.

In comparing the coincidence and singles spectra for the depleted uranium foil we see the increase in peak to Compton ratio in the former relative to the latter is appreciable even for these highly depleted uranium foils. This effect would be even more pronounced for foils with a higher ^{235}U content.

The singles spectra for the highly enriched ^{235}U -Al foil indicates three prominent energy groups at about 28, 144 and 250 keV. Although there are no prominent peaks in the fission product singles spectra in the 90 to 122 keV "window" there is a one in the coincidence spectra at ~ 130 keV which would contribute to the gating pulses and coincidence counts in this window. This relatively broad peak is a backscatter peak resulting from Compton scattering of the 250 keV γ -rays observed in the singles spectrum. These back scattered γ -rays lose ~ 120 keV of energy in one crystal and are absorbed in the other, all within the resolving time of the coincidence circuit. The contribution to the total coincidence activity due to this effect is minimal for highly depleted foils but becomes appreciable as the ^{235}U content (and therefore fission product activity) of the foil is increased. A foil holder, specially designed to minimize this effect, is shown in Reference 18.

Although the coincidence spectra, N_1 , were recorded over the energy range 0 to 512 keV, only those pulses representing γ -rays in the energy range 90 to 122 keV were used to determine the coincidence activity. As mentioned previously this energy discrimination tends to minimize the correction necessary for coincidences due to fission product γ -rays.

There are several corrections which must be applied to this measured coincidence activity to obtain the activity due to ^{239}Np alone. In addition, the order in which corrections are made to the measured coincidence count rate is important. The random coincidence rate as determined in Appendix A is subtracted first since it depends only on the recorded rates in each channel and does not require a dead time correction. (A few true coincidences may be lost in the dead times following random coincidences. This loss can be neglected here since the random rate is always less than 3% of the total coincidence rate and $N_1\tau_1$ and $N_2\tau_2$ are $\ll 1$).

The correction for coincidence losses described in Appendix B is then applied to the random coincidence corrected coincidence rate. Finally this coincidence rate is corrected for the natural coincidence rate measured prior to foil irradiation and the background coincidence rate which is purely due to cosmic ray coincidences and is measured in the absence of the source. The natural coincidence rate ranged from 14 to 19 CPM while the background coincidence rate was < 1 CPM.

The correction for fission product coincidences is applied next. These so-called fission product coincidences result primarily from the detection of ^{239}Np γ -rays or ^{239}Pu X-rays in coincidence with fission product γ -rays, Compton scattered fission product γ -rays, and the uranium X-rays associated with photoelectric events in the foil.

Coincidences between any combination of the latter three items also contribute but generally to a much lesser extent. In the present case the use of highly depleted uranium foils and the afore mentioned energy discrimination reduced this correction to well under 0.1% in all cases and it was therefore neglected in the analysis. Since in general this correction will not be negligible the reader is referred to References 2, 12, 36, and Appendix "C" of Reference 37 for a detailed discussion and methods used for these corrections.

No corrections were necessary for source decay during the counting time since the counting time (10 minutes) is much less than the 2.35 day half life of ^{239}Np . The midpoint of the counting time was used for decay correcting back to reactor shut down.

All depleted uranium foils used in the experiments were cut from the same piece of 0.003 in. nominal thickness material. Based on a γ -ray self attenuation of $\sim 7\%$ per mil⁽¹⁷⁾ for 105 keV γ -rays in thin uranium metal foils it was estimated that a 10% change in foil thickness would be required to change the transmission by 1%. In no case did the weights (which were assumed proportional to the thickness) of any pair of foils used differ by more than 1%. Corrections for differences in self attenuation for the foil pairs were therefore judged to be negligible ($<0.1\%$) and were not applied. Although each pair of foils used was matched in weight as closely as possible a small weight correction was still required.

Two approaches were used to obtain the cadmium ratios from the foil activation data. First, ratios of the bare and cadmium covered foil

activities, decay corrected back to reactor shutdown, were taken for pairs of measurements which were made at approximately the same decay time. These ratios were then averaged to yield the final result. In the second method, the count rates for a given foil were all decay corrected back to reactor shutdown and an average activity obtained. Ratios of these average activities then resulted in the cadmium ratio. The maximum differences found in any of the cadmium ratios obtained by the two approaches was less than 0.1%. The values quoted in this report were obtained using the first approach.

B. NaI SINGLES TECHNIQUE

1. Introduction

The determination of ^{239}Np specific activities via NaI singles spectrometry is probably the most widely used foil counting technique for ^{238}U activation measurements. Its popularity is mainly based on its ease of application, the relatively simple electronics involved, and the availability of NaI detectors at most experimental facilities.

The decay scheme of ^{239}Np to ^{239}Pu shown in Figure 3 and the γ -ray spectrum in Figure 4 indicate many γ -rays in the energy range 44 to 334 keV. The most prominent γ -rays are found at energies of 106, 278, 228, and 210 keV, listed here in order of decreasing intensity. Due to its higher intensity, and a relatively smaller interference effect from fission products of nearly equal energies, the 106 keV photopeak is generally chosen over the other three for specific activity determinations.

2. Description of Counting System

The counting system for NaI singles spectrometry consisted of one channel of the NaI-NaI coincidence system described in the previous section. Referring back to Figure 5, the section labeled channel 1 was utilized as a singles spectrometer by simply opening the switch between channel 2 and the multichannel analyzer. In this mode of operation, the pulse height signals from the photomultiplier pass through a preamplifier and amplifier and are applied to the analog to digital converter at input A of the 512 channel analyzer. Each pulse is analyzed and recorded in one of the 512 channels of the analyzer memory as determined by the pulse height.

In general, the best results utilizing this type of counting system are obtained by using small source-detector separations, efficient detectors, and a source of not-too-high specific activity. A source-detector distance of about 1/4 in. was attained utilizing the foil holder system described in the section on coincidence counting. Room background was minimized by completely enclosing the detector in a 2 in. thick lead cave.

Although the source strength used was primarily designed to optimize the coincidence counting it also provided near optimum conditions for the NaI singles counting. The maximum dead time observed was ~15% which is not an unreasonable operating condition for this type of system.

3. Data Accumulation, Reduction and Analysis

The gamma activity of each depleted and highly enriched uranium foil was measured a minimum of six times during the period 20 to 200 hours following the irradiation. The observed activities in the depleted uranium foils were found to decay with the half life of ^{239}Np (2.35 days)⁽²⁷⁾ during this period of time indicating the absence of gross contaminants in the foils. The high internal consistency observed between the ^{239}Np activity determinations at different decay times testifies as to the reproducibility of the counting geometry. One minute live time counts were found to yield the same counting statistics (<1%) as the ten minute coincidence counts.

Since both the singles and coincidence counting were performed using the same equipment, a foil counting cycle was devised to minimize foil changes. In addition, the cycle was designed to minimize the difference in decay time between counting a given foil and its monitor and/or fission product activity correction foil. This approach has the advantage of minimizing systematic errors due to half life uncertainties and counting system gain shifts and zero drifts.

A typical foil counting cycle for a depleted uranium foil (D_1) and its monitor (D_2) and their corresponding highly enriched, fission product activity correction foils, U_1 and U_2 , is shown in Table 4. The choice of either the coincidence or singles counting mode was made by simply flipping a single switch in the counting circuit. Standard sources (^{241}Am , ^{243}Am , and ^{203}Hg) were used for energy calibration and to check system stability (gain shift and zero drift). As was the case with the coincidence system,

TABLE 4. Typical Counting Cycle for NaI Coincidence and Singles Counting

Sample	Mode	Counting Live Time, min
Standard Sources	Singles	--
Background	Singles	10
^{243}Am	Singles	1
^{243}Am	Coincidence	10
U_1	Coincidence	10
U_1	Singles	1
D_1	Singles	1
D_1	Coincidence	10
D_2	Coincidence	10
D_2	Singles	1
U_2	Singles	1
U_2	Coincidence	10
^{243}Am	Coincidence	10
^{243}Am	Singles	1
Background	Singles	10
Standard Sources	Singles	--

the long term stability of the counting equipment was found to be excellent in that no corrections to foil activities were found to be necessary for these effects.

The singles data consisted of a spectrum of pulses resulting from γ -rays with energies up to 1024 keV. Typical spectra for depleted and enriched uranium foils are shown in Figure 6.

The depleted uranium foil spectrum is characterized by three principle energy groups. The 105 keV peak is a composite of the 106 keV ^{239}Np γ -ray and Pu and U X-rays. The peak near 228 keV results primarily from the 210 and 228 keV ^{239}Np γ -rays, while the peak at 278 keV is due to the ^{239}Np γ -ray at that energy.

The singles spectra for the highly enriched ^{235}U -Al foil is characterized by three prominent energy groups at 28, 144, and 250 keV. Note that this 250 keV peak will contaminate both the 228 and 278 keV peaks in the ^{239}Np spectrum if fission products are present in the depleted uranium foil. The 105 keV composite peak in the ^{239}Np spectrum would thus appear to suffer the least from any direct fission product activity present in the depleted uranium foil.

Although the 105 keV composite peak in a depleted uranium foil spectrum consists primarily of activity associated with the ^{239}Np decay process, it does contain, in general, a significant amount of activity representative of fission product decay. The source of this activity is best illustrated by Figure 9 (see Page 52) which shows the γ -ray spectrum of an irradiated depleted uranium foil for the energy region of interest. This spectrum, taken at a cooling time of 108 hours with a high resolution Ge(Li) detector, indicates the absence of any fission product lines of appreciable intensity between 90 and 122 keV. However, the characteristic X-rays of uranium are found in the spectrum along with the Pu X-rays and the 106 keV ^{239}Np γ -ray.

The presence of the uranium X-rays in the spectrum indicates the photoelectric interaction of higher energy γ -rays with the uranium atoms in the foil (fluorescence radiation). For singles counting with NaI detectors, these X-rays (94 to 115 keV) cannot be resolved from the Pu X-rays (99 to 121 keV) and 106 keV ^{239}Np γ -ray which are characteristic of the ^{239}Np decay process. It is, therefore, important to use highly depleted uranium foils when NaI singles counting techniques are employed so as to minimize the production of uranium X-rays from ^{235}U fission product γ -rays.

Production of uranium X-rays due to photoelectric interaction of the higher energy γ -rays of ^{239}Np (or ^{239}Pu X-rays) and the uranium atoms in the foil is on the other hand beneficial in that it provides additional radiation in the 105 keV composite peak which is characteristic of the ^{239}Np decay. Since uranium X-ray production from ^{238}U fission product γ -ray interactions in the foils cannot be completely eliminated, corrections may still be required for this activity.

Although the singles spectra were recorded over the energy range 0 to 1024 keV, only two portions of these spectra were used to determine the ^{239}Np activity. The first portion, referred to hereafter as the "NP integral", represents those pulses resulting from γ and X-rays in the energy range 90 to 122 keV. In the case of the depleted uranium spectra, this integral is primarily representative of the ^{239}Np activity. The second portion, referred to as the "FP integral", consists of pulses in the energy range 512 to 675 keV and is due only to fission product γ -rays since no ^{239}Np γ -rays have been observed in this energy range.⁽²⁷⁾

The correction for fission product activity in the NP integral of the depleted uranium foil is obtained from the NP and FP integrals of the depleted and highly enriched U-Al foils at the same decay time.⁽¹³⁾ Let f_d be the fission product activity in the NP integral of the depleted uranium foil, f_e the corresponding activity in the enriched U-Al foil and F_d and F_e the fission product activities in the FP integral of the depleted and enriched uranium foils respectively. Now if the fission product γ -ray spectra and decay curves are the same for ^{235}U and ^{238}U fission we can equate the time dependent ratios of fission product activities in the NP and FP integrals of the depleted and enriched

uranium foils. Thus

$$\frac{f_d}{F_d} = \frac{f_e}{F_e}$$

or

$$f_d = \frac{f_e}{F_e} F_d = r(t) F_d$$

which assumes that the contributions to f_e due to ^{239}Np activity can be neglected. Although the assumptions made above in deriving the fission product activity correction are not strictly valid, these assumptions generally produce only a second order affect on the measured value of ρ^{28} since the fission product correction can usually be made to be small by using highly depleted uranium foils and selectively "window counting" over the 105 keV composite peak to obtain the ^{239}Np activity.

Note that in the ^{235}U -Al (7 wt% uranium in aluminum) foil spectrum [Figure 8 (see Page 51)], there are essentially no uranium X-rays produced. As indicated before, no prominent fission product γ -rays were observed in the energy range 90 to 122 keV. Thus in the present case the fission product correction amounts essentially to a fission product Compton background correction. That this correction is sufficient is evidenced by the spectra in Figure 8 which indicate that essentially all uranium X-rays in the depleted uranium spectrum result from interactions with higher energy ^{239}Np γ -rays rather than fission product γ -rays. Since the uranium X-rays produced by ^{239}Np γ -rays are characteristic of the ^{239}Np decay, no correction is required for them. Had natural instead of depleted uranium foils been used, then full density, highly enriched uranium foils of the same

thickness would have to replace the ^{235}U -Al foils for fission product corrections in order to properly account for uranium X-ray production from fission product γ -rays.

Corrections for fission product activity in the ^{239}Np window (90 to 122 keV) followed the same general form as observed in previous experiments by others.^(8,12) Although the absolute value of the percent fission product correction is dependent upon window width and foil enrichment, the minimum value was found to occur at 90 to 100 hours after irradiation, as was observed in References 8 and 12. A theoretical prediction of this minimum assuming a ^{239}Np half life of 56.3 hours and an approximate $t^{-1.2}$ fission product activity decay results in a value of 97.5 hours. During the counting period from 20 to 200 hours after the irradiation the percent fission product correction was typically less than 1%.

Corrections to the NP integral were also made to account for room background and natural decay activity (measured prior to irradiation) which ranged from 0.1 to 1 and 1 to 10%, respectively. Corrections for detector dead time were made automatically by utilizing the live time mode of the multichannel analyzer. A small correction for differences in foil weights was also required. Corrections for differences in self attenuation of 105 keV γ -rays due to different foil thicknesses were not necessary for reasons discussed in the section on coincidence counting techniques. Source decay during counting was also determined to be negligible and the midpoint of the counting interval was used for decay correcting back to reactor shut down.

As in the case of the coincidence data analysis two approaches were used to obtain the cadmium ratios from the NaI singles activation data. First, ratios of the bare and cadmium covered foil activities, decay corrected back to reactor shutdown, were taken for pairs of measurements which were made at approximately the same decay time. These ratios were then averaged to yield the final result. In the second method, the count rates for a given foil were all decay corrected back to reactor shutdown and an average activity obtained. Ratios of these average activities then resulted in the cadmium ratio. The maximum differences found in any of the cadmium ratios obtained by the two approaches was less than 0.2%. The values quoted in this report were obtained using the first approach.

C. Ge(Li) SINGLES TECHNIQUE

1. Introduction

Rapid progress during the last few years in the design and development of semiconductor detectors and associated electronics has made practical Ge(Li) detector systems commercially available for γ and X-ray spectroscopy.* The principal advantage in using Ge(Li) detectors for the present application lies in the high energy resolution which they provide. Typical Ge(Li) detectors yield an order of magnitude better energy resolution than

* The interested reader is referred to References 38-45 for discussion of the basic principles and application of these detectors.

NaI detectors. However, due to the small sensitive volumes of present day Ge(Li) detectors (generally <50 cc) their absolute efficiency is an order of magnitude lower than that for NaI. Although the absolute efficiency of these detectors is relatively low it is peaked at about 100 keV enhancing the detection of 106 keV ^{239}Np γ -rays and Pu X-rays relative to the higher energy fission product γ -rays. Furthermore, the lower efficiency can be compensated for by using higher disintegration rates and longer counting times. Once this compensation has been achieved the high resolution of the Ge(Li) detector can be utilized to separate the ^{239}Np γ -rays and Pu X-rays from fission product γ -rays and uranium X-rays, thus reducing the magnitude of, and greatly simplifying the corrections for these interfering radiations relative to when NaI detectors are employed.

2. Description of Counting System

The counting system for Ge(Li) singles spectrometry used in this experiment is shown schematically in Figure 7. The operation of this system is similar to that of the previously described NaI singles system. The 20 cc Ge(Li) crystal, cooled with liquid nitrogen to a temperature of 77°K, had a measured energy resolution of 2.1 keV (FWHM) at 122 keV.

The source detector distance was maintained by a plexiglass tower fixed to the vertically mounted detector housing. The tower allowed positioning of plexiglass foil holders at several locations from the Be window in the detector housing. Since preliminary checkout and calibration of the system indicated that the live timer was not functioning properly, a source detector distance of 2 in. was chosen to minimize dead time corrections. The detector was surrounded laterally by a 2-in. thick lead wall to minimize room background radiation.

FIGURE 7. Schematic Diagram of Ge(Li) Counting System

3. Data Accumulation, Reduction and Analysis

The γ -ray spectrum of each depleted uranium foil was recorded a total of three times during the period 12 to 80 hours after irradiation. A 10 minute counting interval was found to yield $<1\%$ counting statistics during the major portion of this period. The observed activities were found to decay with the half life of ^{239}Np (2.35 days).⁽²⁷⁾

A standard ^{243}Am source, containing an ^{241}Am impurity, was used for energy calibration and to check system stability to gain shift and zero drift. The counting system was found to be extremely stable with no corrections being necessary for either effect.

Data from the Ge(Li) counting system consisted of the γ and X-ray spectrum from the depleted uranium foils. Typical spectra for both depleted and enriched uranium foils are shown in Figure 8. The full spectra shown in the figure were obtained at a cooling time of 60 hours and the partial spectrum in the insert was obtained at a cooling time of 108 hours.

The upper spectrum in Figure 8 shows the fission product γ -rays from an irradiated, highly enriched U-Al foil. The lower spectrum is from an irradiated 20-fold depleted uranium foil and contains primarily ^{239}Np γ -rays along with fission product γ -rays and plutonium and uranium X-rays. The spectrum in the insert is from the same 20-fold depleted uranium foil, but at a longer cooling time. This spectrum, taken with a smaller volume (0.4 cc) and higher resolution (800 eV at 100 keV) detector than that used for the ^{239}Np activity determinations, shows the details of the spectrum near 100 keV (see also Figure 9). The detail possible with the excellent energy resolution of this spectrometer allows separation and observation

FIGURE 8. Typical γ -Ray Spectra for Irradiated Highly Depleted (Lower Spectrum) and Highly Enriched (Upper Spectrum) Uranium Foils [20 cc Ge(Li) Detector]

FIGURE 9. γ - and X-Ray Spectrum from an Irradiated Depleted Uranium Foil from 0 to 200 keV

of the many plutonium and uranium X-rays from the 106 keV ^{239}Np γ -rays and fission product γ -rays in this complex portion of the spectrum. Note that although there are no appreciable fission product γ -rays in this portion of the spectrum (90 to 120 keV) there are uranium X-rays indicative of interactions of higher energy γ -rays with the uranium foil material. The source and energy of the numbered photopeaks in Figure 8 are given in Table 5.

Although the entire γ -ray spectrum was recorded for each activity determination on the depleted uranium foils only a portion of the spectrum was actually used. This portion covered the energy from 101 to 108 keV and consisted of the photopeaks due to the $\text{Pu}_{\text{k}\alpha 1}$ X-ray at 103.76 keV and the ^{239}Np γ -ray at 106.14 keV, both resulting from the decay of ^{239}Np to ^{239}Pu . The reason for selecting this energy range is illustrated in Figure 10. This figure shows the spectrum from a depleted uranium foil taken with the 20 cc detector (upper curve) and that taken with the higher resolution, 0.4 cc detector (lower curve). Sources for the photopeaks in the spectra are labeled in the latter case only but do apply to both spectra.* Gamma and X-ray energies for the photopeaks labeled in the figure are given in Table 3.

Due to the relatively low efficiency of the Ge(Li) detector it is desirable, if possible, to use the photopeak due to the 106 keV ^{239}Np

* Although the two spectra shown were taken at different decay times (60 and 108 hours, respectively) they are both representative of the spectra in the energy range 90 to 120 keV during foil counting in that no peaks, other than those indicated in the figure, were observed during the period 12 to 80 hours after irradiation.

TABLE 5. Source and Energy of Photopeaks in Figure 8.

Photopeak Number	Energy, keV	Source	Photopeak Number	Energy, keV	Source
1	81	^{133}Xe	32	497	^{103}Ru
2	91	^{147}Nd	33	523	^{132}I
3	94.7	$\text{U}_{\text{K}\alpha 2}$ X-ray	34	531	^{147}Nd
4	99.6	$\text{Pu}_{\text{K}\alpha 2}$ X-ray	35	536	^{140}Ba
5	103.8	$\text{Pu}_{\text{K}\alpha 1}$ X-ray	36	556	
6	106.1	^{239}Np	37	630	^{132}I
7	111	$\text{U}_{\text{K}\beta 1}$ X-ray	38	658	
8	117	$\text{Pu}_{\text{K}\beta 1}$ X-ray	39	668	^{132}I
9	120.6	$\text{Pu}_{\text{K}\beta 2}$ X-ray	40	725	^{95}Zr
10	140	^{99}Mo	41	743	
11	145	^{141}Ce	42	752	^{140}La
12	162	^{140}Ba	43	757	^{95}Zr
13	181	^{99}Mo	44	773	^{132}I
14	186		45	815	^{140}La
15	210	^{239}Np	46	877	
16	228	^{239}Np	47	955	^{132}I
17	228	^{132}Te	48	1026	
18	250		49	1398	^{132}I
19	254	^{239}Np	50	1598	^{140}La
20	278	^{239}Np	51,3	94.66	$\text{U}_{\text{K}\alpha 2}$ X-ray
21	285	^{239}Np	52,4	98.44	$\text{U}_{\text{K}\alpha 1}$ X-ray
22	293	^{143}Ce	53,4	99.55	$\text{Pu}_{\text{K}\alpha 2}$ X-ray
23	304	^{140}Ba	54,5	103.76	$\text{Pu}_{\text{K}\alpha 1}$ X-ray
24	316	^{239}Np	55,6	106.14	^{239}Np
25	319	^{147}Nd	56,7	110.43	$\text{U}_{\text{K}\beta 3}$ X-ray
26	328	^{140}La	57,7	111.30	$\text{U}_{\text{K}\beta 1}$ & $\text{U}_{\text{K}\beta 5}$ X-rays
27	334	^{239}Np		111.98	
28	350	^{143}Ce	58,8	114.33	$\text{U}_{\text{K}\beta 4}$ & $\text{U}_{\text{K}\beta 2}$ X-rays
29	364	^{131}I		114.56	
30	384		59,8	116.27	$\text{Pu}_{\text{K}\beta 3}$ X-ray
31	487	^{140}La	60,8	117.26	$\text{Pu}_{\text{K}\beta 1}$ & $\text{Pu}_{\text{K}\beta 5}$ X-rays
				117.85	
			61,9	120.44	$\text{Pu}_{\text{K}\beta 4}$ & $\text{Pu}_{\text{K}\beta 2}$ X-rays
				120.70

FIGURE 10. Comparison of γ - and X-Ray Spectra from an Irradiated Depleted Uranium Foil Using 20 cc (Upper Spectrum) and 0.4 cc (Lower Spectrum) Ge(Li) Detectors

γ -ray for activity determinations as it is by far the most intense photopeak in the spectrum. However, the spectra in Figure 10 indicate that the 20 cc detector does not quite resolve the $\text{Pu}_{K\alpha 1}$ X-ray from the ^{239}Np γ -ray. This isn't too important if only relative activities are desired since they are both representative of ^{239}Np decays. What is important is that the 20 cc detector was able to resolve this pair from adjacent pairs due to the $\text{Pu}_{K\alpha 2}$ and $\text{U}_{K\alpha 1}$ X-rays (on the lower energy side) and the $\text{U}_{K\beta}$ X-rays (on the high energy side). The reason that this is important is, as discussed previously, that the uranium X-rays are generally not representative of the ^{239}Np decay. Since spectra taken with the high resolution detector indicated no other photopeaks or "shoulders" in this energy range during the period of time after irradiation in which the foils were counted the area (corrected for Compton background) under the $\text{Pu}_{K\alpha 1}$ X-ray - 106 keV ^{239}Np γ -ray "doublet" was taken to represent the ^{239}Np activity. Use of the lower intensity, higher energy ^{239}Np photopeaks, although apparently isolated in the 20 cc detector spectrum, would require much longer counting times. Similarly in the present case requiring only the determination of relative activities, the higher resolution provided by the use of the 0.4 cc detector did not offset the order of magnitude lower efficiency of this detector relative to the 20 cc detector. The small detector did however, prove to be an invaluable tool in determining the components of the complex spectrum in the energy range from 90 to 120 keV. In summary, the 20 cc detector was found to provide adequate energy resolution in the present application and obtaining higher energy resolution at the expense of detector efficiency is not warranted. It is, however, recommended that when initially applying this technique to more highly enriched uranium

foils, high resolution spectra should be taken at several decay times after irradiation to verify the absence of fission product γ -rays in the energy region of interest.

Several corrections were required to convert the measured "doublet" area to a relative ^{239}Np activity. The major correction, amounting to $\sim 15\%$ of the total area, was due to the Compton background under the "doublet". For this correction a linear Compton background was assumed to pass through the minimum at the terminal energies of the region of integration.

Natural and residual activity in this energy region prior to irradiation were determined to be negligible. Since counting was performed in the true time rather than live time mode, corrections for detector dead time were necessary. These detector dead time corrections, amounting to a maximum of 3% of the total "doublet" area, were applied based on data obtained from a dead time meter reading. These corrections tended to cancel out since both the foil and its monitor, whose activities were ratioed, had nearly the same activity.

A correction for differences in foil weights was made but corrections for differences in γ self-attenuation between foils were determined to be negligible. Activity decay during the ten minute counting period is negligible and the mid-point of the counting period was used to decay correct back to reactor shutdown. Ratios of bare and cadmium covered foil activities were determined for pairs of measurements which were made at approximately the same decay time. These ratios were then averaged to yield the final result.

SYSTEMATIC ERRORS AND CORRECTIONS

The experimental methods employed herein were designed to avoid appreciable systematic errors. There are, however, many possible errors of this type which can be associated with ρ^{28} measurements. Several of these errors and corrections for them have been discussed in the section on ^{239}Np counting techniques. No attempt will be made here to discuss all of the other possible errors. The reader is referred to References 2, 4, 6, 25, 46 and 47 for a discussion of many of these types of errors. Justification for experimental design to avoid systematic errors, and corrections for errors which could not be completely eliminated by experimental design, are discussed for the more appreciable systematic errors in the present experiment. Other possible errors mentioned in the above references, but not considered here, are felt either to be unimportant or negligible in the present case.

A. PERTURBATIONS DUE TO THE PRESENCE OF CADMIUM IN THE LATTICE - THE THERMAL ACTIVATION TECHNIQUE

One possible source of systematic experimental error in the measurement of ρ^{28} is the spatial and spectral distortion of the neutron flux by the cadmium in the cadmium covered foil irradiation. The presence of cadmium in the lattice would be expected to reduce the epithermal ^{238}U activation in the fuel rod since the cadmium depresses the thermal flux and thus the source of fast neutrons in the immediate neighborhood of the fuel rod. A "thermal activation technique", designed to eliminate the need for a cadmium covered ^{238}U activation measurement in the lattice, has been described by Egiazorov et al.,⁽⁴⁸⁾ and further developed by

Lewis et al.⁽⁷⁾ The thermal activation technique was applied in this experiment in addition to making the direct ^{238}U cadmium ratio measurements in the lattice, in order to determine the effects of the cadmium on the measured value of ρ^{28} . A brief description of the thermal activation technique and its advantages will now be given along with the working equations needed for its application. A more detailed treatment is given in Reference 7, where these equations are derived.

The thermal activation technique is a method of measuring ρ^{28} through the comparison of total absorption in ^{238}U to thermal absorption in an intermediate material having a much higher probability of thermal absorptions than does ^{238}U . Although the thermal activation technique does not require a cadmium covered ^{238}U activation in the lattice, the cadmium perturbation in the lattice is not completely eliminated, since the cadmium covered activity of an intermediate foil (Dy) is needed. The effect of the cadmium perturbation on the intermediate foil activity can however be minimized by using a material like dysprosium, whose ratio of thermal cross section to resonance integral is large. Thus, the thermal activation technique doesn't eliminate cadmium perturbation in the lattice, but transfers the problem from ^{238}U to an intermediate material where its effect can be minimized and in many cases made negligible.

In addition to eliminating the need for cadmium covered ^{238}U activation measurements in the lattice, the technique significantly reduces the uncertainty in measured values of ρ^{28} as the cadmium ratio of ^{238}U approaches unity ($\rho^{28} \gg 1$). When this is the case and when ρ^{28} is determined from direct measurements of the ^{238}U cadmium ratio, small

uncertainties in the ^{238}U cadmium ratio will propagate large uncertainties in ρ^{28} . For example, if the ^{238}U cadmium ratio were 1.05 ± 0.01 then the uncertainty in ρ^{28} would be $\pm 20\%$. The reason for this large propagated uncertainty is that in the determination of ρ^{28} by direct cadmium ratio measurement, the thermal activity is inferred from the total minus the epithermal capture, which for the case when the ^{238}U cadmium ratio is approximately unity, are nearly equal quantities. Small percentage uncertainties in either quantity will then produce large percentage uncertainties in their difference. However, in the thermal activation technique, the epithermal activity is inferred from the total minus the thermal activity. Since the total activity is much larger than the thermal activity when the ^{238}U cadmium ratio is near unity, and both quantities can be determined with relatively good precision, precise values for ρ^{28} are obtainable.

In applying the thermal activation technique, both ^{238}U and an intermediate material are irradiated in the fuel rod and in a well thermalized reference spectrum. The intermediate detector is irradiated both bare and cadmium covered in the fuel rod and reference spectrum. The ^{238}U detector is irradiated bare in the fuel rod and both bare and cadmium covered in the reference spectrum. The ^{238}U subcadmium activity in the fuel rod is then inferred from the subcadmium activity of the intermediate material in the fuel rod and the subcadmium activities of ^{238}U and the intermediate material in the reference spectrum. The ^{238}U epicadmium activity in the fuel rod is then determined from the difference of the total ^{238}U activity in the fuel rod (from the bare ^{238}U detector irradiation) and this inferred value of the ^{238}U subcadmium activity.

Possible candidates for the intermediate material and desirable properties of the reference spectrum are suggested by consideration of what affects the accuracy of the thermal activation technique. For maximum accuracy in the measurement of the thermal activity of ^{238}U the intermediate detector should have a high cadmium ratio in the fuel rod. Furthermore, it should have a thermal neutron cross section which is approximately $1/v$ so that it is equivalent to ^{238}U in this respect. Due to its high ratio of thermal cross section to resonance integral, ^{164}Dy (28.18% of natural dysprosium) is commonly used as the intermediate material, although its use requires a small correction to account for the absorption cross section falling off slightly faster than $1/v$. Since thermal activities are being compared in the reference spectrum, maximum accuracy will result from a spectrum with a large thermal component, suggesting utilization of a thermal column.

Following the notation in Reference 7, the working equation for the thermal activation technique is

$$\frac{C^{28}_{-1}}{C^{28}} = \frac{1}{1 + \rho^{28}} = \left[\frac{A^B_X}{A^B_U} \right]_R \left[\frac{C^{X-1}_X}{C_X} \right]_R \Phi K$$

where C^{28} is the ^{238}U cadmium ratio in the fuel rod, ρ^{28} is the ratio of epicadmium to subcadmium absorptions in ^{238}U in the fuel rod, A is the saturated specific activity of the product isotope (^{239}Np or ^{165}Dy), U and X are subscripts which refer to ^{238}U and the intermediate material (^{164}Dy), B is a superscript referring to the bare specific activity, C is the cadmium ratio, and R denotes an irradiation in the fuel rod.

With the exception of K, all quantities on the right hand side of the above equation are measurable. The ^{239}Np specific activities are measured by one of the three previously described counting techniques. The ^{165}Dy specific activities are obtained from NaI integral counting (above 32 keV) the γ activity of irradiated Dy-Al foils have the same diameter as the fuel.

The calibration factor Φ combines all terms involved in the thermal reference spectrum irradiation and is given by

$$\Phi = \left[\frac{A_U^B}{A_X^B} \right]_{\phi} \left[\frac{C_U^{-1}}{C_U} \right]_{\phi} \left[\frac{C_X^{-1}}{C_X} \right]_{\phi}$$

where the subscript ϕ denotes the thermal reference spectrum. K is a correction factor to account for the different energy distribution of thermal neutrons in the reference spectrum and lattice irradiations and the different energy dependence of ^{164}Dy and ^{238}U absorption cross sections in the thermal energy region, and is given by (49)

$$K = \frac{\int \sigma_U(E) \phi(E) G_U(E) dE}{\int \sigma_X(E) \phi(E) G_X(E) dE} \cdot \frac{\int \sigma_X(E) \phi^t(E) G_X(E) dE}{\int \sigma_U(E) \phi^t(E) G_U(E) dE}$$

The integrals are evaluated over the energy range from zero to the cadmium cutoff energy [which is 0.56 eV for 20 mil thick cadmium⁽⁵⁰⁾]. Here the superscript t designates values from the thermal reference spectrum. The function G(E) is included to account for self shielding and flux depression in the foil. If foil perturbation effects are accounted for separately, the K factor can be written

$$K = \left(\frac{g_U}{g_X} \right)_R \left(\frac{g_X}{g_U} \right)_\phi$$

where $g \equiv \frac{\bar{\sigma}}{\sigma_0}$, and $\bar{\sigma} \equiv \frac{\int \sigma(E)\phi(E)dE}{\int \phi(E)dE}$

and the integrals are evaluated over the energy range from zero to the cadmium cutoff energy. The σ_0 is the 2200 m/sec cross section for the material of interest. Further, if ^{238}U is assumed to be $1/v$, the expression for K can be written in terms of "non $1/v$ factors"⁽⁷⁾

$$K = (\text{Non } 1/v \text{ factor}_X)_\phi / (\text{Non } 1/v \text{ factor}_X)_R$$

where $(\text{Non } 1/v \text{ factor}_X) \equiv g_X/g_{1/v}$

Although K is in general a function of the neutron spectra and the differential cross sections involved, it is usually close to unity since the ^{164}Dy and ^{238}U absorption cross sections have approximately the same energy dependence in the thermal region and flux perturbations are similar and generally small. For this reason the error introduced by K is generally negligible in comparison with other errors.

Values of the various quantities used to obtain ρ^{28} in the present experiment are tabulated below. The dysprosium cadmium ratios are measured values. Measured specific activities and cadmium ratios for ^{238}U are given in a later section.

TABLE 6. Quantities Used in the Determination of ρ^{28} by the Thermal Activation Technique

Lattice (R)	$g_R^{238} = 1.007$	$g_R^{164} = 0.966$
Thermal Column (ϕ)	$g_\phi^{238} = 1.013$	$g_\phi^{164} = 0.986$

$$K = \left(\frac{g^{238}}{g^{164}} \right)_R \left(\frac{g^{164}}{g^{238}} \right)_\phi = 1.015$$

^{164}Dy Cadmium Ratio

Lattice: 43.6 ± 0.2

Thermal Column: 418 ± 2

B. THE USE OF FUEL SPACERS INSIDE THE CADMIUM BOX

As indicated previously, 0.125 in. thick spacers, of the same composition as the fuel, were used inside the cadmium covered assembly to simulate conditions present in the rod in the absence of the cadmium. This was necessary since the presence of cadmium next to a foil allows streaming of epithermal neutrons into the foil from the moderator through the cadmium without having had a possible interaction in the fuel.

The magnitude of this effect has been measured by many experimenters and indicates an extremely small mean free path for a neutron interaction with ^{238}U in the resonance region. A 20% increase in the ^{238}U cadmium ratio was observed by Klein⁽⁵¹⁾ et al., when 0.100 in. spacers were used relative to the case when they were not used. From the literature^(4,51,52) it appears that 100 mils represent an optimum fuel spacer thickness

although spacers as thick as 150 mils have been used.⁽⁵³⁾ In the present experiment, 125 mil thick pellets were available and were therefore utilized. The effect of these spacers on the value of ρ^{28} in the present experiment was measured and is indicated in a later section.

C. THE USE OF URANIUM METAL RATHER THAN OXIDE FOILS

Ideally, one would like to measure ρ^{28} with foils of the same material as the fuel so as to eliminate possible flux perturbations. This is seldom practical however, since as discussed earlier, foils depleted in ^{235}U are often required to minimize the interfering contribution of fission product activity when counting these foils. Furthermore, thin UO_2 foils are difficult to fabricate and thick foils necessitate large γ -ray attenuation corrections. For these reasons, thin depleted uranium metal foils are generally used, even in UO_2 fuel rods.

The use of uranium metal foils in a uranium oxide rod assumes that these foils will provide experimental data that is characteristic of the lattice and not a function of the detector. Several experiments at Bettis^(4,8,11,51) indicate that this assumption is valid for thin (0.005 in. thick) uranium metal foils and that these foils correctly sample the resonance flux without perturbing it. These conclusions were assumed to be valid for the 3 mil foils used in the present experiments and no corrections were made to the resonance activity of these foils.

D. THERMAL FLUX PEAKING AND DIPPING IN LATTICE AND THERMAL COLUMN FOILS

Foils used to monitor a given reaction rate in a medium, will generally have different nuclear properties than does the medium itself. As a result the flux in the foil will generally be different from the flux in the medium in the absence of the foil. The magnitude of this flux peaking or dipping in the foil will depend on the medium in which it is placed as well as the nuclear properties of the foil itself. Corrections for preferential surface absorption and edge effects in thick or highly absorbing foils may also be required in general but were considered to be negligible in the present case. Several formulas of various complexity have been derived for these corrections and appear in the literature. References 54 and 55 discuss several of these corrections and provide many references to both calculational and experimental methods and data available.

These corrections are not necessary if we are only interested in comparing the three counting techniques since they are independent of the counting technique. However, in order to compare measured and calculated values of ρ^{28} these corrections must be applied. Since this comparison was only of secondary concern here, only a relatively simple and inexpensive means for these corrections was justified in this case. However, the reader is strongly urged to consult References 54 and 55, and the references therein, when more accurate estimates of these corrections are required. Furthermore it is recommended in Reference 54 that methods which have not been evaluated or which do not properly account for neutron scattering be avoided since the results could be a factor of 2 to 3 in error.

A relatively simple formula used here to correct measured activities for thermal flux perturbations is given by⁽⁵⁷⁾

$$A_o = A_{\text{foil}} \left\{ 1 - t(\Sigma_{\text{medium}} - \Sigma_{\text{foil}}) \ln(L/t) \right\}$$

where A_o = Activity of the foil that would occur if the flux in the foil were the same as the flux in the medium

A_{foil} = Measured foil activity

t = Thickness of the foil

L = Diameter (or width) of the foil

Σ_{medium} = Macroscopic absorption cross section of the medium.

Σ_{foil} = Macroscopic absorption cross section of the foil.

This formula was applied to correct measured ^{239}Np and ^{165}Dy thermal activities in both the lattice and thermal column irradiations. Foil and medium thermal absorption cross sections for both cases were obtained from THERMOS⁽⁵⁸⁾ calculations. The thermal activity correction factor for foils in the lattice was 0.988 indicating a 1.2% thermal flux peaking in these foils relative to that in the fuel in which they were irradiated. Corrections for thermal flux dipping in the foils in the thermal column amounted to 0.44% in the case of ^{238}U and 2.0% for the Dy-Al foils. This latter result agrees reasonably well with measured data in Reference 56 which indicates a correction of 1.5% for this case. A two dimensional transport theory calculation would be required to accurately and explicitly account for each foil in the packet irradiated in the fuel rod.

RESULTS AND CONCLUSIONS

Measured ^{239}Np specific activities, ^{238}U cadmium ratios, and the associated values of ρ^{28} are summarized in Table 7 for the three counting techniques. Only the directly measured activities and ratios have been included in this table in order to allow the comparison of these values for the purpose of determining the presence or absence of systematic errors in a given counting technique. Furthermore, the activities and ratios indicated have not been corrected for thermal flux peaking or dipping perturbations, and in this sense can be considered as raw data.

The errors quoted in this section for the measured activities are standard deviations based on the consistency of repeated observations and are due only to random statistical fluctuations. The ratios quoted and their averages were obtained via inverse variance weighting with errors being propagated by standard statistical methods. (59-62)

Considering the specific activities in Table 7 we see that in all cases the extreme values obtained via the three counting techniques do not differ by more than 2% in the worst case and in general, the three values are within $\pm 1\%$ of their average value. In many cases the agreement is even better than this. Even though, in several cases, the values obtained from the three counting techniques do not overlap within the quoted statistical errors, there does not appear to be a consistent bias in any of the techniques relative to another which would positively indicate the presence of a systematic error due to the mode of counting. The same conclusions are drawn when the cadmium ratio or ρ^{28} values are compared.

TABLE 7. Summary of ^{239}Np Specific Activities of Depleted Uranium Foils for Three Counting Techniques. (Activities and ratios are not corrected for foil flux perturbation effects.)

Foil Position	Measured Reaction Rate* or Ratio	Counting Technique		
		NaI	NaI-NaI	Ge(Li)
<u>Lattice**</u>	Bare specific activity	1.0006(± 0.0035)	0.9794(± 0.0022)	0.9827(± 0.0046)
		Average 0.9863(± 0.0017)		
	Cd covered specific activity	0.5561(± 0.0014)	0.5543(± 0.0015)	0.5511(± 0.0023)
		Average 0.5546(± 0.0009)		
^{238}U Cd Ratio		1.799 (± 0.010)	1.767 (± 0.006)	1.783 (± 0.011)
		Average 1.777 (± 0.005)		
	ρ^{28}	1.252 (± 0.016)	1.304 (± 0.011)	1.277 (± 0.018)
		Average 1.285 (± 0.008)		
<u>Lattice***</u>	Bare specific activity	(above)	(above)	
	Cd covered specific activity	0.7940(± 0.0010)	0.7865(± 0.0030)	
		Average 0.7933(± 0.0009)		
	^{238}U Cd Ratio	1.260 (± 0.005)	1.245 (± 0.006)	
ρ^{28}		Average 1.253 (± 0.004)		
		3.846 (± 0.069)	4.082 (± 0.092)	
		Average 3.931 (± 0.055)		
<u>Thermal Column</u>	Bare specific activity	1.1127(± 0.0048)	1.1249(± 0.0025)	1.1074(± 0.0046)
		Average 1.1117(± 0.0030)		
	Cd covered specific activity	0.3288(± 0.0009)	0.3320(± 0.0005)	0.3358(± 0.0018)
		Average 0.3315(± 0.0004)		
^{238}U Cd Ratio		3.384 (± 0.012)	3.388 (± 0.009)	3.298 (± 0.022)
		Average 3.378 (± 0.007)		
	ρ^{28}	0.419 (± 0.002)	0.419 (± 0.002)	0.435 (± 0.004)
		Average 0.421 (± 0.002)		

* Reaction rates are quoted relative to a bare monitor foil of the same type in the same irradiation.

** Cadmium covered irradiation made with 0.125 in. fuel pellets inside cadmium "box".

*** Cadmium covered irradiation made without 0.125 in. fuel pellets inside cadmium "box".

As a result it is felt that the quoted uncertainties which were based on the consistency of repeated observations of the random statistical fluctuating activities, do not in all cases, fully account for the entire source of random statistical errors which are actually present in the experiment. Based on the data in Table 7 a more reasonable value for the total random statistical error in the cadmium ratio in the general case would be $\pm 1\%$. A $\pm 1\%$ random statistical error in the cadmium ratio is not considered unreasonable for these types of measurements. Thus, the general agreement of the cadmium ratio values from all three counting techniques to within 2% is considered statistically good and sufficient to indicate the absence of systematic errors due to the counting mode in the present experiment.

The effect of the fuel spacers inside the cadmium box in the lattice irradiation is evidence by the data in Table 7. The 43% increase in the cadmium covered specific activity in the absence of the fuel spacers decreases the ^{238}U cadmium ratio by 30%. The effect on ρ^{28} is even more dramatic with its value increasing by more than a factor of 3 in the absence of the fuel spacers. The presence of these fuel spacers inside the cadmium box in the lattice irradiation to eliminate streaming of epithermal neutrons from the moderator through the cadmium and into the foils is therefore justified.

Table 8 provides a summary of cadmium ratio and ρ^{28} values for the three counting techniques which have been corrected for thermal flux perturbations in the foils. Cadmium ratio and ρ^{28} values obtained by both direct cadmium ratio measurements in the lattice and via the thermal activation method are given for each counting technique.

TABLE 8. Summary of Measured and Calculated Values
for the ^{238}U Cadmium Ratio and ρ^{28}

		<u>Measured Values</u>			
<u>Foil Position</u>	<u>Measured Ratio</u>	<u>Counting Technique</u>			
		<u>NaI</u>	<u>NaI-NaI</u>	<u>Ge(Li)</u>	
Lattice	^{238}U Cd Ratio (Direct)	1.789 (± 0.010)	1.758 (± 0.006)	1.774 (± 0.011)	
		Average 1.768 (± 0.005)			
	^{238}U Cd Ratio (T.A.)	1.773 (± 0.011)	1.754 (± 0.008)	1.750 (± 0.011)	
		Average 1.758 (± 0.006)			
	ρ^{28} (Direct)	1.267 (± 0.016)	1.320 (± 0.011)	1.293 (± 0.018)	
		Average 1.301 (± 0.008)			
	ρ^{28} (T.A.)	1.293 (± 0.018)	1.326 (± 0.013)	1.333 (± 0.020)	
		Average 1.319 (± 0.009)			
Thermal Column	^{238}U Cd Ratio (Direct)	3.394 (± 0.012)	3.398 (± 0.009)	3.308 (± 0.022)	
		Average 3.388 (± 0.007)			
	ρ^{28} (Direct)	0.418 (± 0.002)	0.418 (± 0.002)	0.433 (± 0.004)	
		Average 0.419 (± 0.001)			
		<u>Calculated Values</u>			
<u>Foil Position</u>	<u>Calculated Ratio</u>	<u>Model*</u>			
		<u>THERMOS-EGGNIT</u>		<u>THERMOS-HRG</u>	
		<u>$E_{\text{co}} = 0.683$</u>	<u>$E_{\text{co}} = 0.414$</u>	<u>$E_{\text{co}} = 0.683$</u>	<u>$E_{\text{co}} = 0.414$</u>
Lattice	^{238}U Cd Ratio	1.799	1.776	1.829	1.804
	(Linear Interpolation at $E_{\text{co}} = 0.555$ eV)	1.788		1.817	
	ρ^{28}	1.251	1.289	1.207	1.244
	(Linear Interpolation at $E_{\text{co}} = 0.555$ eV)	1.269		1.225	

* See Appendix D for details of these calculations and models.

Comparing individual values for a given counting technique or comparing averages for three counting techniques both lead to the results that the thermal activation method yields a higher value for ρ^{28} than does the direct measurement of the cadmium ratio. This indicates that there is a systematic perturbation of the epicadmium flux in the cadmium covered lattice irradiation, but the magnitude of the perturbation, as indicated by the difference in the ρ^{28} values from the two methods, is of the same order of magnitude as the random statistical errors quoted. Thus, although we can see that this systematic error is present from the consistently higher ρ^{28} values from the thermal activation method about all we can say concerning its magnitude in the present case, is that it appears to be less than 1% of ρ^{28} .

Also shown in Table 8 are calculated values of the ^{238}U cadmium ratio and ρ^{28} via two analytical models. Details of these calculations and the models used are discussed in Appendix D. A few words concerning the concept of an effective cadmium cutoff energy are, however, appropriate here.

The comparison of measured and calculated cadmium ratios or ρ^{28} values is facilitated by the concept of an ideal neutron filter which yields the same neutron activation in the detector foil as a given cadmium filter but which transmits all neutrons above a certain cutoff energy, E_{co} , and none below. This cut off energy for a given foil is a function of the cross section variation (with energy) of the foil, filter, and medium in which the foil is placed, as well as the neutron energy spectrum present and the thickness of the filter and the foil. The effect of cadmium thickness on the effective cutoff energy for thin ^{238}U foils is relatively small since the ^{238}U cross section is approximately $1/v$ in this energy range and

is small compared to the resonance integral. A calculated value for the effective cadmium cutoff energy for a thin 1/v detector in a 20 mil cadmium cover in an isotropic flux is 0.555 ev.⁽⁵⁰⁾

The calculations were made using cutoff energies of 0.414 ev and 0.683 ev as these values provided a convenient interface between the code generating the thermal group constants (THERMOS) and those generating the fast group constants [HRG3⁽⁶³⁾ and EGGNIT⁽⁶⁴⁾]. Since the actual thickness of cadmium used in the experiments was 20 mils a linear interpolation between the two calculated values was made to get the cadmium ratio and ρ^{28} for the cutoff energy of 0.555 ev. These interpolated values are also shown in Table 8.

Comparing the measured and calculated values of the cadmium ratios from Table 8, we see that the measured values are from 0 to 2% lower than the THERMOS-EGGNIT calculated values and in greater disagreement with the THERMOS-HRG values. This 2% bias in the cadmium ratio propagates to a 5% bias in the value of ρ^{28} as seen from Table 8.

A possible source of systematic error which would yield too high a measured value for ρ^{28} would be the streaming of epithermal neutrons through the Dy-Al, U(93)-Al, and Al foils which are adjacent to the depleted uranium foil in the foil packet. Unfortunately, no irradiation was made without these supplementary foils in the packet to check this hypothesis out. Due to the size of the foils involved this effect would be expected to be small but could account for part of the difference observed between the measured and calculated values of ρ^{28} .

COMPARATIVE EVALUATION OF THE THREE COUNTING TECHNIQUES

The results of the measurements which have been reported herein indicate that from the standpoint of accuracy all three counting techniques are equivalent and that there is no reason to choose one method over another in the present application. It is recognized however, that this will not generally be true, especially in applications where interfering fission product activities become appreciable relative to the desired ^{239}Np activity. Furthermore, there are several factors which must be considered in addition to accuracy requirements when choosing an optimum counting technique for a given application. These factors, which will carry different weights depending on experimental conditions and requirements, are enumerated below:

1. Accuracy or precision required in the final measured result.
2. Counting equipment cost, complexity, availability, and stability requirements.
3. Foil irradiation and counting time required as dictated by the flux level available for the irradiations.
4. Foil material cost and availability.
5. Scale of the measurements.
6. The presence or absence of systematic errors, their relative magnitudes, and the availability of an experimental means for their correction.

With these factors in mind, we can now compare the relative advantages and disadvantages of the three counting techniques used in this study for certain particular applications and experimental conditions.

1. NaI Singles Counting

The major advantage that this technique has and the major reason for its general popularity, is its ease of application. The counting equipment required is relatively simple and its cost is moderate. Counting systems of this type are generally available at laboratories where activation measurements are made. Counting system stability requirements are no more stringent than for any other activation type measurements. Due to the relatively high efficiency of NaI detectors, foil irradiation and counting times are minimal to moderate, depending on the flux level available for the irradiation, making this technique particularly attractive for large scale experiments.

The foil material is generally limited to thin depleted uranium in order to minimize interfering fission product activities and maintain reasonable accuracy and precision requirements. In addition to being costly and generally difficult to obtain, depleted uranium foils can cause appreciable flux perturbations which must be accounted for. Corrections for these perturbations can be obtained experimentally by irradiating foils of different thicknesses and extrapolating the activities to zero foil thickness.

The only other major source of systematic experimental error is due to the interfering fission product activity. Although corrections for the fission product activity can, in principle, be made experimentally, it does require irradiation of additional foils which may perturb the measured ^{239}Np activity. When fission product activity corrections become appreciable (i.e., when foils with an enrichment greater than natural are

used or when fast reactor neutron spectrum activations are involved) it is generally recommended that the NaI-NaI coincidence technique be employed.

2. NaI-NaI Coincidence Counting

A major advantage of this technique is its general utility, due to its selective separation of ^{239}Np from fission product activity even in cases where an appreciable fission product activity is present. The counting equipment required is more complex than that for NaI singles counting but the system cost is significantly less than double that for the NaI singles method. In addition to doubling the number of detectors, preamplifiers, amplifiers, power supplies and scalers required, a single channel analyzer is needed as well as a multichannel analyzer with coincidence analysis capabilities. Counting system stability requirements are high for this mode of counting although they are not generally hard to meet in practice.

Since the coincidence activity is an order of magnitude lower than the singles activity, longer irradiation and count times are required for a given available flux level. This could limit its application to small scale experiments if adequate flux levels are not available.

Even though depleted uranium foils were used in the present experiments, higher enrichment foils could have been used with no loss of accuracy. Analogously, the technique could be expected to be applied to fast reactor spectrum activations with the same degree of confidence.

Utilizing foils made from the fuel material would eliminate flux perturbations due to the presence of the foil in the fuel rod. Experimental methods requiring auxiliary irradiations, are available for the

determination of the generally small fission product coincidence correction which is the only appreciable systematic error associated with the technique. At present the NaI-NaI coincidence technique is felt to be the best all-around method available for ^{239}Np specific activity determinations.

3. Ge(Li) Singles Counting

This technique has the ease of application of the NaI singles technique with the potential general utility of the NaI-NaI coincidence technique. The counting equipment required is relatively simple although the detectors are presently quite expensive relative to NaI and require liquid nitrogen cooling. The high energy resolution of these detector systems requires the high system stability achievable only with modern day transistorized electronic equipment.

The efficiency at 100 keV of the 20 cc Ge(Li) detector used in the present experiments was close to an order of magnitude lower than that of the NaI detector at the same energy. The specific activities obtained with the Ge(Li) detector were therefore nearly the same as the recorded NaI-NaI coincidence activities. Thus, irradiation and count times required for a given flux level will be very similar to those for the NaI-NaI coincidence technique. Here again, if adequate flux levels are not available, the application of these detectors could be limited to small scale experiments.

Although depleted uranium foils were used in the present experiments, the energy resolution obtained with the Ge(Li) counting system seemed to indicate that higher enrichment foils could have been used with no loss in accuracy. It is strongly recommended that this hypothesis be tested by

irradiating higher enrichment foils (or low enrichment foils in a fast neutron spectrum) and observing the γ -ray spectrum as a function of time after rod drop on both a 20 cc detector, and a smaller, high resolution spectrometer. This type of experiment could provide data which would indicate whether or not the relatively simple Ge(Li) spectrometer could replace the NaI-NaI coincidence system as the best all-round method available for this application.

Since fission product corrections are not expected to be required for this method, no additional foils need be irradiated. Furthermore, since it appears that the fuel material can be used as the foil material, there will be no flux perturbation caused by the presence of the foil in the fuel. The apparent absence of sources of systematic errors associated with this counting technique makes it a strong candidate for further development.

REFERENCES

1. D. R. Oden, "Comparison of Calculated and Measured Values of ρ^{28} and δ^{28} for 26 Lattices of UO_2 Rods in a Light Water Moderator," BNWL-1589 May, 1971.
2. H. R. Lutz and Y. Hachiya, " ρ^{28} Measurements of Tight Water Lattices with Natural Uranium Foils," Appendix B of: G. A. Price, "Uranium-Water Lattice Compilation-Part 1, BNL Exponential Assemblies," BNL-50035, December 30, 1966. (See also, H. R. Lutz and Y. Hachiya, " ρ^{28} Measurements of Tight Water Lattices with Natural Uranium Foils," Trans. ANS 6, No. 1, 1963.)
3. H. A. Risti, "Microscopic Parameters by the Cadmium Ratio Technique and Perturbation Effects," Trans. ANS 5, No. 1, p. 84, 1962.
4. P. W. Davison, J. D. Cleary, B. Jennings, H. A. Risti and G. H. Minton, "Microscopic Lattice Parameters in Single and Multi-Region Cores: A Comparison of Theory and Experiment," WCAP 1434, June 1961.
5. B. Arcipiani, D. Ricabarra and G. H. Ricabarra, "A Note on the Measurement of the U^{238} Cadmium Ratio," NSE 14, 316-317 (1962).
6. C. Ertek, A. Yalcin, and Y. Inel, "An Accurate Determination of the Cadmium Ratio of Neutron Capture Rates in U^{238} in Slightly Enriched Uranium," NSE 36, 209-219 (1969).
7. R. H. Lewis, T. C. Engelder, D. M. Roberts and D. B. Wehmeyer, "Thermal Activation Method for ρ^{28} Measurements in Slightly Enriched Uranium Oxide Lattices, BAW-1268, May 1963.
8. G. G. Smith, J. Hardy, D. Klein and J. A. Mitchel, "Experimental Studies of U^{238} Resonance Neutron Capture in UO_2 Fuel Rods," NSE 9, 421-429 (1961).
9. J. Hardy, Jr., G. G. Smith and D. Klein, "The Effective Resonance Capture Integrals of Uranium Metal and UO_2 Rods," NSE 14, 358-365 (1962).
10. W. G. Davey, "The Ratio of U^{238} Capture and U^{235} Fission Cross Sections in Fast Reactors," NSE 24, 26-41 (1966).
11. W. Baer, M. J. Galper and N. Carbone, "Measurements of the Conversion Ratio of the PWR Critical Facility Blanket," NSE 3, 113-128 (1958).
12. A. Weitzberg, I. Kaplan and T. J. Thompson, "Measurements of Neutron Capture in U^{238} in Lattices of Uranium Rods in Heavy Water," NYO-9659 (January 1962).
13. D. A. Daavettila, "Measurements of Conversion Ratio, Resonance Escape and Fast Fission as Student Experiments," NSE 20, 142-150, (1964). See also D. A. Daavettila, "A Manual of Reactor Laboratory Experiments," ANL-6990, Sections 12-14, 1965.

14. A. M. Bresesti, M. Bresesti and H. Neumann, "A Method for Absolute Determination of U^{238} Capture Rates," Journal of Nuclear Energy, Vol. 23, pp. 379-386, 1969.
15. L. S. Beller, "Beta-Gamma Delayed Coincidence Method for Resonance Escape Measurements," NSE 9, p. 411, 1961
16. R. Sher, " γ - γ Coincidence Method for Measuring Resonance Escape Probability in U^{238} Lattices," NSE 7, 479 (1960).
17. A. Weitzberg and T. J. Thompson, "Coincidence Technique for U-238 Activation Measurements," Trans. ANS 3, 456 (1960).
18. P. R. Tunnicliffe, D. J. Skillings and B. G. Chidley, "A Method for the Accurate Determination of Relative Initial Conversion Ratios," NSE 15, 268-283 (1963).
19. H. Seufert and D. Stegemann, "A Method for Absolute Determination of U^{238} Capture Rates in Fast Zero-Power Reactors," NSE 28, 277-285 (1967).
20. R. Chawla and C. B. Besant, "Absolute Measurements of U^{238} Capture and Pu^{239} Fission Rates in Fast Reactors," The Journal of British Nuclear Energy Society, January 1970, p. 28-34.
21. P. Kafalas and R. R. Heinrich, "Capture and Fission Patterns of U^{238} in EBR-I (Mark II)," NSE 4, 698-702 (1958).
22. Proceedings of the Brookhaven Conference on Resonance Absorption of Neutrons in Nuclear Reactors, BNL-433, pp 5-12, 32 (September 1956).
23. Judson Hardy Jr., G. G. Smith and D. Klein, "Experimental Check of a Monte Carlo-Calculated Distribution of Resonance Neutron Capture in a Gold Rod," NSE 7, 263-267. (1960).
24. D. Klein, W. Baer and G. G. Smith, "Spatial Distribution of U^{238} Resonance Neutron Capture in Uranium Metal Rods," NSE 3, 698-706 (1958).
25. W. H. D'Ardenne, T. J. Thompson, D. D. Lanning and I. Kaplan, "Studies of Epithermal Neutrons in Uranium, Heavy Water Lattices," MIT-2344-2 (MITNE53) August 1964.
26. Eric Hellstrand, "Measurements of the Effective Resonance Integral in Uranium Metal and Oxide in Different Geometries," Journal of Applied Phys. 28, No. 12, December 1957.
27. C. M. Lederer, J. M. Hollander and I. Perlman, "Table of Isotopes," 6th Edition, John Wiley and Sons, 1967.

28. G. T. Ewan, J. W. Knowles and D. R. MacKenzie, "Internal Conversion Coefficients of γ rays in Pu^{239} ," Phys. Rev. Vol. 108, No. 5, p. 1308-1311 December 1957.
29. R. L. Graham and R. E. Bell, "Disintegration of Np^{239} ," Phys. Rev. Vol. 83, p. 222-223 (1951).
30. J. M. Hollander, W. G. Smith and J. W. Mihelich, "Energy Levels of Pu^{239} Populated by the Beta Decay of Np^{239} ," Phys. Rev. Vol. 102, No. 3, p. 740, May 1956.
31. G. C. Nelson, B. G. Saunders and W. John, " $K_{\alpha 1}$ and $K_{\alpha 2}$ X-Ray Energies of Neptunium, Plutonium, and Americium," Phys. Rev. Vol. 188, No. 1, December 1969.
32. J. E. Cline, "Gamma Rays Emitted by the Fissionable Nuclides and Associated Isotopes," IN-1448 Rev, January 1971.
33. R. Gunnink and R. J. Morrow, "Gamma-Ray Energies and Absolute Branching Intensities for ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu and ^{241}Am ," UCRL-51087, July 1971.
34. K. Siegbahn, "Alpha-, Beta- and Gamma Ray Spectroscopy," Vol. 1, Chapters V, VII and VIII, North-Holland Publishing Company, Amsterdam, 1968.
35. B. Crasemann and H. Easterday, "Locating Compton Edges and Backscatter Peaks in Scintillation Spectra," Nucleonics Data Sheet.
36. H. M. Antunez, "Determination of Fission Product Background in the Gamma Coincidence Technique for U^{238} Activation Measurements," NSE 14, pp. 8-10 (1962).
37. P. W. deLange, C. B. Bigham, R. E. Green and T. J. Manuel, "Experimental Initial Conversion and Fast Fission Ratios for Clusters of Natural U and UO_2 in D_2O ," AECL-2636 (1966).
38. D. C. Camp, "Applications and Optimization of the Lithium-Drifted Germanium Detector System," UCRL 50156, March 1967.
39. G. T. Ewan and A. J. Tavendale, "High-Resolution Studies of Gamma-Ray Spectra Using Lithium-Drift Germanium Gamma-Ray Spectrometers," Canadian Journal of Physics, Vol. 42, p. 2286 (November 1964).
40. F. S. Goulding and Y. Stone, "Semiconductor Radiation Detectors," Science, Vol. 170, October 1970, pp. 280-289.
41. W. R. French, Jr., R. L. LaShure and J. L. Curran, "Lithium-Drifted Germanium Detectors," American Journal of Physics, Vol. 37, No. 1, pp. 11-22, January 1969.

42. F. S. Goulding, "Semiconductor Detectors for Nuclear Spectrometry, I," Nuclear Instruments and Methods, Vol. 43, p. 1-54, August 1966.
43. J. W. Mayer, "Semiconductor Detectors for Nuclear Spectrometry, II," Nuclear Instruments and Methods, Vol. 43, p. 55-64, August 1966.
44. J. M. Hollander, "The Impact of Semiconductor Detectors on Gamma-Ray and Electron Spectroscopy," Nuclear Instruments and Methods, Vol. 43, pp. 65-109, August 1966.
45. J. A. Cooper, "Applied Ge(Li) Gamma-Ray Spectroscopy," BNWL-SA-3603, January 1971.
46. L. N. Price, "A Systematic Study of Foil Correction Factors in the Measurement of δ_{28} , δ_{25} , ρ_{28} , and C^* in Slightly Enriched, D₂O-Moderated Lattices," September 1966 (MIT-M.S. Thesis).
47. A. V. Bushuev and L. N. Yurova, "Sources of Systematic Errors in Measuring the Cadmium Ratio in U²³⁸," Soviet Atomic Energy, Vol. 27, No. 4, p. 1095, October 1969.
48. V. B. Egiazarov, V. S. Dikarev and V. G. Madeev, Proc. Conf. Academy of Sci. of USSR on the Peaceful Uses of Atomic Energy (English Translation), Consultants Bureau, N.Y. (1955) pp. 59-67.
49. G. S. Sanford, "Determination of the K-Constant for the Dysprosium Substitution Method," Reactor Physics Division Annual Report, July 1, 1965 to June 30, 1966, ANL-7210, pp. 325-327, December 1966.
50. N. P. Baumann, "Resonance Integrals and Self Shielding Factors for Detector Foils," DP-817, January 1963.
51. D. Klein, A. Z. Kranz, G. G. Smith, W. Baer and J. DeJuren, "Measurements of Thermal Utilization, Resonance Escape Probability, and Fast Effect in Water Moderated, Slightly Enriched Uranium and Uranium Oxide Lattices," NSE 3, 403-427 (1958).
52. V. E. Grob, E. Santandrea and H. Ritz, "Measurements of Parameters Leading to ρ_{28} , f , and ϵ in Light Water Moderated 4.48% and 2.73% Enriched Lattices," NSE 7, 514-524 (1960).
53. P. W. Davison, S. S. Berg, W. H. Bergmann, D. F. Hanlen, B. Jennings, R. D. Leamer and J. E. Howard, "Yankee Critical Experiments-Measurements on Lattices of Stainless Steel Clad Slightly Enriched Uranium Dioxide Fuel Rods in Light Water," YAE-94, April 1959.
54. L. C. Schmid, Critical Assemblies and Reactor Research, Wiley-Interscience, 1971.
55. K. H. Beckurts and K. Wirtz, Neutron Physics, Springer-Verlag, 1964.

56. G. M. Jacks, "A Study of Thermal and Resonance Neutron Flux Detectors," DP-608, August 1961.
57. F. E. Dunn, "Flux Dipping and Peaking in Foils Used in Thermal Disadvantage Factor Measurements," Trans Ans, 9, No. 1, 189 (1966).
58. H. C. Honeck, "THERMOS - A Thermalization Transport Theory Code for Reactor Lattice Calculations," BNL-5826, September 1961.
59. C. Eisenhart, "Expression of the Uncertainties of Final Results," Science, Vol. 160, pp. 1201-1204, June 1968.
60. A. H. Jaffey, "Statistical Tests for Counting," Nucleonics, Vol. 18, No. 11, pp. 180-184, November 1960.
61. C. W. Tittle, "How to Apply Statistics to Nuclear Measurements," Nuclear-Chicago Technical Bulletin No. 14, Nuclear-Chicago Corporation, 333 East Howard Avenue, DesPlaines, Illinois.
62. A. A. Jarrett, "Statistical Methods Used in the Measurement of Radioactivity (Some Useful Graphs)," AECU-262 MONP-126, June 1946.
63. J. L. Carter, "HRG3: A Code for Calculating the Slowing-Down Spectrum in the P_1 or B_1 Approximation," BNWL-1432, June 1970.
64. C. R. Richey, "EGGNIT: A Multigroup Cross Section Code," BNWL-1203, November 1969.
65. C. L. Bennet and W. L. Purcell, "BRT-1: Battelle-Revised-Thermos," BNWL-1434, June 1970.
66. B. J. Toppel and I. Baksys, "The Argonne-Revised THERMOS Code," ANL-7023, March 1965.
67. G. D. Joanou and J. S. Dudek, "GAM-I: A Consistent P_1 Multigroup Code for the Calculation of Fast Neutron Spectra and Multigroup Constants," GA-1850, June 1961.
68. L. L. Carter, C. R. Richey and C. E. Hughey, "GAMTEC II: A Code for Generating Consistent Multigroup Constant Utilized in Diffusion and Transport Theory Calculations," BNWL-35, March 1965.
69. F. T. Adler, G. W. Hinman and L. W. Nordheim, "The Quantitative Evaluation of Resonance Integrals," Paper No. P./1988, Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Vol. 16, pp. 155-171, United Nations, Geneva, 1958.
70. G. F. Kuncir, "A Program for the Calculation of Resonance Integrals," GA-2525, August 1961.
71. I. Carlvik, "The Dancoff Correction in Square and Hexagonal Lattices," NSE 29, 325-336 (1967).

APPENDIX A

CORRECTION FOR RANDOM COINCIDENCES

Due to the finite resolving time of each channel in the coincidence circuit, false or random coincidences will occur in addition to the true coincidences. These random coincidences will result whenever a γ -ray activates one detector and an unrelated γ -ray activates the other detector within the resolving time of the first detector and vice versa. Since both true and random coincidences are registered as true coincidences, a correction to the measured coincidence count rate must be applied to account for the random coincidences.

An expression relating the true coincidence rate to the measured coincidence rate is arrived at by considering the events in each channel which result in random coincidences. If N_1^m and N_2^m are measured count rates in channel 1 and channel 2, N_c' the true coincidence count rate, and N_R the random coincidence rate, then the portion of N_1^m which does not result in true coincidences is $(N_1^m - N_c')$. If the resolving time of channel 1 is τ_1 , then channel 1 is set up to observe random coincidences for the portion $(N_1^m - N_c')\tau_1$ of the running time. Since pulses from channel 2 are arriving at a rate N_2^m , the rate of random coincidences due to these pulses is $(N_1^m - N_c')\tau_1 N_2^m$. This expression then accounts for the random coincidences due to single pulses in channel 1 being followed within its resolving time τ_1 , by single pulses in channel 2. By analogy the random coincidence rate due to single pulses in channel 2 being followed within its resolving time τ_2 , by single pulses in channel 1 is $(N_2^m - N_c')\tau_2 N_1^m$. The total random coincidence rate is therefore

$$N_R = (N_1^m - N_C^m) \tau_1 N_2^m + (N_1^m - N_C^m) \tau_2 N_1^m \quad . \quad (A-1)$$

Noting that

$$N_C^m = N_C^m + N_R$$

where N_C^m is the measured coincidence count rate, we have

$$N_C^m - N_C^m = (N_1^m - N_C^m) \tau_1 N_2^m + (N_2^m - N_C^m) \tau_2 N_1^m \quad .$$

Since two identical NaI detectors were used in the symmetrical coincidence apparatus $\tau_1 = \tau_2 = \tau$ and we have

$$N_C^m = N_C^m [1 - \tau(N_1^m + N_2^m)] + 2\tau N_1^m N_2^m$$

$$N_C^m = \frac{N_C^m - 2\tau N_1^m N_2^m}{1 - \tau(N_1^m + N_2^m)} \quad (A-2)$$

$$N_C \sim N_C^m - 2\tau N_1^m N_2^m = N_C^m - N_R \quad (A-3)$$

$$\text{or} \quad N_R \sim 2\tau N_1^m N_2^m \quad . \quad (A-4)$$

The validity of the approximation resulting in Equation (A-3), i.e., that $\tau(N_1^m + N_2^m) \ll 1$, is demonstrated by consideration of the maximum usable values of N_1^m and N_2^m for a given percent random coincidence correction. Since N_1^m and N_2^m are proportional to the source strength, we see that the random coincidences increase as the square of the source strength. This

puts an upper limit on the values at N_1^m and N_2^m usable in order not to exceed a given random to true coincidence ratio.

The maximum usable source disintegration rate, N_1^m , for a system with a coincidence resolving time 2τ is given by the relation⁽³⁴⁾

$$\frac{N'_C}{N_R} = \frac{f(E_1, E_2)}{2\tau N_1^m} \quad (\text{A-5})$$

where N'_C/N_R is the ratio of true to random coincidence rates and $f(E_1, E_2)$ is the fraction of the pulses in the portion of the spectrum accepted by one analyzer window that is related by coincidence to the radiation being accepted by the other analyzer. For the system used in this experiment it was found that $f(E_1, E_2) \approx N'_C/N_2^m \approx 0.1$. We therefore have

$$\frac{N'_C}{N_R} = \frac{N'_C}{2\tau N_1^m N_2^m}$$

in agreement with Equation (A-4).

Due to the symmetrical detector arrangement $N_2^m \approx N_1^m = N$, so that

$$\frac{N'_C}{N_R} = \frac{N'_C}{2\tau N^2}$$

Thus by establishing the criteria that the maximum random coincidence correction be 5% we have for $2\tau = 0.5 \mu \text{ sec}$ (see Appendix C)

$$N = N_1^m = N_2^m = \frac{N_R}{N'_C} \frac{f(E_1, E_2)}{2\tau} = 0.5 \frac{0.1}{0.5 \times 10^{-6} \text{ sec}} \times \frac{60 \text{ sec}}{\text{min}}$$

$$N = 6 \times 10^5 \text{ counts/min.}$$

In all cases coincidence counting was performed with $N_1^m \approx N_2^m \leq 2 \times 10^5$ counts/min, so that the random coincidence correction was never greater than 3%. Using Equation (A-3) to correct for random coincidences instead of Equation (A-2) would result in an underestimate of N_c by 0.16% in the worst case when $N_1^m \approx N_2^m = 2 \times 10^5$ counts/min.

It should be noted that $f(E_1, E_2)$ in Equation (A-5) depends strongly on the coincidence spectrum and is much less than 0.1 if either window is moved off the 105 keV "composite peak." This is to be expected since the highest intensity peaks in the singles spectrum (see Figure 6) are the coincident peaks due to the ^{239}Np γ -ray at 106 keV and the ^{239}Pu X-ray at 104 keV. It is for this reason that the maximum coincidence efficiency and lowest ratio of random to true coincidences occur with both windows set on the 105 keV "composite peak." With any other window settings, the maximum usable disintegration rate would be smaller requiring longer counting times for a specified ratio of random to true coincidence rates, or alternately for the same counting time a higher random to true coincidence rate would be observed. Thus by the judicious use of energy discrimination, a high disintegration rate can be used to maximize coincidence efficiency even in the presence of many interfering radiations.

APPENDIX B

CORRECTION FOR COINCIDENCE LOSSES

The loss of a pulse in either channel due to its finite resolving time can result in a lost coincidence. Corrections to the measured coincidence count rate must be applied to account for these losses, although coincidence losses due to the resolving time of the coincidence recording circuit were corrected for automatically by utilizing the live time mode of the 512 channel analyzer.

If the true count rate in channel 1 is N_1 , then channel 1 will be "dead" and insensitive to incoming pulses for the portion $N_1\tau_1$ of the running time, where τ_1 is the dead time (or resolving time) of channel 1. The measured count rate N_1^m will therefore be

$$N_1^m = \frac{N_1}{1 + N_1\tau_1} .$$

Solving for the true count rate we get

$$N_1 = \frac{N_1^m}{1 - \tau_1 N_1^m} . \tag{B-1}$$

Similarly, for channel 2

$$N_2 = \frac{N_2^m}{1 - \tau_2 N_2^m} . \tag{B-2}$$

Now if N_1^m pulses result in N'_C measured coincidences then N_1 pulses would result in N_C true coincidences. The ratio of true to measured coincidences, N_C/N'_C , is therefore

$$\frac{N_C}{N'_C} = \frac{N_1}{N_1^m} = \frac{N_1^m / (1 - \tau_1 N_1^m)}{N_1^m}$$

or

$$\frac{N_C}{N'_C} = \frac{1}{1 - \tau_1 N_1^m} \quad (B-3)$$

Similarly for channel 2

$$\frac{N_C}{N'_C} = \frac{1}{1 - \tau_2 N_2^m} \quad (B-4)$$

The total correction is then the product of the factors for the two channels resulting in

$$N_C = \frac{N'_C}{(1 - \tau_1 N_1^m)(1 - \tau_2 N_2^m)} \quad (B-5)$$

Neglecting second order terms in τ which amount to a contribution of only 7×10^{-7} in the denominator of Equation (B-5) for $N_1^m = N_2^m = 2 \times 10^5$ counts/min, we get with $\tau_1 = \tau_2 = \tau$

$$N_C = \frac{N'_C}{1 - \tau(N_1^m + N_2^m)} \quad (B-6)$$

The maximum value of this correction amounted to 0.16% for the case when $N_1^m = N_2^m = 2 \times 10^5$ counts/min.

Combining Equation (B-6) above with Equation (A-2) of Appendix A we get for the true coincidence rate

$$N_C = \frac{N_C^m - 2\tau N_1^m N_2^m}{[1 - \tau(N_1^m + N_2^m)]^2} \quad (B-7)$$

Again, neglecting second order terms in τ in the demoninator, which contribute only 7×10^{-12} for the worst case when $N_1^m = N_2^m = 2 \times 10^5$ counts/min we get for the true coincidence rate

$$N_c = \frac{N_c^m - 2\tau N_1^m N_2^m}{1 - 2\tau(N_1^m + N_2^m)} \quad (B-8)$$

The composite correction for random coincidences and coincidence losses in Equation (B-8) was always less than 3% of N_c in these experiments.

APPENDIX C

MEASUREMENT OF COINCIDENCE RESOLVING TIME

It was pointed out previously that it is desirable to have the coincidence resolving time as low as possible from the standpoint of minimizing random coincidence losses and maximizing usable source strengths. However, if all true coincidences are to be recorded without loss, the resolving time must be much greater than the mean life of the excited state between coincident radiations so that the radiations are truly coincident when compared to the resolving time. In addition, the resolving time of each channel must be long enough to accommodate electronic delays in the separate channels (which cannot be matched perfectly) or true coincidences are lost. In the present case employing NaI (Tl) scintillation detectors the latter consideration is the more important in the design of the coincidence circuit. The resolving time of the detector itself is much smaller than the resolving time and delays in the circuit components which as a result determine the ultimate resolution of the coincidence counting system. It is the determination of this "coincidence system resolving time" that is the subject of this appendix.

There are basically two ways to measure the resolving time of the coincidence circuit. The first is based on the principle that if two detectors are counting at rates N_1^m and N_2^m counts per second then the random coincidence rate, N_R , is given by Equation (A-4) of Appendix A

$$N_R = 2\tau N_1^m N_2^m$$

where 2τ is the resolving time of the coincidence circuit in seconds. Thus by taking two independent sources, placing one over each detector and measuring the singles rates in each detector and the coincidence rate for the combination, 2τ can be determined from the above equation. The two sources used must be well shielded from each other so that no direct or scattered radiation which is in true coincidence can reach both detectors, thus assuring that all coincidences measured are truly random coincidences. By using pairs of sources of different activities or varying the source detector separations, the coincidence resolving time can be determined from the slope of the straight line obtained by plotting the measured coincidence rate N_R versus the product $N_1^m N_2^m$.

Due to the fixed source-detector geometry used in the present experiment it was not possible to get adequate shielding between the two independent sources in order to apply the above technique for measuring the coincidence resolving time. An alternate technique utilizing a double pulse generator was therefore used. This technique is based on the principle that if the coincidence circuit will detect the overlap of two pulses each of duration τ , then the resolving time of the coincidence circuit is 2τ , since a coincidence will be detected if one pulse lies within $\pm\tau$ of the other.

Coincidence resolving time measurements made with the double pulse generator utilized a fast oscilloscope with calibrated sweep. Each oscillator of the double pulse generator produces a square wave pulse whose width and frequency are variable and calibrated. In addition, the set of pulses from one generator can be delayed in time with respect to

the other by a variable and calibrated amount. In performing the resolving time measurement pulses are fed from each of the generators into each of the amplifiers and the delay changed until coincidences are observed. At this point the delay time is measured on the calibrated oscilloscope and taken to be the resolving time by the previous definition. The resolving time obtained with this method was $2\tau = 0.5 \mu \text{ sec.}$

A more precise definition of the resolving time⁽³⁴⁾ results when we consider that pulses from the detectors are not perfectly rectangular, as assumed in the previous definition of resolving time. In the ideal case of rectangular pulses a plot of recorded coincidence rate as a function of an artificial delay time inserted in each channel in turn would result in a rectangular "prompt resolution" curve of full width 2τ seconds. By definition the resolving time of this ideal coincidence circuit is the full width of this curve or 2τ . In the real case of non-rectangular pulses occurring under actual conditions of use the prompt resolution curve will be rounded rather than rectangular and the resolving time is defined as the effective width of the prompt resolution curve which is approximately its full width at half maximum.

In the present case the precision required in the measurement of the resolving time did not warrant the time required to determine the prompt resolution curve. However, in cases where random coincidence corrections are large or the system deviates greatly from the ideal, the distinction between the two definitions becomes important and a precise determination of the resolving time from the prompt resolution curve is required.

APPENDIX D

DESCRIPTION OF CALCULATIONS AND MODELS USED

Calculated values of ρ^{28} were based on a two energy group model. Thermal group $(0-E_{co})^*$ constants were obtained from BRT-1,⁽⁶⁵⁾ a Battelle revised version of the integral transport theory, thermal spectrum code THERMOS,⁽⁵⁸⁾ with a "white" boundary condition. The Nelkin model for H_2O was used for the scattering kernel with a correction applied for linear anisotropic scattering.⁽⁶⁶⁾ Two sets of epithermal $(E_{co}-10 \text{ MeV})$ group constants were generated; one set from HRG3,⁽⁶³⁾ the latest Battelle revised version of GAM-I,⁽⁶⁷⁾ a space independent slowing down spectrum code, and the other from EGGNIT,⁽⁶⁴⁾ which is a revised version of the GAMTEC II code.⁽⁶⁸⁾ The same basic cross section data and energy group structure were used in both models employing the B_1 approximation. This was done to assure that any differences observed in calculated parameters from the two codes could be attributed to differences in the calculational model only and not due in part to differences in cross section libraries. The differences between HRG3 and EGGNIT which affect the ρ^{28} calculations are in the treatment of resonance absorption.

The major difference between the HRG3 and EGGNIT treatments of resonance absorption arises in the resolved resonance region. In HRG3, Doppler broadening and spatial self-shielding effects are computed using an adaptation of the Adler-Hinman-Nordheim method⁽⁶⁹⁾ to include an intermediate resonance approximation for both the absorber nuclide and

* E_{co} = effective cadmium cut off energy (0.414 or 0.683 eV used in these calculations)

an admixed moderator. In EGGNIT, the Adler-Hinman-Nordheim method is replaced by the Nordheim numerical integration technique⁽⁷⁰⁾ which permits treating both absorber and admixed scatterers exactly. In addition, EGGNIT includes a treatment of resonance overlap, i.e., interference absorption due to a nearby resonance line, as well as including interference scattering in the Nordheim method. Neither of these interference effects are presently included in the HRG3 code. The treatment of unresolved resonances is the same in both codes. Dancoff shielding corrections were computed by Carlvik's DASQUE program⁽⁷¹⁾ for both codes.

The calculated values of ρ^{28} were generated from HRG3 and EGGNIT fast group constants and BRT-1 thermal group constants via the standard two group formula

$$\rho^{28} = \left(\frac{\sigma_{c1}}{\sigma_{a2}} \right)_{28} / \left(\frac{\phi_2}{\phi_1} \right)$$

where

$$\frac{\phi_2}{\phi_1} = \frac{\Sigma_1^{\text{out}}}{\Sigma_{a2} + D_2 B^2} ,$$

and other parameters are:

$(\sigma_{c1})_{28}$ = Microscopic fast capture cross section for ^{238}U from HRG3 (or EGGNIT).

$(\sigma_{a2})_{28}$ = Microscopic thermal absorption cross section for ^{238}U from BRT-1.

Σ_1^{out} = Macroscopic cell averaged down scattering cross section
from fast to thermal group from HRG3 (or EGGNIT).

Σ_{a2} = Macroscopic cell averaged thermal absorption cross
section from BRT-1.

D_2 = Cell averaged thermal diffusion coefficient from BRT-1.

B^2 = Geometrical buckling.

DISTRIBUTION

No. of
Copies

OFFSITE

1	<u>AEC Chicago Patent Group</u> G. H. Lee
8	<u>AEC Division of Reactor Development and Technology</u> Director RDT Asst. Dir. for Project Management Chief, Water Reactors Branch (2) Asst. Dir. for Reactor Technology Chief, Reactor Physics Branch (2) Chief, Fuels Recycle Branch
1	<u>AEC Division of Reactor Licensing</u> R. E. Baker
3	<u>AEC Division of Technical Information Extension</u>
5	<u>Argonne National Laboratory</u> R. Avery W. G. Davey E. M. Pennington W. C. Redman W. B. Seefeldt
3	<u>Atomic Energy of Canada, Ltd.</u> Chalk River Nuclear Laboratory Reactor Physics Branch Chalk River, Ontario, Canada M. Duret J. Griffiths P. R. Tunnicliffe
5	<u>Babcock and Wilcox</u> P.O. Box 1260 Lynchburg, VA 24505 M. N. Baldwin R. H. Lewis D. H. Roy J. R. Tulenko W. A. Wittkopf

No. of
Copies

- 1 Bhabha Atomic Research Centre
Theoretical Physics Section/RED
Central Complex Building
Trombay, Bombay-85, India
S. R. Dwivedi
- 1 Brookhaven National Laboratory
G. A. Price
H. Kouts
- 1 Atomic Energy Establishment, Winfrith
Dorchester, Dorset, England
D. J. Skillings
- 2 Cekmece Nuclear Research & Training Center
P.O. Box 1, Hava Alam, Istanbul, Turkey
C. Ertek
A. Yalcin
- 3 Chemistry Department, Nuclear Chemistry Laboratory
CCE EURATOM, Ispra, Italy
A. M. Bresesti
M. Bresesti
H. Neumann
- 5 Combustion Engineering
P.O. Box 500
Windsor, Connecticut
W. P. Chernock
R. Harding
R. L. Hellens
S. Visner
H. H. Windsor
- 1 CNEN - Centro Studi-Nucleaire
Casaccia, Rome, Italy
Ugo Farinelli
- 2 Edison Electric Institute
750 Third Avenue
New York, New York 10017
John J. Kearney
George Watkins
- 2 E. I. du Pont de Nemours & Co., Inc. SRL
G. Dessauer
H. Honeck

No. of
Copies

1	<u>ENEL</u>
Via G. B. Martini
(Piazza Verdi)
Rome, Italy

Mr. Paoletti Gualandi |
| 3 | <u>General Electric Co., San Jose</u>
Nuc. Dev. and Eng.
175 Curtner Avenue
San Jose, California 95112

D. L. Fischer
S. Levy
R. B. Richards |
| 4 | <u>General Electric Co., Vallecitos Atomic Lab.</u>
P.O. Box 846
Pleasanton, California 94566

B. F. Judson
T. M. Snyder
R. E. Harris
E. Kurtz |
| 1 | <u>Idaho Nuclear Corporation</u>

R. Grimsey |
| 2 | <u>Institut für Neutronenphysik und Reaktortechnik,</u>
<u>Kernforschungszentrum, Karlsruhe, Germany</u>

H. Seufert
D. Stegemann |
| 1 | <u>Japan Atomic Energy Research Institute</u>
<u>TCA-JPDR</u>
Tokai-Mura, Naka-Gun, Ibaraki-Ken
Japan

Shojiro Matsuura |
| 1 | <u>Massachusetts Institute of Technology</u>

D. D. Lanning |
| 2 | <u>Nuclear Fuels Services</u>
<u>Suite 600</u>
6000 Executive Blvd.
Rockville, Md. 20852

J. D. McDaniels, Jr.
W. Eich |
| 1 | <u>NUKEM</u>
<u>D-645, HANAU</u>
<u>POSTFACH 869</u>
<u>Germany</u>

Mr. W. K. L. Jager |

No. of
Copies

1	<u>Pakistan Institute of Nuclear Sci. & Tech.</u> P. O. Nilore Rawalpindi, Pakistan M. A. Mannan
1	<u>Power Reactor & Nuclear Fuel Development Corp.</u> 9-13, 1-chome, Akasaka Minato-ku, Yokyo, Japan Setsuo Kobayashi
1	<u>S. C. K. - C. E. N.</u> MOL-DONK Belgium Dr. H. Van den Broeck
3	<u>Gulf United Nuclear Fuels Corporation</u> Research & Engineering Center Grasslands Road Elmsford, New York 10523 G. Hamilton R. D. Schamberger J. R. Tomonto
1	<u>University of Toledo</u> Toledo, Ohio J. J. Turin
4	<u>Westinghouse Electric Corporation</u> Atomic Power Division P.O. Box 355 Pittsburgh, Penn. 15230 R. J. French W. L. Orr J. R. Worden C. L. Bennett
3	<u>Westinghouse Electric Corporation</u> J. Hardy, Jr. B. Klein J. J. Volpe

ONSITE-HANFORD

1	<u>AEC Chicago Patent Group</u> R. M. Poteat
3	<u>RDT, Pacific Northwest Programs</u> W. E. Fry P. G. Holsted J. B. Kitchen

No. of
Copies

2	<u>AEC Richland Operations</u> H. A. House M. R. Scheller
10	<u>Westinghouse Hanford Co.</u> R. A. Bennett R. W. Hardie R. E. Heineman P. L. Hofmann W. W. Little Document Control (5)
39	<u>Battelle-Northwest</u> J. M. Batch J. L. Carter N. E. Carter D. E. Christensen D. E. Deonigi J. C. Fox C. M. Heeb U. P. Jenquin D. A. Kottwitz B. R. Leonard R. C. Liikala C. W. Lindenmeier D. F. Newman D. R. Oden (10) R. S. Paul D. L. Prezbindowski L. C. Schmid R. I. Smith V. O. Uotinen L. D. Williams W. C. Wolkenhauer M. G. Zimmerman BNW Legal (2) Technical Information (5) Technical Publications