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**LMFBR Reaction Rate
and Dosimetry
4th Quarterly Progress Report
March, April, May, 1972**

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Hanford Engineering Development Laboratory

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LMFBR Reaction Rate and Dosimetry 4th Quarterly Progress Report March, April, May, 1972

Compiled by:
W. N. McElroy
June, 1972

NOVA

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ABSTRACT

This report was compiled at the Hanford Engineering Development Laboratory operated by Westinghouse Hanford Company, a subsidiary of Westinghouse Electric Corporation, for the United States Atomic Energy Commission, Division of Reactor Development and Technology, under Contract No. AT (45-1) 2170. It describes technical progress made in the Interlaboratory LMFBR Reaction Rate Program during the reporting period.

ACKNOWLEDGEMENTS

The following organizations are presently participating in the Interlaboratory LMFBR Reaction Rate (ILRR) Program and periodically contributing to this report.

Aerojet Nuclear Company (ANC)
Argonne National Laboratory (ANL)
Atlantic Richfield Hanford Company (ARHCO)
Atomics International (AI)
Brookhaven National Laboratory (BNL)
Hanford Engineering Development Laboratory (HEDL)
Lawrence Livermore Laboratory (LLL)
Los Alamos Scientific Laboratory (LASL)
National Bureau of Standards (NBS)
Oak Ridge National Laboratory (ORNL)
University of California at Santa Barbara (UCSB)

FOREWORD

PURPOSES OF ILRR PROGRAM

The Interlaboratory LMFBR Reaction Rate (ILRR) program has been established by USAEC/RDT to develop a capability to accurately measure neutron-induced reaction rates for LMFBR fuels and materials development programs. The initial goal for the principal fission reactions, ^{235}U , ^{238}U , and ^{239}Pu , is an accuracy to within ± 5 at the 95% confidence level. Accurate measurement of other fission and non-fission reactions will be required, but to a lesser accuracy, between ± 5 to 10% at the 95% confidence level. A secondary program objective is improvement in knowledge of the nuclear parameters involved in fuels and materials dosimetry measurements of neutron flux, spectra, fluence, and burnup.

These accuracy goals for the ILRR program are severe; measurements of fast-neutron-induced reaction rates have not been rapidly moving toward this level of precision. Using a number of techniques in well established neutron environments of current interest for fast reactor development and critically evaluating the results will help establish existing levels of accuracy and indicate the scale of effort required for improvement.

To accomplish the objectives of this program, reliable and documented experimental values of reaction rates and ratios will be determined for various well established and permanent neutron fields. The Coupled Fast Reactivity Measurement Facility (CFRMF) at Aerojet Nuclear Company (ANC) is the first neutron field being studied because of the similarity of its spectrum to that of a fast reactor and the range and reproducibility of flux levels available for track etch, fission chamber, radiochemistry, and helium mass spectrometric measurements. Results of the CFRMF and other ILRR tests coupled with those of past and present EBR-II dosimetry tests will provide the improved and more precise values of nuclear parameters urgently needed for LMFBR fuels and materials development programs.

This report is issued to government agencies, and to their contractors, to provide preliminary, incomplete experimental data and technical calculations relating to the ILRR Program. Since some of the technical data may be unverified or erroneous, the report is issued with the understanding that the contents will not be released, quoted, extracted or referenced without the prior written concurrence of the respective author.

PREVIOUS REPORTS IN THIS SERIES

HEDL-TME 71-131

HEDL-TME 71-179

HEDL-TME 72-45

CONTENTS

	<u>Page</u>
SUMMARY	S-1
AEROJET NUCLEAR COMPANY	
A. CFRMF ILRR Program Irradiations Schedule	ANC-1
B. CFRMF-ILRR Irradiations Descriptions	ANC-2
C. CFRMF Flux Spectral, Power Level, and Gradient Determinations	ANC-3
D. ^{140}Ba Standards Preparation and Measurements	ANC-6
E. Counting of ^{140}Ba - ^{140}La Standard Samples	ANC-9
F. CFRMF Reaction Rates from Power Level Monitor Foils	ANC-12
G. Gamma-Ray Counting of Monitor Foils	ANC-15
H. CFRMF Reaction Rates from ANC-I Foil Package	ANC-17
I. Activities of Radiochemistry Foils	ANC-20
J. Decay Scheme Information for Analysis of Radiochemistry Foil Measurements	ANC-25
K. Analytical Representation of Ge(Li) Detector Efficiency Curves	ANC-27
L. Tests to Determine Time Variations in the Peak Detection Efficiencies of ND-3	ANC-29
ARGONNE NATIONAL LABORATORY	
A. Absolute Fission Rate Measurements by the Track Recorder Technique	ANL-1
B. CFRMF Reaction Rates by Foil Activation	ANL-6
C. Accelerator-Produced Neutron Spectra	ANL-11
ATOMICS INTERNATIONAL	
A. Integral (n, α) Cross Sections by Helium Measurement	AI-1
B. Neutron Cross Section Evaluation - Dosimetry Applications	AI-5
BROOKHAVEN NATIONAL LABORATORY	
A. Evaluated Cross Section Libraries	BNL-1
HANFORD ENGINEERING DEVELOPMENT LABORATORY	
A. Fission and Non-Fission Foil Material Acquisition and Quality Assurance	HEDL-1
B. ^{140}Ba Standard for Fission Rates by Radiochemistry	HEDL-3
C. Experimental Fission Yields for ^{137}Cs and ^{148}Nd Relative to ^{140}Ba from Neutron Dosimetry Fission Foil Irradiations in EBR-II	HEDL-9

CONTENTS (Cont'd)

	<u>Page</u>
D. Absolute Gamma-Ray Counting	HEDL-12
E. CFRMF Reaction Rates by Radiochemistry	HEDL-15
F. Shape and Size Corrections for Small Sample Reaction Rates	HEDL-18
G. Reaction Cross Sections	HEDL-22
H. $\Sigma\Sigma$ Information Relevant to the ILRR Program	HEDL-26
I. Calculated Reaction Rates for Several LASL Critical Assemblies	HEDL-33
LOS ALAMOS SCIENTIFIC LABORATORY	
A. Detector Access Channels in the Flattop and Big-10 Assemblies	LASL-1
NATIONAL BUREAU OF STANDARDS	
A. Intrinsic Scattering Corrections for Activation Detectors in CFRMF	NBS-1
OAK RIDGE NATIONAL LABORATORY	
A. Dosimeter Sample Preparation and Quality Assurance for ILRR, FRMDC, and ANL Dosimetry Programs	ORNL-1
B. Calculations Supporting the National Bureau of Standards Spectrum Facility Design Effort	ORNL-9
C. Evaluation of Dosimetry Cross Sections	ORNL-10

FIGURES

<u>Figure</u>		<u>Page</u>
ANC-1	Composite Analysis of CFRMF Neutron Spectrum	ANC-5
ANC-2	Comparison of Ge(Li) Detector Efficiencies Interpolated by Different Methods	ANC-28
ANL-1	Measured Neutron Spectra and Angular Distributions from the $^9\text{Be}(d,n)^{10}\text{B}$ Reaction at 8.3 MeV Deuteron Energy	ANL-12
BNL-1	Data used in ENDF/B-III Evaluation	BNL-6
BNL-2	Comparison of SAND-II Curve with Data	BNL-8
BNL-3	Comparison of SAND-II and ENDF/B-III for ^{46}Ti	BNL-9
BNL-4	$^{46}\text{Ti}(n,p)$ and $^{47}\text{Ti}(n,np)$ Data used in ENDF/B-III Evaluations	BNL-10
HEDL-1	Reference Standard T 913	HEDL-5
HEDL-2	Relative Gold Capture Reaction Rate (Capture Reactions per Second per Nuclei) as a Function of Foil Thickness	HEDL-20
HEDL-3	Comparison of Measured and SAND-II Evaluated Cross Sections for $^{58}\text{Ni}(n,p)^{58}\text{Co}$	HEDL-25
HEDL-4	Gold Radial Reaction Rate Distribution in $\Sigma\Sigma$	HEDL-29
HEDL-5	Gold Self Shielding in Two Fast Reactor Neutron Spectra	HEDL-29
HEDL-6	Comparison of the CFRMF and LASL Critical Assemblies Neutron Spectra	HEDL-35
ORNL-1	Encapsulated ^{235}U Foil Radiographs	ORNL-6
ORNL-2	Encapsulated ^{238}U Foil Radiographs	ORNL-7
ORNL-3	Encapsulated ^{239}Pu Foil Radiographs	ORNL-8

TABLES

<u>Table</u>		<u>Page</u>
ANC-1	ILRR-CFRMF Irradiations	ANC-1
ANC-2	CFRMF Neutron Spectrum	ANC-4
ANC-3	Amersham/Searle ^{140}Ba Standard Sources	ANC-7
ANC-4	HEDL ^{140}Ba Standard Sources	ANC-7
ANC-5	^{140}Ba Standards Comparison	ANC-8
ANC-6	Determination of the Specific Activity of ^{140}Ba in the HEDL Standard Sample	ANC-11
ANC-7	Determination of the Specific Activity of ^{140}Ba in the Standard Sample Obtained from Amersham/Searle	ANC-11
ANC-8	CFRMF Reaction Rates from Power Level Monitor Foils	ANC-14
ANC-9	Comparison of Saturated Activities for the Indium Monitor Foils	ANC-16
ANC-10	Comparison of Saturated Activities for the Gold Monitor Foils	ANC-16
ANC-11	CFRMF Reaction Rates from ANC-I Foil Set Package	ANC-18
ANC-12	Comparison of Similar Foil Activities from ANC-I Packet	ANC-21
ANC-13	Saturated Activities of Foils from ANC-I Packet	ANC-23
ANC-14	A Set of Recommended Half-Lives, Gamma-Ray Energies and Branching Ratios for the Radiochemistry Foil Measurements	ANC-26
ANC-15	Tests of Long-Term Variation in the Peak Detection Efficiencies of ND-3	ANC-30
ANC-16	Test of Constancy of Peak Detection Efficiency and Reproducibility of the Source-Detector Geometry at the 10 cm Position with ND-3 Using a ^{137}Cs Source	ANC-31
ANC-17	Test of Constancy of Peak Detection Efficiency and Reproducibility of the Source-Detector Geometry at the 10 cm Position with ND-3 Using a ^{60}Co Source	ANC-32
ANL-1	SSTR Results at the Center of CFRMF	ANL-1
ANL-2	Preliminary Comparison of Absolute Fission Rate Measurements of ^{235}U in CFRMF Normalized to a 0.6 kW Power Level	ANL-4
ANL-3	Nuclear Constants Used in Reaction-Rate Measurements	ANL-7
ANL-4	Summary of Measured Reaction Rates for the ANL-1 Nonfission Foil Set	ANL-8
ANL-5	Fission Product Reaction Rates of ^{235}U and ^{238}U	ANL-9
AI-1	Helium Concentration Measurements	AI-2
AI-2	Comparison of Measured and Calculated Helium Production in ^{10}B	AI-3

TABLES (Cont'd)

<u>Table</u>		<u>Page</u>
BNL-1	Energy Levels of $Z = 11-21$ Nuclei (IV)	BNL-5
BNL-2	References for Figure BNL-1	BNL-7
BNL-3	Fission Spectrum Averaged Cross Section Comparison	BNL-11
HEDL-1	Interlaboratory Comparison of Data from Gamma Spectrometric Measurements on HEDL T 913 Barium-Lanthanum-140 Standard	HEDL-6
HEDL-2	HEDL Gamma Spectrometric Measurement Results on HEDL T 913 Barium/Lanthanum-140 Standard	HEDL-7
HEDL-3	Preliminary Fission Yields for Cs-137 and Nd-148 from Analysis of Neutron Dosimetry Foils Irradiated in EBR-II and Based on Assumed Ba-140 Fission Yields Given	HEDL-11
HEDL-4	Summary of HEDL Measured $^{115}\text{In}(n,n')^{115\text{m}}\text{In}$, $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$, and $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$ Reaction Rates	HEDL-16
HEDL-5	Gold Self-Shielding Factors in $\Sigma\Sigma$	HEDL-31
HEDL-6	Estimated Central Flux Spectra for the Big-10 and Flattop Critical Assemblies	HEDL-34
HEDL-7	SAND-II Calculated Saturated Activities for the Big-10	HEDL-37
HEDL-8	SAND-II Calculated Saturated Activities for the Flat-top ^{235}U	HEDL-38
HEDL-9	SAND-II Calculated Saturated Activities for the Flat-top ^{239}Pu	HEDL-39
HEDL-10	SAND-II Calculated Saturated Activities for the Flat-top ^{233}U	HEDL-40
ORNL-1	^{235}U Dosimeters, Batch No. U-235-264-CM	ORNL-3
ORNL-2	^{238}U Dosimeters, Batch No. U-238-ES-Z	ORNL-4
ORNL-3	^{239}Pu Dosimeters, Batch No. Pu-239-453B	ORNL-4

Summary

AEROJET NUCLEAR COMPANY

The AI helium production samples were irradiated in the CFRMF. An analysis of the CFRMF neutron spectrum measurements and calculations is presented. ^{140}Ba standards from Amersham/Searle Corporation and HEDL were received and the results from the preparation and measurements of these standards are given. The reaction rates of power level monitor foils from the CFRMF irradiations have been analyzed and are presented. The reaction rates from the foils in the ANC-I radiochemistry foil package have been obtained using both NaI(Tl) and Ge(Li) counting techniques. An evaluated set of decay scheme information is presented for the analysis of the radiochemistry foil measurements. Analytical representations for Ge(Li) detector efficiency curves are compared and tests to determine any changes in Ge(Li) peak detection efficiencies with time are presented.

ARGONNE NATIONAL LABORATORY

Fission rate results for ^{235}U and ^{237}Np measured in CFRMF by the SSTR method are reported and compared with fission-chamber and radiochemical results. The relative precision between multiple SSTR measurements was $\sim 0.4\%$ for both ^{235}U and ^{237}Np . Reaction rate results from the ANL-I nonfissile foil packet irradiated at 9.75 kW power level in CFRMF are reported. An evaluation of several fission product yields from ^{235}U and ^{238}U is presented. Neutron flux intensities and spectra available from accelerator produced neutron sources are presented in order to provide information for the planning and selection of standard neutron fields for future ILRR program studies.

ATOMICS INTERNATIONAL

Fourteen small specimens of boron and of its two separated isotopes have been irradiated in CFRMF. Mass spectrometric measurements of the helium generated in two of the specimens of ^{10}B have given concentrations in excellent agreement with each other. The results compare well with preliminary values calculated using available energy dependent (n, α) cross section data. Data for the reactions $^{63}\text{Cu}(n, \gamma)^{64}\text{Cu}$, $^{63}\text{Cu}(n, \alpha)^{60}\text{Co}$ and $^{65}\text{Cu}(n, 2n)^{64}\text{Cu}$ were evaluated, formulated for the ENDF/B data file and forwarded to the National Neutron Cross Section Center (NNCSC) at Brookhaven.

BROOKHAVEN NATIONAL LABORATORY

Comparison of the SAND-II and ENDF/-III evaluated energy dependent cross section libraries was continued during the quarter. The activation cross section of Titanium to produce ^{46}Sc was evaluated. This cross section is useful in dosimetry because of its long half-life and simple mode of decay.

HANFORD ENGINEERING DEVELOPMENT LABORATORY

Results are reported of initial QA work on samples of the high purity aluminum used by ORNL to encapsulate the ^{235}U , ^{238}U , ^{237}Np , and ^{239}Pu fission foils. The material was determined to be suitable for use in fast reactor environments, such as CFRMF.

Data from ILRR program participants who have reported their results from gamma spectrometric measurements on the HEDL ^{140}Ba - ^{140}La standard are summarized. The agreement is very good for this first ILRR program effort to standardize ^{140}Ba - ^{140}La counting.

A few preliminary measurements on the determination of ^{137}Cs and ^{148}Nd fission yields relative to ^{140}Ba values (for vanadium encapsulated fission foils being used for routine EBR-II dosimetry) are reported.

Results are documented of high power CFRMF tests related to run-to-run power level normalization for the ANC-I Non-Fission foil set, AI-I natural and enriched boron sample irradiation, and the HEDL recounts of ANC and ANL foils.

Theoretical calculations support recent experimental observations of significant neutron resonance self-shielding effects for thin (0.5 to 6 mil thick) gold foils irradiated in CFRMF. Further tests are being planned to study this effect for gold as well as other materials [$^{235}\text{U}(n,f)$, $^{238}\text{U}(n,\gamma)$, $^{59}\text{Co}(n,\gamma)$, $^{45}\text{Sc}(n,\gamma)$, and $^{63}\text{Cu}(n,\gamma)$] of concern to the ILRR program.

A brief review is presented of progress, relevant to the ILRR program, in the study and use of the $\Sigma\Sigma$ facility. Of particular importance are recent results on gold self-shielding measurements which support the observations in CFRMF.

A Task Group has been established through the Normalization and Standards Subcommittee of CSEWG to develop an "ENDF-B File for Dosimetry Applications."

Results of the first meetings of this group are briefly described.

Comparison of Meadows' and Smith's recent energy dependent measurements of the $^{58}\text{Ni}(n,p)^{58}\text{Co}$ cross section with those of Paulsen's and Wiedera's and those derived on the basis of SAND-II analysis of integral data shows agreement in the 1 to 4 MeV energy region. Such agreement in this energy region had not previously been observed for a threshold dosimetry reaction. This type of agreement is a necessary requirement for cross sections added to the ENDF/B file and used for fuels and materials dosimetry.

The spectra provided by LASL for Big-10 and several Flattop critical assemblies in the 3rd ILRR Quarterly Report were used with the SAND-II code to calculate fission and non-fission foil reaction rates of interest for future ILRR program study. The calculated spectra for the LASL assemblies are compared with the CFRMF spectrum.

LOS ALAMOS SCIENTIFIC LABORATORY

Diameter limitations on foils and chambers are 0.50" for the ^{233}U core and ^{239}Pu core Flattops, 1.00" for ^{235}U Flattop, and 1.50" for Big-10. Fission chamber stem lengths and diameters must be >10" long and <1/8" O.D. for Flattop and >20" long and <3/16" O.D. for Big-10. (A shorter stem length can be accommodated in Big-10 if the pre-amplifier fits into the 1.5" diameter glory hole.)

NATIONAL BUREAU OF STANDARDS

The NBS INTRAN Code which computes disk-detector responses to neutrons scattered from nearby materials is operational. A simple formulation of the energy dependent part of the scattering correction is complete, and three-group energy transfer cross sections for a few materials have been prepared.

OAK RIDGE NATIONAL LABORATORY

A 100-g batch of ^{239}Pu -1% Al metal alloy has been chemically converted to oxide and the aluminum content removed. The oxide will be used in the future preparation of needle-type flux monitors. All metal disk dosimeters of ^{235}U , ^{238}U , and ^{239}Pu have been produced, encapsulated in high purity

aluminum, and tested for leakage. Fabrication of ^{237}Np disks of the same form is now in progress and these samples will be shipped in June 1972. Preparation of an archive sample of each oxide and metal actinide isotope being used in the ILRR Dosimetry Program is now in progress.

ORNL is providing calculational support to assist NBS in the design of a well characterized intermediate neutron spectrum facility and is in the process of completing evaluations on several neutron reactions with cobalt.

Aerojet Nuclear Company

A. CFRMF ILRR PROGRAM IRRADIATIONS SCHEDULE

J. W. Rogers and Y. D. Harker

Objective

- Provide a convenient and expeditious schedule for ILRR experimental investigations in CFRMF.

Summary

The schedules for the preliminary foil irradiations, fission chamber measurements, fission track irradiations, and radiochemistry foil packet irradiations have been made and are summarized in the previous progress report (HEDL-TME-72-45). The AI helium production irradiation was made in April 1972. More irradiations are planned.

Accomplishments and Status

The AI helium production irradiation was completed during this quarter and is summarized in Table ANC-1.

TABLE ANC-1
ILRR-CFRMF IRRADIATIONS

<u>Run Designation</u>	<u>Laboratory</u>	<u>Date</u>	<u>Power</u>	<u>Time</u>
AI-I	AI	4-11-72	~9.75 kW	16 hours

Approximately 121 hours of CFRMF operating time have been used in support of the ILRR program.

Expected Achievements

More irradiations will be scheduled in support of the ILRR program.

B. CFRMF-ILRR IRRADIATIONS DESCRIPTIONS

J. W. Rogers

Objective

- Provide descriptions of irradiation conditions for all CFRMF-ILRR program tests.

Summary

A description of the test conditions for the helium production irradiation is presented. Descriptions of previous CFRMF-ILRR program tests have been presented in the previous progress report (HEDL-TME-72-45).

Accomplishments and Status

The following is a description of the test conditions of the AI helium production irradiation.

AI-I (4-11-72)

Power Level Setpoint - 975 X 1000
Shim Position - 03333
Time In - 15:30:28 MST 4-10-72
Time Out - 07:30:28 MST 4-11-72
Total Exposure - 16 hours
Power Level #1 - 86.0 - 87.7
Power Level #2 - 97.0 - 98.2
Auxiliary Fission Chamber - 2.07 - 2.08

Prior to this test the power level control system CIC chamber was moved to permit reactivity measurements and then returned to its previous position as closely as possible.

The foil containment and positioning arrangement were identical to the ANC-I (2-29-72) test except that the chamber contained only the AI-I helium production sample packet.

After irradiation, two foils (Au-A0 and In-2#15) were retained by ANC and the other two monitor foils and the AI-I helium production sample packet were sent to HEDL.

Expected Achievements

As future tests are completed descriptions following this outline will be presented.

C. CFRMF FLUX SPECTRAL, POWER LEVEL, AND GRADIENT DETERMINATIONS

J. W. Rogers

Objective

- To meet ILRR program objectives an accurate characterization of the neutron environment in the CFRMF test region must be available. Important parameters are the neutron energy spectrum and flux gradient around the central test region, with power levels and the reproducibility of a given power level or multiples of it. The best possible information on these parameters will be obtained and disseminated.

Summary

Previously determined neutron environment characterization information has been disseminated and reported in the previous progress reports. A figure and table of the relative flux is presented.

Accomplishments and Status

In the previous progress report the spectral shape of the CFRMF flux as determined by proton-recoil measurements, foil activation measurement and a diffusion calculation was presented. These results were normalized between 300 keV and 400 keV and a composite spectrum from these measurements and this calculation was determined. These results are shown in Figure ANC-1. The results are also tabulated in Table ANC-2.

Expected Achievements

Efforts will continue to insure that as more accurate or reliable results become available they will be included in the spectrum analysis so as to make the CFRMF neutron spectrum among the most intensely studied and accurately characterized reactor neutron environments.

TABLE ANC-2
CFRMF NEUTRON SPECTRUM

Remaining flux values are
extrapolated or from diffu-
sion theory calculations.

$\Delta u = .25$ Group No.	Lower Energy (ev)	Flux per Unit Lethargy	Relative Uncertainty (Percent)	$\Delta u = .25$ Group No.	Lower Energy (ev)	Flux per Unit Lethargy
1	7.79×10^6	0.311	64.5	46	1.01×10^2	0.03
2	6.07×10^6	0.758	55.4	47	7.89×10^1	0.015
3	4.72×10^6	1.38	37.7	48	6.14×10^1	0.0041
4	3.68×10^6	1.90	36.8	49	4.79×10^1	0.00036
5	2.87×10^6	2.34	6.8	50	3.73×10^1	0.000062
6	2.23×10^6	2.71	6.6	51	2.90×10^1	0.000062
7	1.74×10^6	3.14	7.3	52	2.26×10^1	0.000062
8	1.35×10^6	3.45	10.1	53	1.76×10^1	0.000029
9	1.05×10^6	4.54	5.7	54	1.37×10^1	0.000029
10	8.21×10^5	6.75	7.8	55	1.07×10^1	0.000029
11	6.39×10^5	8.23	9.9			
12	4.98×10^5	9.62	9.1			
13	3.88×10^5	9.96	4.6			
14	3.02×10^5	10.0	4.6			
15	2.35×10^5	9.13	7.1			
16	1.83×10^5	8.0	7.1			
17	1.43×10^5	7.11	6.9			
18	1.11×10^5	6.31	6.2			
19	8.65×10^4	5.21	7.5			
20	6.74×10^4	4.46	7.2			
21	5.25×10^4	3.46	27.2			
22	4.09×10^4	2.78	29.5			
23	3.18×10^4	2.02	9.9			
24	2.48×10^4	1.71	11.1			
25	1.93×10^4	1.29	16.3			
26	1.50×10^4	1.03	16.5			
27	1.17×10^4	0.89	51.7			
28	9.12×10^3	0.84	42.8			
29	7.10×10^3	0.76	71.1			
30	5.53×10^3	0.70	71.4			
31	4.31×10^3	0.54	62.9			
32	3.36×10^3	0.44	70.5			
33	2.61×10^3	0.40	50.0			
34	2.04×10^3	0.38	47.4			
35	1.59×10^3	0.35	51.4			
36	1.23×10^3	0.32	59.4			
37	9.61×10^2	0.30	43.3			
38	7.48×10^2	0.28	35.7			
39	5.83×10^2	0.25	28.0			
40	4.54×10^2	0.20	50.0			
41	3.54×10^2	0.18	38.9			
42	2.75×10^2	0.15	40.0			
43	2.14×10^2	0.11	54.6			
44	1.67×10^2	0.08	87.5			
45	1.30×10^2	0.05	100.0			

Mean Energy = 700 keV

Median Energy = 325 keV

Figure ANC-1. Composite Analysis of CFRMF Neutron Spectrum.

D. ^{140}Ba STANDARDS PREPARATION AND MEASUREMENTS

J. W. Rogers and E. H. Turk

Objective

- Establish an absolute calibration technique for counting the ^{140}Ba - ^{140}La fission product gamma rays. By counting these gamma rays from irradiated fissionable materials the fission rates will be determined.

Summary

Two ^{140}Ba - ^{140}La standard solutions were acquired, one from Amersham/Searle Corporation and one from Hanford Engineering Development Laboratory, and from these counting sources were prepared. These sources were counted, the data analyzed and the results were compared to the supplied certified activities. One solution yielded results in agreement with certification and the other did not.

Accomplishments and Status

One ^{140}Ba - ^{140}La solution standard was obtained from Amersham/Searle Corporation (AmS) and another from Hanford Engineering Development Laboratory (HEDL). Each of these standard solutions was certified by its respective supplier and shipped in a sealed glass ampoule.

The AmS standard consisted of 0.9579 grams of solution containing 95.97 μg of barium and 95.97 μg of lanthanum. The ^{140}Ba activity was certified to be 4.6287×10^6 nuclear transformations per second (ntps) per gram on March 3, 1972 at 1200 hours GMT.

The HEDL standard consisted of 4.9563 grams of solution containing 337 μg of barium and 49.6 μg of lanthanum. The ^{140}Ba activity was certified to be 1.134×10^6 ntps per gram on March 13, 1972 at 1200 hours PST.

In preparation to make the counting sources the glass ampoules were opened by breaking off the top end. The sample size was based on aliquot weight and total weight of solution. The ampoule was weighed immediately before and after the taking of each aliquot for each source. Calibrated pipettes were used to take the aliquots. To insure complete removal of the weighed aliquot from the pipette, three rinses of the pipette were made using demineralized water and adding the rinses to the source. The pipettes were thoroughly dried after the taking and rinsing of each aliquot.

Four sources were prepared from the AmS standard. These sources were deposited on 0.5" diameter blotting paper discs. Each aliquot and its respective rinses were deposited on the discs uniformly and then allowed to air dry under an infrared heat lamp before being sealed between two pieces of plastic tape. Table ANC-3 summarizes the preparation of the AmS sources and the specific activities, relative to the first source, obtained by counting the sources at 10 cm on a 3" x 3" NaI(Tl) crystal.

TABLE ANC-3

AMERSHAM/SEARLE ^{140}Ba STANDARD SOURCES

<u>Identification</u>	<u>Volume</u>	<u>Weight</u>	<u>Size</u>	<u>Relative Specific Activity</u>
AmS #1	~ 50 λ	0.05040 g	0.5" dia.	$\equiv 1.0000 \pm 0.007$
AmS #2	~ 50 λ	0.05060	"	0.9998 ± 0.007
AmS #3	~ 50 λ	0.05055	"	1.0072 ± 0.007
AmS #4	~ 10 λ	0.01045	"	0.9845 ± 0.009

Nine sources were prepared from the HEDL standard. Six of these sources were prepared in the same manner as the AmS sources on 0.5" blotting paper discs. Three point sources were prepared by depositing the aliquots and rinses on thin 0.5" diameter mylar discs and allowing them to air dry letting the surface tension bring them to a point source at the center of the disc. Table ANC-4 summarizes the preparation of the HEDL sources and the specific activities, relative to the first source, obtained by counting the sources at 10 cm on the same system used to count the AmS sources.

TABLE ANC-4

HEDL ^{140}Ba STANDARD SOURCES

<u>Identification</u>	<u>Volume</u>	<u>Weight</u>	<u>Size</u>	<u>Relative Specific Activity</u>
HEDL #1	~ 100 λ	0.10325 g	0.5" dia.	$\equiv 1.0000 \pm 0.007$
HEDL #2	~ 100 λ	0.10230	"	0.9950 ± 0.007
HEDL #3	~ 100 λ	0.10115	"	1.0006 ± 0.007
HEDL #4	~ 25 λ	0.02600	"	0.9970 ± 0.007
HEDL #5	~ 25 λ	0.02635	"	0.9745 ± 0.007
HEDL #6	~ 25 λ	0.02620	"	0.9875 ± 0.007
HEDL #7	~ 100 λ	0.09945	Point	1.0115 ± 0.007
HEDL #8	~ 100 λ	0.10225	"	1.0041 ± 0.007
HEDL #9	~ 100 λ	0.10065	"	1.0099 ± 0.007

As can be seen in these two tables only one source of 13 prepared yielded specific activity outside the estimated limits of uncertainty and can be rejected based on a 90% confidence level rejection quotient test.

The activities of these standards were determined by using the detector system [3" x 3" NaI(Tl)] calibration, counting techniques and data analysis currently being used. All counting was done at a source-to-detector distance of 10 cm and corrections were made for coincidence and random summing. Half-lives, taken from the literature, of 12.79 days for ^{140}Ba and 40.26 hours for ^{140}La were used and the branching ratio of the 1596-keV gamma ray was taken to be 0.9533. The final values obtained for the ^{140}Ba activity of these two standards and the certified values are given in Table ANC-5.

TABLE ANC-5

^{140}Ba STANDARDS COMPARISON

<u>Standard</u>	<u>Certified^a</u>	<u>Measured at ANC</u>
AmS	$(4.6287 \pm 0.0463) \times 10^6$	$(4.354 \pm 0.048) \times 10^6$
HEDL	$(1.134 \pm 0.018) \times 10^6$	$(1.139 \pm 0.007) \times 10^6$

^a in units of nuclear transformations per second per gram.

The result for the HEDL standard is in agreement with the certification within the limits of uncertainty quoted. The result for the AmS standard was 6% lower than the certification. No explanation for the disagreement on the AmS standard has been found. Additional analyses of measurements on these standards are found in section ANC-E.

Expected Achievements

Further investigation will be made in an attempt to determine the reason for the discrepancy in these results.

E. COUNTING OF ^{140}Ba - ^{140}La STANDARD SAMPLES

R. C. Greenwood and R. G. Helmer

Objectives

- Provide a test of our present ability to measure a ^{140}Ba - ^{140}La equilibrium activity utilizing present detector efficiency data.
- Utilize these standard sources to provide a direct measurement of detector efficiency at the 1596-keV gamma-ray energy of ^{140}La .

Summary

Specific activities of ^{140}Ba and ^{140}La have been measured for the HEDL and Amersham/Searle standard ^{140}Ba - ^{140}La samples utilizing the ND-3 and OR-5 Ge(Li) detector systems together with a standard 3" x 3" NaI(Tl) detector. The activity levels measured for each of these three detector systems show good internal consistency, within the limits of the errors assigned ($\sigma \sim \pm 1.5\%$). Furthermore, the activity level measured for the HEDL standard sample is in close agreement with the certified activity level which was determined at HEDL using 4π β - γ coincidence counting. The activity levels which were determined for the Amersham/Searle standard are, however, $\sim 6\%$ lower than the levels which are specified by Amersham/Searle in the certification. Taking into account the similarity of the techniques used to make up the sources from these two standard solutions, the demonstrated ability of this laboratory to measure gamma-ray activity levels with good precision ($< \pm 2\%$), and the good agreement between our measurements and the HEDL 4π β - γ measurements of the HEDL standard, we must conclude that the specific activity quoted by Amersham/Searle for the particular ^{140}Ba sample which we received is grossly in error.

Accomplishments and Status

Various standard sources, containing different weights of sample solution, were made up from both the HEDL and the Amersham/Searle standard solutions of ^{140}Ba - ^{140}La , as described in Section ANC-D. The emission rates of the 1596-keV gamma ray of ^{140}La were measured for several of these standard sources using both the ND-3 and OR-5 Ge(Li) detector systems as well as a standard 3" x 3" NaI(Tl) detector other than the one used to obtain the results presented in Section ANC-D. From these data, and using a branching ratio of $(95.33 \pm 0.16)\%$ for the 1596-keV gamma ray of ^{140}La together with a value of

(1.15096 ± 0.00012) for the ratio of the ^{140}La -to- ^{140}Ba activity levels in the samples, specific activities were determined for the HEDL and Amersham/Searle standard samples. These measured activities are shown in Tables ANC-6 and ANC-7. The principal contribution to the uncertainties quoted on the Ge(Li) detector measurements arises from the expected errors in the interpolated peak efficiencies at 1596 keV ($\sim \pm 1.5\%$). Other contributions arising from uncertainties in the source-detector distance ($\pm 0.5\%$), peak area ($\sim \pm 0.2\%$), estimate of real and random summing corrections (each $\sim \pm (0.1-0.2)\%$), decay correction (up to $\pm 0.3\%$) and gamma-ray branching ratio ($\pm 0.16\%$) were added in quadrature to obtain the total error estimates. Based upon the data contained in Table ANC-XV of HEDL-TME 72-45 we would estimate that in favorable cases (well resolved peaks) gamma-ray activities can be measured with a precision of $\pm 1\%$ with this NaI(Tl) detector system. This uncertainty together with the expected uncertainties in the source-detector distance ($\pm 0.5\%$), peak area ($\pm 0.5\%$), real and random summing corrections ($\sim \pm 0.25\%$ and $\pm 0.5\%$), decay correction (up to $\pm 0.3\%$) and gamma-ray branching ratios ($\pm 0.16\%$) have been combined in quadrature to obtain the uncertainties quoted on the NaI(Tl) data in Tables ANC-6 and ANC-7.

Comments and Expected Achievements

The problems that we have experienced with the Amersham/Searle source of ^{140}Ba demonstrate in a rather dramatic way the potential dangers which are inherent in utilizing a detector efficiency which has been determined using just one standard source emitting the gamma ray of interest (in this case the 1596-keV gamma ray). Ultimately, the credibility of a measured detector efficiency value is based upon how well its value agrees with that obtained off the curve fit made to it and the neighboring efficiency points (in this case for example, 834-keV ^{54}Mn , 898- and 1836-keV ^{88}Y , 1173- and 1332-keV ^{60}Co , 1274-keV ^{22}Na , 2185-keV ^{144}Ce and 2614-keV ^{228}Th). This being the case, it is probably appropriate for the participants in the ILRR program to reconsider the second part of the above objectives, namely to utilize these standard ^{140}Ba sources to provide a direct measurement of detector efficiency at the 1596-keV gamma-ray energy of ^{140}La . Rather, it might be more appropriate to consider the ^{140}Ba standard as providing just one more efficiency value (in this case at 1596 keV) to be fit by the detector efficiency curve, and subsequently to utilize an efficiency value at an energy of 1596 keV which is obtained from a "best" fit to all the available efficiency data.

TABLE ANC-6

DETERMINATION OF THE SPECIFIC ACTIVITY OF ^{140}Ba IN THE HEDL STANDARD SAMPLE

<u>Detector</u> ^a	<u>Source Identification</u>	<u>Sample Mass (mg)</u>	<u>Measured Specific Activity</u> ^b d/sec-gm
3"x3" NaI(Tl)	HEDL-1	103.25	(1.129±0.015) x 10 ⁶
	HEDL-3	101.15	(1.133±0.015) x 10 ⁶
	HEDL-7	99.45	(1.135±0.015) x 10 ⁶
	HEDL-9	100.65	(1.126±0.015) x 10 ⁶
ND-3 Ge(Li)	HEDL-1	103.25	(1.148±0.018) x 10 ⁶
	HEDL-3	101.15	(1.142±0.018) x 10 ⁶
	HEDL-7	99.45	(1.151±0.019) x 10 ⁶
	HEDL-9	100.65	(1.156±0.019) x 10 ⁶
OR-5 Ge(Li)	HEDL-9	100.65	(1.132±0.019) x 10 ⁶

^a All source-to-detector distances were 10 cm.^b Specific activities were all referenced to 1200 PST on 3/13/72 at which time a certified value was determined for the specific activity of ^{140}Ba of 1.134×10^6 d/sec-gm at HEDL using 4π β - γ coincidence counting.

TABLE ANC-7

DETERMINATION OF THE SPECIFIC ACTIVITY OF ^{140}Ba IN THE STANDARD SAMPLE OBTAINED FROM AMERSHAM/SEARLE

<u>Detector</u> ^a	<u>Source Identification</u>	<u>Sample Mass (mg)</u>	<u>Measured Specific Activity</u> ^b nC/mg
3"x3" NaI(Tl)	AmS-1	50.4	116.2 ± 1.6
	AmS-3	50.55	117.0 ± 1.6
ND-3 Ge(Li)	AmS-1	50.4	118.6 ± 1.9
	AmS-2	50.6	117.4 ± 1.9
	AmS-3	50.55	119.2 ± 1.9
OR-5 Ge(Li)	AmS-3	50.55	116.8 ± 2.0

^a All source-to-detector distances were 10 cm.^b Specific activities are all referenced to 1200 GMT on 3/3/72 at which time the specific activity of ^{140}Ba quoted by Amersham/Searle for this sample was (125.1 ± 1.3) nC/mg.

F. CFRMF REACTION RATES FROM POWER LEVEL MONITOR FOILS

J. W. Rogers

Objective

- Present results of power level monitor foil measurements at ANC.

These results provide a monitor of the neutron fluence and a spectral index of the ILRR-CFRMF program tests.

Summary

Power level monitor foils from the various ILRR program tests in the CFRMF have been counted using NaI(Tl) gamma-ray counting techniques. The counting techniques are briefly discussed and the saturated reaction rates calculated from the monitor foils are presented and discussed.

Accomplishments and Status

Saturated reaction rates determined from gamma-ray spectra measurements on several power level monitor foils activated in the ILRR-CFRMF tests have been obtained using standard NaI(Tl) counting techniques. A 3" x 3" NaI(Tl) crystal detector was used at a source-to-detector distance of 10 cm. A 1.024 g/cm² polystyrene absorber was used to stop the source beta radiation. The gamma-ray detection efficiency of the NaI(Tl) system is that computed from the known characteristics of the detector. The system and techniques have been checked against sources calibrated by different techniques and yielded excellent agreement. To eliminate the need to make high counting-rate corrections, all counts were made at decay times sufficiently long to yield acceptably low rates. The monitor foils were the 0.5" diameter 2-mil thick gold metal and 2-mil thick indium metal. The foils were carefully weighed to ± 0.00002 g following irradiation. No corrections have been made for gamma-ray attenuation in the foils.

The 411.8-keV gamma ray from the decay of ¹⁹⁸Au was counted from the gold foils. In all measurements more than 10⁵ counts were collected in the photopeak. In order to have acceptable counting rates at 10 cm, decay times of 16 to 20 days were allowed for the higher exposure foils. A half-life of 2.700 days and a branching ratio of 0.955 were used in the gold data reduction.

The 335-keV gamma ray from the decay of ^{115m}In was counted from the indium foils. In all measurements more than 10⁴ counts were collected in the photopeak (in most cases more than 10⁵ counts). Decay times of between 20 and 28

hours were allowed so that good counting rates and a single gamma-peak spectrum were obtained. A half-life of 4.5 hours and a branching ratio of 0.47 were used in the indium data reduction.

Table ANC-8 summarizes the results from all the power monitor foils measured thus far. (It is noted that the results for indium in this table and those in the previous quarterly are different because those stated in the previous quarterly had not been corrected for the indium isotopic abundance.)

Assuming a 0.6 kW power level for the three tests made on 2/2/72 and 2/4/72 and comparing the gold and indium reaction rates, power levels indicated by the gold foils are about 5% lower than those indicated by the indium foils. The gold-to-indium ratio spectral index shows a shift between the 0.6 kW tests and the higher power tests of about 5%. It is known that the temperatures of the core components and the monitor foils are different at the different power levels. The magnitudes of temperature effects on the CFRMF spectrum and the foils have not been investigated.

The relative power levels of the higher power runs are discussed in section ANC-G.

Expected Achievements

Foils from all future tests will be measured and analyzed. Further investigation of the differences mentioned above will be made.

TABLE ANC-8
CFRMF REACTION RATES FROM POWER LEVEL MONITOR FOILS

Test Identity	Foil	Mass ^a (g)	Saturated Reaction ^b Rate by NaI(Tl)	Number of ^c Measurements	Power ^d Level (kW)	$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$
						$^{115\text{m}}\text{In}(n,n')^{115\text{m}}\text{In}$
Preliminary-III (1/26/72)	Au-MD In-14	0.1229 0.05034	3.173×10^{-15} 3.953×10^{-16}	2(.64%) 2(.51%)		(e)
Fission Chamber NBS (2/2/72)	Au-AA In-2#1	0.1216 0.04878	3.193×10^{-15} 3.956×10^{-16}	3(.33%) 1	0.600	8.071
Fission Tracks ANL (2/4/72)	Au-AC In-2#3	0.1214 0.04802	3.186×10^{-15} 3.920×10^{-16}	2(.25%) 2(.14%)		8.127
Fission Chamber NBS (2/4/72)	Au-AE In-2#5	0.1239 0.04909	3.193×10^{-15} 3.943×10^{-16}	2(.74%) 1		8.098
ARHCO-I (2/8/72)	Au-AG In-2#7	0.1234 0.04976	4.902×10^{-14} 6.250×10^{-15}	3(.26%) 3(1.04%)	9.217 9.518	7.843
ANL-I (2/15/72)	Au-AI In-2#9	0.1216 0.04848	4.708×10^{-14} 6.052×10^{-15}	5(.97%) 3(.27%)	8.852 9.216	7.779
HEDL-I (2/16/72)	Au-AK In-2#11	0.1220 0.04840	4.665×10^{-14} 6.097×10^{-15}	6(.53%) 3(.11%)	8.772 9.285	7.651
ANC-I (2/29/72)	Au-AM In-2#13	0.1221 0.04805	4.764×10^{-14} 6.148×10^{-15}	8(.49%) 4(.77%)	8.958 9.362	7.749
AI-I (4/11/72)	Au-AO In-2#15	0.1202 0.05010	4.701×10^{-14} 6.173×10^{-15}	8(.64%) 8(.54%)	8.839 9.400	7.615

^a Determined by weighing at ANC.

^b Events per atom per second.

^c The number in parentheses is the estimated percent standard deviation on span of measurements (does not include uncertainties in efficiencies).

^d Based on the average of the first three tests at 0.600 kW using the dummy fission chamber foil positioning arrangement.

^e Not applicable because two 2-mil gold foils were sandwiched together for this test.

ANC-14

G. GAMMA-RAY COUNTING OF MONITOR FOILS

R. G. Helmer

Objective

- To provide an accurate measure of the relative total neutron exposure of the radiochemistry foil packets irradiated as part of the ILRR program.

Summary

The gamma-ray spectra from the indium and gold monitor foils were measured with a Ge(Li) detector for the CFRMF irradiations ARHCO-1, ANL-1, HEDL-1, ANC-1 and AI-1. The relative gamma-ray counting rates have been determined with uncertainties of less than 1%. These measurements indicate that the neutron exposure was constant to within $\approx 1\%$ in the last four irradiations, but was higher by 3% in ARHCO-1.

Accomplishments and Status

The procedures used in handling these foils and taking the spectra have been described in the last quarterly. As noted there, only the first measurement of the indium spectrum is used (i.e., decay time of ≈ 8 hours), whereas all gold spectra are included.

The results of these measurements are given in Tables ANC-9 and ANC-10. In both tables the errors include only those terms which contribute to the uncertainty in the relative activity. For the indium foils, these contributions are source position 0.4% (value taken from the data in Table ANC-16), peak area 0.14-0.21%, and the decay correction to 8 hours after the irradiation 0.02-0.23%. For the gold foils, several measurements are combined so the situation is somewhat more complex, but the error contributions from individual runs are source position 0.4%, peak area 0.07-0.13%, decay correction 0.00-0.32%, and random summing correction 0.06 to 1.0%. The combined error on the average is 0.4-0.55%.

The data in Tables ANC-9 and ANC-10 show that for the last four irradiations the spread in activity, gold or indium, is 1.6%. Although the gold foils in the HEDL-1 and AI-1 experiments might indicate a lower flux, the indium activities do not support this possibility. Therefore, it would appear that these fluxes were the same within about 1.0%. The ARHCO-1 sample was exposed to 3% more neutrons.

TABLE ANC-9

COMPARISON OF SATURATED ACTIVITIES FOR THE INDIUM MONITOR FOILS

<u>Experiment</u>	<u>Mass^a</u> <u>(mg)</u>	<u>Saturated Activity^b</u> <u>(10⁻¹⁵ dis/sec-atom)</u>	<u>Ratio^b</u>
ARHCO-1	49.76	6.445 ± 0.033	1.035 ± 0.007
ANL-1	48.48	6.225 ± 0.026	≅1.000
HEDL-1	48.40	6.280 ± 0.027	1.009 ± 0.006
ANC-1	48.05	6.190 ± 0.027	0.994 ± 0.006
AI-1	50.10	6.205 ± 0.028	0.997 ± 0.006

^a ANC values determined after irradiation.

^b These errors are discussed in the text.

TABLE ANC-10

COMPARISON OF SATURATED ACTIVITIES FOR THE GOLD MONITOR FOILS

<u>Experiment</u>	<u>Saturated Activity^a</u> <u>(10⁻¹⁴ dis/sec-atom)</u>	<u>Ratio</u>
ARHCO-1	5.102 ± 0.020	1.027 ± 0.006
ANL-1	4.970 ± 0.022	≅1.000
HEDL-1	4.910 ± 0.022	0.988 ± 0.006
ANC-1	4.962 ± 0.020	0.998 ± 0.006
AI-1	4.891 ± 0.027	0.984 ± 0.007

^a The errors are discussed in the text.

Expected Achievements

Monitor foils from future experiments will be counted and analyzed.

H. CFRMF REACTION RATES FROM ANC-I FOIL PACKAGE

J. W. Rogers

Objective

- Obtain reaction rates from fission and non-fission foils of selected materials irradiated in the CFRMF neutron spectrum. These results are to be used in conjunction with results obtained from other laboratories and thus establish an accepted set of absolute reaction rates for these materials. These reaction rates will then be used to help characterize the CFRMF neutron spectrum and adjust energy dependent cross sections. The use of other techniques in conjunction with the reaction rates in standard neutron spectra such as the CFRMF will be used to improve the accuracies of all techniques and more precisely establish the fission rates of ^{235}U , ^{238}U , ^{239}Pu and ^{237}Np .

Summary

Saturated reaction rates have been obtained from the ANC-I set of foils irradiated in the CFRMF. The foils were counted using standard NaI(Tl) techniques. A brief discussion of the results is given.

Accomplishments and Status

Gamma-ray spectrometry measurements on all the foils in the ANC-I foil package have been made using standard NaI(Tl) techniques. A brief description of the counting equipment and conditions is given in section ANC-F. All foils were counted at a source-to-detector distance of 10 cm.

In the analysis of the measurements the most prominent and clearly defined photopeak of the reaction of interest was analyzed to obtain the reaction rates. The decay scheme data found in HEDL-TME 72-45, Table ANC-V, was used. Corrections for random and coincident summing were made where high counting rates and cascade gammas were present. No corrections were made for gamma attenuation in the foils. The reaction rate results are summarized in Table ANC-11. The foils are listed as they were placed in the package from top to bottom.

The aluminum foils were counted as soon as possible after irradiation in order to observe the activity of the ^{27}Mg but foil Al-1 was not counted soon enough to see this activity. Following these measurements the foils were etched in NaOH to remove any ^{24}Na surface contamination. After etching the foils were reweighed and counted for the ^{24}Na activity. No deviations in the

TABLE ANC-11

CFRMF REACTION RATES FROM ANC-I FOIL SET PACKAGE^a

Foil	Mass (g) ^b	Reaction	Saturated Reaction ^{c,e} Rate by NaI(Tl)	Number of ^d Measurements
Al-3	0.04720	$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	2.017×10^{-17}	4(.81%)
	0.04739	$^{27}\text{Al}(n,p)^{27}\text{Mg}$	1.144×10^{-16}	1
Au-2	0.03551	$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	5.206×10^{-14}	6(.66%)
In-3	0.04800	$^{115\text{m}}\text{In}(n,n')^{115\text{m}}\text{In}$	6.295×10^{-15}	4(.51%)
Cu-1 ^(g)	0.01480	$^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$	5.136×10^{-15}	5(.76%)
Co-3	0.01473	$^{59}\text{Co}(n,\gamma)^{60}\text{Co}$	9.517×10^{-15}	3(.44%)
Co-2	0.01440	$^{59}\text{Co}(n,\gamma)^{60}\text{Co}$	8.919×10^{-15}	3(.33%)
Co-1	0.01439	$^{59}\text{Co}(n,\gamma)^{60}\text{Co}$	9.394×10^{-15}	3(.92%)
Ni-TC	0.29538	$^{58}\text{Ni}(n,p)^{58}\text{Co}$	3.055×10^{-15}	6(.31%)
Fe-A	0.81195	$^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$	7.566×10^{-16}	3(1.56%)
		$^{54}\text{Fe}(n,p)^{54}\text{Mn}$	2.203×10^{-15}	3(.77%)
Al-2	0.04700	$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	1.987×10^{-17}	4(1.04%)
	0.04725	$^{27}\text{Al}(n,p)^{27}\text{Mg}$	1.132×10^{-16}	1
Au-1	0.00338	$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	5.366×10^{-14}	6(.27%)
In-2	0.04800	$^{115\text{m}}\text{In}(n,n')^{115\text{m}}\text{In}$	6.316×10^{-15}	4(.84%)
Ti-N	0.15925	$^{46}\text{Ti}(n,p)^{46}\text{Sc}$	3.244×10^{-16}	2(3.94%)
		$^{47}\text{Ti}(n,p)^{47}\text{Sc}$	5.215×10^{-16}	3(2.14%)
Ti-M	0.15771	$^{46}\text{Ti}(n,p)^{46}\text{Sc}$	3.273×10^{-16}	2(1.50%)
		$^{47}\text{Ti}(n,p)^{47}\text{Sc}$	5.254×10^{-16}	3(1.50%)
Sc-1	0.04925	$^{45}\text{Sc}(n,\gamma)^{46}\text{Sc}$	2.956×10^{-15}	5(.47%)
^{235}U -1	0.24811	$^{235}\text{U}(n,f)^{140}\text{Ba}$	$2.035 \times 10^{-13}\text{f}$	7(1.62%)
^{238}U -1	0.06439	$^{238}\text{U}(n,f)^{140}\text{Ba}$	$9.816 \times 10^{-15}\text{f}$	7(1.05%)
In-1	0.04686	$^{115\text{m}}\text{In}(n,n')^{115\text{m}}\text{In}$	6.333×10^{-15}	4(.26%)
Au-1	0.03733	$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	5.196×10^{-14}	6(.69%)
Al-1	0.04638	$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	2.023×10^{-17}	4(1.10%)
Au-A1	0.10132	$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	5.302×10^{-14}	4(.92%)

^a Described in previous progress report HEDL-TME 72-45.^b Determined by weighing at ANC (fission foils were not weighed at ANC).^c Events per atom per second.^d The number in parentheses is the estimated standard deviation on span of measurements (does not include any uncertainties in efficiencies).^e Results obtained by Ge(Li) counting are found in Tables ANC-12 and ANC-13.^f These are actually fission rates rather than saturated reaction rates.^g This foil was counted without copper covers.

activities from these foils outside the limits of uncertainty were observable, indicating that these reactions do not see any flux gradients or scattering and shielding effects in their locations.

The gold foils and the gold-aluminum wire were counted as soon as their activities were down to desired levels. The wire was wound into a 0.5" diameter flat-coil geometry for counting. The very thin foil at the center of the package and the dilute gold-aluminum wire around the package gave the same activities but were ~3% higher than the activities of the thicker gold foils on the top and bottom of the package. This may be interpreted to suggest that there is some self-shielding in the thicker foils and that there is no observable flux gradient between the inside and outside of the foil package with this reaction.

The indium foils also show no deviations outside the limits of uncertainty leading to the same conclusions suggested by the ^{24}Na activity produced in the aluminum foils.

The cobalt foils yielded activities which imply that the reaction rate in the middle foil is reduced ~6% by the foils above and below it. This result suggests that there may also be considerable self-shielding within each foil.

The reaction rates in the titanium foils agree quite well with one another although large uncertainties were incurred due to low counting rates and the complexity of the gamma-ray spectra.

Since only one foil each of copper, nickel, iron and scandium were in the package not much can be said about the reaction rates from these foils.

The ^{238}U and ^{235}U fission foils were counted at decay times of ~14 days and ~85 days, respectively. ^{140}Ba fission product yields of 6% for ^{238}U and 5.8% for ^{235}U were used in the computation of the reaction rates in order to be consistent with the values used by other laboratories in the last progress report. The $^{235}\text{U}/^{238}\text{U}$ ratio from these measurements is 20.73 with a range of possible values due to uncertainties between 21.29 and 20.20.

Expected Achievements

The reaction rates from these foils may be adjusted when a standard set of decay scheme data is established. Any errors in the analysis will be corrected if and when they are discovered.

Additional studies will be made on the self-shielding effects observed in these measurements.

I. ACTIVITIES OF RADIOCHEMISTRY FOILS

R. G. Helmer and R. C. Greenwood

Objective

- Obtain rates for certain neutron activation reactions for irradiations in the CFRMF.

Summary

The activities in all of the foils from the packet ANC-1 have been measured on the ND-3 Ge(Li) system and analyzed. Differences in the activities for the foils of the same material have been determined. Several of these differences are significant and indicate either self-shielding or other effects.

Accomplishments and Status

The gamma-ray spectra of the foils from the packet ANC-1 were measured for two purposes. The first was to determine any differences in the activity (per atom) across the volume of the packet. Any variation in flux between the position of the packet and that of the monitor foils could also be determined. Second, the "saturated activities" were determined for use in computing the energy distribution of the neutron spectrum.

In the computation of the relative activities of the similar foils, the corrected peak areas P for any one measurement were calculated as follows:

$$P = \frac{C(T, \tau) e^{\lambda \tau} \delta_{\text{pos}}}{(1 - e^{-\lambda T}) \delta_{\text{RS}}}$$

where

T = counting time

τ = decay time before count

λ = decay constant

C = observed peak area

δ_{pos} = correction, if any, for observed deviation of source position from standard position

δ_{RS} = correction for random summing.

The values from individual measurements were averaged with a $1/\sigma$ weighting factor. Where two gamma rays were used, the relative activities were computed and these ratios were averaged. In one case, data were obtained at two

different source-detector distances. Here, also, the relative activities were computed for each distance and then the ratios were averaged.

The results of these relative activity measurements are given in Table ANC-12. The uncertainties quoted include the contributions from the peak area

TABLE ANC-12
COMPARISON OF SIMILAR FOIL ACTIVITIES FROM ANC-1 PACKET

<u>Foil</u>	<u>Location</u>	<u>Isotope</u>	<u>Relative Activity (per unit mass)</u>
A1-3	top	^{27}Mg	$\equiv 1.000$
A1-2	middle		0.993 ± 0.024
A1-1	bottom		
A1-3	top	^{24}Na	$\equiv 1.000$
A1-2	middle		0.995 ± 0.010
A1-1	bottom		1.018 ± 0.010
Au-2	top	^{198}Au	$\equiv 1.00^a$
Au-1	middle		1.029 ± 0.005
Au-1	bottom		0.994 ± 0.006
{dilute wire	around packet		1.021 ± 0.005
Au-AM	monitor position		0.925 ± 0.006
In-3	top	^{116m}In	$\equiv 1.000$
In-2	middle		1.003 ± 0.010
In-1	bottom		0.991 ± 0.010
In-3	top	^{115m}In	$\equiv 1.000$
In-2	middle		1.004 ± 0.005
In-1	bottom		1.010 ± 0.005
In-2-13	monitor position		0.981 ± 0.005
Co-3	top ^b	^{60}Co	1.000
Co-2	middle		0.891 ± 0.004^c
Co-1	bottom		0.988 ± 0.004^c
Ti-N	top ^b	^{46}Sc	1.000
Ti-M	bottom		1.018 ± 0.022

^a The Au samples were of the following thicknesses: Au-2 and Au-1 (bottom) 0.0005", Au-1 (middle) 0.00005", Au-AM 0.002", and the wire was a dilute mixture of Au in Al.

^b These foils were adjacent in this packet.

^c The source position error is not included in error estimate. For all the ^{60}Co sources it was necessary to count the samples directly on top of the detector cap (source-to-detector distance ~ 1 cm) and thus it is rather difficult for us to estimate a relative source position error.

determination, source position, decay correction, and the random summing correction. The data for ^{27}Mg , ^{24}Na , $^{116\text{m}}\text{In}$, $^{115\text{m}}\text{In}$, ^{60}Co , and ^{46}Sc all are consistent with the proposition that the flux is constant across the packet to within 1%. The $^{115\text{m}}\text{In}$ data from the monitor foil indicate that the flux at the monitor position is 2% lower than at the packet.

The interpretation of the gold data is complicated by the use of four different foil thicknesses. The thin foil (0.00005") and the wire (dilute mixture with aluminum) agree within <1%. The foils (0.0005") at the top and bottom of the packet also agree within <1%, but are lower by ~3%. The agreement of the two pairs of similar samples suggests that the flux distribution is uniform (at least for some spacial distributions). The difference between the two pairs of samples is presumed to be due to self-shielding of the neutrons in the thicker samples. The lower activity of the monitor foil is taken to be a combination of even more self-shielding and the lower flux at the monitor foil position (which is indicated by the $^{115\text{m}}\text{In}$ data).

The "saturated activities" per atom were computed for each foil by the following expression

$$A_s = \frac{P}{B\epsilon} \cdot \frac{\lambda}{(1-e^{-\lambda t})} \cdot \frac{a}{mN_0}$$

where

B = absolute gamma-ray intensity (per decay)

ϵ = detector efficiency

t = neutron irradiation time

a = atomic mass of target isotope (g)

m = mass of isotope of interest in foil (g)

N_0 = Avogadro's number = 6.023×10^{23}

The saturated activities are given in Table ANC-13. Generally, these spectra were measured with a source-detector distance of 10 cm. Two exceptions were the ^{24}Na and ^{60}Co spectra, which were measured at 5 cm and 1 cm, respectively. In each case, one foil was measured at 10 cm, where the efficiency is best known, to determine the absolute activities. As indicated in the table, two or three measurements were included for most of the foils.

No uncertainties have been included in Table ANC-13; these are still being evaluated. Also corrections for several effects have not yet been included. These are for real coincidence summing and finite size source correction to the

TABLE ANC-13
SATURATED ACTIVITIES OF FOILS FROM ANC-I PACKET

<u>Foil</u>	<u>Isotope</u>	<u>Number of Measurements</u>	<u>Saturated Activity 10⁻¹⁶ dis/sec-atom</u>
A1-3	²⁷ Mg	2	1.084
A1-2		1	1.076
A1-3	²⁴ Na	2 ^a	0.2041
A1-2		1 ^a	0.2021
A1-1		1 ^a	0.2078
Au-2	¹⁹⁸ Au	2	536.6
Au-1 (.00005")		2	552.2
Au-1 (.0005")		2	533.5
Au-A1 wire		1	548.1
Au-AM monitor		3	496.2
In-3	^{116m} In	2	370.4 per atom ¹¹⁵ In
In-2		2	367.6
In-1		2	365.8
In-3	^{115m} In	3	63.11 per atom ¹¹⁵ In
In-2		3	63.69
In-1		3	64.02
In-2-13 monitor		2	61.91
Co-3	⁶⁰ Co	2 ^b	94.87
Co-2		1 ^b	84.53
Co-1		1 ^b	93.73
Ni-TC	⁵⁸ Co	3	30.81 per atom ⁵⁸ Ni
Fe-A	⁵⁹ Fe	2	7.65
Fe-A	⁵⁴ Mn	2	21.15 per atom ⁵⁴ Fe
Ti-M	⁴⁸ Sc	3	0.08887 per atom ⁴⁸ Ti
Ti-M	⁴⁷ Sc	3	5.441 per atom ⁴⁷ Ti
Ti-N	⁴⁶ Sc	1	3.261 per atom ⁴⁶ Ti
Ti-M		1	3.319
Sc-1	⁴⁶ Sc	3	29.58
²³⁵ U-1	¹⁴⁰ Ba	3	119.1 ^c per atom ²³⁵ U
²³⁸ U-1	²³⁹ Np	3	226.6
²³⁸ U-1	¹⁴⁰ Ba	3	5.793 ^d per atom ²³⁸ U

^a Only one measurement of the ²⁴Na activity was made at a source-detector distance of 10 cm, all other measurements were at ~5 cm.

^b Only one measurement of the ⁶⁰Co activity was made at a source-detector distance of 10 cm, all other measurements were at ~1 cm.

^c Assuming a ¹⁴⁰Ba fission yield of 5.8% (as used previously in HEDL-TME 72-45) this gives a ²³⁵U fission rate of 2.054 x 10⁻¹³ dis/sec-atom.

^d Assuming a ¹⁴⁰Ba fission yield of 6.0% (as used previously in HEDL-TME 72-45) this gives a ²³⁸U fission rate of 9.655 x 10⁻¹⁵ dis/sec-atom.

efficiency (determined for point sources). Since these corrections are all small, the values in Table ANC-13 will suffice for the first interlaboratory comparisons. The computation of these activities is based on the half-life and branching ratio information in Table ANC-V of the last quarterly (HEDL-TME 72-45).

Expected Achievements

The saturated activity data will be re-evaluated in terms of the decay scheme information in Table ANC-14 and corrections for real coincidence summing and finite source size will be included. Based on recent experiments, the Ge(Li) detector efficiency will also be modified slightly. The uncertainties in the results will be evaluated in detail.

J. DECAY SCHEME INFORMATION FOR ANALYSIS OF RADIOCHEMISTRY FOIL MEASUREMENTS

R. G. Helmer and R. C. Greenwood

Objective

- Provide an evaluated set of decay scheme information allowing the various laboratories to carry out consistent analyses of the radiochemistry foil packets.

Summary

For the radioactivities that are to be observed in the foils in these packets, the available decay scheme literature has been reviewed. Values are presented for the half-lives and the gamma-ray energies and absolute intensities for from one to three prominent transitions from each isotope. The uncertainties (1σ value) are given for each quantity.

Accomplishments and Status

Four radiochemistry foil packets have been irradiated and the gamma-ray spectra measured. The first step in the analysis of these data will be to determine the level of agreement between the various laboratories. The second step will involve the use of the activity levels in the analysis of the neutron spectrum. The decay scheme information needed for this comparison is given in Table ANC-14. Uncertainties (1σ level) are given for all of the values.

Expected Accomplishments

This evaluation is complete, unless it is found that more information is needed for future experiments.

TABLE ANC-14

A SET OF RECOMMENDED HALF-LIVES, GAMMA-RAY ENERGIES AND
BRANCHING RATIOS FOR THE RADIOCHEMISTRY FOIL MEASUREMENTS

Radioisotope	Half-Life ^a $t_{1/2}$	Gamma-Ray Energy ^a (keV)	Gamma-Ray Branching Ratio ^a (%)	
²⁷ Mg	9.46(2) m	843.73(4) 1014.44(5)	71.4(5) } 28.6(5) }	sum = 100.0
²⁴ Na	14.99(2) h	1368.52(6) 2753.98(10)	99.99(1) 99.85(2)	
⁴⁶ Sc	83.85(10) d	889.258(18) 1120.516(25)	99.98(1) 99.99(1)	
⁴⁷ Sc	3.39(4) d	159.39(5)	69.0(25)	
⁴⁸ Sc	43.8(1) h	983.4(2) 1037.4(3) 1311.9(3)	99.98(1) 97.7(3) ^b 99.99(1)	
⁵⁴ Mn	312.6(3) d	834.827(21)	99.978(2)	
⁵⁹ Fe	44.6(1) d	1099.224(25) 1291.564(28)	55.5(17) } 44.1(12) }	sum = 99.6(1)
⁵⁸ Co	71.3(2) d	810.757(21)	99.44(2)	
⁶⁰ Co	5.268(5) y	1173.208(25) 1332.491(41)	99.86(2) 99.985(1)	
⁶⁴ Cu	12.78(5) h	511.002 ^c	36.8(16)	
^{115m} In	4.50(2) h	336.2(1)	47.0(20)	
^{116m} In	54.10(20) m	1293.4(3) 2112.1(4)	83.7(15) } 16.2(15) }	sum = 99.93(2)
¹⁴⁰ Ba	12.79(1) d ^d			
¹⁴⁰ La	40.26(2) h ^d	1596.18(5)	95.33(16)	
¹⁹⁸ Au	2.696(2) d	411.794(8)	95.47(10)	
²³⁹ Np	2.355(4) d	228.19(1) ^e 277.60(3) ^f	12.5(15) ^e 14.5(15) ^f	

^a The numbers in parentheses indicate the uncertainty (1 σ) in the last digit(s).

^b The combined intensity of the 1037-keV gamma ray and a 1212-keV gamma ray is (99.98 $\pm$ 0.01)%.

^c The effective line-energy of this peak may be lower than this energy due to electron binding effects and the finite width of the annihilation radiation energy distribution.

^d The ratio $t_{1/2}(\text{Ba})/[t_{1/2}(\text{Ba})-t_{1/2}(\text{La})]$, which is the ratio of the ¹⁴⁰La to ¹⁴⁰Ba activities in a mixed ¹⁴⁰Ba-¹⁴⁰La source at equilibrium, is therefore 1.15096 $\pm$ 0.00012.

^e There is also a gamma-ray transition of 226.4 keV which has an intensity of 0.7%.

^f There are also gamma-ray transitions of 272.8 keV and 280.5 keV, but both have intensities of less than 0.1%.

K. ANALYTICAL REPRESENTATION OF Ge(Li) DETECTOR EFFICIENCY CURVES

R. G. Helmer

Objective

- Evaluate various methods of interpolation of the Ge(Li) detector efficiency for arbitrary energies.

Summary

The efficiency values determined by interpolation from a hand-drawn curve have been compared with the interpolated values from the expression of Mowatt¹. The results indicate deviations of up to 5% between 300 and 2500 keV.

Accomplishments and Status

In the last quarterly report (HEDL-TME 72-45), the efficiency measurements for the OR-5 Ge(Li) detector were described. A comparison of the interpolated efficiency values from a hand-drawn curve and a $\ln \epsilon$ vs $\ln E$ polynomial was also given in tabular form. This comparison is shown graphically in Fig. ANC-2. In the region of 200 to 1500 keV (which includes all but one of the transitions used in the radiochemistry foil experiments), the two interpolation methods agree to 1%. The larger deviations outside this region are due to the sparseness of data points, or their large uncertainties.

The interpolated values from the hand-drawn curve have also been compared with the equation proposed by Mowatt¹. In this expression one tries to relate the efficiency to the photoelectric and Compton cross sections for germanium (and the materials of the detector housing). The parameters in this expression were determined by means of a non-linear, least-squares fit. The results of this comparison are also shown in Fig. ANC-2 for the data above 200 keV. This expression has the same number of variable parameters as the $\ln \epsilon$, $\ln E$ polynomial used in this energy region. It is clear that the Mowatt expression is not as satisfactory since the deviation is often larger than 1%.

In interpolating efficiency values, we will, therefore, take into account the hand-drawn curve and the $\ln \epsilon$ vs $\ln E$ polynomials.

Expected Accomplishments

This project is considered to be complete.

Figure ANC-2. These Curves Give a Comparison of the Ge(Li) Detector Efficiencies Interpolated by Different Methods. The Dashed Curve Represents the Difference between the Values Obtained from a Hand-Drawn Curve and a Fit of the Mowatt Expression to the Measured Values. The Two Solid Curves Show the Difference between $\ln \epsilon$ vs $\ln E$ Polynomials Fit to the Data and the Hand-Drawn Curve.

L. TESTS TO DETERMINE TIME VARIATIONS IN THE PEAK DETECTION EFFICIENCIES OF ND-3

R. C. Greenwood and R. G. Helmer

Objective

- Detect any time variations in the peak detection efficiencies of the ND-3 Ge(Li) detector, and test the reproducibility of the source position (at a source-to-detector distance of 10 cm).

Summary

Recently there has been speculation about the constancy of peak detection efficiencies of Ge(Li) detectors as a function of time; and in fact Wagner et al.² have presented data which indicate that the peak detector efficiencies of many detectors do change significantly with time. Data taken over the past 7 months (i.e., over the period covering our participation in the ILRR program) indicate that no significant change has occurred in the peak detection efficiency of the ND-3 Ge(Li) detector (this detector having principally been used for ILRR related measurements). Quantitatively we can specify that the peak detection efficiency of this detector has remained constant to within 0.3% during this period. Also, these data would indicate that the contribution to the uncertainty in gamma-ray intensity measurements from the reproducibility of the source-detector geometry is no greater than $\pm 0.4\%$ for a source-to-detector distance of 10 cm.

Accomplishments and Status

The peak detection efficiency of the ND-3 Ge(Li) detector has been periodically remeasured using selected standard sources over the past seven months. Standard sources emitting both low (< 150 keV) and higher energy gamma rays were included in this series of measurements in order that we might be able to monitor changes in the detector surface (e.g., dead layer) as well as in the bulk sensitive volume. The results of these measurements are summarized in Table ANC-15. It can be seen from these data that the peak detection efficiency of this detector has remained constant to within $\sim 0.3\%$ over this period of time. Short-term checks on the constancy of the ND-3 detector efficiency during the period in which the ILRR radiochemistry foils were counted were made using both a strong ^{137}Cs source ($\sim 15\%$ dead time) and a weaker ^{60}Co source ($\sim 2\%$ dead time). These data are summarized in Tables ANC-16 and ANC-17. For the ^{60}Co

TABLE ANC-15

TESTS OF LONG-TERM VARIATION IN THE PEAK DETECTION EFFICIENCIES OF ND-3

Source - E_γ (keV)	Source-Detector Distance (cm) ^a	Date of Initial Measurement t_i	Date of Final Measurement t_f	ϵ_i	ϵ_f	$\frac{\epsilon_i - \epsilon_f}{\epsilon_i} \times 100$
						$(\%)^b$
²⁰⁴ Hg - 279	10	10/11/71	11/ 9/71	2.641×10^{-3}	2.655×10^{-3}	$-(0.53 \pm 0.80)$
⁵⁴ Mn - 834	10	10/ 8/71	11/15/71	7.959×10^{-4}	8.007×10^{-4}	$-(0.60 \pm 0.76)$
¹³⁷ Cs - 661	10	11/ 8/71	5/ 9/72	9.968×10^{-4}	9.974×10^{-4}	$-(0.06 \pm 0.73)$
¹³⁷ Cs - 661	25	11/ 8/71	5/ 9/72	1.0000^c	1.0037^c	$-(0.37 \pm 0.37)$
⁶⁰ Co - 1173 1332	10	11/10/71	5/ 9/72	5.811×10^{-4}	5.833×10^{-4}	$-(0.38 \pm 0.75)$
				5.129×10^{-4}	5.126×10^{-4}	$+(0.06 \pm 0.75)$
¹⁵³ Gd - 41 100	10	12/ 1/71	5/ 9/72	4.30×10^{-4}	4.23×10^{-4}	$+(1.6 \pm 1.3)$
				4.348×10^{-3}	4.355×10^{-3}	$-(0.16 \pm 0.78)$

^a The source-position error for each measurement is $\pm 0.5\%$ and 0.2% (1σ uncertainties) for source-detector distances of 10 cm and 25 cm, respectively.

^b The 1σ uncertainties are quoted here.

^c Relative efficiencies.

TABLE ANC-16

TEST OF CONSTANCY OF PEAK DETECTION EFFICIENCY AND REPRODUCIBILITY OF THE SOURCE-
DETECTOR GEOMETRY AT THE 10 CM POSITION WITH ND-3 USING A ^{137}Cs SOURCE

<u>Date</u>	<u>Peak Area*</u> <u>(counts)</u>
2/15	1,026,078
2/16	1,025,569
2/16	1,027,712
2/17	1,024,615
2/18	1,029,054
2/22	1,025,666
2/29	1,023,497
3/ 1	1,019,708
3/ 3	1,022,158
3/ 6	1,023,159
3/ 8	1,025,087
3/10	1,024,086
3/13	1,021,591
3/17	1,025,996
3/24	1,024,048
3/27	1,025,110
4/12	1,019,753
4/24	1,017,535
AVG.	1,023,912

RMS deviation from mean is ± 4031 ($\pm 0.40\%$)

* The areas measured for this 661-keV line have been corrected for the decay of the ^{137}Cs source.

TABLE ANC-17

TEST OF CONSTANCY OF PEAK DETECTION EFFICIENCY AND REPRODUCIBILITY OF THE SOURCE-
DETECTOR GEOMETRY AT THE 10 CM POSITION WITH ND-3 USING A ^{60}Co SOURCE

<u>Date</u>	<u>Peak Area</u> [*] <u>(counts)</u>
2/ 8	59,250
2/ 9	58,872
2/15	59,437
2/16	59,037
2/17	59,257
2/22	59,322
3/ 6	59,001
3/13	58,484
3/17	59,587
3/24	58,907
4/24	58,865
Mean	59,092

RMS deviation from mean is ± 299 ($\pm 0.51\%$)

* The areas measured for the summed 1173- and 1332-keV peaks have been corrected for the decay of the ^{60}Co source.

source, after correcting for decay, the RMS deviation of the measured peak areas from the mean is $\pm 0.51\%$, which is only slightly larger than the expected statistical variation of $\pm 0.42\%$. For the ^{137}Cs source, however, the RMS deviation from the mean is $\pm 0.40\%$ which is significantly larger than the expected statistical variation of $\pm 0.10\%$. It is to be expected for our source mounting arrangement that most of this $\pm 0.40\%$ error is due to small variations of the source position. (In fact, we have usually estimated our source position error to be $\pm 0.5\%$ at a source-detector distance of 10 cm.)

Expected Accomplishments

We will continue to monitor the peak detection efficiencies of all the Ge(Li) detectors used on the ILRR measurement program to check for time variations in these efficiencies.

References for ANC Chapter

1. R. S. Mowatt, Nucl. Instr. & Meth. 70 (1969) p. 237.
2. S. Wagner, R. Trammell and F. J. Walter, "Scintillation and Semiconductor Counter Symposium, Washington, D.C., March 1-3, 1972".

Argonne National Laboratory

A. ABSOLUTE FISSION RATE MEASUREMENTS BY THE TRACK RECORDER TECHNIQUE

R. J. Popek, N. D. Dudey, R. J. Armani, and R. Gold

OBJECTIVE

• To measure the absolute fission rates of ^{235}U , ^{238}U , ^{237}Np , and ^{239}Pu in the CFRMF spectrum by means of solid-state track recorders (SSTR).

SUMMARY

Results from the ^{235}U and ^{237}Np SSTR samples irradiated in CFRMF on February 3 and 4, 1972 are reported. The fission rates by SSTR compared with fission-chamber measurements agree within 2.5% for ^{235}U and 7.5% for ^{237}Np . Deviations between individual SSTR measurements, i.e., experimental reproducibilities, are 1% or less. It is estimated that the time uncertainty in introducing the SSTR packages into and removing them from CFRMF is about 10 sec.

ACCOMPLISHMENTS AND STATUS

Fission rate measurements by the SSTR technique were conducted in CFRMF on February 3 and 4, 1972 at a nominal reactor power level of 600 W. Descriptions of the irradiation package, foil preparation, and a general discussion of the technique can be found in earlier ILRR reports. At present, track counting has been completed for the ^{235}U and ^{237}Np packages; the corrected fission rates are tabulated in Table ANL-1.

TABLE ANL-1
SSTR Results at the Center of CFRMF

Sample Number	Irradiation Number	Isotope	Weight (ng)	Exposure Time (sec)	10^{-15} f/atom-sec
5-10	4	^{235}U	99.7	3605	13.07
5-11	4	^{235}U	98.2	3605	13.21
5-12	7	^{235}U	86.2	3609	13.23
7-1	8	^{237}Np	3410	368	4.94
7-2	9	^{237}Np	3509	371	4.83
7-5	6	^{237}Np	352	2948	4.84
7-6	6	^{237}Np	324	2948	4.88
7-7	7	^{237}Np	322	3609	4.87

The ^{235}U foils were prepared by electrodeposition of a 93% enriched uranium solution tagged with ^{234}U . Alpha counting of the ^{234}U enabled mass determination of each sample. For these samples, fission tracks can occur from ^{234}U (3.16 at. %) and ^{238}U (5.59 at. %) in addition to ^{235}U . Corrections were made for the fissions resulting from the former two sources from assumed fission-rate ratios of $234:235 = 0.417$ and $238:235 = 0.047$. The former ratio was measured in the ZPR-3, Assembly 60 spectra, which was similar to the CFRMF spectra and the latter was measured by Grundl in the CFRMF. Both ratio measurements were made by back-to-back fission chambers. No corrections were required for the ^{237}Np track recorder results. The Np masses were determined by alpha counting of the 99.99% ^{237}Np electrodeposited on platinum plates.

This first CFRMF track-recorder study had several objectives and these will be discussed individually. Firstly, we wanted to ascertain the degree of precision which could be obtained. For this purpose we prepared three ^{235}U recorders and five ^{237}Np recorders. Two each of the ^{235}U and ^{237}Np foils were irradiated together to examine the deviations due to track counting and mass determinations. The ^{235}U foils agreed within 1.07% and the ^{237}Np within 0.82%. Since each foil produced about 15,000 tracks, the statistical fluctuation in each sample can be expected to be 0.83% (1 σ). Thus, the precision of these measurements is actually better than the expected precision due to counting statistics. Next we compared two ^{235}U samples and two ^{237}Np samples irradiated in separate irradiations. In this case the ^{235}U foils agreed within 0.67% and the ^{237}Np foils within 0.62%. Our conclusion is that the masses of the track recorders are very well established, at least on a relative basis.

All of the above irradiations were for 60-min duration. Our second objective was to define the uncertainty resulting from inserting and removing the samples from the reactor. For this test we irradiated two ^{237}Np samples in separate irradiations for 6 min each, again obtaining about 15,000 tracks per exposure. In this case, the deviation between the two samples was 1.85%. We estimate that roughly 1% of this is due to time uncertainty, which corresponds to an uncertainty of about 10 sec in insertion-removal time. Thus, for a 1-hr exposure, this uncertainty is negligible.

The above tests relate to the precision of the technique, that is, the relative uncertainties in the number of atoms of fissile material on each plate, variations in irradiation times, and reactor power normalization, and variations in counting of the tracks. Although a complete error analysis has not been performed, the relative accuracy of the ^{235}U and the ^{237}Np samples is 0.4% (1 σ). Of more importance to the ILRR program, however, is the absolute accuracy. Sufficient data have been accumulated for CFRMF to begin to examine this question. These data are tabulated in Table ANL-2. Because of present uncertainties in the absolute power normalization, we show the ^{235}U fission rates (measured by the three techniques) normalized to the NBS results in which the normalization is computed on the basis of both the gold monitor foils and the stated reactor power levels (reactor instrumentation). The uncertainties assigned to the fission rates in the last two columns of Table ANL-2 are the relative precision of the multiple measurements. In addition to pointing out the difficulties in power level normalization we can get a feeling for the current accuracies in ^{235}U fission rate measurements. Based upon the gold monitor power level normalization we find the SSTR results about 2.5% higher than the fission chamber results and virtually identical to the radiochemical results. The 0.6 kW radiochemical results were measured in the Preliminary I tests and were not intended for absolute accuracy. For this reason and the relatively large spread in the radiochemical data of 2-3%, the agreement of the radiochemical results with the other methods should more appropriately be interpreted as verification of the ^{140}Ba fission yield of ^{235}U rather than an indication of the absolute accuracy of these preliminary results.

Similar comparisons of SSTR results with fission-chamber results for ^{237}Np are not as satisfactory as for ^{235}U . In this case the track-recorder data are about 7.5% higher than the fission-chamber results. The source of this discrepancy is not apparent at this time, although foil weights are the most obvious choice.

Gram quantities of high-isotopic-purity ^{235}U , ^{238}U , ^{237}Np , and ^{239}Pu (reserved for dosimetry purposes) are scheduled for shipment from ORNL on June 2, 1972. These materials are needed to prepare stock solutions for SSTR electroplating. No ^{238}U SSTR will be prepared until this material is

TABLE ANL-2
Preliminary Comparison of Absolute Fission Rate Measurements
of ^{235}U in CFRMF Normalized to a 0.6 kW Power Level

Method	Lab	Reported Fission Rate ^(a) ($\times 10^{14}$)	Reactor Power Level (kW)	Relative Power Level ^(b)	Fission Rate Normalized to 0.6 kW (10^{-14} f/at-sec)	
					Gold Norm.	Reactor Power Norm.
F.C.	NBS	1.284	0.6	1.000	1.284 $\pm$ 0.001	1.284 $\pm$ 0.001
SSTR	ANL	1.317	0.6	0.996	1.322 $\pm$ 0.005	1.317 $\pm$ 0.005
Foil ^(c)	ANL	1.308	0.6	0.966	1.354	1.308
	ARHCO	1.280	0.6	0.966	1.325	1.280
	HEDL	1.248	0.6	0.966	1.292	1.248
	ANL	21.0	9.75	14.83	1.416	1.292
	ARHCO	20.3	9.75	15.40	1.318	1.249
	HEDL	20.3	9.75	14.62	1.389	1.249

^aCorrected for ^{238}U contribution and gamma-ray self absorption but not corrected for neutron self-absorption or scattering.

^bDetermined from gold-monitor foils reported by both HEDL and ANC and normalized to the NBS measurement.

^cFission rates were measured by counting ^{140}Ba and applying a fission yield of 5.8%.

received. Some ^{234}U has been separated on the mass separator and will be used for spiking the ^{235}U and ^{238}U solutions. These solutions will represent the highest possible quality obtainable for SSTR preparation in terms of isotopic purity and the accuracies with which the concentrations are known.

EXPECTED ACHIEVEMENTS

Counting of tracks from the ^{239}Pu samples exposed in the first CFRMF irradiation will be completed. ^{235}U samples exposed at NBS in a back-to-back configuration with Grundl's fission chambers will be analyzed. Final stock solutions of ^{235}U , ^{238}U , ^{239}Pu , and ^{237}Np will be prepared when we receive the high-isotopic-purity materials. A second series of SSTR irradiations will be conducted in CFRMF which will emphasize ^{238}U and include several ^{234}U track recorders in order to establish more accurately the corrections needed for the ^{234}U spike.

B. CFRMF REACTION RATES BY FOIL ACTIVATION

R. J. Popek and N. D. Dudey

OBJECTIVE

- To measure fission and reaction rates by means of activation techniques for those reactions having fast-neutron dosimetry applications.

SUMMARY

Reaction rates from the ANL nonfissile foil packet irradiated on February 15, 1972 in CFRMF at a nominal reactor power level of 10 kW are reported. The HEDL calibrated $^{140}\text{Ba}/^{140}\text{La}$ standard (T913-1) was counted on our Ge(Li) detector systems and found to agree within 0.9% of our previous efficiency calibrations at 1596 keV.

ACCOMPLISHMENTS AND STATUS

Reaction rate results from the ANL-1 nonfissile foil packet irradiated in CFRMF on February 2, 1972 for a nominal exposure of 70 kWh have been completed. Table ANL-3 summarizes the nuclear constants used in the data reduction and Table ANL-4 summarizes the reaction rates together with additional information necessary for evaluating the quality of the results. The additional information includes the type of detector used for counting, the number of times each reaction product was counted, and the relative precision between the multiple counts. The counting statistics of virtually all samples was 0.1-0.3% but the precision was computed on the actual deviations between counts. The final results were corrected for gamma ray self-absorption; the corrections are given in the 6th column of Table ANL-4. The assigned absolute errors were determined from (1) the relative precisions and uncertainties, (2) branching ratios, (3) half-lives, (4) detector efficiency calibrations, and (5) fission yields. No corrections have been made for neutron scattering or self-shielding effects in the foil packets.

Table ANL-5 summarizes the fission product results for the ^{235}U and ^{238}U foils in this packet. The fission yields are those which we have measured in the EBR-II mockup critical assembly experiments. Excluding ^{131}I the fission rate as determined from the individual fission products is quite good, especially for ^{235}U . The average fission rate for each nuclide is shown together with the 1 σ standard deviation of the individual

TABLE ANL-3
Nuclear Constants Used in Reaction-Rate Measurements

Target	Product	Energy (keV)	Atom Abundance (%)	Half-life (day)	Branching Ratio
Al	^{24}Na	1368.5	100	0.625	1.0
Au	^{198}Au	411.8	100	2.70	0.968
In	$^{115\text{m}}\text{In}$	335	95.72	0.1875	0.45
Cu	^{64}Cu	511	69.09	0.533	0.380
Co	^{60}Co	1173.23	100	1920	1.0
	^{60}Co	1332.48	100	1920	1.0
Ni	^{58}Co	810.5	67.88	71.3	0.994
Fe	^{54}Mn	835.3	5.82	303	1.0
	^{51}Cr	319.8	5.82	27.8	0.95
	^{59}Fe	1099.5	0.33	45.6	0.56
	^{59}Fe	1292	0.33	45.6	0.44
Ti	^{46}Sc	889.4	7.93	83.9	1.0
	^{46}Sc	1120.5	7.93	83.9	1.0
	^{47}Sc	160	7.28	3.43	0.73
	^{48}Sc	983	73.94	1.83	1.0
	^{48}Sc	1040	73.94	1.83	1.0
	^{48}Sc	1314	73.94	1.83	1.0
Sc	^{46}Sc	889.4	100	83.9	1.0
	^{46}Sc	1120.5	100	83.9	1.0

TABLE ANL-4

Summary of Measured Reaction Rates for the ANL-1 Nonfission Foil Set

Reaction	Sample Number	Type of Detector	Times Counted	Relative Precision (%)	γ -ray Self Absorption	Reaction Rate ($\bar{\sigma}\phi$) (10^{-15} atoms/atom-sec)	1 σ Absolute Error (%)
$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	6	NaI	3	0.3	1.00	0.0209	5.8
	5	"	2	0.1	"	0.0206	5.8
	4	"	3	0.5	"	0.0209	5.8
	2	Ge	4	0.8	0.998	0.0206	5.5
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	4	Ge	4	0.2	1.00	52.6	2.4
	2	"	3	0.3	"	53.5	2.4
	3	"	4	0.2	"	52.1	2.4
	2	"	4	0.6	0.996	53.5	2.5
$^{115}\text{In}(n,n')^{115\text{m}}\text{In}$	6	Na, Ge	2	0.8	0.998	6.61	3.2
	5	"	2	0.8	"	6.64	3.2
	4	"	3	0.7	"	6.64	3.2
$^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$	2	Ge	3	0.2	1.00	7.30	5.5
$^{59}\text{Co}(n,\gamma)^{60}\text{Co}$	6	"	4	0.5	1.00	9.37	2.4
	5	"	4	0.3	"	8.49	2.4
	4	"	4	0.2	"	9.01	2.5
$^{58}\text{Ni}(n,p)^{58}\text{Co}$	TE	"	3	0.3	0.992	3.07	5.5
$^{54}\text{Fe}(n,p)^{54}\text{Mn}$	B	"	5	1.0	0.978	2.21	2.7
$^{54}\text{Fe}(n,\alpha)^{51}\text{Cr}$	B	"	5	3.3	0.966	0.0292	6.4
$^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$	B	"	4	0.5	0.981	0.750	3.0
$^{46}\text{Ti}(n,p)^{46}\text{Sc}$	P	"	4	0.2	0.996	0.340	2.4
	0	"	6	1.0	"	0.337	2.6
$^{47}\text{Ti}(n,p)^{47}\text{Sc}$	P	"	6	0.8	0.990	0.512	5.5
	0	"	6	0.5	"	0.517	5.5
$^{48}\text{Ti}(n,p)^{48}\text{Sc}$	P	"	7	1.0	0.996	0.00857	5.6
	0	"	8	0.6	"	0.00861	5.5
$^{45}\text{Sc}(n,\gamma)^{46}\text{Sc}$	2	"	6	0.3	0.999	2.94	2.4
$^{238}\text{U}(n,\gamma)^{239}\text{Np}$	2	"	5	0.8	0.985	22.5	5.5
$^{235}\text{U}(n,f)^{140}\text{Ba}$	2	"	6	0.4	0.994	210	4.3
$^{238}\text{U}(n,f)^{140}\text{Ba}$	2	"	5	0.8	0.999	0.987	4.4

TABLE ANL-5
Fission Product Reaction Rates of ^{235}U and ^{238}U

Fission Product	²³⁵ U					²³⁸ U				
	[10 ⁻¹⁴ atoms/(atom ²³⁵ U)(sec)]			F.Y. (%)	Fission Rate (10 ⁻¹³)	[10 ⁻¹⁶ atoms/(atom ²³⁸ U)(sec)]			F.Y. (%)	Fission Rate (10 ⁻¹⁵)
	% Error		% Error							
	Rate	Relative	Absolute			Rate	Relative	Absolute		
⁹⁵ Zr	1.33	0.9	3.1	6.41	2.08	5.09	0.7	3.1	5.44	9.35
¹⁰³ Ru	0.680	0.3	2.4	3.29	2.06	6.33	0.2	2.4	6.29	10.07
¹³¹ I	0.640	1.1	2.7	3.44	1.86	3.11	0.9	2.6	3.64	8.54
¹³² Te	0.962	1.0	3.9	4.77	2.01	5.64	1.2	3.9	5.35	10.5
¹⁴⁰ Ba	1.22	0.4	2.5	5.80	<u>2.10</u>	5.82	0.8	2.6	5.90	<u>9.87</u>
					2.06±0.02					9.95±0.22

rates from the average. From these results we conclude that ^{95}Zr , ^{103}Ru , ^{132}Te , and ^{140}Ba are all equally reliable for measuring ^{235}U fission rates. At this point, ^{140}Ba appears preferable for determining ^{238}U fission rates; ^{103}Ru is reliable but ^{95}Zr requires more study. The discrepancy in the ^{131}I yields has been observed in other measurements and also requires additional study. In practice ^{131}I is considered less reliable than the others because (1) it has high mobility in uranium due to its higher volatility and (2) its softer gamma ray requires larger self-absorption corrections, thereby increasing the uncertainty of results. Work is under way to relate and compare the ^{137}Cs yields with ^{140}Ba , ^{103}Ru , and ^{95}Zr . Confidence in these four fission product yields will enable fission rate measurements to be conducted for virtually any flux, fluence, and time duration encountered in dosimetry situations.

The HEDL calibrated Ba/La standard T-913 has been measured on our Ge(Li) detector systems. The ^{140}Ba disintegration rate measured by HEDL was 1.134 dps/g; our measured value is 1.121 dps/g. This agreement of better than 1% substantiates our detector efficiency calibrations at 1596 keV. This fact, together with the 1% agreement between our ^{235}U fission rates as measured by SSTR and foil activation (see Table ANL-2) strongly supports the accuracy of the ^{140}Ba yield value of 5.80%.

EXPECTED ACCOMPLISHMENTS

Irradiation and counting of the high-isotopic-purity fissile foils of ^{235}U , ^{238}U , ^{239}Pu , and possibly ^{237}Np are expected to begin in the next quarter. Planning of experiments in facilities other than CFRMF will be completed.

C. ACCELERATOR-PRODUCED NEUTRON SPECTRA

N. D. Dudey and M. J. Fluss

OBJECTIVE

- To present information regarding sources of neutron fields which provide alternatives to reactor spectra for use in the ILRR Program.

SUMMARY

Accelerator produced neutron sources are now available which can provide flux intensities approaching 10^{10} n/cm²-sec on target. These neutron spectra may be tailored in mean energy (1-8 MeV) and energy spread so as to enable reaction rate measurements which emphasize neutron energies considerably higher than is convenient in reactor spectra. By providing a means for evaluating differential cross-section data above 2 MeV, they are a potentially valuable addition to the ILRR program's efforts to reduce uncertainties in fast-neutron dosimetry.

ACCOMPLISHMENTS AND STATUS

Recent studies of the $^9\text{Be}(d,n)^{10}\text{B}$ reaction by Krushnir, Elwyn and Mooring¹ with 8 MeV deuterons at ANL and earlier work by Inada et al² with 2 MeV deuterons have demonstrated that adequate neutron intensities can be obtained to utilize foil-activation techniques. The principal feature of this approach is that pulsed deuteron beams may be employed to enable time-of-flight neutron spectral measurements, yet d.c. (continuous) beams may be employed to achieve high fluence levels. Current work has indicated that the combination of variable deuteron energies between 2 and 16 MeV and energy variations as a function of angle relative to the deuteron beam provides a means for tailoring both the mean neutron energy and the shape of the energy spectrum. The energy distribution at several angles relative to an 8 MeV deuteron beam as measured by Krushnir et al. is shown in Fig. ANL-1.¹

The flux intensities measured with 8.3 MeV deuterons was 1.1×10^{10} (neutrons/steradian- μcoul) and 5.5×10^{10} with 16 MeV deuterons.¹ Beam currents of 10 μA are available on the ANL Tandem accelerator for up to 16 MeV deuterons, and currents of 500 μA are available on the ANL Dynamitron for deuteron energies up to 4 MeV. The ANL Fast Neutron Facility (FNG) is capable of several hundred microamperes current for up to 8 MeV deuterons; however, present limitations make it comparable to the Tandem. We can

Fig. ANL-1. Measured Neutron Spectra and Angular Distributions from the ${}^9\text{Be}(d,n){}^{10}\text{B}$ Reaction at 8.3 MeV Deuteron Energy. The integrated neutron yield (0° - 10°) is 1.1×10^{10} (neutrons/ster.- μcoul).

conclude that 10^{10} - 10^{11} n/cm²-sec can be achieved on target (5 cm from the source) for virtually any mean neutron energy between 1 and 8 MeV. Another feature of these spectra is that the energy spread of (typically ± 25 -100% of the mean energy) is sufficiently broad that foil activation techniques may be applied, yet the spectra are considerably more confined than reactor spectra. In effect, this means that the response functions for all reaction rates will peak very near the mean neutron energy, and thus by varying the neutron energy one can critically evaluate differential cross-section data for dosimetry reactions in specifically defined energy regions. This is not often possible in reactor spectra, e.g., (n, γ) reactions are dominated by their response to lower energy neutrons so that little cross-section information can be obtained above 1 MeV by integral measurements. For comparison purposes, we estimate that the flux between 4 and 8 MeV from a 10 μ A accelerator is comparable to that of CFRMF (for that energy range) operated at 10 kW. For higher energies the accelerator neutrons exceed CFRMF in flux by up to a factor of 5. This could be particularly useful in studying (n, α) reactions.

The spectra shown in Fig. ANL-1 are for a thick Be target. The energy resolution was obtained with a 2-meter flight path; however, longer flight paths, shorter beam bursts, and faster detection equipment, all of which are available, could significantly increase this energy resolution. In addition, proton-recoil, and ⁶Li spectrometry techniques can improve the low-energy spectral characterization. Because "infinitely" thick Be targets are used, once a spectrum is measured at a given deuteron energy, any accelerator can be used without the need to remeasure the spectrum. Thus, a permanent, well-established neutron spectrum can be developed for all desired energies.

Neutron scattering effects and flux gradients may be more severe than in reactor environments; however, because the neutrons originate from a point source, corrections for these effects can, in principle, be made much more accurately. In conclusion, the potential of accelerator-produced neutrons to supplement the current ILRR efforts and to extend the neutron energies available for study up to ~ 8 MeV is very attractive. We suggest that incorporation of these neutron sources into the ILRR program be given serious consideration.

EXPECTED ACCOMPLISHMENTS

If possible, a foil packet will be irradiated in a neutron spectrum produced by 8 MeV deuterons on Be. The objective will be to verify the flux intensities and to provide preliminary reaction-rate data for purposes of scoping experiments.

REFERENCES

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Atomics International

A. INTEGRAL (n, α) CROSS SECTIONS BY HELIUM MEASUREMENT

H. Farrar IV

Objective

• To determine integral (n, α) cross sections of ^6Li , ^{10}B , and possibly ^{27}Al and other isotopes, in a variety of well-established and permanent neutron fields, including several that cover the neutron energy range of interest for FTR and LMFBR's. The cross sections, which are obtained by mass spectrometric measurement of the amount of helium generated in milligram-size specimens, will provide an additional and independent absolute base for normalization of available energy dependent (n, α) cross section data.

Summary

Fourteen small specimens of boron and of its two separated isotopes have been irradiated in CFRMF. Mass spectrometric **measurements of the** helium generated in two of the specimens of ^{10}B have given concentrations in excellent agreement with each other. The results compare well with preliminary values calculated using available energy dependent (n, α) cross section data.

Accomplishments and Status

Ten crystals of enriched ^{10}B and two crystals each of natural boron and enriched ^{11}B were irradiated for 16 hours on April 10-11 1972, in the Coupled Fast Reactor Measurement Facility (CFRMF). Details of the irradiation are given in the Aerojet Nuclear Company (ANC) section of this report. After the irradiation, the Al, Au and In monitor foils which were placed adjacent to the boron specimens were removed for radiochemical determinations at HEDL, and the boron specimens were returned to Atomics International for mass spectrometric analysis. The results of the radiochemical analyses of the monitor and power normalization foils are given in the HEDL and ANC sections of this report.

Two of the specimens of 96.5% enriched ^{10}B were reweighed and analyzed for helium content by mass spectrometer. Each boron specimen was vaporized along with some titanium wire, in a resistance-heated graphite crucible. Gases other than helium that were released by the crucible were adsorbed satisfactorily by a charcoal getter. The amount of ^4He released by each specimen was determined with respect to a known number of ^3He atoms added to the system at the time of sample vaporization. The results of the analyses are given in Table AI-1, along with the location of each specimen in the aluminum disk holding them during the irradiation.

TABLE AI-1
HELIUM CONCENTRATION MEASUREMENTS

Sample Number	Location in Disk	Mass (mg)	Number of Atoms of ^4He Released	Helium Concentration (Parts Per Billion Atoms of ^{10}B)
R10B2	5	4.437	3.2144×10^{12}	12.53
R10B8	4	2.989	2.1394×10^{12}	12.38

The values of mass are the weighted averages of values obtained before and after the irradiation and have an absolute accuracy of better than 0.2%. The helium concentrations were calculated with respect to the number of atoms of ^{10}B , in each sample. Assuming an enrichment of 96.50%, a value of 5.7827×10^{22} atoms of ^{10}B per gram was used. The isotopic compositions of this material and those of the enriched ^{11}B and natural boron are currently being verified at HEDL.

The absolute uncertainty of the concentration values shown in Table AI-1 has not yet been calculated but is expected to be better than 2% (1 σ). The two values so far obtained differ by only 1.2%. Final values for the helium production rate in ^{10}B will be calculated from these and future specimens after helium measurements have been made on the enriched ^{11}B and natural boron specimens that were also irradiated. The helium contribution from the ^{11}B is, however, expected to be very small.

The measured values of helium concentration have been compared with preliminary values calculated at HEDL using current energy-dependent cross section data. The shape of the neutron energy spectrum used for the calculations is shown on page HEDL-11 in the 1st ILRR Quarterly Report (HEDL-TME 71-131). This was developed by ANC as the best current approximation to the true spectrum. Preliminary examination of the various normalization foils indicates that the flux level during the AI-1 irradiation was approximately 1.76% higher than that during the HEDL-1 irradiation. The average value for the total flux during the HEDL-1 irradiation, given in the 3rd ILRR Quarterly Progress Report (HEDL-TME 72-45) is 1.090×10^{11} n/cm²-sec ($\pm 4.9\%$ 1 σ). Assuming these numbers are correct, this means that the AI foil set experienced a total neutron flux of 1.109×10^{11} n/cm²-sec and a total fluence of 6.389×10^{15} n/cm².

Table AI-2 presents calculated values of the spectral averaged cross sections, nominal 5% energy limits between which 90% of the reactions take place, and helium concentrations.

TABLE AI-2
COMPARISON OF MEASURED AND CALCULATED HELIUM PRODUCTION IN ¹⁰B

Source of Data	Spectral Averaged (n,α) Cross Section (barns)	Nominal 5% Energy Limits (keV)		Helium Conc. (ppb ¹⁰ B) *
		Lower	Upper	
Calculation, SAND-II (total)	1.914	0.21	630	12.23
Calculation, ENDF/BII (n,α)	1.902	0.23	690	12.15
Calculation, ENDF/BII (total)	1.919	0.23	800	12.26
Measurement, Mass Spectrometry	-	-	-	12.46

* Preliminary calculated helium concentrations assume a total neutron flux of 1.109×10^{11} neutrons/cm²-sec.

The calculations were made using a) the SAND-II total absorption cross section for ¹⁰B, b) the ENDF/BII (n,α) cross section, and c) the ENDF/BII total absorption cross section. The close agreement between these preliminary

calculated and measured values is most encouraging, particularly considering the experimental uncertainties in the flux value at this time. It should be pointed out that the average flux value used above was derived from data from 15 reactions, many of which have energy responses very different from that of ^{10}B . Perhaps a better number to use would have been the total flux based solely on the $^{235}\text{U}(\text{n},\text{f})^{140}\text{Ba}$ reaction. The cross section for this reaction is considered to be among the more reliable, and the calculations based on it are subject to less uncertainty due to spectral shape uncertainties, because of its relatively flat energy response. If this value of total flux had been assumed, the total neutron fluence for the AI-1 foil set would have been $7.010 \times 10^{15} \text{ n/cm}^2$. This would have resulted in calculated values of helium concentration averaging 13.40 ppb.

Whether the calculated values of helium concentration are slightly lower or higher than the measured values will be known after further measurements have been made, both radiochemical and mass spectrometric. The possibility of slight neutron self-shielding in the ^{10}B is being examined, and the possibility of loss of helium by diffusion must be considered. Nevertheless, these results show that for this CFRMF irradiation, the mass spectrometric technique appears to offer a good opportunity to normalize the energy dependent (n,α) cross section data in the neutron energy range 0.2 to 800 keV.

Expected Achievements

Further mass spectrometric analyses of the helium content of the boron specimens irradiated in CFRMF will give the exact amount of helium generated by the ^{10}B isotope. The spectrum-integrated (n,α) cross section thereby obtained will then be compared with values calculated using energy dependent (n,α) cross section data, providing the latter with an independent and absolute normalization.

B. NEUTRON CROSS SECTION EVALUATION - DOSIMETRY APPLICATIONS

H. Alter

Objective

- To prepare a dosimetry data file for the copper isotopes in ENDF/B format.

Summary

Data for the reactions $^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$, $^{63}\text{Cu}(n,\alpha)^{60}\text{Co}$ and $^{65}\text{Cu}(n,2n)^{64}\text{Cu}$ were evaluated, formulated for the ENDF/B data file and forwarded to the National Neutron Cross Section Center (NNCSC) at Brookhaven.

Accomplishments and Status

At the request of the CSEWG Normalization and Standards Subcommittee, available experimental data for the reactions $^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$, $^{63}\text{Cu}(n,\alpha)^{60}\text{Co}$ and $^{65}\text{Cu}(n,2n)^{64}\text{Cu}$ were reviewed, and evaluated data sets for the above reactions were generated. The data were cast into the ENDF/B format, and forwarded to the National Neutron Cross Section Center at BNL for inclusion into the ENDF/B dosimetry file.

Expected Achievements

It is expected that the ENDF/B dosimetry data file, once it has been established and checked out, will undergo extensive testing against integral measurements. Such integral benchmarks are currently being set up for formal review by the CSEWG Data Testing Subcommittee.

Brookhaven National Laboratory

NATIONAL NEUTRON CROSS SECTION CENTER
B. A. Magurno

A. Evaluated Cross Section Libraries

Objective

- Continuation of the comparison of the SAND-II and ENDF/-III evaluated energy dependent cross section libraries.

Summary

Continuing the effort to compare the ENDF/B-III and SAND-II libraries, the newly evaluated (preliminary) ENDF/B production cross section of ^{46}Sc from Titanium is plotted on the same scale as that of the SAND-II version.

The Normalization and Standards Subcommittee of CSEWG is directing a Task Force for the purpose of evaluating all the isotopes of "primary importance", not now in the ENDF/B library, to be included in the creation of a dosimetry library.

The fission spectrum averaged cross sections of the isotopes common to both libraries were calculated and compared.

Accomplishments and Status

The activation cross section of Titanium to produce ^{46}Sc was evaluated in this quarter. This cross section is useful in dosimetry because of its long half-life and simple mode of decay (Table 1). ^{46}Sc is produced from Titanium by four reactions, $^{46}\text{Ti}(n,p)^{46}\text{Sc}$, $^{47}\text{Ti}(n,np)^{46}\text{Sc}$, $^{47}\text{Ti}(n,pn)^{46}\text{Sc}$ and $^{47}\text{Ti}(n,d)^{46}\text{Sc}$. All the ^{47}Ti reactions will hereafter be referred to as $^{47}\text{Ti}(n,np)$. The threshold for $^{46}\text{Ti}(n,p)^{46}\text{Sc}$ is 1.62 MeV and for $^{47}\text{Ti}(n,np)^{46}\text{Sc}$ is 10.58⁽¹⁾ MeV.

The SAND-II version $^{46}\text{Ti}(n,p)^{46}\text{Sc}$ was created by the "unfolding technique"⁽²⁾ utilizing a Hauser-Feshbach calculation and one experimentally determined data set. The above calculations did not include the $^{47}\text{Ti}(n,np)^{46}\text{Sc}$ contribution.

The ENDF/B-III version includes both reactions and the $^{47}\text{Ti}(n,np)^{46}\text{Sc}$ begins to make its presence felt about 13 MeV. The data sets used in the ENDF/B-III evaluation are shown in Fig. 1. [The error bars in Fig. 1 and the reference numbers in Fig. 2 are omitted for clarity.] The 14 MeV cross sections were included in Fig. 1 but the references appear as a separate listing (Table 2). A comparison of the SAND-II curve and the data is shown in Fig. 2 and a comparison of SAND-II and ENDF/B-III curves is shown in Fig. 3.

The first version of the ENDF/B-III evaluation i.e. $^{46}\text{Ti}(n,p)^{46}\text{Sc}$ was presented at a "Task Force" meeting at Hanford, March 28, 1972. The Task Force consisted of the Normalization and Standards Subcommittee of CSEWG and several volunteers interested in dosimetry problems. This was a tentative evaluation since most of the standards used in the experimental data were being reevaluated for this meeting. The point was made that activation detectors generally used natural elements and therefore evaluations such as $^{46}\text{Ti}(n,p)^{46}\text{Sc}$ should include contributions from any other reaction leading to the same final product. Acceptance of the ENDF/B-III evaluation was put off pending final renormalization and inclusion of all Titanium reactions leading to a production cross section of ^{46}Sc . At the regular meeting of CSEWG May 9th and 10th 1972 the updated version of the evaluation (including all provisions above) was presented. The evaluation is still

labeled $^{46}\text{Ti}(n,p)^{46}\text{Sc}$. The $^{47}\text{Ti}(n,np)$ data used is shown plotted on Fig. 4. This evaluation along with 10 other reactions (9-isotopes) will be released for CSEWG distribution in early June.

As a further comparison of the two libraries, the fission spectrum averaged cross sections were calculated where common to both libraries. The spectrum used in the calculation is defined in ENDF 102⁽³⁾ as LF = 7, a simple fission (Maxwellian) spectrum in the form

$$\frac{2}{\sqrt{\pi}} \frac{E_0^{3/2}}{E^3} e^{-E/E_0}$$

where E_0 is the nuclear temperature. ($E_0 = 1.3$ MeV) The results of the comparison are given in Table 3.

Expected Achievements

The evaluation of isotopes not currently in the ENDF/B-III library but of importance as dosimetry cross sections will continue. Group averages of ENDF/B-III for direct comparison to SAND-II and a linear plot of the ratio of SAND-II to ENDF/B-III, and where possible, resonance integrals for $1/E$ and fission spectrums will be calculated.

References

1. R. J. Howerton, UCRL-50400 Vol. 9, 1970.
2. R. L. Simons and W. N. McElroy , BNWL-1312, 1970.
3. M. K. Drake, BNL 50274 (ENDF-102) Vol I, 10.3, 1970.

TABLE BNL-1

ENERGY LEVELS OF $Z = 11-21$ NUCLEI (IV) ^{46}Sc Energy levels of ^{46}Sc

Figure BNL-1 Data Used in ENDF/B-III Evaluation

TABLE BNL-2

REFERENCES FOR FIGURE BNL-1

	<u>E (MeV)</u>	<u>σ(mb)</u>	<u>Reference</u>
6	14.0	203 ± 21	D. L. Allen Nucl. Phys. <u>24</u> , 274, 1961
7	14.0	240 ± 40	Bayhurst and Prestwood In <u>Fast Neutron Physics</u> , <u>Part II</u> , Interscience Publ., N.Y., London 1963. Chap. V.N by Ribe.
8	14.5	268 ± 30	W. G. Cross and H. L. Pai Pri. Comm. to M. D. Goldberg 1963.
9	14.7	324 ± 30	D. R. Koehler and W. L. Alford J. Nucl. En. <u>18 A/B</u> , 81, 1964.
A	14.8	230 ± 50	V. N. Levkovskii et al. Yad Fiz <u>10</u> , 44, 1969. Trans. in Sov. J. Nucl. Phys. <u>10</u> , 25, 1969.
not shown	14.8	~ 520	A. Poularikas and R. W. Fink Phys. Rev. <u>115</u> , 989, 1959.

BNL-8

Figure BNL-2 Comparison of SAND-II Curve with Data

BNL-9

Figure BNL-3 Comparison of SAND-II and ENDF/B-III for ⁴⁶Ti

BNL-10

Figure BNL-4 ^{46}Ti (n,p) and ^{47}Ti (n,np) Data Used in
 ENDF/B-III Evaluation

TABLE BNL-3

FISSION SPECTRUM AVERAGED CROSS SECTION COMPARISON

<u>Isotope</u>	<u>Spectrum Averaged Cross Sections (mb)</u>		
	ENDF/B-III	SAND-II ^(a)	SAND-II ^(b)
⁶ Li(n, total He)	473.8	389.1	-
¹⁰ B(n, total He)	570.5	460.8	-
²⁷ Al(n, p) ²⁷ Mg	4.02	3.76	4.15
²⁷ Al(n, α) ²⁴ Na	0.74	0.72	0.66
⁴⁶ Ti(n, p) ⁴⁶ Sc	13.66	10.97	11.30
⁵⁹ Co(n, α) ⁵⁶ M	0.16	0.16	0.15
¹⁹⁷ Au(n, γ) ¹⁹⁸ Au*	88.59	86.65	82.80
²³⁵ U(n, f)F.P.	1243.0	1233.0	1230.0
²³⁸ U(n, f)F.P.*	291.70	276.8	287.0
²³⁹ Pu(n, f)F.P.	1812.0	1758.0	1762.0

(a) calculated this work

(b) from Ref. 2

*This isotope has been updated in the SAND-II Library. The new calculations will appear in the next report.

Hanford Engineering Development Laboratory

A. FISSION AND NON-FISSION FOIL MATERIAL ACQUISITION AND QUALITY ASSURANCE

L. S. Kellogg, W. N. McElroy, W. Y. Matsumoto, and
J. L. Jackson

Objectives

- Establish reliable and qualified sources of high purity foil materials required for the ILRR program and the Fast Reactor Materials Dosimetry Center (FRMDC) for routine fast and thermal reactor dosimetry measurements and stockpile selected batches of foil materials that meet Quality Assurance (QA) analytical and use tests.
- Perform QA characterization and evaluation of the materials maintained for dosimetry measurements and maintain archive samples.

Summary

High purity aluminum (stated 99.999%) for encapsulation of the fission foils to be fabricated by ORNL was obtained and a sample was sent to HEDL for QA analysis.⁽¹⁾ The preliminary QA work indicates that use of the aluminum in an LMFBR type spectral environment should present no particular difficulties in the direct gamma spectrometric measurement of the fission foil material for ^{140}Ba . Possible problems could be encountered, though, in highly thermalized neutron spectra due to material impurities such as Sc.

Chemical and isotopic analyses of the 1 and 5 mil ^{235}U metal foil used in CFRMF irradiations to date have confirmed previous metal purity and isotopic enrichment statements.

Accomplishments and Status

A pool of highly enriched fission foil material has been allocated by the AEC and is being held at ORNL for use by the ILRR program and for routine LMFBR dosimetry.

A 6.82" x 1.93" x 0.0074" piece of the aluminum to be used for encapsulation of those fission materials was obtained from ORNL. QA efforts on the aluminum are necessary to confirm the absence of impurities which could compromise the accuracy of counting the encapsulated fission materials.

Preliminary dissolution and density measurements were made to confirm the major composition as aluminum and the apparent thickness uniformity was measured and found to be .00741" $\pm$.00036".

Six representative 1/2" discs were prepared, packaged in Al foil wrap, and irradiated for seven hours (total fluence $\sim 3 \times 10^{15}$) in the CFRMF at a stated reactor power of 9.75 KW.⁽¹⁻³⁾

After allowing for decay of the ^{24}Na , the entire set of six was counted as a sample ~ 1 cm from a 3" x 3" NaI(Tl) detector face. Approximately one cpm/mg was observed for the energy region 0.02 to 4.0 MeV.

A second set of six foils was prepared and irradiated for a total of 16 hours (total fluence $\sim 8 \times 10^{17}$) in the NBS reactor RT-4 position (highly thermalized).⁽²⁾ These foils indicated a number of contaminating gamma activities. Those now identified are ^{46}Sc , ^{182}Ta , and ^{60}Co with ^{46}Sc the major contributor to the gamma spectra. Gamma energies of peaks noted but not identified at this time (in $\sim \text{keV}$) are: 54, 65, 99, 147, 316, 606, 324, 486, 135, and 1101. Preliminary calculations assuming $^{45}\text{Sc}(n,\gamma)^{46}\text{Sc}$ indicate an initial concentration of ^{45}Sc of ~ 0.1 ppm. The variation in impurity content (homogeneity) utilizing ^{46}Sc for the measurements was found to be $<3\%$ relative.

Analyses of the 1 and 5 mil ^{235}U metal foil materials utilized in the CFRMF irradiations to date (listed enrichments of 93.25 ^{235}U , and 93.10% ^{235}U , respectively) were made.^(2,3) The U metals, at least after surface etching as was done prior to preparation of the irradiation foils, were found to be 100% U metal and the ^{235}U content was found to be within 0.1% relative of the values listed above.

Expected Achievements

It is anticipated that QA on other foil materials being utilized in the ILRR program will have been initiated, particularly for fissionable materials. Details of the uranium metal QA analysis will also be included in the next quarterly report.

B. ^{140}Ba STANDARD FOR FISSION RATES BY RADIOCHEMISTRY

W. Y. Matsumoto, M. R. Weiler, and L. S. Kellogg

Objectives

- Establish an absolute reaction rate base standard for fission rate measurements and subsequent readjustment of evaluated energy dependent cross sections for fuels and materials dosimetry.^(1,2,3) (At present, most energy dependent cross sections are generally adjusted relative to some absolute reference cross section [$^{235}\text{U}(n,f)\text{F.P.}$, $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$, etc.] rather than to reaction rates.)
- Provide the necessary reference standard calibrated to within $\pm 2\%$ (95% confidence level) to allow radiochemical determinations of the absolute reaction rates within the goals of $\pm 5\%$ (95% confidence level) for fission reaction rates and 5 to 10% (95% confidence level) for integral flux-fluence values required for Fast Reactor Dosimetry and the ILRR program.

Summary

A ^{140}Ba - ^{140}La standard solution (T 913) was prepared with an estimated uncertainty of $\pm 1.6\%$ at the 95% confidence level by the HEDL Radiochemistry Standards Laboratory in March 1972. Weight aliquots (~ 5 grams each) of this standard in flame sealed glass ampoules were distributed to the following ILRR program participants: ANC (J. W. Rogers), ANL (R. J. Armani), ANL (N. D. Dudev), ANL (E. R. Ebersole), ARHCO (W. H. Zimmer), and HEDL (L. S. Kellogg). Two ampoules of the standard were shipped to the National Bureau of Standards (S. B. Garfinkel) for reference measurements in the NBS $4\pi\gamma$ ionization chambers. These measurements will be used to normalize the HEDL standard to a ^{140}Ba - ^{140}La standard to be prepared by the NBS at some future time.

A ^{140}Ba standard from the Amersham/Searle Corporation was checked at HEDL by 4π beta-gamma coincidence counting and calibrated [NaI(Tl)] gamma spectrometry. The HEDL results were 2.1% to 2.4% higher than the Amersham/Searle values but well within the overall limits of error of $+4.4\%$, -1.3% (99.7%

confidence limits) quoted by Amersham/Searle.

Accomplishments and Status

HEDL T 913 ^{140}Ba - ^{140}La Standard Solution

The ^{140}Ba - ^{140}La standard solution, T 913, was prepared and shipped to interested ILRR program participants by the HEDL Radiochemistry Standards Laboratory on March 7, 1972. Absolute standardization of this solution was made March 15, 1972, by 4π beta-gamma coincidence counting of four source mounts from each of four accurately prepared dilutions. The random statistical uncertainty for the sixteen measurements expressed as the relative standard error was .083%. The overall uncertainty of the measurement was estimated to be $\pm 1.6\%$ at the 95% confidence level. Two ampoules of the standard were shipped to the National Bureau of Standards, Radioactivity Section, for reference measurements in the NBS $4\pi\gamma$ ionization chambers.

The final standardization results and data pertinent to the standard were mailed to each participant on May 13, 1972, along with a request for the results from gamma spectrometry measurements made in their laboratories. The data sheet sent each participant is reproduced in Figure 1.

Interlaboratory Comparisons of HEDL T 913 ^{140}Ba - ^{140}La Standard

Data from the ILRR program participants who have reported their results from gamma spectrometric measurements on the HEDL T 913 ^{140}Ba - ^{140}La standard are summarized in Table 1. No formal attempt will be made to evaluate these data until all participants have had the opportunity to review and comment on the summarized results. Each participant is expected to report more detailed data on his measurements of the T 913 standard in his own section of the ILRR Program Quarterly Report. The data summarized in Table 1 have been normalized on a consistent basis (explained in the footnotes) for convenience in making preliminary and cursory interlaboratory comparisons.

Descriptions of the specific methods used by each laboratory to calibrate, for ^{140}Ba measurement, the gamma spectrometry systems used, are of particular interest.

All HEDL gamma spectrometry systems used were calibrated for ^{140}Ba (in transient equilibrium with ^{140}La) by direct calibration with previous ^{140}Ba - ^{140}La standards standardized by 4π β - γ coincidence counting by the HEDL

Ba/La-140 Radioactivity Standard

This standard consists of barium-140, in transient equilibrium with its lanthanum-140 daughter in $\pm .001$ grams of solution in a flame sealed glass ampoule.

The barium-140 plus lanthanum-140 total activity in nuclear transformations* per second (ntps) per gram of solution, as of 12:00 noon PST March 13, 1972 was:

$$\underline{2.43_8 \times 10^6 \pm 1.6\%}$$

comprised of $1.13_4 \times 10^6$ ntps Ba-140 plus $1.30_4 \times 10^6$ ntps La-140 per gram of solution.

The estimated uncertainty in the activity, 1.6 percent, is the sum of 0.2 percent, which is the standard error at the 95-percent confidence level computed from 16 sets of measurements, and 1.4 percent, which is the linear sum of the estimated systematic errors considered to be possible in the measurements.

This solution was standardized by 4 pi beta-gamma coincidence counting of sources prepared from accurately made dilutions of the solution. The standardization is based on the assumptions that barium-140 and its lanthanum-140 daughter both decay 100 percent by beta emission and that the majority of the beta particles emitted are accompanied by coincident gamma emission. A barium-140 half-life of 12.80 days and a lanthanum-140 half-life of 40.20 hours were used where required in calculations.

The chemical composition of the standard is as follows:

Solution matrix:	0.98N nitric acid
Carrier concentrations:	68 ppm Ba, 10 ppm La
Total solids:	330 ppm
Density:	1.0269 grams/ml at 23.9 °C

This standard was prepared and standardized by the Hanford Engineering Development Laboratory, Radiochemistry Section. The material from which the standard was prepared was purchased from the Oak Ridge National Laboratory and chemically purified at HEDL on February 15, 1972, to reduce a strontium-89 impurity. The 4 π β - γ coincidence counting measurements were performed on March 15, 1972.

*The actual aliquot number and weight (to the nearest 0.1 mg) for the specific aliquot shipped to the ILRR program participant were provided to each participant.

FIGURE HEDL-1 Reference Standard T 913

TABLE HEDL-1
INTERLABORATORY COMPARISON OF DATA FROM GAMMA SPECTROMETRIC MEASUREMENTS ON
HEDL T 913 BARIUM/LANTHANUM-140 STANDARD

<u>Lab./Investigator</u>	<u>^{140}Ba Half-Life Used</u>	<u>Date(s) of Counting</u>	<u>Detector Type and System Identification</u>	<u>1596 keV γ's Per ^{140}Ba Dis. (1)</u>	<u>Meas. ^{140}Ba (2) HEDL Calib.</u>
ANC/J. Rogers	12.79 d.	3/22/72	NaI(Tl) Std. 3" x 3" (A)	1.0972	1.0044
ANC/R. Hellmer	"	3/24-3/27	Ge(Li) ND-3	"	1.0132
ANC/R. Greenwood	"	5/10/72	Ge(Li) OR-5	"	.9982
" "	"	5/11-5/19	NaI(Tl) Std. 3" x 3" (R)	1.0988 ⁽³⁾	.9974
ANL/N. Dudey	12.80 d.	3/22/72	Ge(Li) 25 cc coax	1.1098	.9885
ANL/E. Ebersole	12.80 d.	3/16-3/21	Ge(Li) North. Scient. Anal.	1.1103	.9718
" "	"	3/16-3/21	NaI(Tl) 3" x 3" N.D.-180	"	.9594
ARHCO/W. Zimmer	12.80 d.	3/10/72	Ge(Li) 20 cc "A" System	1.1068	1.0115 ⁽⁴⁾
" "	"	3/10/72	Ge(Li) 35 cc "C" System	"	1.0119 ⁽⁴⁾
HEDL/M. Weiler	12.80 d.	3/16/72	NaI(Tl) 3" x 3" Std. RIDL	1.1103	.9979
HEDL/L. Kellogg	12.80 d.	3/17-3/22	Ge(Li) 40 cc VDM 622 i	1.1106	1.0071
" "	"	3/17-4/20	Ge(Li) 48 cc Pack. 4096	"	.9859
" "	"	3/18-3/30	NaI(Tl) 3" x 3" Std. RIDL	"	.9912

(1) Calculated from ^{140}Ba and ^{140}La half-lives and 1596 keV γ /(^{140}La Dis.) reported by participants assuming transient equilibrium conditions.

(2) Ratio of reported measured ^{140}Ba d/s/g to HEDL standardization value of $1.13_4 \times 10^6$ at 12:00 PST 3/13/72.

(3) Includes .0014, 1521 keV γ / ^{140}La Dis.

(4) Reported values from volume aliquots converted to a weight basis using a solution density of 1.0269 g/ml.

Radiochemistry Standards Laboratory. Calibrations are normally maintained in units of disintegrations/count (d/c) for each geometry on each detector. For ^{140}Ba - ^{140}La , the units are in ^{140}Ba dis./count in the 1596 keV gamma photopeak from ^{140}La . Therefore, the gamma branching ratio does not directly enter into the HEDL gamma spectrometry system calibrations for ^{140}Ba . For the 3" x 3" NaI(Tl) detector a calibration based on the composite peak from the 537 keV and 487 keV gammas from ^{140}Ba and ^{140}La , respectively, is also used, particularly for checking consistency between calibrations. The composite peak from ^{140}La gammas around 2520 keV is also used for some counting done using NaI(Tl) detectors. Details not given in Table 1 on HEDL gamma spectrometric measurements of the T 913 ^{140}Ba - ^{140}La standard are summarized in Table 2.

TABLE HEDL-2

HEDL GAMMA SPECTROMETRIC MEASUREMENT RESULTS
ON HEDL T 913 BARIUM/LANTHANUM-140 STANDARD

<u>Dates of Counting</u>	<u>Detector Type and System Identification</u>	<u>^{140}Ba d/s/g at 12:00 hrs PST, 3/13/72</u>	<u>Number of Measurements</u>
3/16/72	NaI(Tl) 3 x 3 Std. RIDL	$(1.1316 \pm .0025) \times 10^6$	19 ⁽²⁾
3/17-3/22	Ge(Li) 40 cc VDM 622i	$(1.142 \pm .006) \times 10^6$	3
3/17-4/20	Ge(Li) 48 cc Pack. 4096	$(1.118 \pm .0045) \times 10^6$	6
3/18-3/30	NaI(Tl) 3 x 3 Std. RIDL	$(1.124 \pm .0035) \times 10^6$	8 ⁽³⁾

(1) Uncertainty limits given are 1 standard error: $\bar{s} = s/\sqrt{n}$

(2) 1596 keV and composite peak from 537 and 487 keV gammas were used.

(3) 1596 keV and composite peak at ~2520 keV were used.

HEDL Checks of ^{140}Ba Standard from Amersham/Searle Corporation

Two ampoules (S2/12/6 and S2/12/7) of a ^{140}Ba standard solution (Item No. 454248, Code: BBZ 72) standardized by the Amersham/Searle Corporation by 4π beta counting at a reference time of 1200 hours GMT (04:00 hours PST), March 3, 1972, were checked at HEDL on March 15 and 16, 1972, by 4π beta-gamma coincidence counting and gamma spectrometry using a calibrated 3" x 3" NaI(Tl)

detector. The HEDL results by 4π beta-gamma coincidence counting, corrected for radioactive decay using a 12.80 day half-life for ^{140}Ba , were 2.4% and 2.1% higher than the Amersham/Searle values for the two ampoules S2/12/6 and S2/12/7, respectively. The HEDL result by gamma spectrometry for ampoule S2/12/6 was 2.3% higher than the Amersham/Searle value. All of these results were within the overall limits of error of +4.4%, -1.3% (99.7% confidence limits) quoted by Amersham/Searle.

Expected Achievements

Analysis and evaluation of the interlaboratory comparison data on the HEDL T 913 ^{140}Ba - ^{140}La standard will be completed. A consistent set of nuclear parameters (half-lives, gamma energies, and gammas emitted per ^{140}Ba disintegration) for the ^{140}Ba - ^{140}La transient equilibrium system will be established with the cooperation of all participants based on intensive work and literature studies to be reported by ANC.

A second ^{140}Ba - ^{140}La standard solution will be prepared by HEDL for distribution to interested ILRR program participants in late July or early August 1972. This distribution will be timed to coincide as closely as practical to the continuation of irradiation tests in the CFRMF. An aliquot of this standard will be sent to the NBS for reference measurements in the NBS $4\pi\gamma$ ionization chambers.

C. EXPERIMENTAL FISSION YIELDS FOR ^{137}Cs AND ^{148}Nd RELATIVE TO ^{140}Ba FROM NEUTRON DOSIMETRY FISSION FOIL IRRADIATIONS IN EBR-II

W. Y. Matsumoto and W. N. McElroy

Objectives

- Establish fission yields for ^{137}Cs and ^{148}Nd relative to ^{140}Ba to within $\pm 2\%$ (95% confidence limits) for ^{235}U , ^{238}U , ^{239}Pu , and ^{237}Np fissions in well characterized neutron environments closely simulating EBR-II and expected FTR neutron spectra.
- Establish absolute fission yields for ^{137}Cs and ^{148}Nd for ^{235}U , ^{238}U , ^{239}Pu , and ^{237}Np in well characterized LMFBR neutron spectra.
- Define and quantify the relationships between the neutron spectra and the fission yields established.

Summary

Fission foils of ^{235}U , ^{238}U , ^{239}Pu , and ^{237}Np are scheduled to be irradiated in various fast reactor irradiation experiments in order to characterize the neutron spectra and fluence for these irradiations. The fission reaction rates are typically established by measuring the fission product ^{140}Ba by direct gamma spectrometry. For some experiments it is also possible to measure ^{137}Cs (radio-metrically) and ^{148}Nd (by isotope dilution mass spectrometry) and thus obtain relative fission yields for these nuclides at minimum cost by taking advantage of foil irradiations already scheduled.

A few preliminary measurements that establish the feasibility of this approach have been made at HEDL with fission foils irradiated in EBR-II.

Accomplishments and Status

Five neutron dosimetry fission foil samples irradiated during EBR-II Run 50H and one fission foil irradiated in the EBR-II irradiated X-135 experimental subassembly were destructively analyzed for ^{137}Cs and ^{148}Nd after ^{140}Ba measurements had been made by direct gamma spectrometry. The apparent fission yields

for ^{137}Cs and ^{148}Nd relative to the base fission yields taken for ^{140}Ba ⁽²⁾ are given in Table 3. Since these values are the results of initial attempts at analysis of this particular type of sample, the probable uncertainty limits may be fairly large. Additional measurements will provide a statistical basis for establishing uncertainty limits as well as to provide practice and experience needed to achieve improved accuracy. Attempts to perform analyses on the two ^{239}Pu foils--irradiated as vanadium encapsulated PuO_2 -- have not yet been successful because of problems encountered in obtaining complete dissolution of the sample.

The data on relative fission yield results obtained from these tests will complement and augment the data from extensive experiments by ANL.^(4,5) Obtaining the fission yield results from fission foils irradiated in fast reactor neutron spectra that will concurrently be characterized by multiple neutron dosimeter foil measurements and SAND-II determinations has a number of inherent benefits such as providing fine definition on neutron energy dependent fission yields.

The apparently low values (compared to current literature values) for the ^{148}Nd fission yields determined for ^{235}U fissions and by inference for ^{237}Np fissions cannot be explained on the basis of currently available data. Although the total quantity of ^{148}Nd available for analysis in any of the foils was significantly less than 0.1 microgram, the fission yield values obtained for two ^{238}U samples are in reasonably good agreement with published data.^(2,6) Additional data are needed before meaningful evaluations can be made. Results from ^{239}Pu foils, when the dissolution problem is resolved, should help in the resolution of anomalous data.

Expected Achievements

A method for dissolution of irradiated PuO_2 foils will be developed so that ^{137}Cs and ^{148}Nd fission yields relative to ^{140}Ba can be measured. Measurements on additional foils will provide a statistical basis for uncertainty estimates and provide practice and experience required to improve the precision and accuracy obtainable.

TABLE HEDL-3

PRELIMINARY FISSION YIELDS FOR CS-137 AND ND-148
FROM ANALYSIS OF NEUTRON DOSIMETRY FOILS IRRADIATED
IN EBR-II AND BASED ON ASSUMED BA-140 FISSION YIELDS GIVEN

<u>Fission Reaction</u>	<u>Sample ID</u>	<u>^{140}Ba ^(a)</u>	<u>^{137}Cs ^(b)</u>	<u>^{148}Nd ^(c)</u>
$^{235}\text{U}(\text{n},\text{f})\text{F.P.}$	W54	$5.8 \pm .2$	$5.93 (6.2 \pm .1)^{(e)}$	$1.57 (1.75 \pm .03)^{(e)}$
"	W60	$5.8 \pm .2$	5.98	1.52
"	W60	$5.8 \pm .2$	5.97*	--
"	UAU ^(d)	$5.8 \pm .2$	6.35	1.66
$^{237}\text{Np}(\text{n},\text{f})\text{F.P.}$	W54	$5.4 \pm .2$	$6.04 (6.0 \pm .5)^{(e)}$	1.58
$^{238}\text{U}(\text{n},\text{f})\text{F.P.}$	W54	$6.0 \pm .2$	$6.13 (6.3 \pm .3)^{(e)}$	$2.13 (1.93 \pm .1)^{(e)}$
"	W62	$6.0 \pm .2$	6.18	2.11
"	W62	$6.0 \pm .2$	6.48*	

(a) Ba-140 fission yields taken from Reference (2) used as the basis for calculating Cs-137 and Nd-148 fission yields.

(b) Cs-137 determined by radiochemical separation and comparison to Nuclear Chicago/ASTM Cs-137 Standard by gamma spectrometry--[NaI(Tl)] well crystal.

*Determined by direct high resolution gamma spectrometry before dissolution of sample.

(c) Nd-148 determined by isotope dilution mass spectrometry.

(d) Fission foil from X-135 subassembly irradiated during the period 8/27/71 through 1/14/72. Since fissions based on ^{140}Ba relied heavily on irradiation time history calculations the uncertainty of this data is significantly greater than that for the other foils which were all irradiated in EBR-II Run 50 H.

(e) Values in current use by FRMDC, ANL, and ILRR program, from Table HEDL-5 of reference 2.

D. ABSOLUTE GAMMA-RAY COUNTING

L. S. Kellogg and W. Y. Matsumoto

Objective

- Obtain radiochemical measurements of fission and non-fission foil reaction rates for the CFRMF and other ILRR tests with a desired measurement accuracy of $\pm 2\%$ (1σ) for fission foils and $\pm 2\%$ to $\pm 5\%$ (1σ) for non-fission foils.

Summary

Previous ILRR Quarterly Reports^(2,3) provide information on the techniques used at HEDL for gamma assay of irradiated foils. Summary type information is provided here which briefly discusses the techniques used at HEDL and unifies past and present information. Specifically, system calibration, capsule absorption and self-shielding corrections, and reference to previous discussions of gamma energies and branching intensities, half lives, and overall estimated calibration accuracy are considered.

Accomplishments and Status

To achieve the desired absolute accuracy entails not only a rigorous evaluation of routine analytical procedures but also consideration of individual problems that may be encountered due to variations encountered in samples prepared for the individual tests involved.

Systems Calibration

Both Ge(Li) and NaI(Tl) detector systems are utilized at this time, dependent upon activity level and purity of gamma spectra. The high resolution of the Ge(Li) detectors is invaluable for complex sample gamma matrices; but because of the low efficiency of Ge(Li) compared to that of NaI(Tl), prolonged counting periods may be required in order to achieve the desired counting statistics. This can be intolerable where multitudes of samples require analysis.

(a) Both the NaI(Tl) and Ge(Li) detector systems are calibrated using radioactive standards that are, for the most part, calibrated in-house. An

example of such a standard would be the ^{140}Ba - ^{140}La Standard prepared for the CFRMF test, section HEDL-B of this report. Comparison is made to plots of efficiency vs. energy prepared from previously run standards (both calibrated in-house), to sources such as NBS and IAEA reference standards, and where realistically possible, to theoretical calculations. These plots are updated as required. Actual isotope sample calculation, where possible, is made with reference to the respective standard rather than the curves to reduce possible error due to decay scheme uncertainty.

(b) Resolution degradation and counting loss have been studied using the "two-source technique." Two different fixed sources were used-- ^{137}Cs and either ^{124}Sb or $^{140}\text{Ba/La}$, and ^{57}Co and ^{60}Co as variable sources. From these data, a system reference deadtime meter reading has been fixed to reduce counting loss to <1%.

(c) Sample configuration corrections may be required because of sample thickness due to the increased sample centroid position in relation to the detector surface and that of the calibration standards. Surface area corrections, though, have been essentially compensated for by calibrations with spread "zero thickness" standards.

Special Considerations

(a) Absorption Corrections - In the case of encapsulated foils or particular situations where use of an external absorber is desired, correction for gamma attenuation is made by use of the equation:

$$I = I_0 \exp [-\mu(E)X]$$

where I and I_0 are the observed and actual gamma-ray intensities, respectively, $\mu(E)$ is the mass absorption coefficient,⁽⁷⁾ and X is the absorber thickness.

(b) Self Absorption - Because of the relatively thick samples required in the ILRR irradiations, corrections are required for self absorption. The detector-to-sample distances are such that the first order approximation is being used:

$$I = I_0 \left(\frac{1 - \exp [-\mu(E)X]}{\mu(E)} \right). \quad (8,9)$$

Where wire samples are utilized, an initial correction for average sample thickness, as compared to a uniform slab, is made.

(c) Uncertainties in decay scheme, gamma energies and their related branching intensities, and half life (where reference standards are unavailable) are also considered. The 3rd Quarterly Report,⁽³⁾ Table HEDL-9, lists gamma energies, branching intensities, and half lives in current use by HEDL.

(d) Overall estimated calibration accuracy at the 95% confidence level for individual isotopes is provided in the 3rd Quarterly Report, Table HEDL-9.

Expected Achievements

More detailed information on methods used and corrections made for conversion of observed gamma-ray activities to absolute reaction rates and an evaluation of the contributing errors will be presented in subsequent reports.

E. CFRMF REACTION RATES BY RADIOCHEMISTRY

W. N. McElroy, L. S. Kellogg, and W. Y. Matsumoto

Objective

● Obtain radiochemical measurements of fission and non-fission foil reaction rates for the CFRMF and other ILRR tests; use the results of these and other absolute measurement techniques to improve the accuracy of radiochemically determined reaction rates (and concurrently fission yields) for EBR-II, future FTR, and other fuels and materials irradiation tests. [The accuracy goals are $\pm 5\%$ (95% confidence level) for ^{235}U , ^{239}Pu , and ^{238}U fission rates and ± 5 to 10% (95% confidence level) for other fission and non-fission reactions.] Establish accepted sets of values of absolute reaction rates for different well established and permanent standard neutron fields for readjustment of evaluated energy dependent cross sections in current use by the FRMDC.

Summary

Results are documented of high power CFRMF tests related to run-to-run power level normalization for the ANC-I Non-Fission foil set, AI-I natural and enriched boron sample irradiations, and HEDL re-counts of ANC and ANL foils.

Accomplishments and Status

Results of HEDL's radiochemistry measurements for the high-power ANC-I Non-Fission foil set and AI-I natural and enriched boron sample irradiations are documented here. Further reactor operations information on these tests and the ANC reactor power level instrumentation and normalization foil activation measurements are given in the ANC chapter of this Quarterly Report. The 3rd Quarterly Report⁽³⁾ provides detailed information on the contents of the ANC-I and AI-I foil packages and placement within the duplicate NBS fission chamber for these two irradiations.

The $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$, $^{115}\text{In}(n,n')^{115\text{m}}\text{In}$, and $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$ measured reaction rates are summarized in Table 4. Reference is made to the discussion of the figures and tables of the 3rd Quarterly Report to complete the documentation

TABLE HEDL-4
SUMMARY OF HEDL MEASURED $^{115}\text{In}(n,n')^{115m}\text{In}$
 $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$, AND $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$ REACTION RATES

Foil Type	Date	Test	Foil No.	Foil Thickness (mils)		10^{15} x Reactions Per Sec.-Nuclei	Ratio ^b	Reactor Power (KW)	Remarks
				Total	Actual				
Au	2/15/72	ANL-I	3	1.05	0.5	52.49 [†]			
Au	2/15/72	ANL-I	4	1.05	0.5	52.09 [†]			
Au	2/29/72	ANC-I	AN	2	2	48.54	Norm. Point	9.104	In norm. foil holder.
Au	2/29/72	ANC-I	2	1.05	0.5	52.79 [†] (3.479) [†]	0.9803	9.104	In NBS chamber.
Au	2/29/72	ANC-I	1	1.05	0.05	55.66 [†] (3.668) [†]	1.034	9.104	In NBS chamber.
Au	2/29/72	ANC-I	1	1.05	0.5	52.74 [†] (3.476) [†]	0.9794	9.104	In NBS chamber.
Au	4/10-11/72	AI-I	AP	2	2	^d	Norm. Point	-	In norm. foil holder.
Au	4/10-11/72	AI-I	2	1.0	0.5	52.14 (-)		-	In top of foil holder. ^c
Au	4/10-11/72	AI-I	1	1.0	0.5	52.29 (-)		-	In bottom of foil holder. ^c
In	4/10-11/72	AI-I	16	2	2	6.859 (0.4438)	Norm. Point	9.273	In norm. foil holder.
In	4/10-11/72	AI-I	2	4	2	6.988 (0.4522)	1.019	9.273	In top of foil holder. ^c
In	4/10-11/72	AI-I	1	4	2	6.987 (0.4521)	1.019	9.273	In bottom of foil holder. ^c
Al	4/10-11/72	AI-I	2	10	5				In top of foil holder. ^c
Al	4/10-11/72	AI-I	1	10	5	0.02028 (± 1.4) ^e		9.093 ^f	In bottom of foil holder. ^c
Au	From Table HEDL-4 of 3rd Quarterly					3.199	1.000	0.600	Reference point.
In	From Table HEDL-3 of 3rd Quarterly					0.4438	1.000	0.600	Reference point.

- a. The In and Au results given here are an extension of results reported previously in Tables HEDL-3 and 4 of the 3rd Quarterly Report. Reference is made to these previous tables for applicable footnotes.
- b. For Au and In only, ratio to values of 3.549×10^{-15} and 4.438×10^{-16} , respectively, taken from the 3rd Quarterly Report as the infinitely dilute reaction rates for the ARHCO-I run for Au and the average value for the low-power run III for In.
- c. The placement order of these foils in the foil set holder (see Figure HEDL-3, Figure AI-1, and Tables AI-1 and 2 of the 3rd Quarterly Report) is from top to bottom: Al-2, Au-2, In-2, AI (B1, Boron specimens), In-1, Au-1, and Al-1, respectively.
- d. Foil was not counted; consequently, ANC result must be used, see ANC Table
- e. Estimated 2 σ percent counting precision.
- f. Relative to HEDL-I values of 0.01996×10^{-15} for aluminum and an estimated power of 8.95 KW - see HEDL chapter of 3rd Quarterly Report.⁽³⁾
- † These foils were counted at HEDL after ANL and ANC had completed their measurements. The estimated relative 1 standard deviation uncertainties (including half life error) for these five samples are $\pm 1.43\%$, $\pm 1.45\%$, $\pm 0.91\%$, $\pm 1.22\%$, and $\pm 0.92\%$, respectively. The total uncertainty for these five samples may actually be higher than stated here due to prolonged decay before counting (~ 22 days for ANL and ~ 17 days for ANC) and possible trace immunity activities not observed.

for these new measurements. The gold and indium results are an extension of results given in Tables HEDL-3 and 4 of the previous Quarterly Report.⁽³⁾

Expected Achievements

Further CFRMF radiochemistry foil irradiations are not anticipated before July 1972. HEDL foil sets II, III, and IV have yet to be irradiated. Work will continue on the analysis of available test data and the definition of special tests to study particular effects, such as foil scattering and self-shielding effects. These tests must be completed prior to developing a final test plan for the CFRMF radiochemistry foil irradiations of the high purity aluminum encapsulated fission foils fabricated by ORNL.

F. SHAPE AND SIZE CORRECTIONS FOR SMALL SAMPLE REACTION RATES

R. B. Kidman, W. N. McElroy, and L. S. Kellogg

Objectives

- In support of ILRR program experimental studies, develop and apply appropriate theoretical methods for correcting measured small sample (dosimetry foils) reaction rates to infinitely dilute values.
- Apply developed methods to EBR-II and FTR fuels and materials dosimetry studies.

Summary

The present report shows the effect of geometric and resonance shielding on the gold capture reaction rate for the CFRMF spectrum and provides theoretical support for recent experimental observations.

Accomplishments and Status

The reaction rate in a small sample is affected by a host of unknown and known effects which must be identified and evaluated before one can properly use measured small sample reaction rates to significantly improve such things as cross sections, flux spectra, dosimetry parameters, burnup results, etc.

The ENDF/B Version III gold data were used to produce infinitely dilute 43-group cross sections* and a set of foil factors for various gold foil thicknesses. The foil factors, through multiplication, correct the calculated infinitely dilute cross sections for the geometric and resonance shielding occurring in a thin gold foil of infinite extent. The foil factors in the unresolved region of gold were found by extrapolation from the calculated values in the resolved region. Finally, the present best estimate for the CFRMF spectrum⁽¹⁾ was used to calculate the gold capture reaction rate for various

* With 1/E weighting.

gold foil thicknesses. These rather idealized results are shown in Figure 2 and are compared with preliminary experimental results from the last Quarterly Report.

For this comparison, previously reported results (Table HEDL-4 of the 3rd Quarterly Report)⁽³⁾ were corrected for gamma-ray attenuation within the different foils (maximum correction of 1%), flux gradients in the CFRMF central core region (maximum correction of 1.1%), and foil edge effects (maximum correction of 0.8%). The approximation used for correcting for the last effect is $(1 + t/\pi r)$, which is independent of the material and the neutron spectrum, where t is the foil thickness and r is its radius.⁽¹⁰⁾ In Figure 2, the corrected experimental values (which apply for thin gold foils of infinite extent) are listed next to the plotted data points for future reference. All values have been normalized to the measured reaction rate for the 0.135% Au-Al alloy foil (20 mil wire wrap), which is assumed to represent the infinitely dilute value.* The experimental results are in reasonable agreement with the shape of the theoretical curve, but are slightly higher in magnitude. The 2 to 5% difference may be the result of both theoretical and experimental uncertainties. Theoretically, weighting of the ENDF/B data with a $1/E$ spectrum rather than the CFRMF spectrum is a source of some uncertainty as well as differences between applied theoretical and actual gold foil densities. Uncertainties in the reference 0.135% Au-Al alloy composition, run-to-run power level normalization, and the definition of the true CFRMF flux-spectrum (including any low energy neutron streaming effects) are sources of experimental errors. This last source of error can have a significant effect on the theoretical predictions.

Additional information is provided by A. Fabry in Section HEDL-H on foil self-shielding effects, but based on studies in the $\Sigma\Sigma$ facility. Fabry's previous and current $\Sigma\Sigma$ studies provided necessary technical justification for the present study.

Expected Achievements

Plans are being formulated for additional measurements in CFRMF which will eliminate many of the present experimental uncertainties. Particular attention will be given to the definition of required corrections for the 1/2" diameter by 5-mil thick encapsulated fission foils being fabricated by ORNL for the ILRR program irradiations.

* The alloy, 0.05, and 0.5 mil foil results are based on the high power ARHCO-I test, all other results are based on low power tests.

FIGURE HEDL-2 Relative Gold Capture Reaction Rate (Capture Reactions per Second per Nuclei) as a Function of Foil Thickness. Normalized to 1 for an Infinitely Thin Foil.

ORNL is preparing five sets of 1/2" diameter foils of ^{235}U , ^{238}U , Sc, Co, and Cu of the following thicknesses: 0.00005", 0.0001", 0.0005", 0.001", 0.002", 0.005", 0.010", and 0.020". These foils, together with Au foils on hand, will be available for use in CFRMF, $\Sigma\Sigma$, and other facilities to experimentally establish relative reaction rate versus foil thickness curves for subsequent comparison with theoretical estimates and correction of measured small sample reaction rates to infinitely dilute values for ILRR program studies.

Effects of such things as Doppler broadening, angular flux distribution, in-foil scattering, and flux depression outside the foil require theoretical study. Probably, however, calculations will first be extended to account for a sample's exact shape and size.

G. REACTION CROSS SECTIONS

W. N. McElroy, R. L. Simons, and L. S. Kellogg

Objective

● Through the National Neutron Cross Section Center (NNCSC) and the Cross Section Evaluation Working Group (CSEWG), establish and maintain an updated set of evaluated energy dependent cross sections for foil reactions used by the Fast Reactor Materials Dosimetry Center (FRMDC) and other laboratories for routine neutron dosimetry. Establish better consistency and accuracy for these cross sections in order to achieve the long range objective of 5 to 10% accuracy (at the 95% confidence level) for measured integral flux spectra and fluence values for LMFBR fuels and materials development programs.

Summary

A Task Group has been established through the Normalization and Standards Subcommittee of CSEWG to develop an "ENDF/B File for Dosimetry Applications." Results of the first meetings of this group are briefly described.

Comparison of Meadows' and Smith's⁽¹¹⁾ recent energy dependent measurements of the $^{58}\text{Ni}(n,p)^{58}\text{Co}$ cross section with those of Paulsen and Widera⁽¹²⁾ and those derived on the basis of SAND-II analysis of integral data shows agreement in the 1 to 4 MeV energy region not previously observed for a threshold type dosimetry foil reaction. This type of agreement is a necessary requirement for cross sections added to the ENDF/B file and used for fuels and materials dosimetry.

Accomplishments and Status

SAND-II determination of flux-spectra and fluence from a set of measured reaction rates requires the use of a consistent set of evaluated energy dependent cross sections $\sigma(E)$.⁽¹³⁾

A reasonably consistent set of $\sigma(E)$ values has been developed and is in current use for the SAND-II code,⁽¹⁴⁾ which is used by HEDL for routine fast reactor dosimetry and will be used to provide multiple foil flux spectral data

for the ILRR program.⁽¹³⁾ Better consistency and accuracy for these cross sections are required. Present accuracy is in the 20 to 40% range at the 95% confidence level.⁽¹²⁻¹⁷⁾ This accuracy can be improved if the sources of existing inconsistencies between calculated integral averaged cross sections $\bar{\sigma}_c$ using the current evaluated energy dependent cross sections $\sigma(E)$ and measured values $\bar{\sigma}_m$ can be identified and effectively eliminated - see previous Quarterly Reports.⁽¹⁻³⁾

ENDF/B File for Dosimetry Applications

A Task Group has been established through the Normalization and Standards Subcommittee (NSS) of CSEWG to develop an "ENDF/B File for Dosimetry Applications." The chairman (B. Leonard of BNW) of this subcommittee held a meeting at BNW in late March 1972 to consider the CSEWG Phase I effort of establishing an initial set of ~20 evaluated cross sections for this data file.⁽¹⁸⁾ Members of the Task Group were asked to document their work and a number of completed reports on initial evaluations were available for the meeting.

A limited number of the members of the Dosimetry Task Group met in conjunction with the regular NSS meetings at the May 9-10, 1972, CSEWG meeting at BNL. Revised and new reports on individual evaluations were distributed and/or reviewed at this meeting.⁽¹⁹⁻³³⁾ Computer plots and output for several reactions which will be added to the ENDF/B File were distributed by BNL. The BNL chapter of this Quarterly provides some information on this subject. Other participants of the ILRR program are encouraged to include summary information on completed evaluations in their contributions to future ILRR Quarterly Reports.

The chairman (H. Alter of AI) of the Data Testing Subcommittee of CSEWG requested specification and selection of a number of benchmarks for Phase II integral testing of the ENDF/B Dosimetry Data File. Documentation was provided in the HEDL chapter of the 3rd Quarterly Report⁽³⁾ on two suggested benchmarks, the fission spectrum and the Mol, Belgium, $\Sigma\Sigma$ facility, for which suitable integral reaction rate and flux spectral data are available. ILRR program integral measurements will be required for these two neutron fields, however, to establish the same absolute base for these integral data as for the CFRMF spectrum.

Neutron fields that have been previously documented and/or discussed in ILRR Quarterly Reports and that should be acceptable as benchmarks are: NBS fission spectra (^{235}U , ^{239}Pu , ^{252}Cf), LASL critical assemblies (Flattops and Big-10), Godiva, Mol $\Sigma\Sigma$, and CFRMF. The ANL chapter of this report provides information on some accelerator derived neutron fields that could also be developed as benchmarks.

$^{58}\text{Ni}(n,p)^{58}\text{Co}$ Cross Section

The SAND-II evaluated energy dependent cross section for $^{58}\text{Ni}(n,p)^{58}\text{Co}$ was compared with Paulsen's and Widera's⁽¹²⁾ mono-energetic results in the last Quarterly Report.⁽³⁾ Recent unpublished mono-energetic measurements of Meadows and Smith (11) are compared with these previous results in Figure 3. The data points of Meadows' and Smith's are connected by straight lines to show the sequence of their data. The agreement between the SAND-II integral results (the histogram) and both sets of mono-energetic measurements is excellent for the region of definition for the integral method, below ~ 4 MeV. Such good agreement between integral and differential data has not been observed before for a threshold type dosimetry foil reaction, but is a necessary requirement in the establishment of acceptable evaluated cross sections for the ENDF/B File for Dosimetry Applications.

Through studies and comparisons of the type presented here and future help and direction from NNCSC, as well as other participants of the ILRR program, the FRMDC hopes to establish a "best" set of evaluated energy dependent reaction cross sections to improve the accuracy of fast reactor dosimetry for fuels and materials problems. The success of this effort will depend on a proper marriage and understanding of existing microscopic data, integral data, theoretical data, and the associated errors. Significantly improved experimental integral data ($\bar{\sigma}_m$ values) are expected from the CFRMF test and other tests of the ILRR program.

Expected Achievements

For the "ENDF/B File for Dosimetry Applications," HEDL will provide initial evaluations for several reactions, three of which are $^{58}\text{Ni}(n,p)^{58}\text{Co}$, $^{54}\text{Fe}(n,p)^{54}\text{Mn}$, and $^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$. HEDL will also provide assistance in making theoretical calculations for the $^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$ reaction, which is being evaluated by A. Fabry.

FIGURE HEDL-3 Comparison of Measured and SAND-II Evaluated Cross Sections for $^{58}\text{Ni}(n,p)^{58}\text{Co}$

H. $\Sigma\Sigma$ INFORMATION RELEVANT TO THE ILRR PROGRAM

A. Fabry^a

Objective

- Document the continuing experimental and theoretical work on the CEN-SCK Secondary Intermediate-Energy Standard Neutron Spectrum Facility $\Sigma\Sigma$ for maintaining an operative link between the Interlaboratory LMFBR Reaction Rate program and the joint NBS/CEN-SCK efforts to develop and/or characterize permanent intermediate-energy standard and reference neutron fields for fast reactor applications.

Summary

A brief review is presented of progress in the study and use of the $\Sigma\Sigma$ facility which is felt particularly relevant to the ILRR program.

Measurements of self-shielding for gold in $\Sigma\Sigma$ are reported which provide additional support for recent observations in CFRMF.

Accomplishments and Status

Fission Rate Measurements

The first measurement of the ^{238}U and ^{235}U fission rates and ratios in $\Sigma\Sigma$ by means of the NBS fission chamber⁽³⁴⁾ has been prepared. A new boron carbide shell⁽³⁵⁾ with an access hole of 5 mm diameter and a corresponding plug for the uranium source shell have been implemented. The measurements will thus be performed using three different shells with access holes of diameters 5, 17 and 27 mm, respectively, and they will be compared to previous solid state track recorder (SSTR) data⁽³⁵⁾ and to measurements by means of Karlsruhe⁽³⁶⁾ and Saclay⁽³⁷⁾ fission chambers.

SSTR irradiations have indicated that intermediate-energy neutrons re-enter the central volume by an access hole of 17 mm diameter and then spread out by

^a. On leave from Reactor Study Department, CEN-SCK, Mol, Belgium, and attached to the Center for Radiation Research, NBS.

collisions with the inner walls to produce a spatially uniform increase of the ^{235}U fission rate by $(8 \pm 0.5)\%$. This measured figure will be reduced by the use of the 5 mm diameter access hole; this effect does not influence the interlaboratory comparison of the various ^{235}U fission rate measurements with each other, but it must be considered as a correction when comparing the data with theory or other integral measurements. Note that no such effect is observed nor expected for ^{238}U fission.

One of the NBS fission chambers has been loaded with calibrated ^{238}U and ^{235}U deposits and the stem was shortened so that the manifold could stay within the 50 cm diameter cavity. Duplicates of this chamber are under construction at CEN-SCK for future systematic applications. A dummy chamber will also be provided for exposure of SSTR and other detectors under physical conditions similar to the ones used in CFRMF.

Gold Reaction Rate Distribution and Self-Shielding in $\Sigma\Sigma$

For various practical reasons, alloy foil detectors as used extensively in $\Sigma\Sigma$ to measure infinitely dilute absolute reaction rates⁽³⁸⁾ are not generally applied in routine neutron dosimetry in fast reactor environments. A capability must therefore be developed to assess corrections for neutron self-shielding within foils of finite thickness in fast reactor spectra.

The reliability of such corrections heavily depends, not only on the availability of suitable theoretical models, but also on the accuracy of nuclear data, i.e., resolved and unresolved resonance parameters, the last ones being especially important for fission reactions without a threshold. Last, but not least, the low energy tail of the neutron spectra in such environments must be adequately known (which is also of relevance to fast reactor safety problems).

Usually, advantage is taken of the self-shielding effect in well-chosen activation foils to experimentally investigate the neutron flux spectrum below 10 keV; this is the well-known sandwich foil technique.^(10,39,40) In many practical neutron dosimetry applications in fast reactor spectra, if dilute alloy foils cannot be used, an iterative interpretation scheme must be considered whenever the neutron spectrum is poorly known; this might be the SAND-II unfolding of a first spectral approximation from multiple foil data, which would be used for computing or specializing self-shielding corrections, and so on until convergence of the procedure.

In order to develop the necessary interpretation capability, reference experimental data are first of all needed. For such purpose, $\Sigma\Sigma$ is an ideal facility because it is simple and clean, with a reasonably uniform and isotropic central neutron flux spectrum currently characterized by sustained interlaboratory work, both experimental and theoretical.⁽³⁸⁾ In terms of spectral shape, it is a useful complement to CFRMF because its low energy tail below a few hundred eV is expected to be significantly softer.

A first self-shielding measurement has been performed for the $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ reaction. Gold foils of 10 mm diameter with thicknesses of 1.5, 8.2, 49, 90, and 232 mg/cm² have been irradiated at the center of the standard $\Sigma\Sigma$ configuration. Two irradiations were performed with three foils in each, at positions +2, 0, and -2 cm on the centerline perpendicular to the axis of the thermal column. The most thin foils, 1.5 mg/cm², were CBNM (Euratom, Geel) 5% Au-Al alloy and one such foil was exposed in each irradiation. The other foils were pure metallic gold. Redundant and precise monitoring was provided as usual.⁽³⁸⁾

The high quality of the CBNM alloys had been previously checked, as well as the fact that they effectively measure infinitely dilute reaction rates in $\Sigma\Sigma$;⁽³⁸⁾ this is illustrated here by Figure 4, which displays a radial traverse along the aforementioned centerline in the $\Sigma\Sigma$ central exposure volume, as measured with 1% Au-Al and 5% Au-Al CBNM alloys. This figure is not only given for completeness, but also because it points out another detail which might be significant for CFRMF; i.e., the central peaking of the low energy radial neutron fluxes within a spherical (or cylindrical*) "void" in an absorber embedded in a medium where the radial flux distribution would otherwise be flat; this neutron transport effect is well reproduced in discrete-ordinates S8 calculations⁽³⁵⁾ and is more marked the greater is the sensitivity of detectors to lower energy neutrons, for instance, the $^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$ reaction in $\Sigma\Sigma$.**

The flux gradient corrections applied in the present self-shielding experiment do not exceed 1.6%.

* Similar trends have been found inside of a boron carbide cylindrical shell exposed in the central horizontal channel of BRI reactor.

** The radial distribution of the fission rates in ^{235}U and ^{238}U along the same centerline is flat.

FIGURE HEDL-4 Gold Radial Reaction Rate Distribution in $\Sigma\Sigma$

FIGURE HEDL-5 Gold Self Shielding in Two Fast Reactor Neutron Spectra

The ^{198}Au activities of all foils have been measured at two distances from a calibrated 3" x 3" NaI(Tl) gamma-ray spectrometer. Most often, the count rate was smaller than 100 cps and it never exceeded 300 cps. Usual corrections such as background, dead time losses, radioactive decay, foil weight, and irradiation profile were applied and their uncertainties, when combined to statistical counting spreads, do not encompass errors greater than ± 0.3 - 0.4% . The four independent monitors for these runs indicated a flux level difference of $(1.3 \pm 0.5)\%$ between the two irradiations.

Corrections for edge effects⁽¹⁰⁾ have also been introduced to derive self-shielding factors for idealized plane detectors of infinite radius; the greatest correction of this type was 0.8%.

Finally, the self absorption of gamma-rays in the foils has been accounted for by means of a phenomenological expression systematically established previously.^(41,42) The maximum self absorption correction at the closest foil - NaI crystal distance reached about 5%, but is not uncertain by more than $\pm 0.5\%$.

The overall accuracy of each experimental point is estimated as $\pm 1.5\%$ or better. The results are gathered in Table 5 and are compared in Figure 5 with CFRMF data, taken from Section F of this report. The trends and orders of magnitude involved are similar, which supports firmly the interpretation of available CFRMF data in terms of self-shielding effects; depending on the accuracy of the available data, the differences observed provide quantitative information on the extent to which the low energy tail in $\Sigma\Sigma$ is softer than in CFRMF.

A theoretical appraisal of these measurements for both $\Sigma\Sigma$ and CFRMF is being investigated at CEN-SCK by J. C. Schepers and will be compared with the calculations of Kidman et al. of this report (see Figure HEDL-2).

It is believed useful to stress here, that for a total self-shielding effect of the order of 15%, the reaction rate difference between the outer and inner foils in an equivalent sandwich of three identical detectors may typically not exceed 2-3% (but is much less sensitive to nuclear data uncertainties, although being of a comparable sensitivity to spectral differences); this closing remark is intended as a warning against the danger to conclude that no self-shielding corrections are needed in situations where no spatial variation of reaction rate is observed within a stack of foils; often also, the bulk of

TABLE HEDL-5
GOLD SELF-SHIELDING FACTORS IN $\Sigma\Sigma$

<u>Gold Thickness (mg/cm²)</u>	<u>Self-Shielding^a Correction Factor</u>
0.3 ^b	1.00 ($\pm 0.5\%$)
1.5 ^b	0.995 ($\pm 1\%$)
8.2	0.979 ($\pm 1\%$)
49	0.917 ($\pm 1.5\%$)
90	0.889 ($\pm 1\%$)
232	0.833 ($\pm 1.5\%$)

- ^a. Corrected for edge effects; i.e., for plane detector foils of infinite radius.
- ^b. CBNM gold/aluminum alloys.

such variations occurs only within the first outer layer of these foils and is hard to observe when the self-shielding is small, but not negligible.

New Transport Calculations of $\Sigma\Sigma$ on the Basis of ENDF/B Versions II and III Nuclear Data

These calculations are in progress at ORNL⁽⁴³⁾ by means of the SUPERTOG-XSDRN code system, in the GAM-II 100-group structure and the S16 P3 approximation. They will be documented in a future ORNL section of the ILRR Quarterly Report.

Expected Achievements

It is anticipated that the first measurement of the ²³⁸U and ²³⁵U fission rates and ratios in $\Sigma\Sigma$ by means of the NBS fission chamber will be documented in the next Quarterly and the results compared with all other previous or new fission rate data.

It is expected that the recently evaluated cross sections for activation detectors developed under the direction of the CSEWG Normalization and Standards Subcommittee will be combined with all or part of the theoretical and experimental spectral information on $\Sigma\Sigma$ for confrontation with the available integral data.

It is expected that a preliminary self-shielding measurement for the $^{235}\text{U}(n,f)$ reaction will be completed for the next Quarterly, together with a first attempt to determine the ^{140}Ba - ^{140}La fission yield in $\Sigma\Sigma$ for ^{235}U fission; it is anticipated that for the final measurements of these data, a set of the high quality foils specially prepared by ORNL for the ILRR program will be made available.

I. CALCULATED REACTION RATES FOR SEVERAL LASL CRITICAL ASSEMBLIES

W. N. McElroy

Objective

- Provide calculated fission and non-fission foil reaction rates per unit flux for the central positions of the Big-10 (U-reflected, 90% ^{238}U - 10% ^{235}U) and Flattop (U-reflected, ^{235}U , ^{233}U , and ^{239}Pu) critical assemblies as background information for the planning and selection of standard neutron fields for future ILRR program study.

Summary

The spectra provided by LASL for Big-10 and several Flattop critical assemblies in the 3rd ILRR Quarterly Report⁽³⁾ were used with the SAND-II code to calculate fission and non-fission foil reaction rates of interest for future ILRR program study. The calculated spectra for the LASL assemblies are compared with the CFRMF spectrum.

Accomplishments and Status

The calculated integral flux values given by Hansen in the LASL chapter of the 3rd ILRR Program Quarterly Report⁽³⁾ were used to generate a set of group integral fluxes and fluxes per unit lethargy, Table 6, for Big-10 and three Flattop assemblies. The spectra for three of these assemblies are compared in Figure 6 to the CFRMF spectrum. Because of its similarity to the Flattop ^{239}Pu spectrum, the Flattop ^{233}U spectrum is not shown in the figure.

Time-of-flight and multiple foil central core spectra for a bare ^{235}U (Godiva) assembly gave a measured spectrum ($\bar{E} = 1.57 \text{ MeV}$) that was $\sim 8\%$ ($\sim 0.1 \text{ MeV}$) harder than that calculated.⁽⁴⁴⁾ The calculated spectrum mean energies of Table 6, therefore, might also be subject to a similar discrepancy if they were to be compared with measurements.

The spectra of Figure 6 and Table 6 were used as input to the SAND-II code to calculate reaction rates (saturated foil activities per unit flux) for

TABLE HEDL-6
ESTIMATED CENTRAL FLUX SPECTRA FOR THE
BIG-10 AND FLATTOP CRITICAL ASSEMBLIES

Group Lethargy Δu	Group Lower Bound E (MeV)	Group Fluxes and Fluxes Per Unit Lethargy ⁽¹⁾							
		Big-10		Ft- ²³⁵ U		Ft- ²³³ U		Ft- ²³⁹ Pu	
		$\Delta\phi$	$\frac{\Delta\phi}{\Delta u}$	$\Delta\phi$	$\frac{\Delta\phi}{\Delta u}$	$\Delta\phi$	$\frac{\Delta\phi}{\Delta u}$	$\Delta\phi$	$\frac{\Delta\phi}{\Delta u}$
0.223	8	0.0009	0.0040	0.0030	0.014	0.0037	0.017	0.0047	0.021
0.288	6	0.0031	0.011	0.011	0.038	0.0133	0.046	0.015	0.053
0.405	4	0.011	0.027	0.045	0.111	0.058	0.143	0.059	0.146
0.288	3	0.014	0.049	0.059	0.205	0.073	0.253	0.076	0.264
0.405	2	0.028	0.069	0.113	0.279	0.137	0.338	0.131	0.323
0.357	1.4	0.028	0.078	0.108	0.303	0.125	0.350	0.120	0.336
0.442	0.9	0.060	0.136	0.140	0.317	0.149	0.337	0.148	0.335
0.811	0.4	0.255	0.314	0.251	0.309	0.226	0.279	0.218	0.269
0.693	0.2	0.250	0.361	0.170	0.245	0.125	0.180	0.133	0.192
0.693	0.1	0.178	0.257	0.064	0.092	0.061	0.0880	0.060	0.087
0.693	0.05	0.110	0.159	0.026	0.038	0.021	0.030	0.025	0.036
1.61	0.01	0.060	0.037	0.010	0.0062	0.008	0.0050	0.010	0.0062
4.61	0.0001	0.002	0.00043	0	0	0	0	0	0
Spectrum Mean ⁽²⁾ Energy $\bar{E}$ (MeV)		0.573		1.36		1.56		1.57	

1. Normalized such that: $\sum_{i=1}^n \Delta\phi_i = 1.$

2. Defined as: $\bar{E} \equiv \int_0^{\infty} E \phi(E) dE / \int \phi(E) dE$

where $\phi(E)$ is the neutron differential flux.

FIGURE HEDL-6 Comparison of the CFRMF and LASL Critical Assemblies Neutron Spectra

foil reactions of current interest to the participants of the ILRR program.

Results of these calculations for the reactions of primary concern (^{235}U , ^{238}U , ^{239}Pu , and ^{237}Np) as well as others of both short- and long-term interest are given in Tables 7 to 10. These reaction rates per unit flux can be used with the fluence data given in the LASL chapter of the Second Quarterly (HEDL-TME 71-79) to provide estimates of anticipated foil activities for the consideration of foils and foil sizes suitable for track recorder, radiochemistry, and helium analysis. The HEDL chapter of the 1st ILRR Quarterly Report lists similar results for the CFRMF spectrum.⁽¹⁾

Expected Achievements

The information provided here and in the LASL chapter of this report completes the preliminary documentation for the LASL standard neutron fields of current interest for future ILRR program study.

TABLE HEDL-7
SAND-II CALCULATED SATURATED ACTIVITIES FOR THE BIG-10

Foil Reaction	Calculated Saturated Activity* (Dps or Fissions per Sec/Nucleus)	Nominal 5% Activity Limits (MeV)	
		Lower	Upper
Primary Standards			
$^{235}\text{U}(n,f)\text{F.P.}$	1.434 -24	1.700 -02	1.900 +00
$^{238}\text{U}(n,f)\text{F.P.}$	4.424 -26	1.300 +00	6.800 +00
$^{238}\text{U}(n,\gamma)^{239}\text{U}$	1.716 -25	1.050 -02	1.000 +00
$^{237}\text{Np}(n,f)\text{F.P.}$	4.193 -25	3.800 -01	4.200 +00
$^{239}\text{Pu}(n,f)\text{F.P.}$	1.605 -24	3.200 -02	2.400 +00
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	3.061 -25	8.000 -03	7.200 -01
$^{10}\text{B}(n,\alpha)^7\text{Li}$	1.263 -24	8.400 -03	7.200 -01
$^6\text{Li}(n,\alpha)^3\text{H}$	9.846 -25	2.200 -02	7.600 -01
$^{115}\text{In}(n,n')^{115\text{m}}\text{In}$	3.334 -26	7.200 -01	5.600 +00
$^{32}\text{S}(n,p)^{32}\text{P}$	8.443 -27	2.400 +00	7.700 +00
$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	1.060 -28	6.500 +00	1.210 +01
$^{127}\text{I}(n,2n)^{126}\text{I}$	1.223 -28	1.010 +01	1.490 +01
Reactions for Dosimetry			
$^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$	3.259 -26	5.750 -03	8.000 -01
$^{59}\text{Co}(n,\gamma)^{60}\text{Co}$	2.001 -26	4.250 -03	6.600 -01
$^{45}\text{Sc}(n,\gamma)^{46}\text{Sc}$	1.975 -26	8.800 -03	7.600 -01
$^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$	4.769 -27	1.050 -02	1.100 +00
$^{109}\text{Ag}(n,\gamma)^{110\text{m}}\text{Ag}$	2.953 -26	1.150 -02	8.400 -01
$^{181}\text{Ta}(n,\gamma)^{182}\text{Ta}$	2.970 -25	6.900 -03	8.000 -01
$^{54}\text{Fe}(n,p)^{54}\text{Mn}$	1.071 -26	2.200 +00	8.000 +00
$^{58}\text{Ni}(n,p)^{58}\text{Co}$	1.443 -26	2.100 +00	7.700 +00
$^{46}\text{Ti}(n,p)^{46}\text{Sc}$	1.588 -27	3.400 +00	9.400 +00
$^{47}\text{Ti}(n,p)^{47}\text{Sc}$	2.447 -27	2.000 +00	7.200 +00
$^{48}\text{Ti}(n,p)^{48}\text{Sc}$	3.834 -29	6.700 +00	1.320 +01
$^{55}\text{Mn}(n,2n)^{54}\text{Mn}$	4.833 -29	1.090 +01	1.560 +01
$^{63}\text{Cu}(n,\alpha)^{60}\text{Co}$	7.455 -29	3.200 +00	1.120 +01
$^{238}\text{Pu}(n,f)\text{F.P.}$	1.416 -24	6.900 -02	3.300 +00
$^{232}\text{Th}(n,f)\text{F.P.}$	1.081 -26	1.400 +00	7.200 +00
Additional Reactions for Dosimetry and Helium Production			
$^{115}\text{In}(n,\gamma)^{116\text{m}}\text{In}$	2.527 -25	1.150 -02	1.200 +00
$^{23}\text{Na}(n,\gamma)^{24}\text{Na}$	6.580 -28	2.800 -03	8.400 -01
$^{31}\text{P}(n,p)^{31}\text{Si}$	4.636 -27	2.200 +00	7.200 +00
$^{56}\text{Fe}(n,p)^{56}\text{Mn}$	1.678 -28	5.500 +00	1.130 +01
$^{27}\text{Al}(n,p)^{27}\text{Mg}$	5.521 -28	3.500 +00	9.500 +00
$^{63}\text{Cu}(n,2n)^{62}\text{Cu}$	1.703 -29	1.190 +01	1.660 +01
$^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$	1.157 -26	8.800 -03	7.200 -01
$^{24}\text{Mg}(n,p)^{24}\text{Na}$	2.393 -28	6.600 +00	1.170 +01
$^{59}\text{Co}(n,\alpha)^{56}\text{Mn}$	2.314 -29	5.900 +00	1.210 +01
$^{14}\text{N}(n,\alpha)^{11}\text{B}$	1.234 -26	1.700 +00	6.100 +00
$^{28}\text{Si}(n,\alpha)$	6.379 -28	6.100 +00	1.030 +01
$^{32}\text{S}(n,\alpha)$	8.090 -27	1.900 +00	6.500 +00
$^{12}\text{C}(n,\alpha)$	1.015 -28	7.100 +00	1.220 +01
$^{16}\text{O}(n,\alpha)$	1.015 -27	4.000 +00	8.400 +00
$^{23}\text{Na}(n,\alpha)$	6.172 -29	6.500 +00	1.210 +01
$\text{Cr}(n,\alpha)$	4.330 -29	5.400 +00	1.180 +01
$\text{Fe}(n,\alpha)$	3.547 -29	5.300 +00	1.180 +01
$\text{Ni}(n,\alpha)$	3.912 -29	5.500 +00	1.100 +01

* Per $1 \text{ n/cm}^2\text{-sec}$; note: these values are also the spectral averaged cross sections for the Big-10 spectrum.

TABLE HEDL-8
SAND-II CALCULATED SATURATED ACTIVITIES FOR THE FLATTOP ^{235}U

Foil Reaction	Calculated Saturated Activity* (Dps or Fissions per Sec/Nucleus)	Nominal 5% Activity Limits (MeV)	
		Lower	Upper
<u>Primary Standards</u>			
$^{235}\text{U}(n,f)\text{F.P.}$	1.268 -24	8.400 -02	4.100 +00
$^{238}\text{U}(n,f)\text{F.P.}$	1.726 -25	1.400 +00	6.600 +00
$^{238}\text{U}(n,\gamma)^{239}\text{U}$	1.141 -25	4.500 -02	2.200 +00
$^{237}\text{Np}(n,f)\text{F.P.}$	9.528 -25	5.500 -01	5.000 +00
$^{239}\text{Pu}(n,f)\text{F.P.}$	1.692 -24	1.200 -01	4.300 +00
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	1.583 -25	3.800 -02	1.800 +00
$^{10}\text{B}(n,\alpha)^7\text{Li}$	6.370 -25	3.400 -02	3.000 +00
$^6\text{Li}(n,\alpha)^3\text{H}$	6.380 -25	8.400 -02	2.100 +00
$^{115}\text{In}(n,n')^{115\text{m}}\text{In}$	1.160 -25	1.000 +00	5.600 +00
$^{32}\text{S}(n,p)^{32}\text{P}$	3.411 -26	2.400 +00	7.400 +00
$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	3.616 -28	6.400 +00	1.210 +01
$^{127}\text{I}(n,2n)^{126}\text{I}$	4.089 -28	1.010 +01	1.490 +01
<u>Reactions for Dosimetry</u>			
$^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$	1.488 -26	2.100 -02	2.800 +00
$^{59}\text{Co}(n,\gamma)^{60}\text{Co}$	8.978 -27	3.200 -02	1.600 +00
$^{45}\text{Sc}(n,\gamma)^{46}\text{Sc}$	9.977 -27	3.200 -02	1.900 +00
$^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$	3.662 -27	8.400 -02	2.400 +00
$^{109}\text{Ag}(n,\gamma)^{110\text{m}}\text{Ag}$	1.630 -26	4.000 -02	3.000 +00
$^{181}\text{Ta}(n,\gamma)^{182}\text{Ta}$	1.600 -25	3.800 -02	1.800 +00
$^{54}\text{Fe}(n,p)^{54}\text{Mn}$	4.266 -26	2.200 +00	7.800 +00
$^{58}\text{Ni}(n,p)^{58}\text{Co}$	5.776 -26	2.100 +00	7.500 +00
$^{46}\text{Ti}(n,p)^{46}\text{Sc}$	6.191 -27	3.400 +00	9.100 +00
$^{47}\text{Ti}(n,p)^{47}\text{Sc}$	9.874 -27	2.000 +00	6.900 +00
$^{48}\text{Ti}(n,p)^{48}\text{Sc}$	1.301 -28	6.600 +00	1.310 +01
$^{55}\text{Mn}(n,2n)^{54}\text{Mn}$	1.615 -28	1.090 +01	1.560 +01
$^{63}\text{Cu}(n,\alpha)^{60}\text{Co}$	2.581 -28	3.500 +00	1.110 +01
$^{238}\text{Pu}(n,f)\text{F.P.}$	2.100 -24	2.400 -01	4.700 +00
$^{232}\text{Th}(n,f)\text{F.P.}$	4.257 -26	1.500 +00	7.100 +00
<u>Additional Reactions for Dosimetry and Helium Production</u>			
$^{115}\text{In}(n,\gamma)^{116\text{m}}\text{In}$	1.754 -25	4.750 -02	2.500 +00
$^{23}\text{Na}(n,\gamma)^{24}\text{Na}$	3.784 -28	4.500 -02	2.900 +00
$^{31}\text{P}(n,p)^{31}\text{Si}$	1.881 -26	2.200 +00	7.000 +00
$^{56}\text{Fe}(n,p)^{56}\text{Mn}$	5.901 -28	5.400 +00	1.120 +01
$^{27}\text{Al}(n,p)^{27}\text{Mg}$	2.103 -27	3.400 +00	9.300 +00
$^{63}\text{Cu}(n,2n)^{62}\text{Cu}$	5.691 -29	1.190 +01	1.660 +01
$^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$	5.395 -27	3.000 -02	2.500 +00
$^{24}\text{Mg}(n,p)^{24}\text{Na}$	8.171 -28	6.600 +00	1.170 +01
$^{59}\text{Co}(n,\alpha)^{56}\text{Mn}$	7.985 -29	5.800 +00	1.210 +01
$^{14}\text{N}(n,\alpha)^{11}\text{B}$	5.017 -26	1.800 +00	5.900 +00
$^{28}\text{Si}(n,\alpha)$	2.202 -27	6.100 +00	1.020 +01
$^{32}\text{S}(n,\alpha)$	3.294 -26	1.900 +00	6.300 +00
$^{12}\text{C}(n,\alpha)$	3.430 -28	7.100 +00	1.220 +01
$^{16}\text{O}(n,\alpha)$	4.016 -27	4.000 +00	8.100 +00
$^{23}\text{Na}(n,\alpha)$	2.101 -28	6.400 +00	1.210 +01
$\text{Cr}(n,\alpha)$	1.513 -28	5.300 +00	1.170 +01
$\text{Fe}(n,\alpha)$	1.236 -28	5.200 +00	1.180 +01
$\text{Ni}(n,\alpha)$	1.379 -28	5.500 +00	1.090 +01

* Per $1 \text{ n/cm}^2\text{-sec}$; note: these values are also the spectral averaged cross sections for the Flattop ^{235}U spectrum.

TABLE HEDL-9
SAND-II CALCULATED SATURATED ACTIVITIES FOR THE FLATTOP ^{239}Pu

Foil Reaction	Calculated Saturated Activity* (Dps or Fissions per Sec/Nucleus)	Nominal 5% Activity Limits (MeV)	
		Lower	Upper
Primary Standards			
$^{235}\text{U}(n,f)\text{F.P.}$	1.264 -24	8.400 -02	4.600 +00
$^{238}\text{U}(n,f)\text{F.P.}$	2.085 -25	1.500 +00	6.900 +00
$^{238}\text{U}(n,\gamma)^{239}\text{U}$	1.067 -25	4.250 -02	2.400 +00
$^{237}\text{Np}(n,f)\text{F.P.}$	1.049 -24	6.000 -01	5.400 +00
$^{239}\text{Pu}(n,f)\text{F.P.}$	1.712 -24	1.275 -01	4.700 +00
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	1.429 -25	3.800 -02	2.000 +00
$^{10}\text{B}(n,\alpha)^7\text{Li}$	6.025 -25	3.400 -02	3.600 +00
$^6\text{Li}(n,\alpha)^3\text{H}$	5.586 -25	7.600 -02	2.500 +00
$^{115}\text{In}(n,n')^{115\text{m}}\text{In}$	1.374 -25	1.100 +00	5.900 +00
$^{32}\text{S}(n,p)^{32}\text{P}$	4.426 -26	2.500 +00	7.600 +00
$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	5.428 -28	6.500 +00	1.220 +01
$^{127}\text{I}(n,2n)^{126}\text{I}$	6.410 -28	1.010 +01	1.490 +01
Reactions for Dosimetry			
$^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$	1.401 -26	2.000 -02	3.300 +00
$^{59}\text{Co}(n,\gamma)^{60}\text{Co}$	8.130 -27	3.000 -02	1.800 +00
$^{45}\text{Sc}(n,\gamma)^{46}\text{Sc}$	9.104 -27	3.200 -02	2.300 +00
$^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$	3.399 -27	8.000 -02	2.700 +00
$^{109}\text{Ag}(n,\gamma)^{110\text{m}}\text{Ag}$	1.532 -26	4.000 -02	3.500 +00
$^{181}\text{Ta}(n,\gamma)^{182}\text{Ta}$	1.457 -25	3.600 -02	2.000 +00
$^{54}\text{Fe}(n,p)^{54}\text{Mn}$	5.576 -26	2.300 +00	8.000 +00
$^{58}\text{Ni}(n,p)^{58}\text{Co}$	7.438 -26	2.100 +00	7.700 +00
$^{46}\text{Ti}(n,p)^{46}\text{Sc}$	8.371 -27	3.400 +00	9.400 +00
$^{47}\text{Ti}(n,p)^{47}\text{Sc}$	1.244 -26	2.100 +00	7.200 +00
$^{48}\text{Ti}(n,p)^{48}\text{Sc}$	1.974 -28	6.700 +00	1.320 +01
$^{55}\text{Mn}(n,2n)^{54}\text{Mn}$	2.532 -28	1.090 +01	1.560 +01
$^{63}\text{Cu}(n,\alpha)^{60}\text{Co}$	3.750 -28	4.000 +00	1.120 +01
$^{238}\text{Pu}(n,f)\text{F.P.}$	2.207 -24	2.550 -01	5.100 +00
$^{232}\text{Th}(n,f)\text{F.P.}$	5.193 -26	1.500 +00	7.300 +00
Additional Reactions for Dosimetry and Helium Production			
$^{115}\text{In}(n,\gamma)^{116\text{m}}\text{In}$	1.684 -25	4.750 -02	2.800 +00
$^{23}\text{Na}(n,\gamma)^{24}\text{Na}$	3.511 -28	4.250 -02	3.500 +00
$^{31}\text{P}(n,p)^{31}\text{Si}$	2.391 -26	2.200 +00	7.200 +00
$^{56}\text{Fe}(n,p)^{56}\text{Mn}$	8.539 -28	5.500 +00	1.130 +01
$^{27}\text{Al}(n,p)^{27}\text{Mg}$	2.879 -27	3.500 +00	9.500 +00
$^{63}\text{Cu}(n,2n)^{62}\text{Cu}$	8.920 -29	1.190 +01	1.660 +01
$^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$	5.058 -27	3.000 -02	3.000 +00
$^{24}\text{Mg}(n,p)^{24}\text{Na}$	1.219 -27	6.600 +00	1.180 +01
$^{59}\text{Co}(n,\alpha)^{56}\text{Mn}$	1.183 -28	5.800 +00	1.220 +01
$^{14}\text{N}(n,\alpha)^{11}\text{B}$	6.282 -26	1.800 +00	6.100 +00
$^{28}\text{Si}(n,\alpha)$	3.233 -27	6.100 +00	1.030 +01
$^{32}\text{S}(n,\alpha)$	4.116 -26	1.900 +00	6.500 +00
$^{12}\text{C}(n,\alpha)$	5.219 -28	7.100 +00	1.220 +01
$^{16}\text{O}(n,\alpha)$	5.374 -27	4.000 +00	8.400 +00
$^{23}\text{Na}(n,\alpha)$	3.164 -28	6.500 +00	1.210 +01
$\text{Cr}(n,\alpha)$	2.224 -28	5.300 +00	1.180 +01
$\text{Fe}(n,\alpha)$	1.825 -28	5.300 +00	1.190 +01
$\text{Ni}(n,\alpha)$	1.992 -28	5.500 +00	1.100 +01

* Per 1 n/cm²-sec; note: these values are also the spectral averaged cross sections for the Flattop ^{239}Pu spectrum.

TABLE HEDL-10
SAND-II CALCULATED SATURATED ACTIVITIES FOR THE FLATTOP ^{233}U

Foil Reaction	Calculated Saturated Activity* (Dps or Fissions per Sec/Nucleus)	Nominal 5% Activity Limits (MeV)	
		Lower	Upper
Primary Standards			
$^{235}\text{U}(n,f)\text{F.P.}$	1.258 -24	1.000 -01	4.500 +00
$^{238}\text{U}(n,f)\text{F.P.}$	2.090 -25	1.500 +00	6.600 +00
$^{238}\text{U}(n,\gamma)^{239}\text{U}$	1.056 -25	5.000 -02	2.500 +00
$^{237}\text{Np}(n,f)\text{F.P.}$	1.063 -24	6.000 -01	5.200 +00
$^{239}\text{Pu}(n,f)\text{F.P.}$	1.714 -24	1.425 -01	4.600 +00
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	1.391 -25	4.250 -02	2.000 +00
$^{10}\text{B}(n,\alpha)^7\text{Li}$	5.836 -25	3.800 -02	3.600 +00
$^6\text{Li}(n,\alpha)^3\text{H}$	5.436 -25	9.200 -02	2.500 +00
$^{115}\text{In}(n,n')^{115\text{m}}\text{In}$	1.384 -25	1.100 +00	5.700 +00
$^{32}\text{S}(n,p)^{32}\text{P}$	4.269 -26	2.400 +00	7.400 +00
$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	4.423 -28	6.400 +00	1.210 +01
$^{127}\text{I}(n,2n)^{126}\text{I}$	5.048 -28	1.010 +01	1.490 +01
Reactions for Dosimetry			
$^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$	1.363 -26	2.700 -02	3.200 +00
$^{59}\text{Co}(n,\gamma)^{60}\text{Co}$	7.852 -27	3.600 -02	1.900 +00
$^{45}\text{Sc}(n,\gamma)^{46}\text{Sc}$	8.833 -27	3.600 -02	2.300 +00
$^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$	3.395 -27	9.600 -02	2.700 +00
$^{109}\text{Ag}(n,\gamma)^{110\text{m}}\text{Ag}$	1.490 -26	4.500 -02	3.500 +00
$^{181}\text{Ta}(n,\gamma)^{182}\text{Ta}$	1.422 -25	4.000 -02	2.000 +00
$^{54}\text{Fe}(n,p)^{54}\text{Mn}$	5.338 -26	2.200 +00	7.700 +00
$^{58}\text{Ni}(n,p)^{58}\text{Co}$	7.201 -26	2.100 +00	7.400 +00
$^{46}\text{Ti}(n,p)^{46}\text{Sc}$	7.788 -27	3.400 +00	9.100 +00
$^{47}\text{Ti}(n,p)^{47}\text{Sc}$	1.223 -26	2.100 +00	6.900 +00
$^{48}\text{Ti}(n,p)^{48}\text{Sc}$	1.595 -28	6.600 +00	1.310 +01
$^{55}\text{Mn}(n,2n)^{54}\text{Mn}$	1.994 -28	1.090 +01	1.560 +01
$^{63}\text{Cu}(n,\alpha)^{60}\text{Co}$	3.157 -28	3.600 +00	1.110 +01
$^{238}\text{Pu}(n,f)\text{F.P.}$	2.227 -24	2.700 -01	4.900 +00
$^{232}\text{Th}(n,f)\text{F.P.}$	5.170 -26	1.500 +00	7.100 +00
Additional Reactions for Dosimetry and Helium Production			
$^{115}\text{In}(n,\gamma)^{116\text{m}}\text{In}$	1.669 -25	5.250 -02	2.700 +00
$^{23}\text{Na}(n,\gamma)^{24}\text{Na}$	3.447 -28	5.000 -02	3.400 +00
$^{31}\text{p}(n,p)^{31}\text{Si}$	2.339 -26	2.200 +00	6.900 +00
$^{56}\text{Fe}(n,p)^{56}\text{Mn}$	7.241 -28	5.400 +00	1.120 +01
$^{27}\text{Al}(n,p)^{27}\text{Mg}$	2.630 -27	3.500 +00	9.300 +00
$^{63}\text{Cu}(n,2n)^{62}\text{Cu}$	7.026 -29	1.190 +01	1.660 +01
$^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$	4.883 -27	3.400 -02	3.000 +00
$^{24}\text{Mg}(n,p)^{24}\text{Na}$	9.969 -28	6.600 +00	1.170 +01
$^{59}\text{Co}(n,\alpha)^{56}\text{Mn}$	9.786 -29	5.800 +00	1.210 +01
$^{14}\text{N}(n,\alpha)^{11}\text{B}$	6.199 -26	1.800 +00	5.900 +00
$^{28}\text{Si}(n,\alpha)$	2.685 -27	6.100 +00	1.020 +01
$^{32}\text{S}(n,\alpha)$	4.069 -26	1.900 +00	6.300 +00
$^{12}\text{C}(n,\alpha)$	4.199 -28	7.100 +00	1.220 +01
$^{16}\text{O}(n,\alpha)$	5.097 -27	4.000 +00	8.100 +00
$^{23}\text{Na}(n,\alpha)$	2.571 -28	6.400 +00	1.210 +01
$\text{Cr}(n,\alpha)$	1.863 -28	5.200 +00	1.170 +01
$\text{Fe}(n,\alpha)$	1.523 -28	5.200 +00	1.180 +01
$\text{Ni}(n,\alpha)$	1.693 -28	5.500 +00	1.090 +01

* Per 1 n/cm²-sec; note: these values are also the spectral averaged cross sections for the Flattop ^{233}U spectrum.

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Los Alamos Scientific Laboratory

A. DETECTOR ACCESS CHANNELS IN THE FLATTOP AND BIG-10 ASSEMBLIES

G. E. Hansen

Objective

- To state present size limitations on foils and fission chambers that can be exposed to central fluences ($\sim 10^{12}$ fissions per gram of core per bombardment).

Summary

Diameter limitations on foils and chambers are 0.50" for the ^{233}U -core and Pu-core Flattops, 1.00" for the ^{235}U Flattop, and 1.50" for Big-10. Fission chamber stem lengths and diameters must be ≥ 10 " long and $\leq 1/8$ " o.d. for Flattop and ≥ 20 " long and $\leq 3/16$ " o.d. for Big-10. (A shorter stem length can be accommodated in Big-10 if the preamplifier fits into the 1.50" diameter glory hole.)

Accomplishments and Status

Not relevant.

Expected Achievements

Not relevant.

National Bureau of Standards

A. INTRINSIC SCATTERING CORRECTIONS FOR ACTIVATION DETECTORS IN CFRMF

C. Eisenhauer

Objective

- Estimate corrections for foil activation detectors in CFRMF due to neutrons scattered in the detector foil materials and in the enclosure surrounding them.

Summary

The NBS INTRAN Code which computes disk-detector responses to neutrons scattered from nearby materials is operational. Options available in the code include disk and cylinder scatterers (with or without anisotropic emission), and thin and thick detector responses. A simple formulation of the energy dependent part of the correction is complete and three-group energy transfer cross sections for this formulation have been prepared for aluminum, carbon, and fluorine.

Accomplishments and Status

As stated in the above summary.

Expected Achievements

Estimates of the scattering correction for fission detectors are to be made available before the primary fission foil irradiations at CFRMF are completed.

Oak Ridge National Laboratory

A. DOSIMETER SAMPLE PREPARATION AND QUALITY ASSURANCE FOR ILRR, FRMDC, AND ANL DOSIMETRY PROGRAMS

E. H. Kobisk and F. R. O'Donnell

Objective

• Preparation and accurate characterization of neutron dosimetry samples containing high purity, isotopically enriched, fissile and stable materials will be performed by the Isotope Target Laboratory of the ORNL Isotopes Division. Archive samples and inventories of each isotopically enriched material will be maintained (as allocated by the USAEC Division of Research) and dispensed exclusively in support of these various programs.

Summary

A 100-g batch of ^{239}Pu -1% Al metal alloy has been chemically converted to oxide and the aluminum content removed. The oxide will be used in the future preparation of needle-type flux monitors. All metal disk dosimeters of ^{235}U , ^{238}U , and ^{239}Pu have been produced, encapsulated in high purity aluminum, and tested for leakage. Delivery of these 60 samples has been made to W. N. McElroy at HEDL. Fabrication of ^{237}Np disks of the same form is now in progress and these samples will be shipped in June 1972. Preparation of an archive sample of each oxide and metal actinide isotope being used in the ILRR Dosimetry Program is now in progress.

Accomplishments and Status

Foils of ^{239}Pu , ^{235}U , and ^{238}U metals having a diameter of 0.5 in. and a thickness of ~ 0.005 in. were produced by rolling metal from the reserved batches of material 453BM, U-235-264CM, and U-ES-ZM, respectively. About 30 disks of each material were precision punched from the rolled sheet. Careful removal of surface oxide by etching and handling of cleaned samples in an inert atmosphere assured little or no oxide impurity. It should be

noted that each batch of metal was pre-treated before rolling by multiple arc melting with subsequent etchings to remove occluded oxide and slag.

Encapsulation of the sample disks was performed using 0.007-in. thick disks of pure aluminum metal sheet (99.999%). Closure technique was that of pressure cold welding the aluminum on the periphery using a sufficient pressure to yield a 50% reduction in thickness of the aluminum sheet. Encapsulated samples were individually leak tested by the ethylene glycol vacuum test method and then were stored for seven days. No radioactive contamination was found on the capsules after storage. Shipment of twenty dosimeters of each isotope was made by secondary encapsulation of the samples in a rigid brass container which prevented distortion of the critical sample dimensions in transit.

At present, similar samples to those described above, but fabricated from cast ^{237}Np (batch NP-21) metal, are being prepared. The highly brittle nature of neptunium metal precludes rolling of the 0.005-in. thickness. Disks of ~ 0.006 -in. thickness have been cast from molten neptunium and will be etched to size. Encapsulation in aluminum by cold-welding will be used.

Following encapsulation and leak testing, all uranium and plutonium samples were subjected to a radiographic check to determine metal sample positioning within the capsule and to detect flaws in the metal disks as well as in the aluminum capsule and weld joint. Radiographs of the ^{235}U , ^{238}U , and ^{239}Pu samples that were shipped are shown in Figs. 1, 2, and 3. Only four encapsulated samples out of a total of 65 were rejected on the basis of "unroundness" or malposition as determined by these radiographs. The critical parameters for each capsule are listed in Tables 1, 2, and 3.

Expected Achievements

By the end of the fiscal year, all disk-type dosimeters will be fabricated (^{237}Np) and shipped to HEDL. Analyses of metals and oxides of these

actinides will be performed to give vital statistics on metal-to-oxygen ratios, trace impurities, water content, and isotopic content. Archive samples will be formed for future reference. Approximately 160 needle-type flux dosimeters will be produced and shipped to HEDL and ANL.

ORNL TABLE 1

^{235}U DOSIMETERS - BATCH NO. U-235-264-CM

Sample Number	Total Weight (mg)	Can Thickness (mil)	Foil Weight (mg)	Foil Thickness (mil)
U5-1	531.67	21.0	286.492	5.1
U5-2	530.73	21.3	281.579	5.1
U5-3	532.19	21.1	278.926	5.3
U5-4	552.52	21.0	297.838	5.4
U5-5	545.51	20.8	296.674	5.3
U5-6	536.22	21.4	283.821	5.1
U5-7	530.28	21.5	277.972	5.1
U5-8	539.80	21.1	286.720	5.1
U5-9	541.95	20.6	288.701	5.2
U5-10	528.64	21.7	276.934	5.1
U5-11	555.94	21.0	302.091	5.3
U5-12	543.88	21.1	290.897	5.2
U5-13	552.75	21.3	299.032	5.3
U5-14	531.45	21.1	278.245	5.1
U5-15	549.51	21.3	297.344	5.3
U5-16	535.15	21.1	283.360	5.1
U5-17	542.91	20.8	291.462	5.2
U5-19	538.16	20.9	285.285	5.1
U5-20	534.46	21.1	282.713	5.1
U5-21	538.51	20.7	287.161	5.2

ORNL TABLE 2

 ^{238}U DOSIMETERS - BATCH NO. U-238-ES-Z

Sample Number	Total Weight (mg)	Can Thickness (mil)	Can Diameter (mil)	Foil Weight (mg)	Foil Thickness (mil)
U8-1	566.1	20.3	750.1	313.963	5.4
U8-2	561.5	21.7	750.2	310.103	5.2
U8-3	570.1	20.6	750.5	316.582	5.4
U8-4	564.4	20.2	750.2	311.585	5.2
U8-5	569.0	20.6	750.1	315.255	5.4
U8-6	559.8	20.6	750.4	307.448	5.2
U8-7	565.5	20.9	750.5	312.806	5.2
U8-8	559.1	20.0	750.1	306.692	5.3
U8-9	561.0	21.1	750.5	308.047	5.3
U8-10	557.7	21.5	750.4	304.007	5.2
U8-11	562.8	21.2	750.2	309.425	5.2
U8-13	568.7	21.3	750.4	315.775	5.4
U8-14	554.6	21.6	750.5	302.705	5.2
U8-15	555.6	20.9	750.3	304.699	5.2
U8-16	561.3	21.2	750.6	309.673	5.3
U8-17	568.4	20.7	750.7	312.875	5.3
U8-18	563.8	21.6	750.2	310.049	5.2
U8-19	563.6	21.5	750.5	308.910	5.2
U8-20	563.6	21.0	750.6	308.973	5.2
U8-21	566.3	20.8	750.5	311.475	5.2

ORNL TABLE 3

 ^{239}Pu DOSIMETERS - BATCH NO. Pu-239-453B

Sample Number	Total Weight (mg)	Can Thickness (mil)	Foil Weight (mg)	Foil Thickness (mil)
P9-1	529.78	21.2	276.204	5.7
P9-2	522.96	21.2	271.344	5.7

ORNL TABLE 3 (Cont'd)

Sample Number	Total Weight (mg)	Can Thickness (mil)	Foil Weight (mg)	Foil Thickness (mil)
P9-3	515.75	20.8	262.825	5.6
P9-4	522.09	21.8	269.643	5.7
P9-5	511.93	21.2	260.078	5.6
P9-6	514.68	21.3	261.030	5.6
P9-7	514.61	21.0	264.317	5.7
P9-8	524.31	20.9	273.543	5.8
P9-10	522.12	21.0	267.977	5.7
P9-11	529.61	20.8	276.503	5.7
P9-12	513.70	21.9	261.677	5.6
P9-14	518.42	21.2	265.057	5.7
P9-15	513.58	21.1	262.062	5.6
P9-16	522.09	21.7	269.078	5.7
P9-17	521.14	21.3	267.593	5.7
P9-18	515.35	21.2	262.710	5.6
P9-19	516.25	21.6	263.373	5.6
P9-20	519.10	21.8	264.875	5.6
P9-21	514.75	21.2	263.870	5.6
P9-22	516.52	21.9	263.707	5.7

ORNL-6

FIGURE ORNL-1 Encapsulated ^{235}U Foil Radiographs

ORNL-7

FIGURE ORNL-2 Encapsulated ^{238}U Foil Radiographs

ORNL-8

FIGURE ORNL-3 Encapsulated ^{239}Pu Foil Radiographs

B. CALCULATIONS SUPPORTING THE NATIONAL BUREAU OF
STANDARDS SPECTRUM FACILITY DESIGN EFFORT

J. D. Jenkins

Objective

- Calculational support was provided to NBS in design studies for their planned intermediate energy neutron field .

Summary

Mr. Albert Fabry of the Center of Nuclear Energy Studies (CEN-SCK), Belgium, and presently on assignment at NBS, visited Oak Ridge National Laboratory from May 30 to June 2, 1972.

During that period, and with the guidance of Dr. J. D. Jenkins of the ORNL staff, he performed a number of transport-theory calculations to aid in the design of a central cavity standard neutron spectrum facility to be constructed at the National Bureau of Standards.

Accomplishments and Status

The required calculations have been completed. In addition a suitably spectrum averaged 40 group cross section set has been constructed which will allow NBS to perform additional transport-theory calculations with their own computing facilities. Details of the calculations and results obtained will be reported in the ILRR quarterly progress report for June through August 1972.

C. EVALUATION OF DOSIMETRY CROSS SECTIONS

J. D. Jenkins

Objective

- To provide an evaluated file of activation cross sections in ENDF/B format under the auspices of the Cross Section Evaluation Working Group (CSEWG).

Summary

ORNL participates in the CSEWG activity and in the effort to provide an evaluated set of activation cross sections. In this capacity we have undertaken the evaluation of specific reaction cross sections for the library.

Accomplishments and Status

An evaluation for the $\text{Co}(n,\alpha)$ reaction has been completed and included on the ENDF/B dosimetry library. An evaluation for the $\text{Co}(n,\gamma)$ reaction is complete except for nuclear model calculations necessary to define the cross section in the MeV region.