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LMFBR REACTION RATE
AND
DOSIMETRY 10th PROGRESS REPORT
JUNE, 1974 THRU OCTOBER, 1975

HANFORD ENGINEERING DEVELOPMENT LABORATORY
Operated by Westinghouse Hanford Company
A Subsidiary of Westinghouse Electric Corporation

P.O. Box 1970 Richland, WA 99352

MASTER

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Compiled by

W. N. McElroy

AUTHORS

J. W. Rogers, E. H. Turk, C. H. Hogg, R. C. Greenwood,
and R. G. Helmer - Aerojet Nuclear Company

R. J. Popek, R. R. Heinrich, and N. D. Dudey -
Argonne National Laboratory

Harry Farrar IV - Atomics International

B. A. Magurno - Brookhaven National Laboratory

W. Y. Matsumoto, L. S. Kellogg, A. I. Davis,
M. R. Weiler, C. J. Ambrose, and J. M. Ruggles -
Hanford Engineering Development Laboratory

R. Gold - Joint Center for Graduate Study

G. E. Hansen, and D. W. Barr - Los Alamos Scientific
Laboratory

D. M. Gilliam, J. A. Grundl, G. E. Hansen, H. H. Helmik,
A. Fabry, and F. Cops - National Bureau of Standards

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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 Energy Research & Development Administration, Division of Reactor Research and Development, under Contract No. E(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.

- Aerojet Nuclear Company (ANC)
- Argonne National Laboratory (ANL)
- Atlantic Richfield Hanford Company (ARHCO)
- Atomics International (AI)
- Brookhaven National Laboratory (BNL)
- Joint Center for Graduate Study (JCGS)*
- 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)

*Richland, Washington

**CEN-SCK Laboratory (MOL) is participating through the joint NBS/CEN-SCK program to develop well-characterized permanent standard neutron fields for fast reactor applications.

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FOREWORD

PURPOSES OF ILRR PROGRAM

The Interlaboratory LMFBR Reaction Rate (ILRR) program has been established by USAEC/RRD 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 $\pm 5\%$ at the 95% confidence level. Accurate measurement of other fission and non-fission reactions will be required, but to a lesser accuracy, between ± 5 and 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/or permanent neutron fields. The neutron fields being used are: the Coupled Fast Reactivity Measurement Facility (CFRMF) at Aerojet Nuclear Company; the 10% enriched ^{235}U critical assembly (BIG-10) at Los Alamos Scientific Laboratory; the Fast Test Reactor (FTR) Engineering Mockup Critical (EMC) at Argonne National Laboratory; the Natural Uranium Secondary Standard Neutron Field ($\Sigma\Sigma$) at Mol, Belgium; the Intermediate-Energy Standard Neutron Fields (ISNF) being developed at the National Bureau of Standards and Mol, Belgium; the fission spectrum and thermal cavities at NBS and Mol; and the californium-252 fission source at NBS. These neutron fields span the range of spectra encountered in fast reactors and are being used because of the range and reproducibility of flux levels available for track etch, fission chamber, radiochemistry, and helium mass spectrometric

measurements. Results of tests in these facilities 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 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 revisions of results in this document are to be expected unless otherwise stated.

PREVIOUS REPORTS IN THIS SERIES

HEDL-TME 71-131	(First Quarterly Progress Report)
HEDL-TME 71-179	(Second Quarterly Progress Report)
HEDL-TME 72-45	(Third Quarterly Progress Report)
HEDL-TME 72-90	(Fourth Quarterly Progress Report)
HEDL-TME 72-129	(Fifth Quarterly Progress Report)
HEDL-TME 72-154	(Sixth Quarterly Progress Report)
HEDL-TME 73-31	(Seventh Quarterly Progress Report)
HEDL-TME 73-54	(Eighth Progress Report)
HEDL-TME 74-45	(Ninth Progress Report)

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SUMMARY

PROGRAM HIGHLIGHTS

A major program milestone was reached with the publication in the February 1975 Special Issue of Nuclear Technology, Volume 25, of a series of articles on ILRR program work. Also included was material from the more general area of fuels and materials dosimetry and reactor physics.

Another significant program highlight was the presentation of a number of invited and contributed papers by ILRR program and other U. S. participants at the First ASTM-Euratom Symposium on Reactor Dosimetry, Petten, Netherlands, September 22-26, 1975. The proceedings of this symposium are being published by Euratom.

It was emphasized at the Petten conference that the most effective utilization of exchanged fuels and materials irradiation effects data would be achieved by international dosimetry and damage analysis standardization. It was pointed out, however, that the success of such standardization would depend on the development and maintenance of well-balanced and properly supported irradiation test facilities and power reactor environmental characterization programs by participating countries. Some stated major benefits of this standardization were:

- Significant lifetime extension of FBR and other type reactor fuels, materials, and components and increased reactor safety by improved data correlation.
- A more rapid lowering of FBR doubling times due to more accurate engineering design data for fuels, materials, and in-vessel components.
- Effective application of fission reactor technology to fusion reactor development programs.
- Major cost savings for both commercial power reactors and nuclear development programs.

In helping to further and coordinate interlaboratory efforts directed towards such dosimetry and damage analysis standardization, the U. S. attendees

to the symposium made a number of separate trips to European Research and Development Establishments.

AEROJET NUCLEAR COMPANY

More complete and accurate neutron flux gradient measurements are reported which define the flux profile in the CFRMF-ILRR test location to $\pm \sim 2\%$. Plugging the ends of the test space with ^{10}B causes no measurable change in the ^{235}U fission rate and ^{198}Au activity at the CFRMF-ILRR test locations. Measurements of the Au Cd-ratio show no spectral perturbation due to the presence of Cd or ^{10}B . Fission product generation rates from fission foils irradiated in CFRMF, NBS Thermal Cavity and BIG-10 are presented. Reaction rates of non-fission foils irradiated in BIG-10 are also reported. Absolute gamma ray intensities for ^{239}Np decay are evaluated for the 228 keV and 277 keV transitions. Comments on gamma-ray spectrometry measurements and analyses employed at ANC are given to completely describe all interpretations, corrections and uncertainties.

ARGONNE NATIONAL LABORATORY

The method of standardization of the ANL Ge(Li) detector calibration is reviewed. Two separate efficiency vs. energy curves are maintained; the K-factor curve remains unchanged and the curve used for nonfission rate analyses periodically is reevaluated as part of the ANL quality assurance program. Also presented are the nuclear and physical constants used in the analysis of ANL counting data.

Final thermal neutron fission yields of ^{235}U have been measured. These values reflect the latest NBS fission rate and the more precise ANL detector calibration. Fission product reaction rates are calculated from the CFRMF and BIG-10 irradiations for ^{235}U , ^{238}U , ^{237}Np , and ^{239}Pu . The production rate of ^{239}Np from ^{238}U for each of these irradiations is also given. Reaction rates for several nonfission foils irradiated in BIG-10 are presented; however, the precision of these data is not as good as that observed for the fission foils.

Fission rates for ^{235}U , ^{238}U , ^{237}Np , and ^{239}Pu have been calculated from CFRMF K-yield factors and six fission-product monitors. Precision of the data indicates these fission products have the potential of being used as reliable monitors.

ATOMICS INTERNATIONAL

Three almost-identical small aluminum assemblies, each enclosing twelve miniature vanadium capsules filled with enriched ^{10}B and ^6LiF (helium accumulation fluence monitors) have been irradiated as a part of the Inter-laboratory LMFBR Reaction Rate (ILRR) program. Two of the assemblies were irradiated consecutively in the BIG-10 facility at Los Alamos during two series of reactor runs. The third was irradiated in the CFRMF facility at Idaho Falls for a 6kw run lasting 29 hours. Additional miniature helium accumulation fluence monitors are being manufactured for irradiation in the $\Sigma\Sigma$ and fission spectrum facilities at Mol-Belgium.

BROOKHAVEN NATIONAL LABORATORY

The dosimetry tape (ENDF/B-IV tape 412) was issued in a general CSEWG distribution, August 1974. The pointwise cross section data file was tested with specified reference spectra. The results are published in the February 1975 issue of Nuclear Science and Technology. A group averaged cross section data file (620 groups based on tape 412) was tested with the above spectra and the results are presented in this report.

HANFORD ENGINEERING DEVELOPMENT LABORATORY

Gamma counting QA analysis on a number of fission foil monitors has been completed. A second $^{140}\text{Ba}/^{140}\text{La}$ standard source was prepared and distributed to ILRR participants. Dissolution and analysis for ^{137}Cs , isotopic Pu and isotopic Nd have been made on 20 EBR-II irradiated foils and neodymium and fission yield changes referenced to neutron mean energies are reported. A possible discrepancy in the ^{239}Pu ENDF/B-IV to measured ^{148}Nd to ^{137}Cs ratio of $\sim 10\%$ has been noted. Reaction rate measurements are reported on the

BIG-10 irradiations and additional determinations made on the NBS thermal irradiation of a ^{235}U foil for ^{137}Cs .

JOINT CENTER FOR GRADUATE STUDY

The variation of the asymptotic sensitivity with charge (Z) and mass (A) of the fissile SSTR source has been estimated using Bohr's formulation of fission-fragment energy-loss in dense media. Numerical values of the asymptotic sensitivity have been generated for ^{232}Th , ^{233}U , ^{235}U , ^{238}U , and ^{239}Pu . These calculations reveal a monotonic increase with increasing A which is small but non-negligible relative to experimental error. This dependence must, therefore, be accounted for in SSTR applications of high accuracy involving asymptotically thick sources.

LOS ALAMOS SCIENTIFIC LABORATORY

The BIG-10 ILRR program tests A through E have been completed and irradiation times and exposures for these tests are documented in this report. Final reaction rate measurements for tests LASL C(74-14) and LASL E(74-21) are reported.

NATIONAL BUREAU OF STANDARDS

An NBS double fission chamber was employed to determine the fission cross section ratios for ^{235}U , ^{238}U , ^{239}Pu , and ^{237}Np in the central test cavity of the LASL BIG-10 critical facility. The accuracy achieved in these measurements ranged from 1.5% to 2.2%. Corresponding cross section ratios were derived from ENDF/B-IV data and a calculated neutron energy spectrum; the agreement with the measured values was good except in the important case of the $^{238}\sigma_f/^{235}\sigma_f$ ratio for which the discrepancy was 10.6%.

The dosimetry foil Tests A, B, C, and E at BIG-10 were monitored continuously by the NBS double fission chamber within the central test cavity. For each irradiation the integrated number of fissions per milligram of ^{235}U was determined, the spectral index $^{238}\sigma_f/^{235}\sigma_f$ was checked, and the power profile, $\phi(t)$, was observed. In spite of a potentially serious

contamination problem, run-to-run normalization accuracy of $\pm 0.3\%$ to $\pm 0.6\%$ was obtained, and absolute fissions/mg values were obtained with an accuracy of $\pm 1.4\%$ to $\pm 1.5\%$. Redundant activation foil monitors and external fission chambers provided an important supplement to the double fission chamber data in overcoming the uncertainties introduced by the contamination problem.

Absolute fission rate measurements were performed in the Mol- $\Sigma\Sigma$ Secondary Standard Neutron Facility in order to prepare for subsequent high-fluence ILRR activation foil irradiations. Fission cross section ratios for ^{235}U , ^{238}U , ^{239}Pu , and ^{237}Np were determined to an accuracy of $\pm 2.1\%$ to $\pm 2.9\%$. Run-to-run fluence monitoring -- based on $\text{In}(n,n')$ activation detectors and peripheral fission chambers -- was established to a precision of $\pm 0.4\%$.

ANC

AEROJET NUCLEAR COMPANY

A. CFRMF NEUTRON FLUX GRADIENT AND SPECTRAL DETERMINATIONS

JW. Rogers, E. H. Turk, C. H. Hogg

Objective

- To meet ILRR program objectives, an accurate characterization of the neutron environment in the CFRMF test region must be available. One of the important parameters in this respect is the gradient of the neutron flux in the test region where the irradiations are performed. Another important parameter is the neutron energy spectrum and factors which may influence this spectrum. The best possible information on these parameters will be obtained and disseminated.

Summary

Previously determined CFRMF neutron flux gradient and neutron energy spectrum information has been given in the previous progress reports. Recently more accurate and complete measurements of the flux gradient have been measured by the activation of ^{235}U and Au samples. Neutron spectrum characteristics were studied by making activation measurements with and without the ends of the CFRMF test region plugged with ^{10}B . These measurements define the flux gradient to $\pm 1\text{-}2\%$ and indicate there is no detectable streaming of thermal or resonance neutrons from the ends in the central 30 cm of the CFRMF test region. Measurements of the Cd ratio of Au foil activations were conducted and these results also indicate there is no streaming of thermal and resonance neutrons into the CFRMF test region.

Achievements and Status

Devices were fabricated from stainless steel that would hold samples for activation along the vertical axis of the CFRMF test region and allow end plugs to be located at the ends of the access hole. A diagram of these devices as located in the CFRMF unit is shown in Figure ANC-1. The larger tubing between the end plugs is for supporting and positioning the plugs and the smaller tubing inside is for holding the activation samples. Two identical sets of end plugs were fabricated, one set (top and bottom) being empty and the other filled with enriched ^{10}B powder. The 99.9% pure B

powder enriched to 92 atom percent ^{10}B was vibracompacted into the end plugs to a density of 1.06 g/cm^3 . The walls of the end plugs were made as thin (0.5 mm) as fabrication procedures would allow in order to minimize any neutron leakage around the plugs. This device allows the vertical position of the activation samples to be known to $\pm 1\text{ mm}$ relative to the bottom of the experimental access tube.

Two materials, pure Au and Al alloy of 10 w/o ^{235}U (93% enriched), have been irradiated (at 6 kW) with the empty and ^{10}B filled end plugs in CFRMF. The activation samples were short wires (6.35 mm long and 1 mm diameter) which were stacked end-to-end the full length of the sample holding tube for each irradiation. In each irradiation 83 samples were exposed and each was counted by gross beta counting techniques. From the $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ reaction an activity due to the capture of resonance energy neutrons was obtained. From the $^{235}\text{U}(n,f)\text{FP}$ reaction an activity due to the fission of a very broad energy range of neutrons was measured.

From these activities profiles of the activation responses of these samples in the CFRMF neutron flux have been drawn and are shown in Figures ANC-2 and 3. From these profiles the peak of the CFRMF flux is found to be $30.6 \pm 0.1\text{ cm}$ from the bottom of the experimental access tube (the geometrical midplane is 31.1 cm from the bottom). The shapes of these profiles are nearly identical and have no irregularities which suggest that the filter materials are very uniformly dispersed. The ^{10}B end plugs cause no detectable changes in either the absolute or relative reaction rates of these materials along the central 30 cm of the fast zone vertical axis. In the end regions beginning 15 cm above or below the peak of the flux the profiles show changes due to the presence of the ^{10}B end plugs. These changes are assumed due to spectral perturbations of the flux in these regions. The reduced reaction rates in the end regions when the ^{10}B end plugs are installed imply that more resonance and/or thermal energy neutrons are present there without the end plugs.

The absolute fission rates at the flux peaks of $1.218 \pm 0.016 \times 10^{-13}$ and $1.232 \pm 0.016 \times 10^{-13}$ fissions/sec-nucleus for the plugged and unplugged measurements respectively are based upon a thermal flux fission rate calibration. The difference ($\pm 0.6\%$) between these fission rates is well within their associated uncertainties. The corresponding absolute Au reaction rates are

$2.515 \pm 0.025 \times 10^{-10}$ and $2.508 \pm 0.025 \times 10^{-10}$ d/sec-atom which agree to $\pm 0.15\%$. These differences are well within statistical uncertainties and are regarded as insignificant. The relative activities used to describe the profiles are of the same accuracy. These measurements have not been corrected for neutron self-shielding and gamma self-absorption. Based upon least squares fitting of these data there is no difference in the shapes of the profiles over the central 30 cm and the scatter of the data relative to the fitted curves is less than the estimated uncertainties.

A study in the CFRMF was made on the effect of cadmium metal of different thicknesses on the activation of gold foils. This study was done to test for the presence of thermal neutrons and to investigate scattering-in-Cd effects on reaction rates in the CFRMF neutron spectrum. This is important because it is necessary to know if there is any thermal neutron reaction contributing to the activities (cross sections) obtained in the CFRMF and if high atomic weight materials cause any moderating effects that are detectable by activation measurements.

For this study a 20 mil cadmium foil strip was rolled to produce strips of various thicknesses. These strips were used to fabricate the cadmium covers. The cadmium cover was made by cutting two pieces 1/2 inch by 3/4 inch from one of these strips. The two pieces were placed one on top of the other with the long axis of the pieces at right angles to each other. A gold foil enclosed in 0.5 mil aluminum foil packet 1/2 inch square was placed between the two cadmium pieces. The overlapping edges of each cadmium piece was bent over the edges of the other cadmium piece. This resulted in a packet with the gold foil in the center. To eliminate any possibility of neutron leakage the corners of the packet were crimped.

A 2 mil gold foil was mounted in a 0.5 mil aluminum foil packet 1-1/2" above the gold packet being tested. This foil was attached using 3.5 mil diameter copper wire. These foils were used to normalize power level variation between the bare and cadmium covered runs. Two irradiations were made using gold foils of the same thickness and both enclosed in 0.5 mil aluminum foil cover. One irradiation was made with the gold foil in the cadmium covered packet and the other without the cadmium cover. For the irradiations the packets were mounted on an aluminum metal strip

(1-1/2" x 5-1/2" x 0.027") using 3.5 mil diameter copper wire. Mounting of the packet was done so that when the mounting strip rested on the pedestal in the CFRMF experimental access tube the gold was located at the reactor centerline with the plane of the sample vertically oriented. Runs were made using 2.0 mil gold and varying the thickness of the cadmium. One run was made using a 0.02 mil gold foil to study the effect of gold thickness. One run was also made with the ^{10}B end plugs installed.

Results of the irradiations are expressed as a cadmium ratio. This is the ratio of the bare gold foil activity (normalized to the cadmium covered run) to the cadmium covered gold foil activity. Table ANC-1 is a summary of these measurements. The results are estimated to have an uncertainty of $\pm 2\%$. Most of this arises from the normalization procedures used. Reproducibility was of the order of $\pm 0.5\%$.

Cadmium thickness and gold thickness gave no indication of having any effect on the cadmium ratio for gold in the CFRMF experimental access facility tube. The plugging of the ends of the test region with ^{10}B end caps showed no appreciable effect.

TABLE ANC-1
Cd RATIO MEASUREMENTS IN THE CFRMF

<u>Cd (mils)</u>	<u>Au (mils)</u>	<u>Cd Ratio</u>
5.0	2.0	1.02
9.0	2.0	1.00
20.0	2.0	1.00
20.0	0.02	1.02
20.0*	0.02*	0.98*

* With ^{10}B end plugs installed.

These measurements define the CFRMF flux gradients as well as current techniques permit. They also indicate there is no thermal or resonance neutron streaming into any portions of the CFRMF test region used in ILRR experiments.

Expected Achievements

Neutron flux profile measurements with threshold type detectors may be obtained.

ANC-B-3115

FIGURE 1. Diagram of Device Used to Position End Plugs and Flux Monitors in CFRMF Fast Neutron Spectrum Zone.

FIGURE 2. Profiles of ^{235}U Fission Rate Along the Vertical Axis of CFRMF Fast Neutron Spectrum Zone With and Without ^{10}B End Plugs.

FIGURE 3. Profiles of ^{198}Au Activity Along the Vertical Axis of CFRMF Fast Neutron Spectrum Zone With and Without ^{10}B End Plugs.

B. ^{140}Ba - ^{140}La GAMMA-RAY STANDARD

J W Rogers and E. H. Turk

Objective

- To prepare counting sources with very accurately known contents and to use these sources for the calibration of gamma-ray detectors.

Summary

A ^{140}Ba - ^{140}La standard solution (#T-1083) was received from HEDL and counting sources were prepared from the solution. These sources were counted to check the efficiency of detectors used in ILRR related measurements. The counting source contents were determined by relative masses and activities.

Accomplishments and Status

HEDL supplied ANC with 4.8313 grams of the ^{140}Ba - ^{140}La standard solution #T-1083 containing 10 ppm of Ba and 10 ppm La. In preparation to make the counting sources, the sealed glass ampoule containing the solution was opened by breaking off the top end. Calibrated pipettes were used to take the aliquots. The ampoule and solution were weighed immediately before and after the taking of each aliquot for each source. The sample size was based on aliquot weight and total weight of the solution and its container. 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. The sources were deposited on 1.27 cm diameter blotting paper discs to match the size of the ILRR foils. Each aliquot and its respective rinses were deposited on the discs uniformly and then allowed to air dry before being sealed between two pieces of plastic tape. Table ANC-2 summarizes the preparation of the sources. Corrections were applied for changes in concentration of the solution due to evaporation during preparation.

The relative weights and activities of a given sample disagree by no more than 0.77% except for the 10 λ sources. Consequently, the 10 λ sources

were not used for checking efficiencies. From these mass and activation comparisons it is determined that the uncertainty of the contents of the counting sources used to obtain the activity from this standard is $\sim \pm 0.5\%$.

Based on these measurements the ^{140}Ba activity of this standard solution is $1.110 \pm .015 \times 10^6$ d/s/g as of 3/20/74 PST.

Expected Achievements

No further work is anticipated.

TABLE ANC-2

^{140}Ba - ^{140}La STANDARD #T-1083 SOURCES

<u>Identification</u>	<u>Volume</u>	<u>Weight</u>	<u>Relative Weight</u>	<u>Relative Activity</u>	<u>Relative Act. Relative Wt.</u>
200 λ A	$\sim$ 200 λ	206.000 g	$\equiv$ 1.0000	$\equiv$ 1.0000	$\equiv$ 1.0000
200 λ B	$\sim$ 200 λ	208.791 g	1.01355	1.01391	0.9996
100 λ A	$\sim$ 100 λ	103.512 g	.50248	.50113	1.0027
100 λ B	$\sim$ 100 λ	104.742 g	.50845	.51015	.9967
50 λ A	$\sim$ 50 λ	52.178 g	.25329	.25254	1.0030
50 λ B	$\sim$ 50 λ	52.994 g	.25725	.25527	1.0077
10 λ A	$\sim$ 10 λ	11.308 g	.05489	.05377	1.0208
10 λ B	$\sim$ 10 λ	11.239 g	.05456	.05191	1.0510

C. CFRMF FISSION PRODUCT GENERATION RATES

J W Rogers

Objective

- Obtain fission product generation rates from foils of fissionable materials irradiated in the CFRMF neutron field. These results are to be used with results obtained from other laboratories to establish the most accurate values for the generation rates. The use of other methods to obtain fission rates and fission product yields will be used to improve foil dosimetry methods in fast breeder reactors.

Summary

Fission product generation rates in high quality foils of ^{235}U , ^{238}U , ^{239}Pu and ^{237}Np have been measured by gamma ray activation analyses. These foils were irradiated in the CFRMF and the $^{140}\text{Ba-La}$ activity was measured by NaI counting techniques.

Accomplishments and Status

Gamma-ray spectrometry measurements on all the fission foils in ILRR tests ANC-74-4&5 have been made using standard NaI(Tl) techniques. A 3"x3" NaI(Tl) crystal detector was used with source-to-detector distances of 10, 20 and 30 cm. All source-to-detector distances were intercalibrated with standard sources at the 1.596 MeV gamma energy. All counting was done with a 0.674 g/cm² polystyrene beta absorber covering the surface of the detector. Additional absorbers of Cd and Cd+Pb were necessary to reduce counting rates for the ^{239}Pu and ^{237}Np respectively. Attenuation factors for these absorbers placed on the surface of the polystyrene were measured at 1.596 MeV with standard sources. Decay times of approximately 22 days were allowed for optimum counting of the $^{140}\text{Ba-La}$ activity at 1.596 MeV. The decay scheme data adopted by the ILRR participants were used in the data reduction. All data reduction was done by consistent manual methods.

The results of these measurements and analyses are given in Table ANC-3. One of the foils, $^{235}\text{U}\#4$ was irradiated in the power level normalization

position for the purpose of comparison with similar foils sent to Mol and LASL. The other foils were irradiated in the fission chamber position.

Expected Achievements

Additional irradiations of fission foils in CFRMF will be analyzed in this manner.

TABLE ANC-3
CFRME FISSION PRODUCT GENERATION RATES
(Tests ANC-74-4&5)

ANC-13	<u>Foil ID</u>	<u>Reaction</u>	<u>Target Mass g</u>	<u>γ Self Absorption</u>	<u>External Absorber</u>	<u>Coincidence Summing</u>	<u>Random Summing</u>	<u>¹⁴⁰Ba-La Activity d/s/atom</u>
	U5-11	$^{235}\text{U}(n,f)^{140}\text{Ba}$	0.3000688	0.9917	1.000	0.9938	0.9900	$8.075 \pm 0.153 \times 10^{-15}$
	U8-16	$^{238}\text{U}(n,f)^{140}\text{Ba}$	0.308056	0.9910	1.000	0.9798	0.9950	$0.392 \pm 0.007 \times 10^{-15}$
	P9-14	$^{239}\text{Pu}(n,f)^{140}\text{Ba}$	0.2581	0.9920	0.9643	0.9938	0.9900	$7.966 \pm 0.159 \times 10^{-15}$
	N7-16	$^{237}\text{Np}(n,f)^{140}\text{Ba}$	0.364462	0.9885	0.7994	0.9988	0.9812	$2.766 \pm 0.061 \times 10^{-15}$
	$^{235}\text{U}\#4$	$^{235}\text{U}(n,f)^{140}\text{Ba}$	0.121044	0.9980	1.000	0.9798	0.9850	$7.867 \pm 0.134 \times 10^{-15}$

D. BIG-10 FISSION PRODUCT GENERATION AND REACTION RATES

J W Rogers

Objective

- Obtain fission product generation rates from foils of fissionable materials and reaction rates from nonfissionable materials irradiated in the BIG-10 neutron field. These results are to be used with results obtained from other laboratories to establish the most accurate values possible. These values will then be applied, in conjunction with other parameters, to the development of dosimetry methods in fast breeder reactors.

Summary

Fission product generation rates for high quality fission foils and reaction rates of nonfission foils have been measured by gamma ray activation analyses. These foils were irradiated in the BIG-10 facility and the activities were measured by NaI counting techniques.

Accomplishments and Status

Gamma-ray spectrometry measurements on BIG-10 irradiated foils in ILRR tests ANC-74-15 and 22 have been made using standard NaI(Tl) techniques. The techniques used on the fission foils are described in Section ANC-C. Most of the nonfission foils were counted at the 10 cm source-to-detector distance but due to the low activity levels the Co and Al foils were counted at 3 cm. These source-to-detector distances were intercalibrated with standard sources. The results of these measurements and analyses are given in Tables ANC-4 and 5. No detectable activity was found in the Mn and In foils due to the delay in transit of the foils.

Expected Achievements

Measurements and analyses of foils from future irradiations will be performed as they become available.

TABLE ANC-4

BIG-10 FISSION PRODUCT GENERATION RATES

(Test ANC-74-15)

<u>Foil ID</u>	<u>Reaction</u>	<u>Target Mass</u>	<u>γ Self Absorption</u>	<u>External Absorber</u>	<u>Coincidence Summing</u>	<u>Random Summing</u>	<u>$^{140}\text{Ba-La}$ Activity d/s/atom</u>
U5-8	$^{235}\text{U}(n,f)^{140}\text{Ba}$	0.284810	0.9917	1.0000	0.9938	0.996	$7.114 \pm 0.135 \times 10^{-15}$
U8-11	$^{238}\text{U}(n,f)^{140}\text{Ba}$	0.307810	0.9910	1.0000	1.0000	0.9990	$0.2577 \pm 0.0362 \times 10^{-15}$
P9-11	$^{239}\text{U}(n,f)^{140}\text{Ba}$	0.269275	0.9920	0.9643	0.9938	0.9940	$7.359 \pm 0.146 \times 10^{-15}$
N7-13	$^{237}\text{U}(n,f)^{140}\text{Ba}$	0.365693	0.9885	0.7994	0.9988	0.9750	$2.147 \pm 0.047 \times 10^{-15}$

ANC-15

TABLE ANC-5
BIG-10 REACTION RATES (Test ANC-74-22)

Foil ID	Reaction	Target Mass	γ Self-Absorption	Coincidence Summing	Random Summing	Reaction Rate x 10 ⁻¹⁵ d/s/atom		
Mn-3	No detectable activity							
Al-7	²⁷ Al(n, α) ²⁴ Na	0.04623	0.999	0.915	1.000	0.0083	± 0.0003	
Au-5	¹⁹⁸ Au(n, γ) ¹⁹⁸ Au	0.02949	0.998	1.000	0.998	18.55	± 0.24	
In-7	No detectable activity							
Cu-3*	⁶³ Cu(n, γ) ⁶⁴ Cu	0.01507	0.998	1.000	1.000	1.783	± 0.037	
Co-7	⁵⁹ Co(n, γ) ⁶⁰ Co	0.01447	1.000	0.918	1.000	1.241	± 0.037	
Co-8	⁵⁹ Co(n, γ) ⁶⁰ Co	0.01420	1.000	0.918	1.000	1.230	± 0.037	
Co-9	⁵⁹ Co(n, γ) ⁶⁰ Co	0.01437	1.000	0.918	1.000	1.179	± 0.037	
Ni-MO	⁵⁸ Ni(n,p) ⁵⁸ Co	0.19606	0.992	1.000	0.999	1.360	± 0.023	
ANC-16	Fe-C	⁵⁴ Fe(n,p) ⁵⁴ Mn	0.04388	0.979	1.000	1.000	1.057	± 0.020
		⁵⁸ Fe(n, γ) ⁵⁹ Fe	0.002672	0.982	1.000	1.000	0.3568	± 0.0115
	Al-8	²⁷ Al(n, α) ²⁴ Na	0.04634	0.999	0.915	1.000	0.0087	± 0.0003
	Au-3	¹⁹⁷ Au(n, γ) ¹⁹⁸ Au	0.003003	1.000	1.000	0.999	18.66	± 0.28
	In-8	No detectable activity						
	Ti-L0	⁴⁷ Ti(n,p) ⁴⁷ Sc	0.01120	0.990	1.000	1.000	0.2258	± 0.0044
		⁴⁸ Ti(n,p) ⁴⁸ Sc	0.11619	0.997	0.983	1.000	0.003754	± 0.000119
	Ti-LP	⁴⁷ Ti(n,p) ⁴⁷ Sc	0.01119	0.990	1.000	1.000	0.2342	± 0.0046
		⁴⁸ Ti(n,p) ⁴⁸ Sc	0.11611	0.997	0.983	1.000	0.003986	± 0.000126
	Sc-4	⁴⁵ Sc(n, γ) ⁴⁶ Sc	0.04705	0.999	0.982	0.999	1.500	± 0.021
²³⁵ U-3	²³⁵ U(n,f) ¹⁴⁰ Ba	0.22603	0.995	0.980	0.987	7.101	± 0.140	
²³⁸ U-5	²³⁸ U(n,f) ¹⁴⁰ Ba	0.29314	0.994	0.980	0.998	0.2540	± 0.0050	
In-9	No detectable activity							
Au-6	¹⁹⁷ Au(n, γ) ¹⁹⁸ Au	0.03155	0.998	1.000	0.998	18.64	± 0.24	
Al-9	²⁷ Al(n, α) ²⁴ Na	0.04606	0.999	0.915	1.000	0.0082	± 0.0003	

*This foil was counted between 12 mil Cu covers to annihilate the positrons.

E. ABSOLUTE GAMMA-RAY INTENSITIES IN DECAY OF ^{239}Np

R. C. Greenwood and R. G. Helmer

Objective

- To determine the absolute intensities of the 228- and 277-keV γ rays in the decay of ^{239}Np .

Summary

The absolute γ -ray intensities have been measured for the 228- and 277-keV transitions in the decay of ^{239}Np . These measurements were made on two ^{243}Am sources supplied by Dr. A. Fabry, Mol, for which the disintegration rates had been measured by α counting.

Accomplishments and Status

A precise knowledge of the branching ratios of the most prominent γ -rays in the 2.35-d ^{239}Np decay is of special import for neutron dosimetry, since the uncertainty in these branching ratios constitutes the limit on the precision with which $^{238}\text{U}(n,\gamma)$ reaction rates can be measured. Until recently these branching ratios have only been poorly known (uncertainty $\geq \pm 12\%$) primarily because of the complexity of the ^{239}Np decay scheme and the uncertainties in the internal-conversion-electron intensities of many of these low-energy transitions.

In a recent work⁽¹⁾, a substantial improvement in precision was achieved by taking advantage of the decay chain

Since the ^{239}Np is in equilibrium with the ^{243}Am , the disintegration rate of the ^{239}Np was determined by absolute counting of the α particles emitted by ^{243}Am and the γ -ray emission rates of ^{239}Np were determined by absolute γ counting using a calibrated Ge(Li) detector system. Hence, a simple division of the latter values by the former value provided the γ -ray branching ratios.

In the present work, this method was repeated using two ^{243}Am sources, each having $\sim 10^3$ disintegrations/sec, supplied to us by Dr. A. Fabry, C.E.N./S.C.K., Mol, Belgium. The ^{243}Am α -particle emission rates of these

sources had been measured independently at three laboratories: C.E.N./S.C.K., Mol, Belgium, K.F.K., Karlsruhe, West Germany, and R.C.N., Petten, The Netherlands. The uncertainty quoted on the average source disintegration rates is $\pm 0.7\%$ (1σ uncertainty). Absolute γ -ray counting was done at this laboratory using the OR-5 Ge(Li) detector system with a source-to-detector distance of 10 cm. The γ -ray branching ratios determined using these data are given in Table ANC-6, and compared therein to those reported in Reference 1.

The average value in Table ANC-6 has been reported in Reference 2 as the recommended ILRR value.

Expected Achievements

This measurement is complete unless other well calibrated ^{243}Am sources become available.

References

1. I. Ahmad and M. Wahlgren, Nuclear Instr. and Meth. 99, 333 (1972).
2. R. G. Helmer and R. C. Greenwood, Nucl. Tech. 25, 258 (1975).

TABLE ANC-6

GAMMA-RAY BRANCHING RATIOS IN DECAY OF ^{239}Np

Gamma-Ray Energy (keV)	Absolute Branching Ratio (%)		
	<u>Present Work</u>	<u>Reference 1</u>	<u>Average</u>
228.19 ^a	11.14 $\pm$ 0.19	11.4 $\pm$ 0.3	11.3 $\pm$ 0.2
277.60	14.07 $\pm$ 0.24	14.5 $\pm$ 0.4	14.3 $\pm$ 0.2

^aThere is a γ ray with energy 226.4 keV and branching ratio of $\sim 0.7\%$ which is not resolved from the stronger 228 keV line in a Ge(Li) detector system. Therefore the absolute intensity value obtained for the "228-keV" line will depend to some extent on the resolution of the particular Ge(Li) detector system and on the specific manner in which the peak area is determined.

F. SOME COMMENTS ON MEASUREMENTS OF REACTION RATES BY γ -RAY SPECTROMETRY

R. G. Helmer

Objective

- To comment on some factors that relate to the accuracy and comparability of the γ -ray spectrometry results obtained in the ILRR program.

Summary

Several features of the methods used in the analysis of our γ -ray spectral data are reviewed. Specifically, a list is given of the expressions used to compute the relative and total uncertainties in the reaction rates. The formulae used to obtain the averages from several spectra are listed. The status of data on atomic abundances is discussed. The magnitude of our random summing correction is tabulated. Data on interferences with the γ -ray peaks at 134 and 696 keV for $^{144}\text{Ce-Pr}$ and 724 keV for ^{95}Zr are given.

Accomplishments and Status

Introduction

At the recent (March 1975) ILRR program review meeting, several items were discussed among those doing γ -ray spectrometry that relate to the accuracy and comparability of the various sets of measurements. These items included atomic abundances, magnitude of random summing corrections, error contributions and interferences with γ -ray peaks of interest. At that meeting it was also agreed that up-to-date results would be provided for this progress report for all counting of the good fission foils from CFRMF and BIG-10, the ^{235}U foils from the NBS thermal facility and the nonfission foils from BIG-10. Therefore, several features of our γ -ray counting and analysis procedures have been reviewed and improved. These items will be commented on here.

Reaction Rates

The reaction rates, or saturated activities, that we compute are given by

$$R = \frac{A}{\epsilon \cdot B} \cdot \frac{a}{mpN_o} \cdot \frac{\lambda e^{\lambda \tau}}{f(1-e^{-\lambda T})(1-e^{-\lambda t})} \cdot \frac{\delta_1}{\delta_2 \delta_3 \delta_4 \delta_5 \delta_6 \delta_7} \quad (1)$$

where

A = measured peak area

ϵ = detector efficiency for point source

B = absolute γ -ray intensity or branching ratio

a = atomic mass of isotope of interest

m = mass of foil

p = weight abundance of mass of interest

$N_0 = 6.023 \times 10^{23}$

$\lambda = \ln 2 / T_{1/2}$, $T_{1/2}$ = half-life

T = count time (live)

f = T/T', T' = count time (clock)

t = irradiation time

τ = decay time - end of irradiation to start of count

δ_1 = correction for finite sample diameter

δ_2 = parent/daughter activity ratio

δ_3 = correction for coincidence summing

δ_4 = correction for random summing

δ_5 = correction for γ -ray absorption in sample

δ_6 = correction for external γ -ray absorbers

δ_7 = correction for reactor shutdown.

Calculated Uncertainties

At the recent ILRR program review meeting, a list of error contributions was agreed on. Our methods of treating these items are as follows. Here all errors are fractional contributions which are added in quadrature to obtain $\left[\frac{\sigma(R)}{R} \right]^2$.

peak area	- $\frac{\sqrt{a+2b}}{a}$ where a = net peak area and b = area of spectral background under peak
shelf factor	- no contribution included routinely, since almost all measurements are at directly calibrated distances; the special cases are noted explicitly.
random summing/live timer corrections	- $\frac{0.2(1-\delta_4)}{\delta_4}$ where δ_4 is the correction factor used in Eq. (1).
coincidence summing	- no error contribution included

finite source size	- no error contribution included
γ -self absorption	- no error contribution included
half-life	$- \frac{\sigma(T_{1/2})}{T_{1/2}} \left[1 + \lambda\tau - \frac{\lambda T e^{-\lambda T}}{1 - e^{-\lambda T}} - \frac{\lambda t e^{-\lambda t}}{1 - e^{-\lambda t}} \right]$ <p>with quantities defined as for Eq. (1) and $\sigma(T_{1/2})$ $T_{1/2}$ values from Ref. 1.</p>
foil mass	- 0.005
external γ absorption	<p>- 0.0025 at all energies for 0.044" Cd absorber</p> <p>- for an 8 gm/cm² Pb absorber</p> <p>0.01 for energies > 900 keV</p> <p>0.015 for 650 < E < 900</p> <p>0.02 for 550 < E_{γ} < 650</p> <p>0.03 for 450 < E_{γ} < 550</p>
peak efficiency	<p>- 0.015 for energies above 200 keV</p> <p>0.030 for energies below 200 keV</p> <p>(i.e., 134-keV line of ¹⁴⁴Ce decay)</p>
branching ratio	- $\frac{\sigma(B)}{B}$ with $\sigma(B)$ and B from Ref. 1.

The computer programs used for these analyses automatically provides two uncertainties; an absolute error including all of the non-zero terms above except the foil mass and external absorber contributions, and a relative error which also excludes the peak efficiency and branching ratio terms. In the following articles, we give this relative error (as σ_1), a second relative error including the foil mass and external absorber terms (σ_2) and the total error including all terms.

Averages

The above error contributions apply to the analysis of one peak in one spectrum. When results are obtained from several spectra they must be averaged. An exact treatment of the error propagation for this average would be very complex since some contributions to the individual errors are correlated and some are not. For the data reported in this progress report, the procedure has been as follows. The average is weighted by $1/\sigma^2$, or

$$\bar{R} = \frac{\sum_{i=1}^N \frac{R_i}{\sigma_i(R)^2}}{\sum_{i=1}^N \frac{1}{\sigma_i(R)^2}} \quad (2)$$

The uncertainty in $\bar{R}$ is taken as the larger of

$$\sigma_{in}(\bar{R}) = \left[\sum_{i=1}^N \frac{1}{\sigma_i(R)^2} \right]^{-1/2}$$

and

$$\sigma_{ext}(\bar{R}) = \epsilon \sigma_{in}(\bar{R})$$

where

$$\epsilon^2 = \frac{1}{N-1} \sum_{i=1}^N \left[\frac{R_i - \bar{R}}{\sigma_i(R)} \right]^2$$

Here $\sigma_{in}(R)$ is an internal error estimate, σ_{ext} is an external error estimate and ϵ^2 is the reduced- χ^2 value. If the set of values (R_i, σ_i) is consistent, ϵ^2 should be about 1.0. In carrying out this calculation, the $\sigma_i(R)$ values used should include only those contributions that are not common to each value. That is, the uncertainties in the branching ratio, efficiency, foil mass and external γ absorption should be excluded. This means that our computer calculated relative errors are appropriate for this analysis. Some samples of the results of this averaging will be discussed below in terms of the peak interferences.

Atomic Masses and Abundances

In the reaction rate expression, Eq. (1), the atomic masses and abundances are needed. It has been noted that for natural abundance foils one can use either the average elemental mass and the abundance in atom percent, or the isotopic mass and the abundance in weight percent. As noted in ANL section , they have used the former set, while we have used the later. For completeness, the atomic masses and abundances used will be included in the following sections on the counting results.

The accuracy and precision of the abundances was also questioned at the recent program meeting. For example, the GE Chart⁽²⁾ lists the ^{58}Fe abundance (in atom%) as 0.31. This implies an error of at least ± 0.005 or an uncertainty of 1.6% in any absolute reaction rate. In order to determine the best available values and errors, we have contacted a member of the US Atomic Weights Committee. If better values are available, they will be circulated.

Random Summing Magnitude

There has been some indication that our random summing corrections are larger than others have measured. Therefore, for comparison purposes we have given in Table ANC-7, the corrections used as a function of counting rate. These Ge(Li) systems use TC-202 amplifiers and Nuclear Data 2200 and 50/50 4096-channel analyzers.

Peak Interferences

One of the purposes of the interlaboratory effort in γ -ray spectrometry is to improve the methods used. In this regard, we wish to present some data on interferences within the peaks that are listed in the evaluated set of decay scheme data⁽¹⁾ used in this program.

A trivial case is the 228-keV lines from ^{239}Np and ^{132}Te . This means that for ^{238}U foils a ^{132}Te reaction rate cannot be obtained and conversely this line should not be used in the ^{239}Np analysis.

Secondly, the ANL group have suggested that the 724-keV line from ^{95}Zr should not be used for the first 30 days after the irradiation. The interference is mainly from the 723-keV line of ^{131}I (8 d). Some information on this question is given in Table ANC-8 for foils irradiated in CFRMF. The apparent reaction rates from the 724-keV line decreases between ~ 25 days and ~ 46 days, while that from the 756-keV line does not. The difference in the reaction rates probably indicates an error in the branching ratios. Because of the later possibility, it is desirable to use both γ rays in the analysis. The individual γ rays have quoted branching ratio uncertainties of $\sim 1\%$, but the sum of the branching ratios has an uncertainty of 0.5%.

In the past we have observed discrepancies between the ^{144}Ce -Pr production rates computed from the 696- and 2185-keV lines. We now have some

instructive data on this problem. And we computed this rate from the 134-keV line. Table ANC-9 gives the reaction rates obtained from these three lines for the ^{235}U foil irradiated in the NBS thermal facility. These data indicate that at 44 days after the irradiation $\sim 20\%$ of the 696-keV peak is from an activity with a half-life of ~ 40 days. Also it appears that at 44 days $\sim 10\%$ of the 134-keV peak may be from a shorter-lived activity. The data for the 2185-keV peak do not indicate any interferences, but here the statistical quality of the data is poor. (Note that the ϵ^2 values clearly indicate which sets of data are inconsistent.) The reaction rates from the later data indicate that the three branching ratios are not consistent. Some unpublished data support this conclusion. It is expected that improved branching ratios will be available for our next evaluation of the decay scheme parameters.

Expected Achievements

Information on the isotopic abundances and their uncertainties will be obtained.

References

1. R. G. Helmer and R. C. Greenwood, Nucl. Tech. 25, 258 (1975).
2. N. E. Holden and F. W. Walker, Chart of Nuclides, Eleventh Edition, 1972.

TABLE ANC-7

RANDOM SUMMING CORRECTIONS FOR ANC SYSTEMS

Count Rate (c/s)	Random Summing Correction Factor	
	OR5	ND5
	ND-2200	ND 50/50
0	1.000	1.000
1000	0.9946	0.9943
2000	0.9890	0.9888
3000	0.9834	0.9824
4000	0.9775	0.9755
5000	0.9717	0.9681

TABLE ANC-8

APPARENT REACTION RATES FROM 724-keV LINE OF ^{95}Zr

Results are given for foils of ^{239}Pu , ^{237}Np and ^{235}U for the 724- and 756-keV lines. Relative errors are given.

Decay Time (days)	Calculated Reaction Rate (10^{-15} atoms/atom-sec)					
	724-keV			756-keV line		
	P9-14	N7-16	U5-11	P9-14	N7-16	U5-11
21	7.45(7)	2.81(4)	8.49(7)	6.92(6)	2.66(3)	8.14(7)
28	7.16(6)	2.73(2)	8.36(7)	6.90(6)	2.64(2)	8.13(7)
48	7.13(5)	2.70(2)	8.25(6)	7.02(5)	2.61(2)	8.17(6)
49	6.97(6)	2.69(2)	8.34(6)	6.89(5)	2.62(2)	8.19(6)
All 4 values -						
average	7.14(9)	2.72(2)	8.35(5)	6.94(3)	2.63(1)	8.16(3)
ϵ^2	11.1	2.8	2.4	1.5	0.73	0.20
Last 2 values -						
average	7.05(8)	2.69(2)	8.29(5)			
ϵ^2	5.4	0.01	1.3			

TABLE ANC-9

CALCULATED REACTION RATES FOR $^{144}\text{Ce-Pr}$

Data are for ^{235}U foils irradiated in NBS thermal facility. Errors are relative values.

Decay Time (days)	Calculated Reaction Rate (10^{-12} atoms/atom-sec)		
	134-keV line	696-keV line	2185-keV line
44	2.744(12)	2.894(66)	2.219(68)
83	2.588(10)	2.516(46)	2.182(50)
85	2.571(9)	2.518(64)	2.248(75)
135	2.502(8)	2.304(42)	2.068(47)
136	2.471(16)	2.252(21)	2.169(23)
141	2.505(14)	2.309(37)	2.221(50)

All 6 values -

average	2.563(36)	2.342(70)	2.170(20)
ϵ^2	70	22	1.48

Last 3 values -

average	2.498(8)	2.272(19)	2.160(32)
ϵ^2	1.67	0.82	2.74

G. FISSION PRODUCT REACTION RATES FOR ^{235}U IN NBS THERMAL FACILITY

R. G. Helmer and R. C. Greenwood

Objective

- Measurement of reaction rates for several fission products for a ^{235}U foil irradiated in the 12 inch cavity in the thermal column of the NBS reactor.

Summary

Reaction rates have been determined for the production of ^{95}Zr , ^{132}Te , ^{140}Ba , ^{103}Ru , ^{95}Zr , ^{144}Ce and ^{137}Cs for thermal neutron irradiation of ^{235}U . The quality of the data is generally good, except for ^{144}Ce -Pr where discrepancies probably exist in the three branching ratios used.

Accomplishments and Status

A high-purity, thin ^{235}U foil (^{235}U -1) was irradiated October 15-16, 1973 in the 12 inch cavity in the thermal column of the NBS reactor.

The physical parameters of this foil are given in Table ANC-10.

Over a period of 3 to 141 days after this irradiation, 14 γ -ray spectra were measured for this foil. The ranges of some of the counting parameters and of the corrections to the measured data are also given in Table ANC-10. Two of the 14 spectra gave reaction rates that were low by 2-8% and have been omitted from the results quoted here.

The individual data for ^{103}Ru are given in Table ANC-11. This shows a typical level of consistency, $\epsilon^2 = 1.76$. In this case, a small discrepancy between the two detectors is indicated since the separate reaction rates differ by $\sim 0.7\%$. The uncertainties shown for the individual values are relative errors (i.e., no branching ratio or efficiency contribution) that do not include any contribution from the foil mass.

Because of the interferences discussed in the preceding article, the following data were rejected: 724-keV line of ^{95}Zr before 30 days decay and 134- and 696-keV lines of ^{144}Ce before 120 days decay. Care must be exercised in the use of the remaining data for these lines since some peak impurity contribution may still be present. The resulting reaction rates are given in Table ANC-12. The ϵ^2 values for the average calculations

indicate that the sets of reaction rates are reasonably consistent. The three uncertainties (in %) represent the total error (σ_3), the relative error (σ_2) and the relative error excluding the foil mass and external absorber contributions (σ_1). Where more than one γ ray is used, σ_1 is given for each value and σ_2 and σ_3 for the average.

These ^{95}Zr data suggest that a small (0.3%) adjustment in each of the branching ratios would produce much better agreement. This would be legitimate since the errors in these values are 0.5%. The same result is obtained by averaging the two values. Similarly, the spread in the three values for ^{144}Ce may be due in part to errors in the branching ratios.

Our counting system has a ^{137}Cs contribution in the room background. This had to be subtracted from the ^{137}Cs observed in the foil spectra and it amounted to $\frac{1}{2}$ to 2% of the total. An 0.5% error contribution has been included in σ_2 for this subtraction.

Expected Achievements

More spectra will be taken in order to get more data on the ^{137}Cs and ^{144}Ce -Pr reaction rates. These results will also be updated after the next evaluation of the decay scheme parameters, especially for ^{144}Ce -Pr.

TABLE ANC-10
PARAMETERS OF ^{235}U -1 FOIL AND THE γ -RAY COUNTING

<u>Parameter</u>	<u>Value</u>
foil mass	0.006330 grams
atomic mass	235.044
abundance ^{235}U	0.99336 - isotopic and purity
foil thickness	0.00011 inches
number of spectra	12 - excluding two rejected ones
Ge(Li) detectors	ND5 and OR5
source - detector distances	10, 25 and 35 cm
correction factor	
- random summing	0.942-0.999
- real summing	0.9988-1.0000-1.0008
- γ self absorption	0.9915-0.9999
- external absorber	1.000 except ^{97}Zr at 0.444

TABLE ANC-11

CONSISTENCY OF REACTION RATES - ^{103}Ru

Both uncertainties are relative values excluding foil mass contribution.

<u>Decay Time (days)</u>	<u>Reaction Rate (10^{-12} atoms/atom-sec)</u>	<u>Error (%)</u>
17.09	1.271(16)	1.27
23.09	1.287(12)	0.96
41.41	1.301(4)	0.32
44.41	1.286(4)	0.32
83.41	1.294(4)	0.32
85.41	1.297(4)	0.29
135.37	1.285(6)	0.43
136.37	1.304(9)	0.72
141.35	1.301(9)	0.67
Average	1.294(2)	0.18
$\epsilon^2 = 1.76$		

TABLE ANC-12

 ^{235}U REACTION RATES FOR NBS THERMAL IRRADIATION

Isotope	Decay Range (days)	No. of Spectra	E_γ (keV)	ϵ^2	Reaction Rate (10^{-12} atom/ atom-sec)		Uncertainty (%)		
					Value	Average	σ_1^a	σ_2^b	σ_3^c
^{97}Zr	3.2	1	743	--	2.547 ^d		1.10	3.2 ^e	3.6
^{132}Te	9-15	2	228	0.6	1.804		0.86	1.00	7.0
$^{140}\text{Ba-La}$	9-41	5	1596	1.6	2.664		0.32	0.59	1.6
^{103}Ru	17-141	9	497	1.8	1.294		0.18	0.53	3.8
^{95}Zr	41-141	7	724	1.6	2.787	2.778	0.19	0.52	1.7
	23-141	8	756	1.0	2.771		0.15		
^{137}Cs	44-141	6	661	0.5	2.588		0.47	0.85 ^f	1.8
$^{144}\text{Ce-Pr}$	135-141	3	134	1.7	2.498	2.31 ^g	0.33	5. ^g	7. ^g
	135-141	3	696	1.2	2.272		0.82		
	44-141	6	2185	1.5	2.170		0.94		

^aRelative uncertainty excluding contribution from foil mass (and for ^{97}Zr , the external absorber).

^bRelative uncertainty, with foil mass error taken to be 0.5%.

^cTotal uncertainty.

^dNo ILRR evaluated decay scheme parameters are available for this nuclide.

^eIncludes 3% uncertainty in external absorption correction ($\delta_6 = 0.444$).

^fIncludes a contribution of 0.5% for the subtraction of the ^{137}Cs observed in the room background.

^gUnweighted average and arbitrary assignment for σ_2 and σ_3 .

H. FISSION PRODUCT REACTION RATES FOR CFRMF IRRADIATION OF ^{235}U , ^{238}U , ^{239}Pu AND ^{237}Np

R. G. Helmer and R. C. Greenwood

Objective

- To determine the production rates for several fission products from the irradiation of ^{235}U , ^{238}U , ^{239}Pu and ^{237}Np in the CFRMF at INEL.

Summary

From γ -ray spectrometry measurements, reaction rates have been determined for the production of ^{97}Zr , ^{132}Te , $^{140}\text{Ba-La}$, ^{103}Ru , ^{95}Zr , ^{137}Cs and ^{144}Ce for most of these fissile materials. The production rate for ^{239}Np was determined for ^{238}U . The results are of reasonable quality except for the long-lived ^{137}Cs and ^{144}Ce ; more measurements will be needed for these isotopes for all four materials.

Accomplishments and Status

High-purity foils of ^{235}U and ^{239}Pu were irradiated at the center of the CFRMF at INEL for ~ 13 hr on January 15, 1974. Similar foils of ^{238}U and ^{237}Np were irradiated in the same place for 14 hr on January 17, 1974. The physical parameters of these foils are given in Table ANC-13. Another ^{235}U foil, $^{235}\text{U-4}$ was irradiated at the monitor foil position. The results from this foil will be reported elsewhere.

Over a period of 1 to 49 days after the irradiation, six γ -ray spectra were measured for each foil. The Ge(Li) detectors ND5 and OR5 were used. The ranges of some of the counting parameters and of the corrections involved are given in Table ANC-14.

The results for the analysis of these data are given in Table ANC-15. As discussed in section ANC-F, the 724-keV from ^{95}Zr has interferences. The data for this peak was used only after 30 days of decay. Even after 30 days, the reaction rate from the 724-keV line is higher than that from the 756-keV line. For $^{144}\text{Ce-Pr}$ the data from the 696-keV is not included because it contains another significant contribution throughout this counting period. A smaller interference occurs in the 134-keV line from ^{144}Ce so this result is given where it is available, but it is not used in the average value.

The data for the long-lived ^{137}Cs and ^{144}Ce generally have poor statistical quality and more experiments are planned to improve this situation. The room background contribution to the ^{137}Cs γ -ray peak is 3-4% for the ^{235}U and ^{239}Pu foils, $\sim 20\%$ for the ^{237}Np foil and $\sim 50\%$ for the ^{238}U foil. Since this correction could not be made accurately for the ^{238}U , no reaction rate is given in this case.

The values of ϵ^2 indicate the degree of consistency of the measurements that were averaged. The results seem quite consistent except for the ^{239}Pu foil where four lines have $\epsilon^2 \geq 2.9$. However, the number of values in the averages (generally 2 or 4) is too small to draw any significant conclusions.

It should be noted that external absorbers were used for all of the ^{239}Pu spectra (i.e., Cd) and the ^{237}Np spectra (i.e., Pb and Cd). These are desirable to reduce the intensity of the natural γ rays from these isotopes. The presence of the thick Pb absorber introduces an estimated 1-2% errors in the absolute reaction rates for the ^{237}Np . However, since this same absorber will be used in measurements with other ^{237}Np samples, it will not introduce large uncertainties in relative measurements. For example, to compare these results with those from BIG-10, the appropriate uncertainties are $\sqrt{\sigma_1^2 + 0.5^2}\%$ where the latter term is from the foil mass. We have included in σ_2 and σ_3 the contributions for the external absorbers as described in Section ANC-F.

Expected Achievements

More spectra will be taken in order to improve the data for the ^{137}Cs and ^{144}Ce -Pr reaction rates.

TABLE ANC-13
PARAMETERS OF FISSION FOILS FOR CFRMF IRRADIATION

<u>Material</u>	<u>Foil Number</u>	<u>Foil Mass (g)</u>	<u>Atomic Weight</u>	<u>Isotopic Abundance^a (weight-fraction)</u>	<u>Foil Thickness (inches)</u>
²³⁵ U	U5-11	0.30209	235.044	0.99336	0.0053
²³⁸ U	U8-16	0.30967	238.051	0.9948	0.0053
²³⁹ Pu	P9-14	0.26506	239.052	0.9738	0.0057
²³⁷ Np	N7-16	0.37385	237.048	1.0000	0.0083

^aIncludes both isotopic abundance and purity.

TABLE ANC-14
PARAMETERS OF γ -RAY COUNTING OF FISSION FOILS
IRRADIATED IN CFRMF

Foil Number	<u>U5-11</u>	<u>U8-16</u>	<u>P9-14</u>	<u>N7-16</u>
Number of Spectra	6	6	6	6
Decay Times (days)	1-48	1-46	1-49	1-47
Source-Detector Distances (cm)	10,35	10,25	10,35	10,25,35
External Absorbers	none	none	Cd-0.044"	Cd-0.044" Pb-8 g/cm ²
Range of Correction Factors				
random summing	.968-.994	.986-1.000	.972-.996	.979-.998
real summing	.993-1.001	.993-1.001	.993-1.001	.993-1.001
γ -self absorption ^a	.662-.994	.662-.994	.891-.993	.957-.989
external absorption ^a	--	--	.807-.966	.265-.687

^aThe lowest values correspond to the following γ rays:

U5-11 134 keV from ¹⁴⁴Ce

U8-16 134 keV from ¹⁴⁴Ce

P9-14 228 keV from ¹³²Te

N7-16 497 keV from ¹⁰³Ru

TABLE ANC-15

REACTION RATES FOR CFRMF IRRADIATION

Foil	Isotope	Decay Range (days)	No. of Spectra	E _γ (keV)	ε ^{2a}	Reaction Rate (10 ⁻¹⁵ atoms/atom-sec)		Uncertainty (%)		
						Value	Average	^b σ ₁	^c σ ₂	^d σ ₃
U5-11	⁹⁷ Zr	1-2	2	743	0.2	7.665 ^e		0.49	0.70	1.7
	¹³² Te	1-2	2	228	0.1	6.187		0.40	0.64	7.0
	¹⁴⁰ Ba-La	22-48	4	1596	0.2	7.752		0.29	0.58	1.6
	¹⁰³ Ru	22-48	4	497	0.2	4.223		0.29	0.58	3.8
	⁹⁵ Zr	45-58	2	724	1.3	8.292	8.207 ^f	0.55	0.66	1.7
		22-48	4	756	0.2	8.164		0.37		
	¹³⁷ Cs	45-48	2	661	0.3	7.77 ^h		3	3	3
U8-16	¹⁴⁴ Ce-Pr	45-48	2	134	2.1	(7.56) ^g	7.33 ^g	1.3	9	10
		45-48	2	2185	2.6	7.33		9		
	²³⁹ Np	1-4	2	278	0.5	14.67		0.23	0.55	2.1
	¹⁴⁰ Ba-La	21-46	4	1596	1.0	0.3741		0.34	0.60	1.6
	¹⁰³ Ru	21-46	4	497	0.2	0.3956		0.29	0.58	3.8
	⁹⁵ Zr	43-46	2	724	4.9	0.3279	0.3213 ^f	1.38	0.8	1.8
		21-46	4	756	1.8	0.3181		0.60		
P9-14	¹⁴⁴ Ce	43-46	2	134	0.2	(0.343) ^g	0.416 ^g	2.2	34.	34.
		43-46	2	2185	6.	0.416		34.		
	⁹⁷ Zr	1-2	2	743	0.5	7.921 ^e		0.43	0.71	1.7
	¹³² Te	1-2	2	228	0.0	7.949		0.37	0.67	7.0
	¹⁴⁰ Ba-La	21-49	4	1596	2.9	7.765		0.53	0.77	1.7
	¹⁰³ Ru	21-49	4	497	3.5	10.22		0.54	0.78	3.8
	⁹⁵ Zr	48-49	2	724	5.4	7.048	6.975 ^f	1.18	0.73	1.9
21-49		4	756	1.5	6.939	0.47				
	¹³⁷ Cs	48-49	2	661	4.7	9.34 ^h		6.1	6.1	6.3
	¹⁴⁴ Ce	48-49	2	2185	0.4	7.72		6.5	6.5	8.5

TABLE ANC-15 (Continued)

Foil	Isotope	Decay Range (days)	No. of Spectra	E_{γ} (keV)	ϵ^2 ^a	Reaction Rate (10^{-15} atoms/atom-sec)		Uncertainty (%)		
						Value	Average	σ_1 ^b	σ_2 ^c	σ_3 ^d
N7-16	⁹⁷ Zr	1-4	2	743	0.3	2.860 ^e		0.79	1.77	2.4
	¹⁴⁰ Ba-La	19-47	4	1596	0.3	2.566		0.28	1.15	1.9
	¹⁰³ Ru	19-47	4	497	0.0	2.738		0.32	3.06	4.8
	⁹⁵ Zr	46-47	2	724	0.1	2.693	2.652 ^f	0.63	1.64	2.4
		19-47	4	756	0.7	2.631		0.45		
	¹³⁷ Cs	46-47	2	661	0.1	2.87 ⁱ		10.6	10.7	11.
	¹⁴⁴ Ce	47	1	2185	--	2.24		8.	8.1	9.7

^a ϵ^2 value for average of reaction rates from one γ ray.

^bRelative error excluding terms from foil mass and external absorbers.

^cRelative error with foil mass uncertainty of 0.5%. See text for discussion of external absorber contribution.

^dTotal uncertainty.

^eNo ILRR evaluated decay scheme parameters are available.

^fAverage weighted by number of spectra.

^gAs noted in the text, the average is based on the data for the 2185-keV γ ray.

^hRoom background subtraction is 3-4% of gross rate; σ_1 includes 1% from this subtraction.

ⁱRoom background subtraction is 19-22% of gross rate; σ_1 includes 7% from this subtraction.

I. REACTION RATES FOR BIG-10 IRRADIATION OF NONFISSION FOIL PACKET

R. G. Helmer and R. C. Greenwood

Objective

- To determine the reaction rates for the various foils in a nonfission foil packet irradiated in the BIG-10 facility at LASL.

Summary

From γ -ray spectrometry measurements, the following reaction rates have been determined for an irradiation in BIG-10: $^{198}\text{Au}(n,\gamma)$, $^{63}\text{Cu}(n,\gamma)$, $^{58}\text{Ni}(n,p)$, $^{54}\text{Fe}(n,p)$, $^{58}\text{Fe}(n,\gamma)$, $^{45}\text{Sc}(n,\gamma)$, $^{46}\text{Ti}(n,p)$, $^{47}\text{Ti}(n,p)$, $^{48}\text{Ti}(n,p)$, $^{235}\text{U}(n,f)^{140}\text{Ba}$, $^{238}\text{U}(n,\gamma,\beta^-)^{239}\text{Np}$ and $^{238}\text{U}(n,f)^{140}\text{Ba}$. The activity levels for several foils were too low to be determined with usable accuracy, and the accuracy of the results for some of the above reactions were limited by the counting statistics.

Accomplishments and Status

A packet containing 21 foils of 12 different materials was irradiated for 220 minutes on August 13, 1974 in the BIG-10 facility at LASL. The physical parameters of these foils are listed in Table ANC-16. The isotopic abundances are based on the Chart of the Nuclides⁽¹⁾. Although several of these abundances are quoted to only two significant figures and therefore are uncertain by $\sim 1\%$, this error contribution has not been included in the following analyses.

These foils were shipped to the INEL and γ -ray counting began on August 16. Generally two spectra were obtained for each foil. The Ge(Li) detectors ND5 and OR5 were used. The ranges of the counting parameters and of the corrections involved are given in Table ANC-17. These data were obtained over a short period of time (i.e., no more than 17 days decay time). This was possible because of the simplicity of the spectra involved and the lack of interfering γ -ray lines. The only external absorbers used were 0.012" Cu on each side of the ^{64}Cu sample to stop the positrons.

The reaction rates computed from these data are given in Table ANC-18. For the isotopes reported, the only γ -ray lines in the ILRR evaluated decay scheme data⁽²⁾ that were not used are: 228 keV of ^{239}Np which includes a ^{132}Te contribution and the 1120-keV line of ^{46}Sc in the Ti foils which has a room background contribution. No useful data were obtained for ^{24}Na and ^{27}Mg from the Al foil, ^{54}Mn from Mn, and $^{115\text{m}}\text{In}$ and $^{116\text{m}}\text{In}$ from In. These activities were too weak following the three day decay time to measure on the Ge(Li) detectors. The ^{60}Co results are also not included in Table ANC-18. Due to their low activity level and a room background contribution, these three foils were each counted on the face of a Ge(Li) detector. A crude determination of the detector efficiency has been made for this source position. This gives a reaction rate of $\sim 1.13 \times 10^{-15}$ atoms/atom-sec, averaged over three foils and two γ rays. This result is noted here only as an aid in resolving possible discrepancies between other measurements; it should not be included in any summary analysis.

The results in Table ANC-18 were computed on the basis that the power during the irradiation was constant. Since the first counting was done three days after the irradiation, and the shortest half-life involved is 12.7 hr, this should be reasonably accurate.

The several large values of σ_1 in Table ANC-18 result primarily from the low activity level of these foils. This is illustrated in Table ANC-17 by the fact that there is no random summing correction, except for the ^{238}U and ^{235}U foils.

Expected Achievements

This work is complete.

References

1. N. E. Holden and F. W. Walker, Chart of Nuclides, Eleventh Edition, 1972.
2. R. G. Helmer and R. C. Greenwood, Nucl. Tech. 25, 258 (1975).

TABLE ANC-16
PARAMETERS OF FOILS IN BIG-10 NONFISSION PACKET

<u>Material</u>	<u>Number</u>	<u>Foil Mass (g)</u>	<u>Atomic Weight</u>	<u>Isotope Abundance (weight fraction)</u>	<u>Elemental Purity (fractional)</u>	<u>Foil Thickness (inches)</u>
Au	AU-5	0.02947	196.970	1.0000	1.0000	0.0005
	AU-3	0.00298				0.00005
	AU-6	0.03154				0.0005
Cu	CU-3	0.01503	62.9296	0.6843	1.0000	0.0005
Ni	NI-MO	0.29363	57.935	0.6743	0.9962	0.010
Fe	FE-C	0.78235	57.9333	0.00322	0.9977	0.031
			53.9396	0.05605		
Sc	SC-4	0.04713	44.9559	1.0000	0.999	0.005
Ti	TI-LO	0.15741	47.9479	0.7381	0.9973	0.010
			46.9518	0.0736		
			45.9526	0.0768		
	TI-LP	0.15723	47.9479	0.7381	0.9973	0.010
			46.9518	0.0736		
			45.9526	0.0768		
²³⁸ U	²³⁸ U-5	0.29314	238.051	0.9948 ^a	1.0000	0.005
²³⁵ U	²³⁵ U-3	0.24279	235.044	0.99336 ^a	0.9372	0.0048

^aIncludes some contribution from the elemental purity

TABLE ANC-17

PARAMETERS OF γ -RAY COUNTING OF BIG-10 NONFISSION PACKET

Foil	Isotope	Source-Detector Distance (cm)	Range of Correction Factors		
			Random Summing	Real Summing	γ -Self Absorption
AU-5	^{198}Au	10	1.0000	1.0000	0.9975
AU-3		10			0.9997
AU-6		10			0.9975
CU-3	^{64}Cu	10	1.0000	1.0000	0.9996
NI-MO	^{58}Co	10	1.0000	0.9999	0.9920
FE-C	^{54}Mn	5	1.0000	1.0000	0.9785
	^{59}Fe			~ 0.999	0.9816
SC-4	^{46}Sc	10	1.0000	0.9925-0.9941	0.9990
TI-L0	^{48}Sc	10	1.0000	0.9832-0.9932	0.9960-0.9980
	^{47}Sc			1.0000	0.9900
	^{46}Sc			0.9925-0.9941	0.9970
TI-LP	^{48}Sc	10	1.0000	0.9832-0.9932	0.9960-0.9980
	^{47}Sc			1.0000	0.9900
	^{46}Sc			0.9925-0.9941	0.9970
^{238}U	^{239}Np	10	0.9883-1.0000	0.9996	0.9235
	$^{140}\text{Ba-La}$			0.9934-0.9943	0.9935
^{235}U	$^{140}\text{Ba-La}$	10	0.9917-0.9941	0.9934-0.9943	0.9943

TABLE ANC-18

REACTION RATES FOR NONFISSION FOIL PACKET IRRADIATED IN BIG-10

Foil	Isotope	Decay Range (days)	Number of Spectra	E _γ (keV)	ε ²	Reaction Rate (10 ⁻¹⁵ atoms/atom-sec)		Uncertainty (%) ^a		
						Value	Average ^b	σ ₁	σ ₂	σ ₃
AU-5	¹⁹⁸ Au	3-6	2	411	1.8	18.42		0.54	0.74	1.7
AU-3	¹⁹⁸ Au	3-6	2	411	3.2	18.74		0.75	0.90	1.8
AU-6	¹⁹⁸ Au	3-6	2	411	1.0	18.39		0.41	0.65	1.6
CU-3	⁶⁴ Cu	3	1	511	--	1.962		1.3	1.71	4.9
NI-MO	⁵⁸ Co	9-15	2	810	0.1	1.336		0.43	0.66	1.6
FE-C	⁵⁴ Mn	14-17	2	834	0.3	0.986		0.82	1.78	2.3
	⁵⁹ Fe	14-17	2	1099 1291	0.0 0.9	0.343 0.321	0.334	2.34 2.81	2.40	4.0
SC-4	⁴⁶ Sc	8-9	2	889	1.1	1.445	1.442	0.52	0.62	1.6
				1120	0.7	1.440		0.50		
TI-L0	⁴⁶ Sc	3-6	2	889	1.3	0.1437		2.36	2.41	2.8
	⁴⁷ Sc	3-6	2	159	11.2	0.2267		1.66	1.73	4.3
	⁴⁸ Sc	3-6	2	983	4.5	0.003716	0.003751	2.85	1.06	1.8
				1037	0.0	0.003791		1.39		
1312				0.5	0.003716	1.44				
TI-LP	⁴⁶ Sc	3-7	2	889	1.4	0.1414		2.56	2.61	3.0

ANC-42

TABLE ANC-18 (Continued)

Foil	Isotope	Decay Range (days)	Number of Spectra	E_{γ} (keV)	ϵ^2	Reaction Rate (10^{-15} atoms/atom-sec)		Uncertainty (%) ^a		
						Value	Average ^b	σ_1	σ_2	σ_3
TI-LP	⁴⁷ Sc	3-7	2	159	2.4	0.2276		0.70	0.86	4.0
	⁴⁸ Sc	3-7	2	983	0.2	0.003837	0.003838	1.72	1.23	1.9
				1037	0.0	0.003835		1.82		
				1312	1.9	0.003848		2.56		
²³⁵ U-3	¹⁴⁰ Ba-La	13	2	1596	3.2	6.822		0.79	0.93	1.8
²³⁸ U-5	²³⁹ Np	3-6	2	278	0.5	12.42		0.41	0.65	2.2
	¹⁴⁰ Ba-La	10-13	2	1596	0.0	0.2448		0.55	0.74	1.7

^aSee Table ANC-12, page ANC-31 for definitions. For ⁶⁴Cu the σ_2 includes a 1% contribution for the external absorber correction; no other external absorbers were used. For ⁵⁹Fe and ⁵⁴Mn, the σ_2 value includes 1.5% contribution for use of an indirectly calibrated geometry.

^bAverage weighted by $\frac{1}{\sigma_1^2}$.

J. PRODUCTION RATES FOR FISSION PRODUCTS FOR IRRADIATION OF ^{235}U , ^{238}U , ^{239}Pu , ^{237}Np IN BIG-10

R. G. Helmer and R. C. Greenwood

Objective

- To determine the reaction rates for several fission products for foils of ^{235}U , ^{238}U , ^{239}Pu and ^{237}Np irradiation in the BIG-10 facility at LASL.

Summary

From γ -ray spectrometry measurements, reaction rates for the production of ^{97}Zr , ^{132}Te , ^{239}Np , $^{140}\text{Ba-La}$, ^{103}Ru and ^{95}Zr have been measured for neutron irradiation of ^{235}U , ^{238}U , ^{239}Pu and ^{237}Np in BIG-10. The quality of the results is somewhat limited by the activity level of the foils, especially for the ^{238}U .

Accomplishments and Status

A packet containing foils of ^{235}U , ^{238}U , ^{239}Pu and ^{237}Np was irradiated for 220 minutes on July 23, 1974 in the BIG-10 facility at LASL. The physical parameters of these foils are listed in Table ANC-19. It should be noted that for ^{237}Np an isotopic abundance and chemical purity value of 97.5% was used. Whereas in the CFRMF analysis a value of 100% was used. No uncertainty in this quantity is included in the analysis.

The measurements on these foils were begun two days after the irradiation. The ranges of the various counting parameters and correction factors are given in Table ANC-20. As noted, an external Cd absorber was used for the ^{239}Pu foil and Pb and Cd were used with the ^{237}Np foil. The uncertainties included for the absorption correction are given in article ANC-F.

The reaction rates computed for these foils are given in Table ANC-21. As discussed in section ANC-F, the data from the 724-keV line of ^{95}Nb is included only after 30 days of decay. In spite of this, the 724-keV result is generally higher than the value from the 756-keV line. Some consideration needs to be given to the method of obtaining the best ^{95}Zr reaction rate.

The ϵ^2 values indicate that several of the ^{235}U results are less consistent than usual (i.e., three ϵ^2 values ≥ 4).

The relative uncertainties, σ_1 , are generally larger than the values for other irradiations. This is primarily due to the lower activity levels of these foils. As a result of this low activity, the spectra used to obtain the ^{103}Ru values (i.e., all those taken after 3 days decay) were counted for between 11 and 66 hours. This means we will have some difficulty obtaining good reaction rates for the longer lived ^{144}Ce and ^{137}Cs .

Expected Achievements

More spectra will be measured in order to obtain the reaction rates for ^{144}Ce and ^{137}Cs .

TABLE ANC-19
PARAMETERS OF FOILS IN BIG-10 FISSION FOIL PACKET

<u>Foil</u>	<u>Foil Mass (g)</u>	<u>Atomic Weight</u>	<u>Isotope Abundance^a (weight-fraction)</u>	<u>Foil Thickness (inches)</u>
U5-8	0.28672	235.044	0.99336	0.0051
U8-11	0.30943	238.051	0.9948	0.0052
P9-11	0.27650	239.052	0.9738	0.0057
N7-13	0.37512	237.048	0.975	0.008

^aIncludes both isotopic and chemical purity.

TABLE ANC-20

γ-RAY COUNTING PARAMETERS FOR BIG-10
FISSION FOIL PACKET

Foil Number	<u>U5-8</u>	<u>U8-11</u>	<u>P9-11</u>	<u>N7-13</u>
Number of Spectra	6	7	6	5
Decay Times (days)	2-69	2-73	2-71	2-64
Source-Detector Distances (cm)	10,25	5,10	10,25	10,25
External Absorbers	none	none	Cd-0.044"	Cd-0.044" Pb-8 g/cm ²
Range of Correction Factors				
random summing	.982-.998	.987-1.000	.989-.991	.973-.996
real summing	.993-1.000	.984-1.000	.993-1.000	.993-1.000
γ-self absorption	.890-.994	.920-.993	.891-.994	.958-.989
external absorption	---	---	.807-.961	.265-.654

TABLE ANC-21

REACTION RATES FOR FISSION FOILS IRRADIATED IN BIG-10

Foil	Isotope	Decay Range (days)	No. of Spectra	E_{γ} (keV)	ϵ^2	Reaction Rate (10^{-15} atoms/atom-sec)		Uncertainty ^a		
						Value	Average ^b	σ_1	σ_2	σ_3
ANC-48	U5-8	⁹⁷ Zr	2-3	2	743	0.0	6.657	0.48	0.69	1.7
		¹³² Te	2-14	3	228	8.7	5.381	0.82	0.96	7.0
		¹⁴⁰ Ba-La	14-62	3	1596	4.0	6.749	0.66	0.83	1.7
		¹⁰³ Ru	14-69	4	497	5.0	3.754	0.63	0.80	3.8
		⁹⁵ Zr	62-69	2	724	2.2	7.318	0.61	0.71	1.9
			14-69	4	756	2.2	7.171	0.50		
	U8-11	⁹⁷ Zr	2-3	2	743	0.5	0.2374	0.66	0.83	1.8
		²³⁹ Np	2-3	2	278	0.0	12.42	0.40	0.64	2.1
		¹⁴⁰ Ba-La	15-65	3	1596	1.5	0.2495	0.59	0.77	1.7
		¹⁰³ Ru	15-73	5	497	1.5	0.2645	0.49	0.70	3.8
		⁹⁵ Zr	64-73	3	724	2.0	0.2182	1.60	1.31	2.2
			15-73	5	756	3.1	0.2134	1.21		
P9-11		⁹⁷ Zr	2-3	2	743	0.0	7.206	0.46	0.72	1.7
		¹³² Te	2-3	2	228	12.7	7.112	1.06	1.20	7.1
		¹⁴⁰ Ba-La	16-63	3	1596	0.2	7.109	0.35	0.66	1.6

TABLE ANC-21 (Continued)

Foil	Isotope	Decay Range (days)	No. of Spectra	E_{γ} (keV)	ϵ^2	Reaction Rate (10^{-15} atoms/atom-sec)		Uncertainty ^a		
						Value	Average ^b	σ_1	σ_2	σ_3
P9-11	^{103}Ru	16-71	4	497	0.7	9.360		0.28	0.63	3.8
	^{95}Zr	63-71	2	724	0.0	6.393	6.395	0.46	0.68	1.9
		16-71	4	756	1.1	6.396		0.39		
N7-13	^{97}Zr	2-3	2	743	1.7	2.294		0.95	1.84	2.4
	$^{140}\text{Ba-La}$	14-64	3	1596	0.1	2.068		0.46	1.21	1.9
	^{103}Ru	14-64	3	497	0.8	2.178		0.54	3.09	4.8
	^{95}Zr	64	1	724	--	2.190	2.117	0.98	1.71	2.5
		14-64	3	756	0.6	2.093		0.65		

^aSee Table ANC-15, p. ANC-36 for definitions of σ_i . The σ_2 and σ_3 values for ^{239}Pu and ^{237}Np do include contributions from the external absorbers.

^bAverage is weighted by the number of spectra used.

ANL

ARGONNE NATIONAL LABORATORY

A. RECALIBRATION OF Ge(Li) DETECTOR EFFICIENCY

R. J. Popek and R. R. Heinrich

Objective

- To maintain the most accurate detector efficiency based on absolute calibrated gamma-ray standards.

Summary

The method of standardization is reviewed. Two separate efficiency *vs.* energy curves are maintained; the K-factor curve remains unchanged while the curve used for nonfission rate analyses periodically (annually) is reevaluated. Also presented are the nuclear and physical constants currently being used in the analysis of the Ge(Li) counting data.

Accomplishments and Status

The accuracy limitation of absolute gamma-ray counting is in the detector efficiency determination for the gamma-ray detector. Detector efficiency calibrations are commonly made by counting radioactive standards whose disintegration rates have been determined in an absolute manner by techniques other than gamma counting. Our laboratory utilizes absolute standards supplied by the National Bureau of Standards (NBS) and the Amersham-Searle Corporation. In general, the precision between the absolute standards has been $\pm 1.2\%$ and the quoted accuracy ranges from 0.8 to 1.8%.

In our laboratory we have established two efficiency curves for each detector, one being used for K-factors and the other used for all nonfission-rate analyses. The K-calibration (efficiency) curve and the nuclear constants are never changed once fission yields (which are, in effect, K-factors) are established. Every fission-rate measurement on that detector uses its own fission yield and the permanent efficiency curve. Each detector has somewhat different K-factor yields, but they all give the same value for a fission-rate measurement since any biases are cancelled. Currently, we have established K-factor yields for ^{235}U from the NBS thermal irradiation and for ^{235}U , ^{238}U , ^{237}Np , and ^{239}Pu applicable to CFRMF. These factors will remain in use until a specific detector is found to exhibit a change in efficiency. We monitor the efficiency every week with a pair of standards counted in a defined manner. The advantage of the K-factor technique is that

uncertainties in branching ratios and detector efficiency calibrations cancel, leaving the dominate error being the uncertainty in the NBS fission-rate measurement.

The detector efficiency curves used for the measurement of nonfission rates is required by the ANL Quality Assurance Division to be evaluated at least once annually to assure the utmost accuracy in reaction-rate measurements. This efficiency curve for each Ge(Li) detector has been recently reevaluated and as a result is now more precisely defined by the use of new standards and more extensive measurements. Data reported here for measured fission product yields and reaction rates reflect these recent changes in the detector efficiencies.

For completeness and general information, we have included the ANC¹ recommended nuclear data in Tables ANL-I and -II being used in the evaluation of our fission-product and nonfission nuclei. In Table ANL-III are listed the physical constants for the calculation of nuclear target atoms based on the formula:

$$N_o = \frac{mnpq}{a}$$

N_o = target atoms

m = foil mass

n = foil chemical purity

p = isotopic abundance in atom percent for isotope of interest

q = 6.023×10^{23}

a^* = atomic weight of element

Expected Accomplishments

Additional absolute gamma-ray standards will be evaluated as they become commercially available.

*Must be calculated if other than natural occurring isotopic composition.

¹R. G. Helmer and R. C. Greenwood, Nucl. Technol. 25, 258 (1975).

TABLE ANL-I
NUCLEAR DECAY DATA FOR NONFISSION NUCLEI

Isotope	Half-Life	Gamma-Ray Energy (keV)	Gamma-Ray Intensity (%)
^{24}Na	15.00(2) h	1368.60(3) 2753.98(10)	99.993(2) 99.84(3)
^{27}Mg	9.46(2) min	843.73(4) 1014.44(5)	71.4(5) 28.6(5) } sum = 99.964(4)
^{46}Sc	83.85(10) days	889.258(18) 1120.516(25)	99.984(6) 99.987(6)
^{47}Sc	3.39(4) days	159.39(5)	69.0(25)
^{48}Sc	43.8(1) h	983.4(2) 1037.4(2) 1311.8(4)	99.987(2) 97.5(3) 99.992(2)
^{54}Mn	312.6(3) days	834.827(21)	99.97(2)
^{58}Co	71.23(15) days	810.757(21)	99.44(5)
^{59}Fe	44.6(1) days	1099.224(25) 1291.564(28)	55.5(17) 44.1(12) } sum = 99.6(1)
^{60}Co	5.268(5) yr	1173.208(25) 1332.464(30)	99.86(2) 99.986(2)
^{64}Cu	12.701(7) h	511.002	36.8(16)
$^{115\text{m}}\text{In}$	4.50(2) h	336.2(1)	47.(2)
$^{116\text{m}}\text{In}$	54.03(20) min	1293.4(3) 2112.1(4)	83.4(15) 16.5(15) } sum = 99.94(1)
^{198}Au	2.696(2) days	411.794(8)	95.48(10)
^{239}Np	2.355(4) days	228.19(1) 277.60(3)	11.3(2) 14.3(2)

TABLE ANL-II
NUCLEAR DECAY DATA FOR FISSION-PRODUCT NUCLEI

Isotope	Half-Life		Gamma-Ray Energy (keV)	Gamma-Ray Intensity (%)
⁹⁵ Zr	64.4(5)	days	724.184(18) 756.715(19)	44.2(5) 54.8(5) sum = 99.0(5)
¹⁰³ Ru	39.43(10)	days	497.08(1)	89.(3)
¹³¹ I	8.05	days	364.5	82.(3) ^a
¹³² Te	77.9(5)	h	228.16(6)	88.5(60)
¹³⁷ Cs	30.03(15)	yr	661.638(19)	85.0(3)
¹⁴⁰ Ba	12.79(1)	days ^b		
¹⁴⁰ La	40.26(2)	h ^b	1596.18(5)	95.33(16)
¹⁴¹ Ce	32.5	days	145.6	50.(2) ^a
¹⁴³ Ce	1.37	days	293.0	46.(2) ^a
¹⁴⁴ Ce	284.4(4)	days	133.53(3)	10.7(4)
¹⁴⁴ Pr	17.28(5)	min	696.492(19) 2185.608(46)	1.49(15) 0.77(4)

^a Nominal 3% error assigned.

^b The ratio $T_{1/2}(\text{Ba})/[T_{1/2}(\text{Ba}) - T_{1/2}(\text{La})]$, which is also the ratio of the ¹⁴⁰La to ¹⁴⁰Ba activities at equilibrium, is therefore 1.15096(12).

TABLE ANL-III
PHYSICAL CONSTANTS FOR TARGET ATOM DETERMINATIONS

Reaction	Atomic Weight ^a of Element (grams)	Isotopic ^a Abundance (atom %)
$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	26.982	100
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	196.97	100
$^{115}\text{In}(n,n')^{115\text{m}}\text{In}$	114.82	95.7
$^{59}\text{Co}(n,\gamma)^{60}\text{Co}$	58.93	100
$^{58}\text{Ni}(n,p)^{58}\text{Co}$	58.71	68.3
$^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$	55.847	0.31
$^{54}\text{Fe}(n,p)^{54}\text{Mn}$	55.847	5.8
$^{48}\text{Ti}(n,p)^{48}\text{Sc}$	47.90	73.7
$^{47}\text{Ti}(n,p)^{47}\text{Sc}$	47.90	7.5
$^{46}\text{Ti}(n,p)^{46}\text{Sc}$	47.90	8.0
$^{45}\text{Sc}(n,\gamma)^{46}\text{Sc}$	44.96	100
$^{238}\text{U}(n,f)\text{FP}$	238.04	
$^{235}\text{U}(n,f)\text{FP}$	235.04	
$^{237}\text{Np}(n,f)\text{FP}$	237.05	
$^{239}\text{Pu}(n,f)\text{FP}$	239.05	

^aN. E. Holden and F. W. Walker, Chart of the Nuclides, Eleventh Ed. (1972).

B. ^{235}U THERMAL NEUTRON FISSION YIELDS RELATIVE TO NBS FISSION MEASUREMENTS
TEST 74-3

R. J. Popek, R. R. Heinrich, and N. D. Dudey

Objective

- To measure fission yields by means of activation techniques relative to the NBS fission chambers for the purpose of establishing energy dependence of fission yields used for fast-neutron dosimetry applications.

Summary

Thermal neutron fission yields of ^{235}U measured relative to the NBS fission chamber have been revised to reflect the latest NBS fission rate and our more precise detector calibrations. In general, the yields are in good agreement with ENDF/B recommended thermal neutron yields.

Accomplishments and Status

A ^{235}U foil was irradiated in the NBS thermal cavity, Test 74-3, for the purpose of measuring fission-product yields of ^{235}U in a thermal neutron spectrum. These irradiations were described in a memo to ILRR participants on January 22, 1974, from D. M. Gilliam *et al.* The fission-product reaction rates were determined by Ge(Li) counting over a time period of 1.3-176 days after the irradiation. Each fission product was counted at least four times, and the reaction rates and total fission-product atoms were obtained using the most recently revised detector efficiencies described previously and represent an average of the replicate counts. The total number of atoms for each fission product was divided by $6.08 \times 10^{13} \pm 2.0\%$, which represents the total number of fissions occurring in the sample, as measured by NBS and reported in the 9th ILRR Progress Report. These revised measured fission yields are tabulated in Table ANL-IV together with the different measures of uncertainty. The relative error represents the counting precision of replicate counts, the absolute error is the uncertainty associated with the determination of the fission-product atoms and excludes any uncertainty in the number of fissions, and the total error represents all uncertainties including the 2.0% assigned to the total number of fissions.

The significance of these results will be discussed in detail when the results from each of the four participating laboratories are compared during the next reporting period.

TABLE ANL-IV
²³⁵U THERMAL NEUTRON FISSION YIELDS RELATIVE
 TO THE NBS FISSION MEASUREMENTS.^a TEST 74-3

Isotope	Energy (keV)	Counting Time (days) ^b	Fission Yield (%)	Error (%)		
				Relative ^c	Absolute ^d	Total ^e
⁹⁵ Zr	724,757	126-176	6.62	0.2	2.3	3.0
⁹⁹ Mo	777	1.3-1.9	6.07	1.7	2.7 ^f	3.4 ^f
¹⁰³ Ru	497	35-176	3.06	0.4	4.0	4.5
¹³¹ I	364	35-48	2.86	0.6	2.1 ^f	2.9 ^f
¹³² Te(¹³² I)	773	1.3-1.9	4.22	0.2	2.0 ^f	2.9 ^f
¹³³ I	530	1.3-1.9	6.58	0.2	2.0 ^f	2.9 ^f
¹³⁷ Cs	662	126-176	6.11	1.0	2.3	3.0
¹⁴⁰ Ba	1596	35-66	6.20	0.2	2.0	2.9
¹⁴¹ Ce	146	35-176	5.61	1.3	2.4 ^f	3.1 ^f
¹⁴³ Ce	293	1.3-1.9	5.48	0.2	2.0 ^f	2.9 ^f
¹⁴⁴ Ce	134	126-176	5.64	1.1	2.3 ^f	3.0 ^f

^a6.08 x 10¹³ ± 2.0% total fissions.

^bPeriod of time, in days, during which the sample was counted measured relative to the end of the irradiation.

^cCounting reproducibility, precision of replicate counts.

^dAll known sources of error, except as noted in footnote f and excluding error in the NBS fission-rate measurement.

^eAll known errors associated with the fission yield, except as noted in footnote f.

^fNo error assigned to branching ratio.

Expected Accomplishments

Thermal neutron irradiations of ^{235}U and ^{239}Pu are scheduled. Fission-product yields and K-factors will be determined for ^{239}Pu and repeated for ^{235}U .

C. FISSION-PRODUCT REACTION RATES FROM CFRMF TESTS ANL 74-7A AND 74-7B

R. J. Popek, R. R. Heinrich, and N. D. Dudey

Objective

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

Summary

Final fission-product reaction rates have been determined by Ge(Li) γ -ray spectrometry for the fissile nuclides of ^{235}U , ^{238}U , ^{237}Np , and ^{239}Pu . Irradiation of these nuclides were performed at the CFRMF facility in Tests 74-7A and 74-7B. The production rate of ^{239}Np was also determined for ^{238}U .

Accomplishments and Status

High purity foils of ^{235}U and ^{239}Pu were irradiated in the CFRMF facility on January 24, 1974, for a period of 7 hr (Test 74-7A). ^{238}U and ^{237}Np foils were irradiated later on the same day for the same length of time (Test 74-7B). Foil parameters have been described previously in the 4th and 5th ILRR Progress Reports.

Each foil was gamma counted a minimum of two times over a period of 1-380 days utilizing each of our three Ge(Li) detectors. In the case of ^{95}Zr , the 724-keV gamma ray was not used until after 35 days of decay; the 756-keV line was used after 20 days of decay. The analysis of these data is presented in Table ANL-V. A thick (1503 mg/cm^2) Pb absorber was used for the ^{237}Np spectra to reduce the naturally occurring gamma rays. The use of this absorber introduces to the absolute reaction rate a 0.4% error for the high energy reaction product (1596-keV ^{140}Ba) and a 1.1% error for the low energy reaction product (497-keV ^{103}Ru).

Expected Accomplishments

Additional counting will be made to possibly improve the precision of the ^{137}Cs and to accumulate data on other long-lived fission products.

TABLE ANL-V
FISSION-PRODUCT RATES FROM CFRMF TESTS ANL 74-7A AND 74-7B

Reaction	Sample	Fission Product	10 ⁻¹⁵ atoms/ atom-sec	Reaction Rate	
				% Error (1σ)	
				Relative ^a	Absolute ^b
²³⁵ U(n,f)	U5-13	⁹⁵ Zr	8.15	0.5	2.3
		¹⁰³ Ru	4.17	1.7	4.3
		¹³¹ I	4.15	1.6	3.9 ^c
		¹³⁷ Cs	7.72	1.2	2.4
		¹⁴⁰ Ba	7.60	1.3	2.4
		¹⁴³ Ce	6.74	1.0	3.7 ^c
²³⁸ U(n,f)	U8-18	⁹⁵ Zr	0.326	0.5	2.3
		¹⁰³ Ru	0.395	0.5	4.0
		¹³¹ I	0.203	0.7	3.7 ^c
		¹⁴⁰ Ba	0.374	1.5	2.5 ^c
		¹⁴³ Ce	0.268	2.1	4.2 ^c
²³⁸ U(n,γ)		²³⁹ Np	14.1	1.0	3.7
²³⁷ Np(n,f)	N7-18	⁹⁵ Zr	2.52	4.0	4.6
		¹⁰³ Ru	2.61	1.9	4.4
		¹⁴⁰ Ba	2.58	1.9	2.8
²³⁹ Pu(n,f)	P9-16	⁹⁵ Zr	6.91	0.6	2.4
		¹⁰³ Ru	10.37	0.9	4.0
		¹⁴⁰ Ba	7.61	1.3	2.4

^aStandard deviation of the mean from three detectors.

^bError analysis includes the relative error, the detector calibration error, and the ILRR recommended branching ratio error.

^cAn arbitrary 3% branching ratio error assigned.

D. FISSION-PRODUCT REACTION RATES FROM BIG-10 TEST C-ANL(74-16)

R. J. Popek, R. R. Heinrich, and N. D. Dudey

Objective

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

Summary

Fission-product reaction rates are reported for the fissile nuclides of ^{235}U , ^{239}Pu , ^{238}U , and ^{237}Np which were irradiated in the Big-10 facility at LASL. Irradiation of these nuclides was conducted in Test C-ANL(74-16). Also reported is the production rate of ^{239}Np from ^{238}U .

Accomplishments and Status

A packet of high purity foils consisting of ^{235}U , ^{238}U , ^{237}Np , and ^{239}Pu was irradiated in the LASL Big-10 facility on July 16, 1974, for a period of 220 minutes. This irradiation was designated Test C-ANL(74-16) by LASL. Specific foil parameters are not discussed and can be found in the 4th and 5th ILRR Progress Reports.

Each fissile foil was Ge(Li) counted a minimum of two times over a period of 1-80 days and each of our three detectors was used. The analysis of these data is presented in Table ANL-VI. The reaction rate of ^{95}Zr is an average of both the 724 and 756-keV gamma rays and the results represent counting data accumulated during a period of 30-80 days after the irradiation. The same Pb absorber (1503 mg/cm²) was used during counting of the ^{237}Np foil to reduce the activity from daughter products, and the same error (0.4-1.1%) is reflected in the absolute reaction rate.

Expected Accomplishments

Additional counting will be done to possibly improve the precision of the ^{137}Cs and to accumulate data on other long-lived fission products.

TABLE ANL-VI
FISSION-PRODUCT RATES FROM BIG-10 TEST C-ANL(74-16)

Reaction	Sample	Fission Product	10 ⁻¹⁵ atoms/ atom-sec	Reaction Rate	
				% Error (1σ)	
				Relative ^a	Absolute ^b
²³⁵ U(n,f)	U5-10	⁹⁵ Zr	7.13	0.8	2.4
		¹⁰³ Ru	3.70	0.7	4.0
		¹³¹ I	3.72	1.1	3.8 ^c
		¹³⁷ Cs	6.82	1.7	2.7
		¹⁴⁰ Ba	6.64	0.7	2.1
		¹⁴³ Ce	5.84	0.8	3.7 ^c
²³⁸ U(n,f)	U8-13	⁹⁵ Zr	0.213	0.6	2.4
		¹⁰³ Ru	0.270	0.8	4.0
		¹³¹ I	0.137	1.1	3.8 ^c
		¹⁴⁰ Ba	0.250	0.6	2.1
		¹⁴³ Ce	0.181	1.5	3.9 ^c
²³⁸ U(n,γ)	U8-13	²³⁹ Np	12.3	0.6	3.7
²³⁷ Np(n,f)	N7-14	⁹⁵ Zr	2.12	3.0	3.8
		¹⁰³ Ru	2.04	1.0	4.1
		¹⁴⁰ Ba	1.96	2.6	3.3
²³⁹ Pu(n,f)	P9-12	⁹⁵ Zr	6.35	0.6	2.4
		¹⁰³ Ru	9.57	0.6	4.0
		¹⁴⁰ Ba	7.00	2.0	2.8
		¹⁴³ Ce	5.33	1.0	3.7 ^c

^aStandard deviation of the mean from three detectors.

^bError analysis includes the relative error, the detector calibration error, and the ILRR recommended branching ratio error.

^cAn arbitrary 3% branching ratio error.

E. REACTION RATES FOR NONFISSION FOILS IRRADIATED IN BIG-10 TEST E-ANL(74-23)

R. J. Popek, R. R. Heinrich, and N. D. Dudey

Objective

- To measure nonfission foil reaction rates by the activation technique for those reactions having fast-neutron application.

Summary

Reaction rates of nonfission foils irradiated in the LASL Big-10 facility have been determined by Ge(Li) gamma-ray spectrometry. The following reaction rates are reported: $^{27}\text{Al}(n,\alpha)$, $^{197}\text{Au}(n,\gamma)$, $^{115}\text{In}(n,n')$, $^{59}\text{Co}(n,\gamma)$, $^{58}\text{Ni}(n,p)$, $^{58}\text{Fe}(n,\gamma)$, $^{54}\text{Fe}(n,p)$, $^{47}\text{Ti}(n,p)$, $^{46}\text{Ti}(n,p)$, $^{45}\text{Sc}(n,\gamma)$, and $^{238}\text{U}(n,\gamma)$. Also reported are the reactions $^{235}\text{U}(n,f)^{140}\text{Ba}$ and $^{238}\text{U}(n,f)^{140}\text{Ba}$. In general, these data are not as precise as previously reported fission reaction rates because of poorer counting statistics.

Accomplishments and Status

The nonfission foil packet was irradiated in the Big-10 facility at LASL on August 6, 1974, for a period of 220 minutes. This irradiation was designated Test E-ANL(74-23). The foil packet was prepared by HEDL, designated ANL 74-23, and contained 21 foils of 12 different materials. Isotopic abundances of the materials were taken from the Chart of the Nuclides.²

Ge(Li) counting of the samples was started approximately 1.5 days after the end of the irradiation. A minimum of two spectra was accumulated for each reaction, although in most cases three to five spectra were obtained. No external absorbers were used for any of the foil spectra.

The reaction rates for this foil set are presented in Table ANL-VII. No data are presented for the Mn and Cu foils because of the low activity levels. The location column indicates the relative position of the foil within the packet and the thickness of each foil counted is also given. Self absorption corrections have been made for the reaction rates presented. In general, the uncertainty of these reaction rates is about a factor of two greater than fission-product reaction rates of previous tests. This is a result of the low activity levels and poor counting statistics of the spectra.

²N. E. Holden and F. W. Walker, Chart of the Nuclides, Eleventh Edition (1972).

TABLE ANL-VII
REACTION RATES FOR THE NONFISSION FOIL SETS BIG-10 TEST E-ANL(74-23)

Foil	Location	Foil Thickness (mil)	Reaction	Reaction Rate (10^{-15} atoms/atom-sec)	
Al-10	Top	5	$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	0.00804	(3.7)
11	Middle	5		0.00794	(7.6)
12	Bottom	5		0.00822	(3.1)
Au-7	Top	0.5	$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	18.7	(2.5)
4	Middle	0.05		19.0	(2.6)
8	Bottom	0.5		18.7	(2.5)
In-10	Top	2	$^{115}\text{In}(n,n')^{115\text{m}}\text{In}$	3.00	(4.9)
11	Middle	2		2.93	(5.0)
12	Bottom	2		2.90	(5.6)
Co-10	Top	0.5	$^{59}\text{Co}(n,\gamma)^{60}\text{Co}$	1.02	(3.1)
11	Middle	0.5		1.01	(3.6)
12	Bottom	0.5		1.06	(3.0)
Ni-MP	Middle	10	$^{58}\text{Ni}(n,p)^{58}\text{Co}$	1.32	(2.3)
Fe-D	Middle	31	$^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$	0.329	(4.3)
Fe-D	Middle	31	$^{54}\text{Fe}(n,p)^{54}\text{Mn}$	0.979	(2.7)
Ti-LR	Top	10	$^{47}\text{Ti}(n,p)^{47}\text{Sc}$	0.239	(4.8)
LS	Bottom	10		0.239	(4.9)
Ti-LR	Top	10	$^{46}\text{Ti}(n,p)^{46}\text{Sc}$	0.136	(5.4)
LS	Bottom	10		0.137	(3.0)
Sc-5	Bottom	5	$^{45}\text{Sc}(n,\gamma)^{46}\text{Sc}$	1.46	(3.7)
U-6	Bottom	5	$^{238}\text{U}(n,\gamma)^{239}\text{U}$	11.9	(3.8)
U-4	Bottom	4.8	$^{235}\text{U}(n,f)\text{Ba}$	6.61	(2.3)
U-6	Bottom	5	$^{238}\text{U}(n,f)\text{Ba}$	0.238	(2.9)

F. FISSION RATES IN BIG-10 TEST C-ANL(74-16) BASED ON K-YIELD FACTORS

R. J. Popek, R. R. Heinrich, and N. D. Dudey

Objective

- To determine whether fission-product rates vary with neutron energy.

Summary

Fission rates have been determined from six fission-product reaction rates using K-yield factors for ^{235}U , ^{238}U , ^{237}Np , and ^{239}Pu which were determined in the CFRMF Test 74-7A and 74-7B irradiations. Precision of the data indicates these fission products are reliable fission monitors.

Accomplishments and Status

Fission rates have been calculated for ^{235}U , ^{238}U , ^{237}Np , and ^{239}Pu irradiated in Big-10 facility at LASL (Test C-ANL 74-16) described previously. Fission products of ^{143}Ce , ^{99}Mo , ^{131}I , ^{140}Ba , ^{103}Ru , and ^{95}Zr were Ge(Li) counted and fission rates calculated using our K-factor libraries. These libraries were compiled from the CFRMF Test 74-7A and 74-7B data and the NBS fission counter data for the same test.

These fission rates are presented in Table ANL-VIII. Accumulation of the fission-product spectra was done over a period of 1-80 days. Precision of the data indicates these monitors are reliable for the uranium isotopes. In the case of ^{237}Np and ^{239}Pu , naturally occurring gamma rays apparently interfere with the ^{99}Mo and ^{143}Ce monitors.

Comparison of these fission rates to the NBS fission chamber will be made when the final fission rate is calculated by NBS. However, preliminary comparisons (ILRR Meeting at LASL, March 19-21, 1975) indicate $\pm 2\%$ agreement. The implication of this is that no apparent change in yield due to neutron energy has occurred in the monitors reported here. When comparing the neutron spectra of CFRMF to that of Big-10, each spectra are similar and therefore these fission rates reported here should check with the NBS fission rate. If there is no significant fission yield change, these results will be incorporated into our K-yield factors.

Expected Accomplishments

When final NBS fission rates are reported, final calculation of K-yield fission rates will be made.

TABLE ANL-VIII
FISSION RATE BIG-10 TEST C-ANL BY K-YIELD FACTORS^a

Fission Product	Time Counted after Irrad. (Days)	Reaction Rate (10^{-13} fissions/atom-sec)			
		²³⁵ U	²³⁸ U	²³⁷ Np	²³⁹ Pu
¹⁴³ Ce	0-10	1.114(1.4) ^b	0.0407(2.4)		1.327(2.2)
⁹⁹ Mo	0-10	1.120(3.3)	0.0414(9.0) ^d		
¹³¹ I	10-40	1.130(1.1)	0.0418(1.4)		
¹⁴⁰ Ba	10-40	1.134(0.5)	0.0405(1.0)	0.350(2.6)	1.356(0.5)
¹⁰³ Ru	20-80	1.141(0.8)	0.0419(1.6)	0.361(1.1)	1.387(1.1)
⁹⁵ Zr	30-80	1.127(1.1)	0.0412(2.2)	0.366(3.4)	1.368(1.1)
Average		1.128(1.6) ^c	0.0412(2.0)	0.359(2.3)	1.360(1.8)

^aRelative to NBS fission chamber results from CFRMF Test 74-7A and 74-7B.

^bError analysis only includes relative counting errors.

^cError analysis includes the NBS fission counter error and standard deviation of the mean.

^dNot included in the average because of the relative high error.

AI

ATOMICS INTERNATIONAL

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

Objective

- Determine integral (n, α) cross sections of ^6Li and ^{10}B and possibly other isotopes in a variety of well-established and permanent neutron fields, including several that cover the neutron energy ranges of interest for LMFBR's and CTR's. The cross sections, which are obtained by mass spectrometric measurement of the amounts of helium generated in miniature capsules containing enriched ^{10}B and ^6LiF , will provide independent, absolute bases for normalization of available energy-dependent (n, α) cross-section data. This will improve the accuracy for using these isotopes as helium accumulation fluence monitors (HAFM's) both in LMFBR and CTR neutron environments.

Summary

Three almost-identical small aluminum assemblies, each enclosing twelve miniature vanadium capsules filled with enriched ^{10}B and ^6LiF (helium accumulation fluence monitors) have been irradiated as a part of the Interlaboratory LMFBR Reaction Rate (ILRR) program. Two of the assemblies were irradiated consecutively in the BIG-10 facility at Los Alamos during two series of reactor runs. The third was irradiated in the CFRMF facility at Idaho Falls for a 6kw run lasting 29 hrs. Additional miniature helium accumulation fluence monitors are being manufactured for irradiation in the $\Sigma\Sigma$ and fission spectrum facilities at Mol-Belgium.

Accomplishments and Status

A. Capsule Preparations: The AI section of the LMFBR Reaction Rate and Dosimetry 9th Progress Report (HEDL-TME 74-45) described three small aluminum assemblies that were inserted into the 10% enriched ^{235}U critical assembly "BIG-10" at Los Alamos, and the Coupled Fast Reactivity Measurements Facility (CFRMF) at Aerojet Nuclear Company, Idaho Falls. Each of the 1.82cm diameter, 3.3mm thick aluminum assemblies was loaded with seven miniature capsules containing ^6LiF and five capsules containing enriched ^{10}B . In addition to these, twelve cylindrical holes were loaded with single bare (unencapsulated) crystals of natural boron, ^{10}B , ^6LiF , and vanadium. The total elemental contents of the capsules were listed, including the total amounts of the isotopes ^{10}B (~30 mg) and ^6Li (~13 mg). The amounts of these two isotopes were limited to these approximate values because it has been experimentally determined that 35 mg of ^{10}B did not appreciably affect the neutron flux spectrum during the first AI irradiation in CFRMF; and HEDL has calculated that the effect will be less in BIG-10.

The three loaded aluminum assemblies were shipped to HEDL where they were integrated with suitable neutron dosimetry foils, and then sent on to Los Alamos or Idaho Falls for irradiations as a part of the Interlaboratory LMFBR Reaction Rate (ILRR) program. In the case of the BIG-10 irradiations, HEDL included flux normalization foils of ^{235}U , ^{239}Pu , Ni, Fe, and Co, as described in detail in a letter from W. N. McElroy to G. E. Hansen & H. H. Helmick of May 30, 1974. The AI capsules are described in AI letter 74AT-3694, "AI Capsule for BIG-10 Irradiations," from H. Farrar IV to G. E. Hansen & H. H. Helmick, dated June 12, 1974.

B. Irradiations in BIG-10: The two capsules were placed consecutively in a special position adjacent to the main test location in BIG-10 during two series of reactor runs performed specifically for the ILRR program. By having the AI capsule in the reactor for several maximum-length runs, a maximum fluence that was necessary to generate sufficient helium for accurate mass spectrometric measurement was obtained. The first AI capsule was in the BIG-10 for the duration of several test irradiations during which time ANC, ANL, HEDL, ARHCO, and LASL made fission product measurements. The second capsule was substituted during a later test series, while nonfission reaction product measurements were conducted by the same laboratories, and including CEN/SCK. Having identical AI capsules present in the cavity for the whole series of runs meant that any perturbation to the flux spectrum due to the presence of the ^{10}B and ^6Li , although calculated by LASL to be negligible, would be consistent for all the irradiation experiments. The flux level at the AI capsule location (Test Region III) is very well-known with respect to the flux level of the location of the NBS fission chamber (Test Region I) less than 1-in. away. Preliminary details of the irradiations are given in two Los Alamos Scientific Laboratory office memoranda by Gordon Hansen: "Record of Tests A, C, and E to September 23, 1974" (September 23, 1974), and "Power Profiles and Monitor Data for the BIG-10 Runs Following Test E-HEDL/ARHCO" (April 29, 1975).

C. Third Irradiation in CFRMF: The irradiation in CFRMF was the third irradiation of an AI capsule containing boron and lithium in this facility. The third aluminum assembly (essentially identical to that in BIG-10) was shipped to HEDL for integration with flux-fluence radiometric foils. Details of the package were included in AI letter 75AT-763, "CFRMF Irradiation AI-III of ^{10}B and ^6Li ," from H. Farrar IV to J W Rogers, dated January 31, 1975. Following this, HEDL included Ni, Au, ^{235}U , and ^{239}Pu normalization foils as described in HEDL letter W/FFTF-C751275 from W. N. McElroy & G. E. Shook to ILRR program participants, dated February 24, 1975. The 6kw irradiation on 25-26 June 1975 lasted approximately 29 hrs.

D. Preparations for Helium Analyses of ^{10}B and ^6LiF : When a sample is vaporized in a graphite crucible of the mass spectrometer high temperature furnace, a small amount of helium is released from the graphite and from the furnace walls as well as from the sample itself. If this level of background helium is precisely known, a correction can be made for it as part of the data reduction process. Past measurements made by vaporizing empty graphite crucibles of the type currently in use have shown that this background level can be of the order of 10^{10} atoms of helium or less. There is some variation in these measurements, and this variation can be ascribed to different degassing histories of the crucibles, different cleaning procedures being used on the furnace walls, and possible inhomogeneity in the graphite from which the crucibles were made. A precise study of the phenomena has not been made until now because the samples being analyzed contained so much helium that the background correction was small and the uncertainty in this correction was negligible. For the capsules that have been irradiated in the CFRMF and BIG-10 facilities, however, it will be necessary to make measurements on samples containing rather low levels of helium. For these samples, the present background correction would not be small, and the present uncertainty in this correction would form a major part of the overall uncertainty in the measurement. Accordingly, a major series of tests was initiated to study the factors which contribute to the helium background, and to find materials and procedures which reduce the uncertainty in the background correction.

Initially, an internal AI document, "Lowering the Helium Background from Graphite Crucibles," was drawn up outlining a plan of action. This is now being implemented and significant success has been achieved. One rather surprising result of the work so far has been a test demonstrating that less than 20% of the total helium background can be ascribed to the graphite crucibles themselves. Accordingly, current efforts to reduce the background are concerned primarily with finding more effective methods of cleaning and degassing the furnace and furnace parts.

Expected Achievements

The direction of future AI contributions to the ILRR program will be as follows:

1. After helium background studies are completed for the mass spectrometer system, all the ^{10}B and ^6LiF miniature capsules (helium accumulation fluence monitors) irradiated in the BIG-10 and CFRMF will be analyzed for helium content.
2. Approximately thirty helium accumulation fluence monitors will be manufactured and filled with ^{10}B and ^6LiF , and shipped to Mol-Belgium for irradiation in the $\Sigma\Sigma$ and fission cavities of the BR1 reactor.

BNL

BROOKHAVEN NATIONAL LABORATORY

A. EVALUATED CROSS SECTION LIBRARIES
B. A. Maurno
(NATIONAL NEUTRON CROSS SECTION CENTER)

Objective

- Testing of ENDF/B-IV dosimetry library (tape 412) with specified reference spectra.

Summary

The dosimetry tape (ENDF/B-IV tape 412) was issued in a general CSEWG distribution, August 1974. The pointwise cross section data file was tested with specified reference spectra. The results are published in the February 1975 issue of Nuclear Science and Technology.⁽¹¹⁾ A group averaged cross section data file (620 groups based on tape 412) was tested with the above spectra and the results are presented in this report.

Accomplishments and Status

Data testing on the pointwise cross section data file (tape 412) was done only at Brookhaven National Laboratory. The tests are those described in the P. C. McElroy to Alter⁽¹⁾ and modified by the Normalization and Standards Subcommittee⁽²⁾ i.e., the resonance integrals of all exoergic reactions on Tape 412 and the spectrum averaged cross sections of all reactions on Tape 412. The reference spectrum for $\Sigma\Sigma$ was supplied by Fabry^{(3)*}, CFRMF by Harker⁽⁴⁾. A simple fission spectrum of U-235 (defined by CSEWG⁽⁵⁾) was also used. The resonance integrals of the exoergic materials were calculated at BNL using the code INTER⁽⁶⁾ with the data from Tape 412. The spectrum averaged cross sections were calculated using INTEND** with data from Tape 412. Reference cross sections were supplied by Fabry*, McElroy[†] and Fabry⁽⁸⁾.

*Data supplied by A. Fabry of CEN/SCK January 1974.

**Computer Code supplied by O. Ozer of BNL.

[†]Data supplied by W. N. McElroy of HEDL June-July 1974.

Some of the reactions on Tape 412 have their resonance regions represented by resonance parameters and background cross sections. These parametric representations were converted to pointwise data for the above calculations by the code RESEND⁽⁹⁾. Because of the voluminous output of RESEND for some isotopes (e.g. U-238) all reactions were converted into 620 group average cross sections by the code INTEND*. These group averaged cross sections facilitated ease in handling for some testers. The BNL tests were also rerun with the 620 group cross set and compared to the calculations done by HEDL, ANC and ORNL. Differences have been noted due in part to modifications of the reference spectra by some of the participants.

Table 1 compares the resonance integrals calculated at BNL and HEDL using 620 group averaged cross sections as input with the resonance integrals listed in BNL-325⁽⁷⁾ and also compares the U-235 fission spectrum averaged cross sections calculated at BNL, HEDL⁽¹⁰⁾ and ORNL** using 620 group averaged cross sections as input with the reference cross sections as supplied by Fabry⁽⁸⁾.

Table 2 compares the Σ ^{(3)†} spectrum averaged cross sections calculated at BNL, HEDL⁽¹⁰⁾ and ORNL** using 620 group averaged cross sections as input with reference cross sections as supplied by Fabry[†] and also compares the CFRMF spectrum averaged cross sections calculated by BNL, HEDL⁽¹⁰⁾ and ANC^{††} using 620 group averaged cross sections as input with reference cross sections supplied by McElroy^{†††}.

* Computer Code supplied by O. Ozer of BNL.

** Data supplied by F. B. K. Kam and B. A. Magurno, October 14, 1974.

† Data supplied by A. Fabry of CEN/SCK, January 1974.

†† Data supplied by Y. Harker and B. A. Magurno October 1974.

††† Data supplied by W. N. McElroy of HEDL July 1974.

The Lippincott and McElroy's data testing letter⁽¹⁰⁾ is attached in its entirety to the end of this report. The table from Kam's* private communication is also attached.

* Data supplied by F. B. K. Kam and B. A. Magurno October 14, 1974.

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R.I.

TABLE 1

Calculated Fission

Measured Fission

Spectrum Averaged Cross Sections

Spectrum Averaged

 U^{235} Fission Spectrum

Cross Sections

CalculatedMeasuredBNLHEDLBNL-325BNL(mb)HEDL(mb)ORNL(mb)Ref. 1 (mb)ISOTOPE/REACTION

(B)

(B)

(B)

(T=1.29)

(T=1.32)

(T=1.29)

(T=1.32)

(T=1.29)

 ^6Li (n, total He)

425.72

425.8

487.0

486.1

487.1

486.2

487.7

 ^{10}B (n, total He)

1721.69

1722.

1722±5

514.4

511.5

515.0

512.2

515.5

 ^{23}Na (n, γ)

0.346

0.346

0.311±0.010

0.294

0.291

0.295

0.291

0.295

 ^{27}Al (n, p)

3.930

4.226

3.93

4.22

3.916

4.0±0.4

 ^{27}Al (n, α)

0.710

0.801

0.710

0.801

0.7058

0.73±0.02

 ^{32}S (n, p)

61.35

63.89

61.35

63.89

61.24

69±2

 ^{45}Sc (n, γ)

11.29

11.29

11.3±1.0

5.98

5.87

5.98

5.88

5.99

 ^{46}Ti (n, p)

9.513

10.25

9.51

10.24

9.477

12.3±0.3

 ^{47}Ti (n, np)

-

-

 4.95×10^{-3} 6.3×10^{-3} 4.9×10^{-3} ^{47}Ti (n, p)

20.65

21.43

20.64

21.42

20.60

20.0±2.0

 ^{48}Ti (n, np)

-

-

 2.62×10^{-3} 3.3×10^{-3} 2.6×10^{-7} ^{48}Ti (n, p)

0.177

0.200

0.177

0.200

0.1758

0.32±0.02

 ^{55}Mn (n, p)

0.300

0.364

0.300

0.363

0.2971

0.25±0.01

 ^{56}Fe (n, p)

74.09

77.72

74.07

77.69

73.91

82.5±2.0

 ^{56}Fe (n, p)

1.034

1.15

1.033

1.145

1.029

1.07±0.06

BNL-6

R. I.

TABLE 1

Calculated Fission
Spectrum Averaged Cross Sections
 U^{235} Fission Spectrum

Measured Fission
Spectrum Average
Cross Sections

ISOTOPE/REACTION	<u>Calculated</u>		<u>Measured</u>						
	<u>BNL</u>	<u>HEDL</u>	<u>BNL-325</u>	<u>BNL(mb)</u>		<u>HEDL(mb)</u>		<u>ORNL(mb)</u>	
	(B)	(B)	(B)	(T=1.29)	(T=1.32)	(T=1.29)	(T=1.32)	(T=1.29)	<u>Ref. 11 (mb)</u>
^{58}Fe (n, γ)	1.595	1.60	1.19 $\pm$ 0.07	1.708	1.695	1.71	1.69	1.713	
^{59}Co (n, 2n)				0.213	0.260	0.213	0.259	0.2112	
^{59}Co (n, γ)	76.74	76.73	75.5 $\pm$ 1.5	6.513	6.429	6.59	6.50	6.590	
^{59}Co (n, α)				0.149	0.168	0.149	0.168	0.1486	0.156 $\pm$ 0.006
^{58}Ni (n, 2n)				3.8 $\times 10^{-3}$	4.7 $\times 10^{-3}$	3.75 $\times 10^{-3}$	4.7 $\times 10^{-3}$	3.71 $\times 10^{-3}$	
^{58}Ni (n, p)				97.27	101.6	97.24	101.5	97.05	113 $\pm$ 3
^{60}Ni (n, p)				2.415	2.660	2.414	2.659	2.404	
^{63}Cu (n, γ)	5.517	5.52	4.9 $\pm$ 0.4	11.14	11.0	11.18	11.04	11.19	10.1 $\pm$ 1.3
^{63}Cu (n, α)				0.354	0.396	0.354	0.396	0.352	0.50 $\pm$ 0.05
^{65}Cu (n, 2n)				0.381	0.460	0.380	0.460	0.377	
^{115}In (n, n')				163.9	165.9	163.8	166.7	163.5	188 $\pm$ 4
^{115}In (n, γ)	3240.8	3240	3300 $\pm$ 100	138.3	136.5	138.5	136.7	138.6	
^{127}I (n, 2n)				1.378	1.636	1.377	1.635	1.367	1.09 $\pm$ 0.05

TABLE 1

ISOTOPE/REACTION	R.I.			Calculated Fission Spectrum Averaged Cross Sections					Measured Fission Spectrum Averaged Cross Sections
				<u>U²³⁵ Fission Spectrum</u>					
	<u>Calculated</u>		<u>Measured</u>						
	<u>BNL</u>	<u>HEDL</u>	<u>BNL-325</u>	<u>BNL(mb)</u>		<u>HEDL(mb)</u>		<u>ORNL(mb)</u>	<u>Ref 11 (mb)</u>
	(B)	(B)	(B)	(T=1.29)	(T=1.32)	(T=1.29)	(T=1.32)	(T=1.29)	
¹⁹⁷ Au (n,γ)	1567.3	1567	1560±40	86.28	84.85	86.58	85.15	86.69	88±5
²³² Th (n, f)				67.28	69.04	67.26	69.02	67.19	
²³² Th (n, γ)	85.39	85.4	85±3	105.4	103.8	105.5	103.9	105.7	83.0±3.5
²³⁵ U (n, f)	283.44	283.4	275±5	1242.99	1242.98	1243	1243	1243	1250 (Ref)
²³⁸ U (n, f)		1.92		289.0	2955	288.9	295.4	288.6	328±10
²³⁸ U (n, γ)	277.06	277.0	275±5	76.74	75.85	76.87	75.68	76.97	
²³⁷ Np (n, f)		6.54		1313.5	1323.1	1313	1323	1312	1370±75
²³⁹ Pu (n, f)	316.66	316.7	301±10	1780.3	1782.7	1780	1783	1780	

TABLE 2

ISOTOPE/REACTION	<u>ΣΣ</u>			Measured Spectrum Averaged Cross Sections Ref. 9 (mb)	<u>CFRMF</u>			Measured Spectrum Averaged Cross Sections Ref. 10 (mb)
	Calculated Spectrum Averaged Cross Sections				Calculated Spectrum Averaged Cross Sections			
	<u>BNL</u> (mb)	<u>HEDL</u> (mb)	<u>ORNL</u> (mb)		<u>BNL</u> (mb)	<u>HEDL</u> (mb)	<u>ANC</u> (mb)	
⁶ Li (n, total He)	914.30	923.3	922.9		986.7	989.4	988.9	900±39
¹⁰ B (n, total He)	1497.5	1514	1507		1692.7	1695	1695	1940±60
²³ Na (n,γ)	1.391	1.405	1.405		1.843	1.83	1.831	
²⁷ Al (n,p)	1.047	0.869	0.8699	1.033±0.103	0.917	0.860	0.894	0.931±0.033
²⁷ Al (n,α)	0.208	0.153	0.1537	0.182±0.005	0.156	0.144	0.171	0.172±0.053
³² S (n,p)	16.32	14.53	14.50		13.89	13.56	13.67	
⁴⁵ Sc (n,γ)	18.79	19.01	18.96		20.08	20.11	20.11	25.0±0.1
⁴⁶ Ti (n,p)	2.524	2.075	2.073		2.196	2.09	2.168	2.78±0.97
⁴⁷ Ti (n,np)	5x10 ⁻⁴	0.96x10 ⁻³	2.75x10 ⁻⁴		—*	0.95x10 ⁻³	1.43x10 ⁻³	
⁴⁷ Ti (n,p)	5.713	5.174	5.174		4.78	4.67	4.73	4.44±0.02
⁴⁸ Ti (n,np)	5x10 ⁻⁴	4.9x10 ⁻⁴	2.93x10 ⁻⁴		—*	.50x10 ⁻⁴	7.2x10 ⁻⁴	
⁴⁸ Ti (n,p)	0.0510	0.0379	0.0377		0.034	0.037	0.0425	0.073±0.003
⁵⁵ Mn (n,2n)	0.080	0.0554	0.0493		—*	.0574	0.7638	
⁵⁴ Fe (n,p)	19.56	17.20	17.22		16.86	16.36	16.35	18.60±0.58
⁵⁶ Fe (n,p)	0.296	0.230	0.2299	0.273±0.008	0.238	0.233	0.2450	

*Because of the sharp cutoff at 10 MeV of CFRMF spectrum, high energy threshold reaction cross sections cannot be calculated

TABLE 2 (Cont.)

	<u>Σ</u>			Measured Spectrum Averaged Cross Sections <u>Ref. 9</u> (mb)	<u>CFRMF</u>			Measured Spectrum Averaged Cross Sections <u>Ref. 10</u> (mb)
	Calculated Spectrum Averaged Cross Sections				Calculated Spectrum Averaged Cross Sections			
	<u>BNL</u> (mb)	<u>HEDL</u> (mb)	<u>ORNL</u> (mb)		<u>BNL</u> (mb)	<u>HEDL</u> (mb)	<u>ANC</u> (mb)	
<u>ISOTOPE/REACTION</u>								
⁵⁸ Fe (n, γ)	4.874	5.377	5.433		6.23	6.02	6.13	6.52±0.22
⁵⁹ Co (n, 2n)	0.056	0.0392	0.0339		—*	0.0408	0.0546	
⁵⁹ Co (n, γ)	55.60	46.21	40.61		86.49	74.10	85.51	97.6±3.6
⁵⁹ Co (n, α)	0.043	0.0326	0.0326		0.033	0.031	0.0357	
⁵⁸ Ni (n, 2n)	1.89x10 ⁻⁴	7.07x10 ⁻⁴	3.8x10 ⁻⁴		—*	7.17x10 ⁻⁴	10.2x10 ⁻⁴	
⁵⁸ Ni (n, p)	26.23	23.33	23.34	28.8±0.7	22.29	21.77	21.97	25.6±0.8
⁶⁰ Ni (n, p)	0.683	0.5376	0.5389		0.569	0.522	0.5683	
⁶³ Cu (n, γ)	39.39	38.76	39.16	40.0±2.0	47.08	47.62	47.26	48.4±2.6
⁶³ Cu (n, α)	0.104	0.0779	0.0782		0.080	0.0740	0.0846	
⁶⁵ Cu (n, 2n)	0.103	0.0708	0.0635		—*	0.0729	0.0967	
¹¹⁵ In (n, n')	53.47	50.79	50.71	58.6±1.2	44.09	43.57	43.59	53±3
¹¹⁵ In (n, γ)	276.74	283.6	278.0	261±9	300.97	300.8	301.0	300±11
¹²⁷ I (n, 2n)	0.393	0.2645	0.2537		0.068	0.264	0.3433	

*Because of the sharp cutoff at 10 MeV of CFRMF spectrum, high energy threshold reaction cross sections cannot be calculated

BNL-10

TABLE 2 (Cont.)

ISOTOPE/REACTION	<u>ΣΣ</u>			Measured Spectrum Averaged Cross Sections <u>Ref. 9</u> (mb)	<u>CFRME</u>			Measured Spectrum Averaged Cross Sections <u>Ref. 10</u> (mb)
	Calculated Spectrum Averaged Cross Sections				Calculated Spectrum Averaged Cross Sections			
	<u>BNL</u> (mb)	<u>HEDL</u> (mb)	<u>ORNL</u> (mb)		<u>BNL</u> (mb)	<u>HEDL</u> (mb)	<u>ANC</u> (mb)	
¹⁹⁷ Au (n,γ)	336.7	371.2	348.7	442±8	412.33	416.3	413.2	457±14
²³² Th (n,f)	19.78	18.50	18.51	21.0±1.3	16.12	15.84	15.93	
²³² Th (n,γ)	261.67	263.3	259.8		286.73	285.1	285.5	
²³⁵ U (n,f)	1516.1	1518	1518	1589±55	1587.8	1587	1588	1659±53
²³⁸ U (n,f)	86.47	81.57	81.59	87.4±3.1	70.13	69.22	69.48	81.0±0.3
²³⁸ U (n,γ)	222.68	220.8	214.1	194±8	232.9	230.2	229.4	196±11
²³⁷ Np (n,f)	625.62	607.4	607.2	634±22	551.2	547.5	547.9	591±21
²³⁹ Pu (n,f)	1753.2	1750	1754	1875±66	1771.7	1771	1771	1930±60

TABLE 3
ORNL CALCULATED AVERAGE CROSS SECTIONS

Reaction	Average Cross Section MOL-ΣΣ ^a	Average Cross Section FISSION ^b
NA23(N,G)NA24	1.405E-27	2.949E-28
AL27(N,P)MG27	8.699E-28	3.916E-27
AL27(N,A)NA24	1.537E-28	7.058E-28
MN55(N,2N)MN54	4.927E-29	2.971E-28
CO59(N,2N)CO58	3.392E-29	2.112E-28
CO59(N,G)CO60	4.061E-26	6.590E-27
CO59(N,A)MN56	3.255E-29	1.486E-28
U235(N,F)FP	1.518E-24	1.243E-24
U238(N,F)FP	8.159E-26	2.886E-25
U238(N,G)U239	2.141E-25	7.697E-26
NP237(N,F)FP	6.072E-25	1.312E-24
PU239(N,F)FP	1.754E-24	1.780E-24
LI6(N,A)H3	9.229E-25	4.877E-25
B10(N,A)LI7	1.507E-24	5.155E-25
AU197(N,G)AU198	3.487E-25	8.669E-26
TH232(N,F)FP	1.851E-26	6.719E-26
TH232(N,G)TH233	2.598E-25	1.057E-25
IN115(N,N')M	5.071E-26	1.635E-25
S32(N,P)P32	1.450E-26	6.124E-26
FE56(N,P)MN56	2.299E-28	1.029E-27
CU63(N,G)CU64	3.916E-26	1.119E-26
CU63(N,A)CO60	7.824E-29	3.519E-28
CU65(N,2N)CU64	6.352E-29	3.773E-28
I127(N,2N)I126	2.537E-28	1.367E-27
SC45(N,G)SC46	1.896E-26	5.994E-27

^aMOL-ΣΣ spectrum normalized such that

$$\int_{4.14E-7}^{14.92 \text{ MeV}} \phi(E) dE = 1.$$

^bFISSION spectrum (Maxwellian) normalized such that

$$\int_{1.0E-10}^{10 \text{ MeV}} \phi(E) dE = 1.$$

TABLE 3 (Cont.)

Reaction	Average Cross Section MOL-ΣΣ	Average Cross Section FISSION
IN115(N,G)IN116	2.780E-25	1.386E-25
FE54(N,P)MN54	1.722E-26	7.391E-26
FE58(N,G)FE59	5.433E-27	1.713E-27
NI58(N,2N)NI57	3.859E-31	3.714E-30
NI58(N,P)CO58	2.334E-26	9.705E-26
NI60(N,P)CO60	5.389E-28	2.404E-27
TI46(N,P)SC46	2.073E-27	9.477E-27
TI47(N,NP)SC46	2.758E-31	4.898E-30
TI47(N,P)SC47	5.174E-27	2.060E-26
TI48(N,NP)SC47	2.931E-31	2.597E-30
TI48(N,NP)SC48	3.766E-29	1.758E-28

P. O. Box 1970
Richland, Washington
99352

Hanford Engineering Development Laboratory

October 21, 1974

Mr. B. A. Magurno
Brookhaven National Laboratory
Upton, Long Island,
New York 11973

Dear Ben:

ENDF/B-IV DOSIMETRY FILE PHASE II DATA TESTING

The 36 cross sections on the ENDF/B-IV dosimetry file (Tape 412) have been subjected to preliminary data testing using the five dosimetry benchmarks. The actual cross sections used were those reduced or expanded to the 620 point SAND-II group structure by BNL using the code INTEND. The SAND-II code was used to calculate the spectral averaged cross sections by expanding each of the five benchmark neutron spectra to 620 groups, and using the equation

$$\bar{\sigma}_R = \sum_{i=1}^{620} \sigma_{Ri} \phi_i \quad (1)$$

The comparison of these calculated spectral average cross sections with measured values are described in the attached discussion and tables. The HEDL results must be considered as preliminary since checking of procedures, numbers, etc., at HEDL for errors is still in progress.

Sincerely yours,

E. P. Lippincott and W. N. McElroy
Irradiation Analysis Section

EPL/WNM:bdw

Attach.

Benchmark 1 - Thermal Spectrum

The calculated thermal cross sections for those reactions with non-zero values are given in Table 4. The approximation of the thermal spectrum in the SAND-II group structure is indicated to give an 0.4% bias for $1/v$ cross sections. This is indicated by comparing the calculated value of σ_0 with σ_{2200} , the actual dosimetry file 2200 m/sec cross section given in the last column of Table 4. A comparison of the ENDF/B values with BNL-325⁽¹⁾ indicates that the former cross section falls within 2σ of the evaluated experimental values in all cases.

Benchmark 2 - Resonance Integrals

The SAND-II resonance integral calculation is defined by:

$$RI = \sum_i \sigma_i \log (E_{i+1}/E_i), \quad (2)$$

where i sums over the range of energies 0.5 eV to 18 MeV. If σ_i is defined by:

$$\sigma_i = \frac{\int_{E_i}^{E_{i+1}} \frac{\sigma(E)}{E} dE}{\log (E_{i+1}/E_i)}, \quad (3)$$

then Equation (2) is exact. Values calculated using Equation (2) are given in Table 5 for those reactions with finite resonance region cross sections.

In the third column of Table 5, the BNL-325 resonance integral values are presented and are compared with the ENDF/B values. Good agreement is obtained except for $^{23}\text{Na}(n,\gamma)$, $^{58}\text{Fe}(n,\gamma)$, and $^{63}\text{Cu}(n,\gamma)$ for which the ENDF/B cross section is high.

In the last column of Table 5 are presented resonance integrals for the dosimetry file calculated independently by Magurno.⁽⁴⁾ These values are in good agreement except for $^{239}\text{Pu}(n,f)$ and $^{237}\text{Np}(n,f)$. Since these two reactions have resonances close to 0.5 eV, the difference probably arises in the cutoff region. The calculation of σ_i in this region by INTEND must be investigated and any differences between Magurno's procedure and SAND-II should be looked at to resolve this discrepancy.

Benchmark 3 - $\Sigma\Sigma$ Spectrum

The 136 point differential spectrum supplied by Fabry⁽⁵⁾ was used in SAND-II to calculate spectral average cross sections as given in Table 6. These are compared

with measured values⁽⁵⁾ which are normalized to ^{235}U . The new cross section for ^{235}U is calculated to be 1518 mb vs. 1589* (difference 4%) and ratios of calculation to measured values renormalized to the new ^{235}U cross section are also presented. Of the broad range monitors, $^{115}\text{In}(n,\gamma)$, $^{197}\text{Au}(n,\gamma)$, and $^{238}\text{U}(n,\gamma)$ show some discrepancy. The threshold monitors are all low by $12 \pm 4\%$. Values calculated by Magurno⁽⁴⁾ for the $\Sigma\Sigma$ spectrum are also presented. These apparently disagree with the SAND-II values due to a different method used to calculate the integral of cross section times flux. These discrepancies should be investigated.

Benchmark 4 - CFRMF Spectrum

The calculated spectral average cross section values for CFRMF are compared with measurements in Table 7. The spectrum used was the 69 point SCAMP calculation⁽⁶⁾ integral spectrum. An updated SPLINE procedure was introduced into SAND-II to expand this spectrum to 620 points. The high end of the spectrum (above 7.79 MeV) was normalized to a fission form.

Two non-threshold monitors show low measured results. The $^6\text{Li}(n,\alpha)$ reaction is a preliminary measurement and the $^{238}\text{U}(n,\gamma)$ reaction is suspected to have resonance self-shielding from ^{238}U in the surroundings of the CFRMF cavity. Calculated cross sections of $^{10}\text{B}(n,\alpha)$, $^{45}\text{Sc}(n,\gamma)$, $^{58}\text{Fe}(n,\gamma)$, $^{59}\text{Co}(n,\gamma)$, and $^{197}\text{Au}(n,\gamma)$ are all low relative to ^{235}U , and ^{63}Cu was indicated previously to have a high resonance integral.

The threshold monitors are again calculated consistently low relative to experiment. The $^{48}\text{Ti}(n,p)$ cross section appears to be outside the range defined by the other reactions and should be investigated.

Benchmark 5 - Fission Spectra

The fission spectrum form used is given by:

$$\phi(E) = 0.77 E^{1/2} e^{-(E/T)}, \quad (4)$$

where two temperatures were used for T, 1.29 and 1.32 MeV. The fission spectral average cross sections are presented in Table 8 and the T = 1.32 calculated is compared with experimental values.⁽⁸⁾ The calculated values are in good agreement with Magurno.⁽⁴⁾ Reactions which are indicated to need investigation are $^{232}\text{Th}(n,\gamma)$, $^{48}\text{Ti}(n,p)$, $^{55}\text{Mn}(n,2n)$, $^{63}\text{Cu}(n,\alpha)$, and $^{127}\text{I}(n,2n)$.

*Measured.

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TABLE 4

BENCHMARK 1 - THERMAL SPECTRUM

Reaction	Calculated $\bar{\sigma}$ (b)	σ_0 (b)*	σ_{2200} (b) ⁽¹⁾	σ_0/σ_{2200}	σ_{2200}^{**}
${}^6\text{Li}(n,\alpha){}^3\text{T}$	836.6	944.0	940 $\pm$ 4	1.004	940.0
${}^{10}\text{B}(n,\alpha){}^7\text{Li}$	3414.	3852.	3837 $\pm$ 9	1.004	3836.5
${}^{23}\text{Na}(n,\gamma){}^{24}\text{Na}$	.475	.536	.530 $\pm$.005	1.011	.534
${}^{45}\text{Sc}(n,\gamma){}^{46}\text{Sc}$	23.47	26.5	26.5 $\pm$ 1.0	1.000	26.4
${}^{58}\text{Fe}(n,\gamma){}^{59}\text{Fe}$	1.051	1.19	1.15 $\pm$ 0.02	1.035	1.18
${}^{59}\text{Co}(n,\gamma){}^{60}\text{Co}$	33.15	37.44	37.2 $\pm$ 0.2	1.006	37.2
${}^{63}\text{Cu}(n,\gamma){}^{64}\text{Cu}$	4.00	4.51	4.5 $\pm$ 0.1	1.002	4.50
${}^{115}\text{In}(n,\gamma){}^{116\text{m}}\text{In}$	150.9	167.	157 $\pm$ 5	1.06	166.4
${}^{197}\text{Au}(n,\gamma){}^{198}\text{Au}$	88.39	99.24	98.8 $\pm$ 0.3	1.004	98.8
${}^{232}\text{Th}(n,\gamma){}^{233}\text{Th}$	6.56	7.44	7.40 $\pm$ 0.08	1.005	7.40
${}^{235}\text{U}(n,f)\text{F.P.}$	509.2	588.2	582.2 $\pm$ 1.3	1.010	585.4
${}^{238}\text{U}(n,\gamma){}^{239}\text{U}$	2.41	2.71	2.70 $\pm$.02	1.004	2.69
${}^{239}\text{Pu}(n,f)\text{F.P.}$	696.2	744.7	742.5 $\pm$ 3.0	1.003	741.7
${}^{237}\text{Np}(n,f)\text{F.P.}$	1.43×10^{-2}	0.016	0.019 $\pm$.003	0.84	.017
${}^{238}\text{U}(n,f)\text{F.P.}$	1.12×10^{-9}	1.3×10^{-9}	$[(3\pm 5) \times 10^{-6}]^{(4)}$	--	5.5×10^{-10}

* $\sigma_0 = \frac{2\bar{c}}{\sqrt{\pi}g}$ where g is the Westcott g factor⁽²⁾ obtained from Ref. 3 except for ${}^{115}\text{In}$ (Ref. 2)

and for ${}^{45}\text{Sc}(n,\gamma)$, ${}^{58}\text{Fe}(n,\gamma)$, ${}^{237}\text{Np}(n,f)$, and ${}^{238}\text{U}(n,f)$ for which g was not available and assumed to be 1.0.

** Cross Section at .0253 ev from dosimetry file.

TABLE 5

Reaction	BENCHMARK 2 - RESONANCE INTEGRALS			
	Calculated Resonance Integral (b)	Measured ⁽¹⁾ Resonance Integral (b)	Calc./Meas.	Calculated Magurno (5)
${}^6\text{Li}(n,\alpha){}^3\text{T}$	425.8	425.8 ± 2.2	1.00	425.9
${}^{10}\text{B}(n,\alpha){}^7\text{Li}$	1722	1722 ± 5	1.00	1722
${}^{23}\text{Na}(n,\gamma){}^{24}\text{Na}$	0.346	0.311 ± 0.010	1.11	.346
${}^{45}\text{Sc}(n,\gamma){}^{46}\text{Sc}$	11.29	11.3 ± 1.0	1.00	11.29
${}^{58}\text{Fe}(n,\gamma){}^{59}\text{Fe}$	1.60	1.19 ± 0.07	1.34	1.58
${}^{59}\text{Co}(n,\gamma){}^{60}\text{Co}$	76.73	75.5 ± 1.5	1.02	76.67
${}^{63}\text{Cu}(n,\gamma){}^{64}\text{Cu}$	5.52	4.9 ± 0.4	1.13	5.55
${}^{115}\text{In}(n,\gamma){}^{116\text{m}}\text{In}$	3240	3300 ± 100*	0.98	3243.
${}^{197}\text{Au}(n,\gamma){}^{198}\text{Au}$	1567	1560 ± 40	1.00	1565.
${}^{232}\text{Th}(n,\gamma){}^{233}\text{Th}$	85.4	85 ± 3	1.00	85.56
${}^{235}\text{U}(n,f)\text{F.P.}$	283.4	275 ± 5	1.03	282.0
${}^{238}\text{U}(n,\gamma){}^{239}\text{U}$	277.0	275 ± 5	1.01	277.5
${}^{239}\text{Pu}(n,f)\text{F.P.}$	316.7	301 ± 10	1.05	303.9
${}^{237}\text{Np}(n,f)\text{F.P.}$	6.54	--	--	6.82
${}^{238}\text{U}(n,f)\text{F.P.}$	1.92	--	--	--

* Includes ${}^{115}\text{In}(n,\gamma){}^{116\text{g}}\text{In}$

TABLE 6
BENCHMARK 3 - $\Sigma\Sigma$ SPECTRUM*

Reaction	Calculated $\bar{\sigma}$ (mb)	Measured ⁽⁶⁾ $\bar{\sigma}$ (mb)	Calc/Meas.	Norm to ²³⁵ U	Magurno ⁽⁵⁾
⁶ Li(n, α) ³ T	923.3	--			924.1
¹⁰ B(n, α) ⁷ Li	1514	--			1605.
²³ Na(n, γ) ²⁴ Na	1.405	--			1.392
⁴⁵ Sc(n, γ) ⁴⁶ Sc	19.01	--			18.79
⁵⁸ Fe(n, γ) ⁵⁹ Fe	5.377	--			5.093
⁵⁹ Co(n, γ) ⁶⁰ Co	46.21	--			55.60
⁶³ Cu(n, γ) ⁶⁴ Cu	38.76	40.0 $\pm$ 2.0	0.97	1.01	41.53
¹¹⁵ In(n, γ) ^{116m} In	283.6	261 $\pm$ 9	1.09	1.14	277.0
¹⁹⁷ Au(n, γ) ¹⁹⁸ Au	371.2	442 $\pm$ 8	0.84	0.88	331.4
²³² Th(n, γ) ²³³ Th	263.3	--			261.4
²³⁵ U(n,f)F.P.	1518	1589 $\pm$ 55	0.96	1.00	1598.
²³⁸ U(n, γ) ²³⁹ U	220.8	194 $\pm$ 8	1.14	1.19	223.2
²³⁹ Pu(n,f)F.P.	1750	1875 $\pm$ 66	0.93	0.98	1753
²³⁷ Np(n,f)F.P.	607.4	634 $\pm$ 22	0.96	1.00	625.5
²³⁸ U(n,f)F.P.	81.57	87.4 $\pm$ 3.1	0.93	0.98	86.46
²⁷ Al(n,p) ²⁷ Mg	0.869	1.033 $\pm$ 0.103	0.84	0.88	1.047
²⁷ Al(n, α) ²⁴ Na	0.153	0.182 $\pm$ 0.005	0.84	0.88	0.208
³² S(n,p) ³² P	14.53				16.32
⁴⁶ Ti(n,p) ⁴⁶ Sc	2.075				2.523
⁴⁷ Ti(n,np) ⁴⁶ Sc	0.000955	--			5 x 10 ⁻⁴
⁴⁷ Ti(n,p) ⁴⁷ Sc	5.174	--			5.712
⁴⁸ Ti(n,np) ⁴⁷ Sc	0.000488	--			5 x 10 ⁻⁴
⁴⁸ Ti(n,p) ⁴⁸ Sc	0.0379	--			0.0510
⁵⁵ Mn(n,2n) ⁵⁴ Mn	0.0554	--			0.080
⁵⁴ Fe(n,p) ⁵⁴ Mn	17.20	--			19.56
⁵⁶ Fe(n,p) ⁵⁶ Mn	0.230	0.273 $\pm$ 0.008	0.84	0.88	0.296
⁵⁹ Co(n, α) ⁵⁶ Mn	0.0326	--			0.043
⁵⁹ Co(n,2n) ⁵⁸ Co	0.0392	--			0.056
⁵⁸ Ni(n,2n) ⁵⁷ Ni	0.000707	--			6.88 x 10 ⁻⁴
⁵⁸ Ni(n,p) ⁵⁸ Co	23.33	28.8 $\pm$ 0.7	0.81	0.85	26.23
⁶⁰ Ni(n,p) ⁶⁰ Co	0.5376	--			0.683
⁶³ Cu(n, α) ⁶⁰ Co	0.0779	--			0.104
⁶⁵ Cu(n,2n) ⁶⁴ Cu	0.0708	--			0.102
¹¹⁵ In(n,n') ^{115m} In	50.79	58.6 $\pm$ 1.2	0.87	0.91	54.37
¹²⁷ I(n,2n) ¹²⁶ I	.2645	--			0.393
²³² Th(n,f)F.P.	18.50	21.0 $\pm$ 1.3	0.88	0.92	19.77

* 136 Point Differential Spectrum⁽⁶⁾

TABLE 7
BENCHMARK 4 - CFRMF SPECTRUM (7)

Reaction	Calculated $\bar{\sigma}$ (mb)	Measured ⁽⁸⁾ $\bar{\sigma}$ (mb)	Calc/ Meas.	Norm. to 235U
${}^6\text{Li}(n,\alpha){}^3\text{T}$	989.4	900 $\pm$ 39	1.10	1.15
${}^{10}\text{B}(n,\alpha){}^7\text{Li}$	1695.	1940 $\pm$ 60	0.87	0.91
${}^{23}\text{Na}(n,\gamma){}^{24}\text{Na}$	1.83	--	--	--
${}^{45}\text{Sc}(n,\gamma){}^{46}\text{Sc}$	20.11	25.0 $\pm$ 0.9	0.80	0.83
${}^{58}\text{Fe}(n,\gamma){}^{59}\text{Fe}$	6.02	6.52 $\pm$ 0.22	0.92	0.97
${}^{59}\text{Co}(n,\gamma){}^{60}\text{Co}$	74.10	77.6 $\pm$ 3.6	0.76	0.79
${}^{63}\text{Cu}(n,\gamma){}^{64}\text{Cu}$	47.62	48.4 $\pm$ 2.5	0.98	1.03
${}^{115}\text{In}(n,\gamma){}^{116\text{m}}\text{In}$	300.3	300 $\pm$ 11	1.00	1.05
${}^{197}\text{Au}(n,\gamma){}^{198}\text{Au}$	416.3	452 $\pm$ 14	0.92	0.96
${}^{232}\text{Th}(n,\gamma){}^{233}\text{Th}$	285.1	--	--	--
${}^{235}\text{U}(n,f)$ F.P.	1587.	1659 $\pm$ 53	0.96	1.00
${}^{238}\text{U}(n,\gamma){}^{239}\text{U}$	230.2	196 $\pm$ 11	1.17	1.23
${}^{239}\text{Pu}(n,f)$ F.P.	1771.	1930 $\pm$ 60	0.92	0.96
${}^{237}\text{Np}(n,f)$ F.P.	547.5	591 $\pm$ 21	0.93	0.97
${}^{238}\text{U}(n,f)$ F.P.	69.22	81.0 $\pm$ 0.3	0.85	0.89
${}^{27}\text{Al}(n,\gamma){}^{28}\text{Al}$	0.860	0.931 $\pm$ 0.033	0.92	0.97
${}^{27}\text{Al}(n,\alpha){}^{24}\text{Na}$	0.144	0.172 $\pm$ 0.053	0.84	0.88
${}^{32}\text{S}(n,p){}^{32}\text{P}$	13.56	--	--	--
${}^{46}\text{Ti}(n,p){}^{46}\text{Sc}$	2.09	2.78 $\pm$ 0.10	0.75	0.79
${}^{47}\text{Ti}(n,np){}^{46}\text{Sc}$	.000946	--	--	--
${}^{47}\text{Ti}(n,p){}^{47}\text{Sc}$	4.69	4.44 $\pm$ 0.22	0.95	0.99
${}^{48}\text{Ti}(n,np){}^{47}\text{Sc}$	.000502	--	--	--
${}^{48}\text{Ti}(n,p){}^{48}\text{Sc}$	.0366	0.073 $\pm$ 0.003	0.50	0.52
${}^{55}\text{Mn}(n,2n){}^{54}\text{Mn}$	0.0574	--	--	--
${}^{54}\text{Fe}(n,n){}^{54}\text{Mn}$	16.36	18.5 $\pm$ 0.6	0.88	0.92
${}^{56}\text{Fe}(n,p){}^{56}\text{Mn}$	0.223	--	--	--
${}^{59}\text{Co}(n,\alpha){}^{56}\text{Mn}$	0.0310	--	--	--
${}^{59}\text{Co}(n,2n){}^{58}\text{Co}$	0.0408	--	--	--
${}^{58}\text{Ni}(n,2n){}^{57}\text{Ni}$	0.000717	--	--	--
${}^{58}\text{Ni}(n,p){}^{58}\text{Co}$	21.77	25.6 $\pm$ 0.8	0.85	0.89
${}^{60}\text{Ni}(n,p){}^{60}\text{Co}$	0.522	--	--	--
${}^{63}\text{Cu}(n,\alpha){}^{60}\text{Co}$	0.0740	--	--	--
${}^{65}\text{Cu}(n,2n){}^{64}\text{Cu}$	0.0729	--	--	--
${}^{115}\text{In}(n,n'){}^{115\text{m}}\text{In}$	43.57	53 $\pm$ 3	0.82	0.86
${}^{127}\text{I}(n,2n){}^{126}\text{I}$	0.264	--	--	--
${}^{232}\text{Th}(n,f)$ F.P.	15.84	--	--	--

TABLE 8

BENCHMARK 5 - FISSION SPECTRUM

Reaction	Calculated Cross Section		Measured $\bar{\sigma}$ (9)	Calc (T=1.32)/ Measured
	T = 1.29 $\bar{\sigma}$ (mb)	T = 1.32 $\bar{\sigma}$ (mb)		
Li(n, α) ³ T	487.1	486.2	--	
⁶ B(n, α) ⁷ Li	515.0	512.2	--	
²³ Na(n, γ) ²⁴ Na	.295	.291	--	
⁴⁵ Sc(n, γ) ⁴⁶ Sc	5.98	5.88	--	
⁵⁵ Fe(n, γ) ⁵⁶ Fe	1.71	1.69	--	
⁵⁹ Co(n, γ) ⁶⁰ Co	6.59	6.50	--	
⁶³ Cu(n, γ) ⁶⁴ Cu	11.18	11.04	10.1 $\pm$ 1.5	1.09
¹¹⁵ In(n, γ) ^{116m} In	138.5	136.7	--	
¹⁹⁷ Au(n, γ) ¹⁹⁸ Au	86.58	85.15	88 $\pm$ 5	0.97
²³² Th(n, γ) ²³³ Th	105.5	103.9	83 $\pm$ 3.5	1.25
U(n,f)F.P.	1243	1243	1250 (REF)	0.99
²³⁵ U(n, γ) ²³⁶ U	76.87	75.68	--	
²³⁸ U(n,f)F.P.	1780	1783	--	
²³⁵ Mo(n,f)F.P.	1313	1323	1370 $\pm$ 75	0.97
²³⁵ U(n,f)F.P.	288.9	295.4	328 $\pm$ 10	0.90
²⁴ Mg(n,p) ²⁷ Mg	3.93	4.22	4.0 $\pm$ 0.4	1.06
²³ Na(n, α) ²⁴ Na	.710	.801	0.73 $\pm$ 0.02	1.10
³¹ P(n,p) ³² P	61.35	63.89	69 $\pm$ 2	0.93
⁴⁴ Ti(n,p) ⁴⁶ Sc	9.51	10.24	12.3 $\pm$ 0.3	0.83
⁴⁴ Ti(n,np) ⁴⁶ Sc	.00495	.00629	--	
⁴⁷ Ti(n,p) ⁴⁷ Sc	20.64	21.42	20.0 $\pm$ 2.0	1.07
⁴⁷ Ti(n,np) ⁴⁷ Sc	.00262	.00327	--	
⁴⁸ Ti(n,p) ⁴⁸ Sc	.177	.200	0.32 $\pm$ 0.02	0.62
⁵⁵ Mn(n,2n) ⁵⁴ Mn	.300	.363	0.25 $\pm$ 0.01	1.45
⁵⁵ Mn(n,p) ⁵⁵ Mn	74.07	77.69	82.5 $\pm$ 2.0	0.94
⁵⁵ Mn(n,p) ⁵⁵ Mn	1.033	1.145	1.07 $\pm$ 0.06	1.07
⁵⁹ Co(n,2n) ⁵⁸ Co	.213	.259	--	
⁵⁵ Mn(n, γ) ⁵⁶ Mn	.149	.168	0.156 $\pm$ 0.006	1.08
⁵⁸ Ni(n,n) ⁵⁸ Ni	.00375	.00470	--	
⁵⁹ Ni(n,p) ⁵⁹ Co	97.24	101.5	113 $\pm$ 3	0.90
⁵⁹ Ni(n,p) ⁵⁹ Co	2.414	2.659	--	
⁶⁰ Ni(n, γ) ⁶⁰ Co	.354	.396	0.50 $\pm$ 0.05	0.79
⁶³ Cu(n,2n) ⁶² Cu	.380	.460	--	
¹¹⁵ In(n,n') ^{115m} In	163.8	166.7	188 $\pm$ 4	0.89
¹²⁷ I(n,2n) ¹²⁶ I	1.377	1.635	1.09 $\pm$ 0.05	1.50
²³² Th(n,f)F.P.	67.26	69.02	83.0 $\pm$ 3.5	0.83

HEDL

HANFORD ENGINEERING DEVELOPMENT LABORATORY

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

W. Y. Matsumoto, L. S. Kellogg and A. I. Davis

Objectives

- Establish reliable and qualified sources of high purity foil materials required for the ILRR program and the 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

QA analyses were completed on mixed plutonium-uranium oxide powder prepared from five of one hundred Fast Test Reactor (FTR) fuel pellets reserved for use in miniature neutron dosimeter capsules. Inadequate analyses on the powder sample at HEDL and poor agreement between HEDL analyses (on the powder and separate pellet samples) and analyses at HNL (ORNL) on the powder leaves a large uncertainty in the correct plutonium and uranium assays for this material.

Evaluations of gamma counting mass assay QA check data on fission dosimeters are still in progress. Procedural standards for counting and evaluating data are being evolved and improved. Definite improvement was noted in the reliability of mass assay values for fission foils fabricated since the gamma QA checks were begun.

Accomplishments and Status

The ninety-five pellets remaining from the one hundred $\text{PuO}_2\text{-UO}_2$ mixed oxide FTR fuel pellets from KM Lot. No. 20 reserved for use in neutron dosimetry tests were shipped to HNL (ORNL) to be held for processing into dosimeters as required. Five of the pellets had previously been oxidized and crushed into powder at HEDL and sent to ORNL for additional QA analyses and fabrication into miniature neutron dosimeters. Fifteen vanadium encapsulated dosimeters have been prepared, nine were used in an EBR-II dosimetry test. Analytical results thus far completed on the FTR fuel material leave large uncertainties in the plutonium and uranium assays of the powder material used in the dosimeters. These results are summarized in Table HEDL-1. This table gives corrected values for the data originally given in Table HEDL-2, Reference 9, in which the plutonium isotopic composition values were inadvertently mixed up. Additional HEDL analyses which were to have been run on the powdered material sent to ORNL were not done because of unfortunate intralaboratory miscommunications coupled with the wish to preserve as much material as possible to send to ORNL. The Table HEDL-1 data show large unexplainable discrepancies in the plutonium and uranium assay data.

Since only nine dosimeters prepared from this material have been used, it is not worthwhile to spend the effort required to establish accurate assay data on this small batch of material. (The nine dosimeters used can be individually analysed destructively by isotope dilution mass spectrometric methods).

It is recommended that future dosimeters to be made using FTR fuel use a new batch of material prepared from the ninety-five remaining FTR fuel pellets. This material should be prepared and mixed in a single batch, thoroughly analysed by at least two independent laboratories, and stored under carefully defined controlled conditions. Analytical results from two laboratories should agree to better than 0.5% before the assay data are considered acceptable. Consideration should be given to conversion of this material to stoichiometric $\text{PuO}_2\text{-UO}_2$, particularly if the oxide wire technique is used to prepare dosimeters.

Tables HEDL-2 through HEDL-13 summarize some of the data from gamma counting mass assay QA checks on encapsulated fission dosimeters. At the start of this program no reference dosimeters were designated so that one or more arbitrarily selected dosimeters from each production batch were used for "reference dosimeters" at HEDL. In these cases only single gamma counting measurements on the HEDL selected reference dosimeters had been made at ORNL. (Tables HEDL-2 through HEDL-11). For instances where the ORNL gamma measurement happened to be atypical, a consistent bias would be expected in the ORNL data column. If the ORNL weight measurement for the selected reference dosimeter was atypical, a consistent bias would be expected in both the HEDL and ORNL data columns. There is some indication that this may have occurred in a few cases, but the data have not yet been evaluated thoroughly enough to reach a definite conclusion.

For recent and future production batches of dosimeters ordered, designated reference dosimeters that have been measured several times by gamma counting at ORNL are being used for the reference dosimeters. (Tables HEDL-10 and 11). This should decrease the chance of bias in inter-laboratory comparisons due to use of an atypical measurement. The values tabulated in Tables HEDL-2 through HEDL-11 were determined as follows:

Weight ratios for each dosimeter to the reference dosimeter were calculated from values for dosimeter material obtained at ORNL by precision weighing. Counting rate ratios (count ratios) for each dosimeter to the reference dosimeter were measured at HEDL and calculated from data supplied by ORNL. The tabulated values are the weight ratios (common to both columns) divided by the count ratios obtained at HEDL and ORNL respectively.

For an ideal case in which the reference dosimeter is an accurate standard, the tabulated values should all fall close to 1.0 with a mean of 1.0 if all weighing and counting data are unbiased and sufficiently precise. A value differing significantly from 1.0 indicates that there is a significant error in either the weight or count data. If the mean of

the tabulated values differs significantly from 1.0, the weight or count data for the reference dosimeter used is indicated to be atypical and probably in error. Although these measurements cannot provide an accurate mass assay at least until accurate reference dosimeter standards have been established, they have been highly useful in detecting anomalous weights in encapsulated fission dosimeter materials.

Expected Achievements

Destructive analysis will be made on the nine vanadium FTR fuel dosimeters used in the EBR-II 75D mapping test. Analysis of additional QA gamma counting data for both aluminum and vanadium encapsulated dosimeters will be completed and where possible correlation will be made to reference foils for which several counting measurements were made by both ORNL and HEDL.

TABLE HEDL-1
ANALYTICAL RESULTS ON FTR FUEL MATERIAL FROM KM PELLET LOT NO. 20

Laboratory:	HEDL	VENDOR	CALC'D ^(a)	HEDL	ORNL ^(b)
Material Form:	(Pellets)	(Pellets)		(Powder) ^(c)	(Powder) ^(c)
Weight % Pu:	24.43±.06 ^(d)	24.30±.06	23.72	23.82	20.96
Weight % U:	63.87±.05 ^(e)	64.24±.18	62.02	63.56	58.86
O/M ^(f) (Range):	(1.962-1.967)	(1.963-1.968)	2.482	—	—
Isotopic Composition					
	(Atom %)				
Pu-238:	0.0606±.003	(Pu-239+	—	—	0.062
Pu-239:	86.365 ±.02	Pu-241=	—	—	86.08
Pu-240:	11.660 ±.02	88.148_	—	—	11.86
Pu-241:	1.721 ±.01		—	—	1.802
Pu-242:	0.193 ±.01		—	—	0.193
Pu-244:	—		—	—	<0.0005

- (a) Calculated values for material oxidized to the form PuO₂ + U₃O₈, based on the HEDL assay values for the pellets and assuming an average value of 1.9645 for the O/M ratio in the pellets (before oxidation).
- (b) Results from H. L. Adair, HNL (ORNL).
- (c) Powder from oxidation and crushing five KM Lot. No. 20 pellets.
- (d) Average value from HEDL Analysis of eleven KM Lot No. 20 pellets.
- (e) Average value from HEDL Analysis of three KM Lot No. 20 pellets.
- (f) Oxygen to metal ratio--FTR fuel pellets are specified to be slightly substoichiometric.

TABLE HEDL-2
 GAMMA COUNTING MASS ASSAY QA CHECK DATA
 ALUMINUM WRAPPED 0.1 MIL ²³⁵U FOILS
 U-235-264-CM, PO #Y4B-S44-14953

Dosimeter Number	ORNL Reported U Wt. (mg)	Weight Ratio ^(a) ÷ Count Ratio ^(b)		Remarks
		HEDL	ORNL	
6	7.229	1.000	1.020	
7	6.723	.981	.982	
8	7.660	1.000	.974	
9	7.400	1.001	.970	
10	7.122	1.002	.982	
11	6.890	.997	1.004	
12	7.261	.999	.971	
13	7.606	.998	.991	
14	6.419	.999	.990	
15	6.716	.978	.987	
16	7.125	.998	.992	
17	6.771	.997	.998	
18	7.007	1.000	.990	
19	6.877	1.000	.997	
20	7.468	1.002	1.004	
21	6.924	.999	.977	
22	7.019	1.002	.981	
23	7.624	.999	.992	
24	6.544	1.000	.996	
25	7.495	.999	.985	
57	7.043	1.0	1.0	Reference Foil
58	7.613	.996	.985	
59	6.314	.994	.976	

- (a) Ratio of the weight of the dosimeter material to the weight of the dosimeter material in the reference dosimeter-values determined by weighing at ORNL.
- (b) Ratio of the gamma counting rate of the dosimeter to the counting rate of the reference dosimeter-measurements made at both HEDL and ORNL.

TABLE HEDL-3
GAMMA COUNTING MASS ASSAY QA CHECK DATA
ALUMINUM WRAPPED 5 MIL ²³⁵U FOILS
U-235-264-CM, PO #Y4B-S44-14953

Dosimeter Number	ORNL Reported U Wt. (mg)	Weight Ratio ^(a) ÷ Count Ratio ^(b)		Remarks
		HEDL	ORNL	
26	326.475	0.999	1.035	
27	332.033	1.008	0.992	
28	332.853	1.004	1.003	
29	333.376	1.007	0.971	
30	317.455	1.000	0.975	
31	327.145	1.001	0.991	
32	331.980	1.000	1.014	
33	318.455	0.991	1.000	
34	329.254	1.000	0.990	
35	325.666	1.005	1.002	
36	313.925	1.005	0.974	
37	330.152	1.008	0.992	
38	327.510	1.007	1.015	
39	320.543	1.006	1.027	
40	323.055	1.004	0.995	
41	322.265	1.003	1.001	
42	330.155	1.014	0.986	
43	331.685	1.003	1.014	
44	327.188	0.999	1.016	
45	330.550	1.004	1.005	
60	303.655	0.989, 0.987	0.998	, recount
61	321.255	1.000	1.000	Reference foil
62	331.925	1.008, 1.009	0.989	, recount

- (a) Ratio of the weight of the dosimeter material to the weight of the dosimeter material in the reference dosimeter-values determined by weighing at ORNL.
- (b) Ratio of the gamma counting rate of the dosimeter to the counting rate of the reference dosimeter-measurements made at both HEDL and ORNL.

TABLE HEDL-4
 GAMMA COUNTING MASS ASSAY QA CHECK DATA
 ALUMINUM WRAPPED 10 MIL ²³⁵U FOILS
 U-235-264-CM, PO #Y4B-S44-14953

<u>Dosimeter Number</u>	<u>ORNL Reported U Wt. (mg)</u>	<u>Weight Ratio (a) ÷ Count Ratio (b)</u>		<u>Remarks</u>
		<u>HEDL</u>	<u>ORNL</u>	
46	622.150	1.006	1.012	
47	631.915	1.002	1.012	
48	618.907	1.019	1.002	
49	633.445	1.028	0.988	
50	631.045	1.002	0.995	
51	624.890	1.000	1.000	Reference foil
52	610.582	0.998	0.985	
53	631.715	1.000	1.005	
54	625.873	1.000	0.998	
55	626.050	1.001	1.002	
63	608.238	0.985	0.996	
64	622.325	0.997	0.996	
65	641.163	1.007	1.021	

(a) Ratio of the weight of the dosimeter material to the weight of the dosimeter material in the reference dosimeter-values determined by weighing at ORNL.

(b) Ratio of the gamma counting rate of the dosimeter to the counting rate of the reference dosimeter-measurements made at both HEDL and ORNL.

TABLE HEDL- 5
GAMMA COUNTING MASS ASSAY QA CHECK DATA
ALUMINUM ENCAPSULATED 5 MIL ²³⁹PU FOILS
PU-239-453-BM, PO #Y4B-S44-14952

Dosimeter Number	ORNL Reported Pu Wt. (mg)	Weight Ratio (a) ÷ Count Ratio (b)		Remarks
		HEDL	ORNL	
43	242.477	1.000*	0.992	*Ave of 4, Range .48%
44	238.213	1.003*	0.999	*Ave of 5, Range .56%
45	243.664	1.030	1.003	
46	242.656	0.998	0.988	
47	249.205	0.999*	1.004	*Ave of 2, Range .21%
48	246.114	1.002	0.991	
49	244.790	1.001*	1.019	*Ave of 2, Range .56%
50	243.824	0.999	1.004	
51	244.509	1.004	1.004	
52	247.777	0.997	1.000	
53	245.605	0.998*	0.993	*Ave of 2, Range .17%
54	243.562	1.001	0.996	
55	247.852	1.001	0.993	
56				Not received
57	243.331	0.997	0.995	
58	242.323	1.011	1.027	
59				Not received
60	244.150	1.002	1.006	
61				Not received
62	240.603	0.998	0.991	
43A	242.961	1.000	1.000	Reference foil
44A	247.835	1.002	0.991	
45A	246.242	0.998	1.003	
46A	238.334	0.980	0.999	
48A	244.685	1.000	0.997	
49A	241.959	1.002	1.003	

- (a) Ratio of the weight of the dosimeter material to the weight of the dosimeter material in the reference dosimeter-values determined by weighing at ORNL.
- (b) Ratio of the gamma counting rate of the dosimeter to the counting rate of the reference dosimeter-measurements made at both HEDL and ORNL.

TABLE HEDL- 6
 GAMMA COUNTING MASS ASSAY QA CHECK DATA
 ALUMINUM ENCAPSULATED 10 MIL ²³⁹PU FOILS
 PU-239-453-BM, PO #Y4B-S44-14952

Dosimeter Number	ORNL Reported Pu Wt. (mg)	Weight Ratio ^(a) ÷ Count Ratio ^(b)		Remarks
		HEDL	ORNL	
64	497.704	1.003	1.012	
65	477.663	1.000	1.014	
66	483.304	0.992	1.011	
67	499.175	0.994	1.009	
68	491.605	1.001	1.005	
69	481.001	0.997	1.006	
70	499.510	0.999	1.003	
71	490.219	1.002	1.040	
72	493.638	0.994	1.008	
73				
74	497.505	1.003	1.005	Not received
75	494.961	0.999	1.014	
76	499.086	0.999	1.004	
77	496.504	1.000	1.000	Reference foil

- (a) Ratio of the weight of the dosimeter material to the weight of the dosimeter material in the reference dosimeter-values determined by weighing at ORNL.
- (b) Ratio of the gamma counting rate of the dosimeter to the counting rate of the reference dosimeter-measurements made at both HEDL and ORNL.

TABLE HEDL-7
 GAMMA COUNTING MASS ASSAY QA CHECK DATA
 ALUMINUM ENCAPSULATED 6-7 MIL ^{237}Np FOILS
 NP-237-24-HP-M, PO #Y4B-S44-14959

Dosimeter Number	ORNL Reported Np Wt. (mg)	Weight Ratio (a) $\div$ Count Ratio (b)		Remarks
		HEDL	ORNL	
22	338.835	1.000	1.000	Reference foil
23	331.711	0.992	0.987	
24	326.726	0.993	0.975	
25	353.978	1.003	1.004	
26	331.506	0.960	0.985	
27	349.767	1.005	0.998	
28	340.068	0.998	0.992	
29	349.520	1.006	0.999	
30	344.531	1.002	0.996	
31	379.744	1.008	1.028	
32	377.073	1.008	1.029	
33	304.720	0.993	0.964	
34	328.199	0.991	0.985	
35	335.375	1.000	0.990	
36	352.498	1.001	1.006	
37	352.781	1.001	0.998	
38	356.046	1.005	1.017	
39	369.244	1.007	1.026	
40	372.177	1.005	1.029	
41	361.642	1.008	1.022	
42	372.148	1.007	1.028	
43	369.301	1.003	1.026	
44	334.504	0.999	0.981	

- (a) Ratio of the weight of the dosimeter material to the weight of the dosimeter material in the reference dosimeter-values determined by weighing at ORNL.
- (b) Ratio of the gamma counting rate of the dosimeter to the counting rate of the reference dosimeter-measurements made at both HEDL and ORNL.

TABLE HEDL-8
 GAMMA COUNTING MASS ASSAY QA CHECK DATA
 VANADIUM ENCAPSULATED ^{237}Np DOSIMETERS (SERIES 1)
 BATCH NP-237-24HP (NpO_2), PO # Y4B-S44-14965

Dosimeter Number	ORNL Reported NpO_2 Wt. (mg)	Weight Ratio ^(a) ÷ Count Ratio ^(b)		Remarks
		HEDL	ORNL	
1	5.899	0.995	1.000	
2	6.590	1.003	1.015	
3	5.642	0.992	0.993	
4	6.904	0.992	1.015	
5	5.937	1.000	1.000	Reference foil
6	6.908	0.982	0.989	
7	6.079	0.993	1.003	
8	6.461	0.987	1.002	
9	6.636	0.987	0.987	
10	6.040	0.992	1.005	
11	5.157	0.981	0.999	
12	6.753	1.016, 1.013	1.026	,recount
13	5.817	0.986	0.994	
14	6.095	0.997	1.010	
15	5.632	0.982	0.974	
16	6.339	0.996	1.000	
17	6.979	0.995, .994	1.012	
18	6.870		1.013	No HEDL data
19	6.207	1.003	1.001	
20	6.858	0.990	1.004	
21	5.760	0.994	0.980	
22	6.597	0.985	0.988	
23	5.112	0.987	0.979	
24	6.395	0.997	1.003	
25	6.444	0.981	0.984	
26	6.657	0.991	0.999	
27	6.052	0.978	0.963	
28	6.752	0.991	1.001	
29	5.689	0.987	1.002	
30	6.785	0.987	0.994	

- (a) Ratio of the weight of the dosimeter material to the weight of the dosimeter material in the reference dosimeter-values determined by weighing at ORNL.
- (b) Ratio of the gamma counting rate of the dosimeter to the counting rate of the reference dosimeter-measurements made at both HEDL and ORNL.

TABLE HEDL- 9
GAMMA COUNTING MASS ASSAY QA CHECK DATA
VANADIUM ENCAPSULATED ^{237}Np DOSIMETERS (SERIES X)
BATCH NP-237-24HP (NpO_2), PO #Y4B-S44-14965

Dosimeter Number	ORNL Reported NpO_2 Wt. (mg)	<u>Weight Ratio (a) ÷ Count Ratio (b)</u>		Remarks
		HEDL	ORNL	
1X	5.780	1.011	1.009	(Dosimeter No. 5 from Series 1 was used as the refer- ence).
2X	6.033	0.991	0.995	
3X	5.272	1.020	1.024	
4X	6.841	0.996	1.003	
5X	5.961	0.980, 0.983	0.967	
6X	6.818	0.993	0.996	,recount (rotated 180°)
7X	6.276	0.998	1.019	
8X	6.596	1.007	1.020	
9X	6.510	0.998	1.018	
10X	6.200	1.029	1.021	
11X	5.795	1.012	1.016	
12X	6.638	0.994	1.000	
13X	5.573	0.982	0.981	
14X	5.209	1.011	0.969	
15X	6.673	0.993	0.997	
16X	6.162	1.008	0.995	
17X	6.046	1.016	1.025	
18X	6.697	0.987	0.992	
19X	6.265	0.995	0.998	
20X	5.268	1.024	1.002	
21X	5.555	1.010	0.985	
22X	5.963	1.024	1.010	
23X	6.712	1.011	0.976	
24X	5.303	0.985	0.979	
25X	5.855	1.025	0.989	
26X	5.775	1.005	0.967	
27X	6.435	1.007	1.022	
28X	6.197	0.990	0.996	
29X	6.584	0.992	0.992	
30X	5.437	0.983	0.984	

- (a) Ratio of the weight of the dosimeter material to the weight of the dosimeter material in the reference dosimeter-values determined by weighing at ORNL.
- (b) Ratio of the gamma counting rate of the dosimeter to the counting rate of the reference dosimeter-measurements made at both HEDL and ORNL.

TABLE HEDL-10
 GAMMA COUNTING MASS ASSAY QA CHECK DATA
 VANADIUM-51 ENCAPSULATED ^{237}Np DOSIMETERS (SERIES A)
 BATCH NP-237-24HP (NpO_2), PO #Y4B-S44-14965

Dosimeter Number	ORNL Reported NpO_2 Wt. (mg)	Weight Ratio ^(a)		Count Ratio ^(b)		Remarks
		HEDL		ORNL		
1A	6.240	1.037	1.034	1.022		, recount Not received
2A						
3A	6.134	0.979		0.975		
4A	7.656	1.001		1.001		(Dosimeter No 5 from Series 1 was used as the reference).
5A	5.797	0.986		0.981		
6A	6.518	1.006		0.991		
7A	6.786	1.021		1.020		
8A	5.717	1.001		0.990		
9A	4.916	0.991		0.991		
10A	6.700	1.023	1.024	1.010		, recount
11A	6.635	0.995		0.992		
12A	5.657	1.009		1.007		
13A	6.616	1.014		1.011		
14A	5.412	0.993		0.993		
15A	5.842	0.991		0.975		
16A	6.420	1.005		1.001		
17A	6.994	1.011		1.008		
18A	7.016	0.990		0.985		
19A						Not received
20A	6.018	0.996		0.980		
21A	5.374	0.979	0.980	0.974		, recount
22A	6.489	1.017		0.999		
23A	6.696	0.990		0.986		
24A	6.359	0.992		0.987		
25A	6.533	0.993		0.987		
26A	5.651	1.007		1.009		
27A	6.085	0.992		0.981		
28A	6.469	1.032	1.031	1.022		, recount
29A	6.936	0.997		0.997		

- (a) Ratio of the weight of the dosimeter material to the weight of the dosimeter material in the reference dosimeter-values determined by weighing at ORNL.
- (b) Ratio of the gamma counting rate of the dosimeter to the counting rate of the reference dosimeter-measurements made at both HEDL and ORNL.

(continued)

TABLE HEDL-10 (cont.)
 GAMMA COUNTING MASS ASSAY QA CHECK DATA
 VANADIUM ENCAPSULATED ^{237}Np DOSIMETERS (SERIES A)
 BATCH NP-237-24HP (NpO_2), PO #Y4B-S44-14965

Dosimeter Number	ORNL Reported NpO_2 Wt. (mg)	Weight Ratio ^(a) ÷ Count Ratio ^(b)		Remarks
		HEDL	ORNL	
30A	5.829	0.992	0.985	
31A	6.194	1.029, 1.028	1.014	, recount
32A	6.936	1.009	0.990	
33A	5.903	1.000	0.995	
34A	6.739	0.988	0.990	
35A	6.646	1.000	0.995	
36A				Not received
37A	6.869	0.989	0.988	
38A	6.695	1.017	1.018	
39A	6.546	1.006	1.000	
40A	5.971	0.996	0.986	
41A	6.082	0.998	0.994	
42A	6.031	0.985	0.972	
43A	5.158	0.986	0.974	

(Dosimeter No. 5
from Series 1 was
used as the refer-
ence).

- (a) Ratio of the weight of the dosimeter material to the weight of the dosimeter material in the reference dosimeter-values determined by weighing at ORNL.
- (b) Ratio of the gamma counting rate of the dosimeter to the counting rate of the reference dosimeter-measurements made at both HEDL and ORNL.

TABLE HEDL-11
 GAMMA COUNTING MASS ASSAY QA CHECK DATA
 VANADIUM ENCAPSULATED ^{237}Np DOSIMETERS (SERIES W)
 BATCH NP-237-24HP (NpO_2) PO #Y51-444-26534

Dosimeter Number	ORNL Reported Weight (mg)		Weight Ratio ^(a) : Count Ratio ^(b)		Remarks
	NpO_2	Np	HEDL	ORNL	
1W	8.025	7.017	0.995	0.994	
2W	6.201	5.422	1.013	0.986	
3W	6.786	5.934	0.997	0.988	
4W	7.889	6.898	0.998	1.010	
5W	6.444	5.635	0.982	0.987	
6W	7.710	6.742	0.991	0.989	
7W	7.200	6.296	0.987	0.997	
8W	7.037	6.153	0.985	0.996	
9W	8.414	7.357	1.003	0.978	
10W	5.806	5.077	0.964	0.977	
11W	5.477	4.789	1.073	0.981	To be reduced
12W	7.043	6.158	0.998	0.996	
13W	7.761	6.786	0.979	0.980	
14W	8.276	7.237	0.985	0.999	
15W	7.401	6.471	0.999	0.985	
16W	6.615	5.784	0.992	1.002	
17W	7.739	6.767	1.012	0.984	To be reduced
18W	8.554	7.480	0.977	1.005	
19W	6.680	5.841	1.006	0.982	
20W	7.424	6.492	1.013	0.979	To be reduced
21W	5.466	4.779	0.999	0.986	
22W	6.770	5.920	1.000	1.000	Reference foil ^(c)
23W	7.495	6.554	1.004	0.999	
24W	8.347	7.299	0.996	1.001	
25W	7.781	6.804	0.980	0.997	

- (a) Ratio of the ^{237}Np reported weight of the dosimeter to the ^{237}Np reported weight of the reference dosimeter-values calculated and reported by ORNL from weighings made at ORNL.
- (b) Ratio of the gamma counting rate of the dosimeter to the counting rate of the reference dosimeter-measurements made at both HEDL and ORNL.
- (c) Multiple counts made on this sample by ORNL.

TABLE HEDL-11 (cont.)
 GAMMA COUNTING MASS ASSAY QA CHECK DATA
 VANADIUM ENCAPSULATED ^{237}Np DOSIMETERS (SERIES W)
 BATCH NP-237-24 HP (NpO_2) PO #Y51-444-26534

<u>Dosimeter Number</u>	<u>ORNL Reported Weight (mg)</u>		<u>Weight Ratio (a) ÷ Count Ratio (b)</u>		<u>Remarks</u>
	<u>NpO_2</u>	<u>Np</u>	<u>HEDL</u>	<u>ORNL</u>	
26W	7.591	6.638	0.985	1.006	
27W	7.532	6.586	0.995	1.013	
28W	7.452	6.516	1.001	1.008	
29W	7.042	6.158	1.012	1.003	
30W	7.342	6.420	0.986	1.006	
31W	6.929	6.059	0.983	1.003	
32W	7.296	6.380	0.994	0.998	
33W	7.191	6.288	0.989	0.997	
34W	6.953	6.080	0.995	0.994	
35W	7.536	6.589	1.003	0.995	
36W	6.873	6.010	0.985	0.994	
37W	7.642	6.682	0.999	1.000	
38W	6.826	5.969	1.005	0.997	
39W	6.556	5.733	0.992	0.995	
40W	6.899	6.032	1.000	0.996	

- (a) Ratio of the ^{237}Np reported weight of the dosimeter to the ^{237}Np reported weight of the reference dosimeter-values calculated and reported by ORNL from weighings made at ORNL.
- (b) Ratio of the gamma counting rate of the dosimeter to the counting rate of the reference dosimeter-measurements made at both HEDL and ORNL.

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

M. R. Weiler

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.
- 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-1083) was prepared with an estimated uncertainty of $\pm 1.6\%$ at the 95% confidence level by the HEDL Radiochemistry Standards Laboratory in March, 1974. 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. Dudey), ANL (E. R. Ebersole), ANL (K. G. Porges), ARHCO (W. H. Zimmer), HEDL (L. S. Kellogg), LASL (D. W. Barr) and MOL (A. Fabray). One ampoule of the standard and two ampoules prepared to special counter specifications were shipped to the National Bureau of Standards (J. A. Grundl) 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.

Accomplishments and Status

HEDL T-1083 ^{140}Ba - ^{140}La Standard Solution

The ^{140}Ba - ^{140}La standard solution, T-1083, was prepared and shipped to interested ILRR program participants by the HEDL Radiochemistry

Standards Laboratory on March 15, 1974. Absolute standardization of this solution was made March 20, 1974, by 4π beta-gamma coincidence counting of four source mounts from each of two accurately prepared dilutions. The random statistical uncertainty for the eight measurements expressed as the relative standard error was $\pm 0.11\%$. The overall uncertainty of the measurement was estimated to be $\pm 1.6\%$ at the 95% confidence level. One ampoule of the standard and two ampoules prepared to special counter specifications 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 April 18, 1974, 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 comparison of this standard is not being made at this time as insufficient data has been received from the participants. Preliminary information received from a number of participants would indicate that the HEDL calibration of this standard is high by $\sim 1.3\%$.

Expected Achievements

Analysis and evaluation of the interlaboratory comparison data on the T-1083 ^{140}Ba - ^{140}La standard will be made upon receipt of data from the participants. A relative comparison of HEDL Standards T-913 and T-1083 will be made by the National Bureau of Standards, Radioactivity Section, from measurements made in the NBS $4\pi\gamma$ ionization chambers.

Correlation of analytical data from participants will be made as soon as all results have been reported.

REFERENCE STANDARD T 1083-
 $^{140}\text{Ba}/^{140}\text{Ba}$ Radioactivity Standard

This standard consists of barium-140, in transient equilibrium with its lanthanum-140 daughter, in + 0.001 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 20, 1974 was:

$$2.44_8 \times 10^6 \pm 1.6\%$$

comprised of 1.138×10^6 ntps ^{140}Ba plus $1.31_0 \times 10^6$ ntps ^{140}Ba per gram of solution.

The estimated uncertainty in the activity, 1.6 percent, is the sum of 0.3 percent which is the standard error at the 95-percent confidence level computed from 8 sets of measurements, and 1.3 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 12.79 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:	1.012N nitric acid
Carrier concentrations:	11 ppm Ba, 11 ppm La
Chloride:	0.003N (as HCl)
Density:	1.0293 grams/ml at 21.5°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 used without further purification. The 4 pi beta-gamma coincidence counting measurements were performed on March 20, 1974.

C. EBR-II DERIVED FISSION YIELDS FROM VANADIUM
ENCAPSULATED FISSION MONITORS

L. S. Kellogg, W. Y. Matsumoto, C. J. Ambrose and J. M. Ruggles

Objective

- Utilize the results of the CFRMF and other ILRR tests coupled with those of past and present EBR-II dosimetry tests to provide improved and more precise values of absolute fission yields for LMFBR fuels and materials development programs.
- Compare the results of ILRR program derived absolute fission yields with those reported elsewhere in order to discern possible parametric trends and reduce systematic errors.

Summary

A procedure for the complete dissolution of vanadium encapsulated plutonia foils was developed by C. J. Ambrose (Columbia Engineering) and J. M. Ruggles of the HEDL Radiochemistry Section. The procedure is now being applied to the dissolution of plutonium foils for neodymium burnup measurements. Eleven samples from the EBR-II Run 50H test and nine samples from the Run 75D test have been dissolved, and analysis made for ^{137}Cs , Pu and Pu isotopic as well as Nd isotopic utilizing gamma spectrometry and mass spectrometry techniques. Comparison of the $^{148}\text{Nd}/^{137}\text{Cs}$ ratios to the ENDF/B IV ^{239}Pu fission yield ratios indicate a possible 10% discrepancy in fission yield ratio. Comparison of the change in the isotopic ratio of ^{150}Nd to ^{143}Nd as a function of $\bar{E}$ derived from the dosimetry foil data has also been made. Nine EBR-II Run 75D vanadium encapsulated ^{235}U foils have also been dissolved and are in the process of being analyzed.

Accomplishments and Status

Eleven EBR-II Run 50H and nine EBR-II Run 75D vanadium encapsulated ^{239}Pu samples have been analyzed for ^{137}Cs , total Pu, Pu isotopic and Nd isotopic content. Dissolution of the samples has been accomplished using a two step dissolution procedure. After cleaning and etching the capsule

surface the vanadium is dissolved in 1-2 ml 8M HNO_3 with Cs carrier and in a leached quartz tube. After approximately 2 hours at room temperature, the vanadium solution is carefully transferred to a leached volumetric flask and 1-2 ml con HNO_3 added to the sample particles in the tube. Four drops of 1M HF is added and the sample refluxed in an oil bath at 105° to 115°C, additional 1M HF is added as required throughout the dissolution. Twenty four or more hours are typically required. The dissolved sample is quantitatively transferred to the volumetric flask with 2M HNO_3 and made to volume. Aliquots are removed for the Cs separation and for Pu and Nd separations using routine isotope dilution methods. The Nd isotopic ratios to $\bar{E}$ relationships are shown in Table HEDL-12 and the ^{151}Nd to ^{143}Nd ratio vs $\bar{E}$ in Figure HEDL-1 . A possible serious yield discrepancy is indicated by the ^{148}Nd to ^{137}Cs ratios as compared to ENDF/B-IV ^{239}Pu fission yield ratios tabulated in Table HEDL-13.

All ratios are based on atoms/sample thereby eliminating possible weight bias problems.

Expected Achievements

Additional correlations will be made to other fission products as ^{140}Ba , ^{95}Zr and ^{103}Ru . Analysis will also be completed on EBR-II Run 75D ^{235}U and ^{238}U , and will be started on FTR encapsulated driver fuel monitors.

FIGURE HEDL-1. Comparison of $^{150}\text{Nd}/^{143}\text{Nd}$ Isotopic Ratio Change With $\bar{E}$

TABLE HEDL-12

^{239}Pu FISSION PRODUCT NEODYMIUM ISOTOPIC RATIOS
EBR-II RUNS 50H and 75D

Run	Sample	Calculated $\bar{E}^*$	143/148	145/148	146/148	150/148	150/143
50H	W-55	0.943	2.6327	1.8197	1.4918	0.5972	0.2268
	W-60	0.923	2.6232	1.8156	1.4888	0.5971	0.2276
	W-53	0.923	2.6155	1.8103	1.4858	0.5987	0.2289
	W-51	0.888	2.6321	1.8184	1.4898	0.5997	0.2278
	W-5	0.853	2.6123	1.8116	1.4855	0.5973	0.2286
	W-9	0.852	2.6218	1.8151	1.4859	0.5970	0.2277
	W-8	0.834	2.6295	1.8184	1.4896	0.5999	0.2281
	W-4	0.734	2.6232	1.8124	1.4882	0.5971	0.2276
	W-15	0.714	2.6479	1.8204	1.4901	0.5935	0.2241
	W-29	0.586	2.6679	1.8287	1.4960	0.5918	0.2218
75D	8A	0.710	2.6793	1.8413	1.5019	0.5894	0.2232
	8B		2.6699	1.8353	1.4960	0.5911	0.2248
	8C		2.6706	1.8347	1.4974	0.5918	0.2249
	7A	0.535	2.7130	1.8468	1.5034	0.5845	0.2188
	7B		2.7078	1.8448	1.5049	0.5863	0.2197
	7C		2.7350	1.8555	1.5074	0.5828	0.2188
	6A	0.280	2.7669	1.8595	1.5120	0.5782	0.2121
	6B		2.7641	1.8611	1.5154	0.5782	0.2122
	6C		2.7463	1.8512	1.5092	0.5804	0.2144

HEDL-24

* Based on SAND II multiple foil flux-spectral measurements.

TABLE HEDL-13

^{239}Pu FISSION YIELD RATIO ^{148}Nd TO ^{137}Cs

EBR-II Run 50H	EBR-II Run 75D	ENDF/B-IV
0.2400	0.2452	0.2650
0.2392	0.2434	
0.2332	0.2407	
0.2436	0.2405	
0.2368	0.2411	
0.2405	0.2413	
0.2394	0.2399	
0.2319	0.2411	
0.2316	0.2385	
0.2323		
Mean 0.2368	0.2413	
Std. Dev 0.0043	0.0019	
Mean 50H+75D 0.2391		
Ratio: Mean to ENDF/B-IV=1.1083		

D. REACTION RATE MEASUREMENTS

L. S. Kellogg and W. N. McElroy

Objective

- Obtain measurements of fission and non-fission foil reaction rates for the CFRMF and other ILRR test environments; use the results of these and other absolute measurement techniques to improve the accuracy of radiochemically determined reaction rates (and concurrent 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 $\pm 5\%$ to 10% (95% confidence level) for other fission and non-fission reactions.] Establish accepted sets of values of infinitely dilute 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

Documentation is provided on HEDL's radiochemical measurements for both fission and nonfission test foils as well as the run-to-run normalization foils irradiated in BIG 10 in FY 75. Additional measurements on the 0.05 mil ^{235}U foil irradiated in the NBS thermal column has resulted in a corrected ^{137}Cs result. Normalization foil data from the CFRMF irradiated 74-AI-III test indicates a power level change from the previous tests 74-5 through 74-7B. Necessary information on nuclear data, foil elemental and isotopic weights, used to correct the measured reaction rates is either documented in this report or has been documented in previous ILRR Progress Reports.

Accomplishments and Status

Data utilized in the calculation of absolute activities and errors for this report, such as counting system calibration, γ -ray absorption calculations, and nuclear parameters, etc., are documented in previous Progress Reports. Updated values of nuclear parameters given in the ANC chapter of this report were used where appropriate.

Results of HEDL's radiochemistry measurements for fission foils, nonfission foils and run-to-run normalization foils irradiated in FY-75 BIG-10, NBS Thermal neutron field and CFRMF tests follow.

1. BIG-10 Measured Reaction Rates

The results of HEDL's radiochemistry measurements for the Au, Al and In power normalization foils for irradiation tests 74-17 and 74-19B through 74-24 are reported in Table HEDL-14. With the exception of 74-19B, all comparative results fall within a maximum of $\leq 2\%$ thus adding to the confirmation of the reproducibility and stability of run-to-run reactor power operation.

Data from the HEDL 74-20 nonfission foil irradiation test are documented in Table HEDL-15. Analysis of ^{56}Mn could not be made due to prolonged decay prior to receipt of samples. Because of the extremely low activities observed in some of the samples, a 3" x 3" solid NaI(Tl) and a 4" x 5" NaI(Tl) well detector were utilized for some of the analysis.

Reaction rate data for the HEDL fission foil set, test 74-13 are reported in Table HEDL-16. Data from the ^{237}Np foil N7-10 are not reported as system counting difficulties have been noted during a portion of the decay counting. These problems have been resolved and necessary corrections are being made on the data.

Results from the Region III, Al foil package test 74-19A are documented in Table HEDL-17. Data from Al test 74-19B is not reported at this time as a review of the time history data and use in calculation of the reaction rates is being checked. Both tests 74-19A and 74-19B were in-reactor for multiple irradiations. Comparison of the 74-19A reaction rate data to HEDL's non-fission foil 74-20 and fission foil 74-13 reported data again indicate the apparent absence of gradient or perturbation effects between regions II and III.

TABLE HEDL-14
BIG 10 TEST NORMALIZATION FOIL
MEASURED REACTION RATES AND RATIOS

TEST	TEST REGION	LABORATORY TEST NO.	IRRADIATION COMPLETION DATE	REACTION	FOIL NO.	FOIL THICKNESS	FOIL (a) WT. (mg)	SS FACTOR	MEASURED REACTION RATE (b) (c) AND 10^{15}	% ERROR RELATIVE	EST. (d) ABSOLUTE	RUN TO RUN RATIOS (e)	SPECTRAL Au/In	INDEX Au/Al
E	II	HEDL 74-20	8-27-74	$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	21	0.5	28.736 (28.71)	0.9982	18.30	.25	1.7	0.9855	6.45	2082
	II	LASL 74-21	9-17-74	"	23	0.5	33.864 (33.84)	0.9979	18.34	.29	1.9	0.9876	6.46	2101
	II	ANC 74-22	8-13-74	"	20	0.5	30.870 (30.80)	0.9976	18.23	.26	1.7	0.9817	6.44	2083
	II	ANL 74-23	8-6-74	"	16	0.5	32.195 (32.16)	0.9980	18.39	.24	1.6	0.9903	6.48	2064
	II	MOL 74-24	10-9-74	"	25	0.5	23.884 (23.85)	0.9985	18.32	.25	1.7	0.9865	6.44	2084
	III	AI 74-19B	10-9-74	"	17	0.5	22.905 (22.88)	0.9985	18.17	.25	1.7	0.9785	6.29	2067
	C	II	MOL 74-17	11-19-74	"	27	36.131 (36.085)	0.9977	18.55	.23	1.6	0.9989		
	E	II	HEDL 74-20	8-27-74	$^{115}\text{In}(n,n')^{115\text{m}}\text{In}$	21	48.947 (48.92)	0.9975	2.836	.31	2.8	0.9803		
	II	LASL 74-21	9-17-74	"	23	2	49.555 (49.51)	0.9973	2.839	.36	3.0	0.9813		
	II	ANC 74-22	8-13-74	"	20	2	51.002 (50.93)	0.9969	2.829	.35	2.9	0.9779		
E	II	ANL 74-23	8-6-74	"	16	2	47.400 (47.38)	0.9977	2.837	.35	3.8	0.9806		
	II	MOL 74-24	10-9-74	"	25	2	49.081 (49.03)	0.9973	2.846	.36	3.1	0.9838		
	III	AI 74-19B	10-9-74	"	17	2	47.575 (50.49)	0.9971	2.891	.36	2.8	0.9931		
	C	II	MOL 74-17	11-19-74	"	27	47.54	0.9977	Sample delayed data invalid.					

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TABLE HEDL-14 (Con't)
BIG 10 TEST NORMALIZATION FOIL
MEASURED REACTION RATES AND RATIOS
(Continued)

TEST	TEST REGION	LABORATORY TEST NO.	IRRADIATION COMPLETION DATE	REACTION	FOIL NO.	FOIL THICKNESS	FOIL (a) WT. (mg)	SS FACTOR	MEASURED REACTION RATE (b) (c) AND 10^{15}	% ERROR RELATIVE	EST. (d) ABSOLUTE	RUN TO RUN RATIOS (e)	SPECTRAL Au/In	INDEX Au/Al
E	II	HEDL 74-20	8-27-74	$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	21	5	45.170	0.9991	0.00879	.29	1.8	0.9826		
	II	LASL 74-21	9-17-74	"	23	5	45.685	0.9991	0.00873	.34	1.9	0.9759		
	II	ANC 74-22	8-13-74	"	20	5	46.646	0.9991	0.00875	.31	1.8	0.9781		
	II	ANL 74-23	8-6-74	"	16	5	45.964	0.9991	0.00891	.37	2.0	0.9960		
	II	MOL 74-24	10-9-74	"	25	5	46.672	0.9991	0.00879	.28	1.7	0.9826		
	III	AI 74-19B	10-9-74	"	17	5	46.885	0.9991	0.00879	.29	1.9	0.9826		
C	II	MOL 74-17	11-19-74	"	27	5	45.700	0.9991	Sample delayed, data invalid.					

- a) Weights listed in brackets are those originally listed. All foils were weighed prior to counting and the observed weight was used in calculations.
- b) Corrections to saturated activity are based on assumed square wave functions from data reported by LASL.
- c) No corrections have been made for either foil neutron self shielding or flux gradient effects.
- d) Relative errors include both counting and partial $t_{1/2}$ errors while absolute errors include total $t_{1/2}$, counting, wt., γ attenuation summary and calibration errors.
- e) Run to run ratios are normalized back to Test A Region II results.

TABLE HEDL-15
BIG 10 TEST FOILS MEASURED REACTION RATES

TEST 74-20 REGION II NON-FISSION

REACTION	FOIL NO.	FOIL THICKNESS (mil)	FOIL WT. (mg)(a)	SS FACTOR	MEASURED REACTION RATE x 10 ¹⁵ (b)(c)	PERCENTAGE ABSOLUTE ERROR EST. 1σ(d)
⁵⁵ Mn(n,γ) ⁵⁶ Mn	1	0.5	13.445	-	Unable to measure	
²⁷ Al(n,α) ²⁴ Na	1	5	46.880 23.815	0.9994	0.00879	2.1
¹⁹⁷ Au(n,γ) ¹⁹⁸ Au	1	0.5	(23.80) 48.595	0.9982	18.34	1.6
¹¹⁵ In(n,n ¹) ^{115m} In	1	2	(48.58)	0.9975	2.809	2.3
⁶³ Cu(n,γ) ⁶⁴ Cu	1	0.5	15.122 14.303	-	1.871	3.1
⁵⁹ Co(n,γ) ⁶⁰ Co	1	0.5	(14.269) 14.578	0.9997	1.047	2.4
⁵⁹ Co(n,γ) ⁶⁰ Co	2	0.5	(14.547) 14.530	0.9997	1.053	2.1
⁵⁹ Co(n,γ) ⁶⁰ Co	3	0.5	(14.501) 294.305	0.9997	1.053	2.3
⁵⁸ Ni(n,p) ⁵⁸ Co	MM	10	(295.5) 785.318	0.9921	1.342	1.9
⁵⁸ Fe(n,γ) ⁵⁹ Fe	A	31	(785.3) 785.318	0.9842	0.340	3.8
⁵⁴ Fe(n,p) ⁵⁴ Mn	A	31	(785.3)	0.9801	0.975	2.5
²⁷ Al(n,α) ²⁴ Na	2	5	46.091 3.220	0.9994	0.00868	2.4
¹⁹⁷ Au(n,γ) ¹⁹⁸ Au	1	0.05	(3.198) 48.852	0.9998	19.02	1.3
¹¹⁵ In(n,n ¹) ^{115m} In	2	2	(48.84) 156.725	0.9975	2.804	2.5
⁴⁶ Ti(n,p) ⁴⁶ Sc	LK	10	(156.8) 156.725	0.9968	0.1440	3.1
⁴⁷ Ti(n,p) ⁴⁷ Sc	LK	10	(156.8) 156.725	0.9911	0.231	4.0
⁴⁸ Ti(n,p) ⁴⁸ Sc	LK	10	(156.8) 157.165	0.9966	0.00379	2.1
⁴⁶ Ti(n,p) ⁴⁶ Sc	LL	10	(157.2) 157.165	0.9968	0.1426	3.0
⁴⁷ Ti(n,p) ⁴⁷ Sc	LL	10	(157.2) 157.165	0.9913	0.237	4.1
⁴⁸ Ti(n,p) ⁴⁸ Sc	LL	10	(157.2)	0.9966	0.00392	2.0

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TABLE HEDL-15 (Con't)
BIG 10 TEST FOILS MEASURED REACTION RATES
(Continued)

REACTION	FOIL NO.	FOIL THICKNESS (mil)	FOIL WT. (mg)(a)(e)	SS FACTOR	MEASURED REACTION RATE $\times 10^{15}$ (b)(c)	PERCENTAGE ABSOLUTE ERROR EST. 1σ (d)
$^{45}\text{Sc}(n, \gamma)^{46}\text{Sc}$	1	5	47.715 (47.742)	0.9988	1.449	2.1
$^{235}\text{U}(n, f)^{140}\text{Ba}$	1	4.8	228.463 (246.782)	0.9933	6.793	1.8
$^{235}\text{U}(n, f)^{103}\text{Ru}$				0.9581	3.782	4.1
$^{235}\text{U}(n, f)^{95}\text{Zr}$				0.9862	7.281	2.5
$^{238}\text{U}(n, f)^{140}\text{Ba}$	3	3	310.953 (312.625)	0.9956	0.2559	2.3
$^{238}\text{U}(n, f)^{103}\text{Ru}$				0.9663	0.2738	4.7
$^{238}\text{U}(n, f)^{95}\text{Zr}$				0.9910	0.2312	2.8
$^{238}\text{U}(n, \gamma)^{239}\text{Np}$					12.13	2.8
$^{115}\text{In}(n, n')^{115\text{m}}\text{In}$	3	2	48.762 (48.77)	0.9975	2.834	2.4
$^{197}\text{Au}(n, \gamma)^{198}\text{Au}$	2	0.5	27.887 (27.88)	0.9979	18.23	1.8
$^{27}\text{Al}(n, \alpha)^{24}\text{Na}$	3	5	46.210 (46.41)	0.9991	0.00875	1.7

- a) Weights listed in brackets are those originally listed. All foils were weighed prior to counting and the observed weight was used in calculations.
- b) Corrections to saturated activity are based on assumed square wave functions from data reported by LASL.
- c) No corrections have been made for either foil neutron self shielding or flux gradient effects.
- d) The absolute errors estimate includes $t_{1/2}$, counting, wt., γ attenuation, summing and calibration errors.
- e) Fission foils - total metallic foil weight, the corrected isotopic (target) weight is listed above the total metallic foil weight (in brackets).

TABLE HEDL-16
BIG 10 TEST FOILS MEASURED REACTION RATES

TEST 74-13 REGION II FISSION FOIL

REACTION	FOIL NO.	FOIL THICKNESS (mil)	FOIL WT. (mg)(a)(e)	SS FACTOR	MEASURED REACTION RATE $\times 10^{15}$ (b)(c)	PERCENTAGE ABSOLUTE ERROR EST. 1σ (d)
$^{235}\text{U}(n,f)^{140}\text{Ba}$	6	5.1	281.911 (283.821)	0.9918	6.855	2.0
$^{235}\text{U}(n,f)^{103}\text{Ru}$				0.9743	3.814	3.9
$^{235}\text{U}(n,f)^{95}\text{Zr}$				0.9840	7.176	2.7
$^{238}\text{U}(n,f)^{140}\text{Ba}$	9	5.3	306.400 (308.047)	0.9913	0.2527	1.9
$^{238}\text{U}(n,f)^{103}\text{Ru}$				0.9726	0.2628	4.2
$^{238}\text{U}(n,f)^{95}\text{Zr}$				0.9830	0.2237	2.0
$^{239}\text{Pu}(n,f)^{140}\text{Ba}$	8	5.8	266.335 (273.543)	0.9919	7.134	1.9
$^{239}\text{Pu}(n,f)^{103}\text{Ru}$				0.9743	9.512	4.1
$^{239}\text{Pu}(n,f)^{95}\text{Zr}$				0.9841	6.414	2.2

- a) Weights listed in brackets are those originally listed. All foils were weighed prior to counting and the observed weight was used in calculations.
- b) Corrections to saturated activity are based on assumed square wave functions from data reported by LASL.
- c) No corrections have been made for either foil neutron self shielding or flux gradient effects.
- d) The absolute errors estimate include $t_{1/2}$, counting, wt., γ attenuation, summing and calibration errors.
- e) fission foils - the corrected isotopic (target) weight is listed above the total metallic foil weight (in brackets).

TABLE HEDL-17
BIG 10 TEST FOILS MEASURED REACTION RATES

REACTION	FOIL NO.	FOIL THICKNESS (mil)	FOIL WT. (mg)(a)(e)	SS FACTOR	MEASURED REACTION RATE $\times 10^{15}$	PERCENTAGE ABSOLUTE ERROR EST. 1σ
<u>TEST 74-19A REGION II</u>						
$^{59}\text{Co}(n, \gamma)^{60}\text{Co}$	1	.5	14.568 (14.530)	0.9991	1.073	1.8
$^{59}\text{Co}(n, \gamma)^{60}\text{Co}$	2	.5	14.618 (14.587)		1.049	1.7
$^{59}\text{Co}(n, \gamma)^{60}\text{Co}$	3	.5	14.415 (14.392)		1.047	1.7
$^{58}\text{Ni}(n, p)^{58}\text{Co}$	MS	10		0.9921	1.383	2.1
$^{58}\text{Fe}(n, \gamma)^{59}\text{Fe}$	N	31	126.757 (126.815)	0.9832	0.334	2.7
$^{58}\text{Fe}(n, \gamma)^{59}\text{Fe}$	P	31	127.615 (127.57)	0.9826	0.332	2.6
$^{54}\text{Fe}(n, p)^{54}\text{Mn}$	N		126.757	0.9800	0.943	2.0
$^{54}\text{Fe}(n, p)^{54}\text{Mn}$	P		127.615	0.9787	0.9453	1.9
$^{239}\text{Pu}(n, f)^{140}\text{Ba}$	2	5.7	264.194 (271.344)	0.9919	7.140	1.8
$^{239}\text{Pu}(n, f)^{103}\text{Ru}$				0.9743	9.388	2.6
$^{239}\text{Pu}(n, f)^{95}\text{Zr}$				0.9841	6.401	4.0
$^{235}\text{U}(n, f)^{140}\text{Ba}$	1	5.1	284.564 (286.492)	0.9917	6.791	1.9
$^{235}\text{U}(n, f)^{103}\text{Ru}$				0.9742	3.803	2.7
$^{235}\text{U}(n, f)^{95}\text{Zr}$				0.9839	7.199	3.9

- a) Weights listed in brackets are those originally listed. All foils were weighed prior to counting and the observed weight was used in calculations.
- b) Corrections to saturated activity are based on assumed square wave functions from data reported by LASL.
- c) No corrections have been made for either foil neutron self shielding or flux gradient effects.
- d) The absolute errors estimate include $t_{1/2}$, counting, wt., γ attenuation, summing and calibration errors.
- e) Fission foils - total metallic foil weight, the corrected isotopic (target) weight is listed above the total metallic foil weight (in brackets).

2. NBS Thermal Cavity ^{235}U Measured Reaction Rates

Continued decay counting of the HEDL ^{235}U #3 0.05 mil foil has resulted in a correction of $\sim 6\%$ on the ^{137}Cs data previously reported.* The measured reaction rate as determined from the three most recent counts is $2.662 \times 10^{-12} \pm 1.3\%$ (1σ) dps/nucleus at TOR. It was anticipated that subsequent counting would result in some modification of the reported value but a change of this degree was not expected. All data is again being reviewed.

3. CFRMF Measured Reaction Rates

The Au and In normalization foils for the CFRMF 74-AI-III test irradiated 6-25/26-75 have been analyzed. The reaction rates observed would indicate a power level reduction of slightly more than 3% from that indicated from the previous tests 74-5 through 74-7B. This is consistent with the information given by J. W. Rogers and I. E. Stephan** assuming the same power level instrument readings were used in all tests. The normalization foil results will be reported later with the foil package data.

Expected Accomplishments

Fission product data for the BIG-10 irradiated 74-13 ^{237}Np foil #10 and the AI 74-19 foil package will be reported, as well as reaction rate data on both the CFRMF irradiated 74-AI-III foil set and the $\Sigma\Sigma$ irradiated foil sets 74-Mol Fission -1 and MOL- $\Sigma\Sigma$ -2.

*LMFBR Reaction Rate and Dosimetry, 9th Progress Report, October, 1973 thru May, 1974, pg HEDL-38, HEDL-TME 74-45.

**LMFBR Reaction Rate and Dosimetry, 9th Progress Report, October, 1973 thru May, 1974, pg ANC-7, HEDL-TME 74-45.

JCGS

JOINT CENTER FOR GRADUATE STUDY

A. MASS AND CHARGE DEPENDENCE OF
SSTR ASYMPTOTIC SENSITIVITY

R. Gold

Objective

- To establish the dependence of the asymptotic sensitivity on charge (Z) and mass (A) so that absolute fission rates by the Solid State Track Recorder (SSTR) method can be extended to different actinide elements and isotopes of interest in the ILRR program.

Summary

The variation of the asymptotic sensitivity with charge (Z) and mass (A) of the fissile SSTR source has been estimated using Bohr's formulation of fission-fragment energy-loss in dense media. Numerical values of the asymptotic sensitivity have been generated for ^{232}Th , ^{233}U , ^{235}U , ^{238}U , and ^{239}Pu . These calculations reveal a monotonic increase with increasing A which is small but non-negligible relative to experimental error. This dependence must, therefore, be accounted for in SSTR applications of high accuracy involving asymptotically thick sources.

Accomplishments and Status

Basic investigations of the SSTR method of absolute fission rate measurements have demonstrated that two types of sources are particularly advantageous¹. If μ denotes the thickness of the SSTR source in g/cm^2 units, then these two advantageous conditions can be expressed as:

$$\mu \ll R_{\min} \quad (1a)$$

and

$$\mu \gg R_{\max} \quad , \quad (1b)$$

where $R_{\min}$ and $R_{\max}$ are the respective minimum and maximum range of fission fragments in the source. Sources which satisfy condition (1a) are said to lie in the region near the origin, where the SSTR efficiency is constant and the sensitivity is linear in μ . Sources which satisfy condition (1b) are said to be asymptotically thick. In this region, the efficiency varies as μ^{-1} and the sensitivity is constant. It has become customary to refer to this constant sensitivity as the asymptotic sensitivity and denote it by the symbol s_{∞} . It is important to recognize that s_{∞} is the maximum sensitivity available with the SSTR method.

Since it is very difficult to prepare acceptable fission sources in the intermediate region, i.e., in between the regions defined by conditions (1a,b), these circumstances have led to the fortuitous result that the SSTR method can be fully exploited in almost all applications with sources confined to these two disjoint regions. Acceptable fission sources are readily obtained in these two regions. Moreover, both the efficiency and sensitivity behave quite simply in these regions.

In applications where asymptotically thick sources are used for absolute fission rate measurements, it is necessary to determine the number of fissionable atoms at the surface that are observed with 100% efficiency. The effective mass per unit area from which all fission fragments are observed is s_{∞} , the asymptotic sensitivity. Extensive experimental measurements¹ have established that $s_{\infty} = 4341 \pm 61 \mu\text{g}/\text{cm}^2$ for ^{235}U in metallic uranium. Consequently, absolute SSTR measurements with asymptotically thick uranium sources will possess a current limiting accuracy of 1.4%.

Analytical Representation of s_{∞}

Under the assumptions of uniform source and no neutron flux depression, geometrical arguments can be used to show that

$$s_{\infty}(\rho) = \eta_{\infty} \langle R_{\rho} \rangle / 2 \quad , \quad (2)$$

where η_{∞} is the optical efficiency and $\langle R_{\rho} \rangle$ is average fission fragment range in the SSTR fissile source. The subscript $\rho = A/Z^{1/3}$ is a parameter indicative of the given SSTR fissile isotope. It must be stressed that the optical efficiency appropriate for Eq. (2) must be that ascribed to asymptotically thick sources. While the optical efficiency has been accurately determined for sources in the region near the origin,¹ viz., condition (1a), these results should not be arbitrarily applied to asymptotically thick sources. Indeed, the dependence of the optical efficiency upon the distribution of observed fission track areas has already been explored at length². These considerations would imply that the optical efficiency for asymptotically thick sources should be somewhat lower than that of thin sources in the region near the origin.

Support for this qualitative argument can be generated from Eq. (2). The optical efficiency of asymptotically thick sources, η_{∞} , can be roughly estimated from Eq. (2) by using the measured value of s_{∞} together with independently observed values of $\langle R \rangle$. For uranium, Niday³ has found $\langle R \rangle \cong 10 \text{ mg/cm}^2$. Consequently, Eq. (2) yields $\eta_{\infty} \cong 0.87$, which is indeed lower than the value $\eta = 0.95$ observed for sources in the region near the origin.

For ^{235}U , Eq. (2) can be written as

$$s_{\infty}(\rho_0) = \eta_{\infty} \langle R_0 \rangle / 2 \quad , \quad (3)$$

where ρ_0 and R_0 are specific values of ρ and R_ρ for ^{235}U , respectively. The ratio of Eqs. (2) and (3) then yields

$$s_\infty(\rho) = s_\infty(\rho_0) \cdot \left\{ \langle R_\rho \rangle / \langle R_0 \rangle \right\} \quad . \quad (4)$$

The dependence of s_∞ upon Z and A can then be estimated from Eq. (4) using the experimental value of $s_\infty(\rho_0)$ for ^{235}U , together with theoretical estimates for the ratio of the average fission fragment ranges.

The mean fission fragment range $\langle R_\rho \rangle$ can be expressed in the form

$$\langle R_\rho \rangle = \iiint R(m, z, v_i) P(m, z, v_i; \rho) dm dz dv_i \quad , \quad (5)$$

where $R(m, z, v_i)$ is the range of a fission fragment of mass m , charge z , and initial velocity v_i in a given SSTR fission source of charge Z and mass A . The distribution of fission fragments in mass m , charge z , and initial velocity v_i is given by $P(m, z, v_i; \rho)$ where the explicit dependence of this distribution upon SSTR fissile isotope is exhibited through the parameter ρ .^{*}

The range R_ρ can be expressed in quadrature form as

$$R_\rho = \int_0^\epsilon dE / (dE/d\mu) \quad , \quad (6)$$

where $(dE/d\mu)$ is the specific energy-loss of fission fragments per g/cm^2 of stopping medium and ϵ is the initial energy of the fission fragment. Since fission fragments are non-relativistic, the conventional Beth-Livingston-Bloch formula for specific energy-loss, which is based upon

* It will be assumed that the distribution $P(m, z, v_i; \rho) dm dz dv_i$ represents the probability that fission fragments possess a mass between m and $m + dm$, a charge between z and $z + dz$, and an initial velocity between v_i and $v_i + dv_i$.

the Born approximation, is not applicable. However, Bohr⁴ has formulated a description which is applicable for fission fragments in heavy media (i.e., large A) and the validity of this description has been confirmed by the catcher-foil work of Niday³. Using Bohr's description, Niday has found that the range of fission fragments can be expressed as

$$R(m, z, v_i) = \rho \cdot \frac{m(v_i - v_0)}{N_a C_1 \cdot f(z)} \quad , \quad (7)$$

where

$$f(z) = K + 4.7622K^{5/3} \quad . \quad (8)$$

Here C_1 is a suitable constant, N_a is Avogadro's number, v_0 is the velocity at which ionization becomes negligible and the function $f(z)$ depends on a parameter K , which is a measure of effective average charge of the fission fragment. Niday has found remarkably good agreement between Bohr's theory and experiment for the choice $K = z^{1/3}$.

It is instructive to partition the range in a form which separates the fission fragment variables (m, z, v_i) and the variables (Z, A) of the medium. Consequently, let

$$R(m, z, v_i) = \frac{\rho}{N_a \cdot C_1} \cdot r(m, z, v_i) \quad , \quad (9)$$

where

$$r(m, z, v_i) = \frac{m(v_i - v_0)}{f(z)} \quad , \quad (10)$$

and

$$f(z) = z^{1/3} + 4.7622z^{5/9} \quad (11)$$

Since $r(m, z, v_i)$ is independent of ρ , it will be called the reduced fission fragment range. In this notation, Eq. (5) becomes

$$\langle R_\rho \rangle = \frac{\rho}{N_a \cdot C_1} \langle r_\rho \rangle , \quad (12)$$

where

$$\langle r_\rho \rangle = \iiint \frac{m(v_i - v_0)}{f(z)} P(m, z, v_i; \rho) dm dz dv_i . \quad (13)$$

Consequently, Eq. (12) reveals that $\langle R_\rho \rangle$, hence $s_\infty(\rho)$, depends upon (Z, A) through the factors ρ and $\langle r_\rho \rangle$. The dependence of fragment range on the (Z, A) of the SSTR fissile (stopping) medium is linear in ρ for all fission fragments. In addition, a dependence on ρ arises through $\langle r_\rho \rangle$ as exhibited in Eq. (13) and this dependence can be attributed to the isotopic variation of the fission fragment distribution $P(m, z, v_i; \rho)$ for heavy fissile elements.

Introducing this notation in Eq. (4), one has

$$s_\infty(\rho) = s_\infty(\rho) \cdot \left(\frac{\rho}{\rho_0} \right) \cdot \left\{ \frac{\langle r_\rho \rangle}{\langle r_0 \rangle} \right\} , \quad (14)$$

which reveals that $s_\infty(\rho)$ depends upon the ratio of the average reduced ranges. To evaluate this ratio, it is convenient to introduce the quantities

$$\bar{r}_\rho = r_\rho(\bar{m}, \bar{z}, \bar{v}_i) , \quad (15a)$$

and

$$\bar{r}_0 = r_0(\bar{m}, \bar{z}, \bar{v}_i) , \quad (15b)$$

where $\bar{m}$, $\bar{z}$, and $\bar{v}_i$ are the most probable values of fission fragment mass, charge and initial velocity, respectively. Consequently, $\bar{r}_\rho$ and $\bar{r}_0$ are reduced ranges evaluated at the most probable values of the fission fragment variables m , z , and v_i . Since $\langle r_\rho \rangle$ is a slowly varying function, $\langle r_\rho \rangle$ and $\bar{r}_\rho$ will not differ significantly. Hence, one can write

$$\langle r_\rho \rangle - \bar{r}_\rho = \delta_\rho \quad , \quad (16a)$$

and

$$\langle r_0 \rangle - \bar{r}_0 = \delta_0 \quad , \quad (16b)$$

where δ_ρ and δ_0 are small. Using Eqs. (16a,b), the ratio of the average reduced ranges can be written as

$$\frac{\langle r_\rho \rangle}{\langle r_0 \rangle} = \frac{\bar{r}_\rho}{\bar{r}_0} \left[1 + \left(\frac{\delta_\rho}{\bar{r}_\rho} - \frac{\delta_0}{\bar{r}_0} \right) - \frac{\delta_0}{\bar{r}_0} \left(\frac{\delta_\rho}{\bar{r}_\rho} - \frac{\delta_0}{\bar{r}_0} \right) + \left(\frac{\delta_0}{\bar{r}_0} \right)^2 \left(\frac{\delta_\rho}{\bar{r}_\rho} - \frac{\delta_0}{\bar{r}_0} \right) - \dots \right] \quad (17)$$

Equation (17) reveals that the difference between $\langle r_\rho \rangle / \langle r_0 \rangle$ and $\bar{r}_\rho / \bar{r}_0$ is only of second order, since $\delta_\rho / \bar{r}_\rho$ and $\delta_0 / \bar{r}_0$ are of the same sign and same order of magnitude. The cancellation of first order terms in these ratios rests upon the systematics of asymmetric fission for the fissile isotopes and range of neutron energies of interest in the ILRR program. Using this approximation, Eq. (14) becomes

$$s_\infty(\rho) = s_\infty(\rho) \cdot \left(\frac{\rho}{\rho_0} \right) \cdot \left\{ \frac{\bar{r}_\rho}{\bar{r}_0} \right\} \quad . \quad (18)$$

Actually, it must be recognized that the asymmetric fission process is characterized by two distinct fission fragment distributions, rather than just one distribution; namely, a light mass distribution and a heavy mass distribution. Each of these distributions has, in turn, most probable values of the fission fragment variables m , z , and v_i . Consequently, one actually has

$$\frac{\bar{r}_\rho}{\bar{r}_0} = \frac{\bar{r}_{\rho,\ell} + \bar{r}_{\rho,h}}{\bar{r}_{0,\ell} + \bar{r}_{0,h}} \quad , \quad (19)$$

wherein the subscripts ℓ and h indicate that the arguments used for m , z and v_i are the most probable values of the light and heavy fission fragment mass distributions, respectively.

The relative magnitude of δ_ρ can be estimated from Niday's experimental data for uranium.³ These data show that $R(m,z,v_i)$ is very slowly varying in the region of the light mass peak for ^{235}U . In fact, numerical calculations of $\langle R_0 \rangle$ for the light fragment mass distribution give $\langle R_0 \rangle_\ell = 11.42 \text{ mg/cm}^2$. On the other hand, evaluating R_0 at the most probable mass for the light fragment distribution in ^{235}U , one finds $\bar{R}_{0,\ell} = 11.36 \text{ mg/cm}^2$ at $\bar{m} = 96$. Hence, $(\langle R_0 \rangle_\ell - \bar{R}_{0,\ell})/\bar{R}_{0,\ell} \cong \delta_{0,\ell}/\bar{r}_{0,\ell}$ is roughly $\frac{1}{2}\%$. In the region of the heavy fragment distribution, the variation of $R(m,z,v_i)$ is somewhat larger, so that one might anticipate a somewhat larger value of $\delta_{0,h}/\bar{r}_{0,h}$. In any event, these data show that a representative value of $\delta_\rho/\bar{r}_\rho$ can be no more than 1% or so. Consequently, the error entailed by use of the approximate ratio $\{\bar{r}_\rho/\bar{r}_0\}$ in Eq. (18) is considerably less than 1% and is, therefore, negligible in comparison with the 1.4% experimental error of $s_\infty(\rho_0)$.

Numerical Results

Equation (18) has been used to calculate $s_\infty(\rho)$ for ^{232}Th , ^{233}U , ^{235}U , ^{238}U , and ^{239}Pu . The most probable fission fragment variables used for these nuclides are listed in Table JCGS-I. Values of $\bar{m}$ and $\bar{v}_i$ were obtained from textbook tabulations.⁵ Where time-of-flight measurements have been performed, i.e., for ^{233}U , ^{235}U , and ^{239}Pu , $\bar{m}$ and $\bar{v}_i$ are known with considerably higher accuracy. The values of $\bar{z}$ in Table JCGS-I were obtained from the most recent compilation (ENDF/B-IV) of fission product yields.⁶ Here $\bar{z}$ corresponds to the most probable charge for a fission product chain of mass $\bar{m}$.

The value $s_{\infty}(\rho_0)$ used in Eq. (18) should correspond to the asymptotic sensitivity of ^{235}U . The experiments of Gold et al.,¹ provide s_{∞} for neutron-induced fission in ^{235}U , but within a ^{238}U stopping medium. To determine $s_{\infty}(\rho_0)$ from this experimental result, namely, $s_{\infty} = 4.341 \pm 0.061$ mg/cm², one need only multiply by $\rho_0/\rho = 235/238$, cf. Eq. (14). Consequently, one finds $s_{\infty}(\rho_0) = 4.2862 \pm 0.0600$ mg/cm² for ^{235}U .

Calculations based on these specific values are summarized in Table JCGS-II. In addition to $s_{\infty}(\rho)$, the factors ρ/ρ_0 and $\bar{r}_{\rho}/\bar{r}_0$, which enter into Eq. (18), are also listed. It can be seen that these two factors are all within a few percent of unity and consequently the error in $s_{\infty}(\rho)$ is essentially that of $s_{\infty}(\rho_0)$, namely 1.4%. While $s_{\infty}(\rho)$ increases monotonically with increasing A , the total variation is small and is, in fact, less than 6% over the range of fissile isotopes considered.

Since the asymptotic sensitivity in units of atoms/cm² is given by

$$a_{\infty}(\rho) = \frac{N_a}{A} s_{\infty}(\rho) \quad , \quad (19)$$

one finds even less variation in $a_{\infty}(\rho)$. The values of $a_{\infty}(\rho)$ in Table JCGS-II exhibit a total variation of less than 3% and are all within the experimental uncertainty of the ^{235}U measurement, $a_{\infty}(\rho_0) = (1.0985 \pm 0.0154) \cdot 10^{19}$ atoms/cm². Clearly, the value of $a_{\infty}(\rho_0)$ can be used for any of these fissile isotopes in experimental work not requiring the limiting accuracy of the SSTR method. However, a systematic variation of $a_{\infty}(\rho)$ is evident and is of the same order as experimental error. Hence, the values tabulated in Table JCGS-II are recommended for SSTR work of the highest possible accuracy with asymptotically thick sources.

Other fissile isotopes of interest could not be included in these calculations due to lack of detailed experimental data on the light and

heavy fission fragment distributions. In particular, from the viewpoint of the ILRR program, it would have been highly desirable to include $^{237}\text{Np}_{93}$. Unfortunately, appropriate values of $\bar{m}$, $\bar{z}$, and $\bar{v}_i$ are not currently available for this nuclide.^{5,6} In lieu of direct calculation, it is recommended that the value $a_{\infty} = (1.1056 \pm 0.0155) \cdot 10^{19}$ atoms/cm² be used for $^{237}\text{Np}_{93}$. This value has been obtained by linear interpolation of the a_{∞} values of ^{235}U and ^{239}Pu cited in Table JCGS-II.

TABLE JCGS-1
MOST PROBABLE FISSION FRAGMENT VARIABLES

<u>Isotope</u>	<u>Light-Mass Distribution^a</u>			<u>Heavy-Mass Distribution^a</u>		
	$\bar{m}$	$\bar{z}$	$\bar{v}_i$	$\bar{m}$	$\bar{z}$	$\bar{v}_i$
$^{232}\text{Th}_{90}$	92	36.59	1.424	139	53.92	0.942
$^{233}\text{U}_{92}$	94.38	38.19	1.423	139.62	54.92	0.967
$^{235}\text{U}_{92}$	95.93	38.50	1.415	140.07	54.67	0.970
$^{238}\text{U}_{92}$	98	38.75	1.383	139	53.44	0.975
$^{239}\text{Pu}_{94}$	100.23	40.51	1.398	139.77	54.72	1.005

^a Mass is given in a.m.u. units and initial velocity in cm/nanosecond.

TABLE JCGS-2
ASYMPTOTIC SENSITIVITY OF SELECTED ACTINIDES

Isotope	ρ	ρ/ρ_0	$\left(\frac{\bar{r}_{\rho,\ell} + \bar{r}_{\rho,h}}{\bar{r}_{0,\ell} + \bar{r}_{0,h}} \right)$	$\begin{matrix} a \\ s_{\infty} \end{matrix}$	$\begin{matrix} b \\ a_{\infty} \end{matrix}$
$^{232}\text{Th}_{90}$	51.7695	0.9945	0.9797	4.1761	1.0341
$^{233}\text{U}_{92}$	51.6131	0.9915	0.9952	4.2294	1.0932
$^{235}\text{U}_{92}$	52.0561	1.0000	1.0000	4.2862	1.0985
$^{238}\text{U}_{92}$	52.7207	1.0128	0.9997	4.3397	1.0981
$^{239}\text{Pu}_{94}$	52.5640	1.0098	1.0202	4.4155	1.1127

^aUnits of mg/cm².

^bUnits of 10¹⁹ atoms/cm².

References

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LASL

LOS ALAMOS SCIENTIFIC LABORATORY

A. LASL-ILRR IRRADIATIONS DESCRIPTION

G. E. Hansen

Objective

- Provide descriptions of irradiation conditions for the Big-Ten ILRR program tests.

Summary

The Big-Ten ILRR program tests A through E are completed and irradiation records sent to the participating laboratories.

Accomplishments and Status

The relative fluences for all Big-Ten ILRR irradiations are documented in the NBS section of this report. The start and stop times for these irradiations are given below.

Test A	05:11:06 to 08:51:06	AM	MDT	May 29, 1974
Test C-LASL	08:01:40 to 11:41:40	AM	MDT	July 9, 1974
Test C-ANL	06:37:08 to 10:17:08	AM	MDT	July 16, 1974
Test C-ANC	07:54:59 to 11:34:59	AM	MDT	July 23, 1974
Test C-HEDL	06:26:00 to 10:06:00	AM	MDT	July 31, 1974
Test E-ANL	06:38:40 to 10:18:40	AM	MDT	Aug. 6, 1974
Test E-ANC	06:28:00 to 10:08:00	AM	MDT	Aug. 13, 1974
Test E-HEDL	06:27:58 to 10:07:58	AM	MDT	Aug. 27, 1974
Test E-LASL	06:35:51 to 10:15:51	AM	MDT	Sept. 17, 1974
Test E-MOL	02:32:28 to 06:12:28	AM	MDT	Oct. 9, 1974
Test C-MOL	02:34:35 to 06:14:35	AM	MDT	Nov. 19, 1974
Test B-ANL	Not applicable (solid state track recorder SSTR-4 was irradiated on Nov. 12, 1974 while SSTR-5, 6, 7, and 8 were irradiated on Dec. 10, 1974)			

Expected Achievements

The relative fluence numbers now available will permit detailed inter-comparisons of the absolute reaction rate data obtained by the various laboratories from the various Big-Ten irradiations.

B. REACTION RATE MEASUREMENTS IN BIG-10

LASL RADIOCHEMISTRY GROUP (D. W. BARR)

Objective

- To provide LASL Radiochemical support of the ILRR program as requested by DRRD through DMA.

Summary

LASL Radiochemical results for the BIG-10 irradiations (tests C and E) conducted in July - September 1974 as part of the ILRR program are reported here.

Accomplishments and Status

Irradiation: Test C-LASL

total exposure: 220 minutes in BIG-10 @ ~1.3 kw

time out (End of Bombardment): 11:41:40 MDT on 7/9/74

materials: The LASL-designated fission foil set (LASL 74-14) detailed in the 5/30/74 memo "Documentation for BIG-10 Fission and Non-fission Foil Set Irradiations."

Results: LASL results obtained for the fission foil set are summarized in Table 1.

TABLE 1
LASL RESULTS FOR FISSION FOILS - TEST C

	^{235}U	^{238}U	^{237}Np	^{239}Pu
$Q^{\dagger}$ -factors used in fission rate determinations:				
^{95}Zr	0.969	0.773	1.07	0.699
^{99}Mo	1.00	1.03	1.10	0.966
^{137}Cs	0.993	0.931	1.02	0.986
^{140}Ba	0.942	0.931	0.933	0.803
^{147}Nd	0.967	1.19	1.05	0.874
fission rate ($\times 10^{15}$)	112 (1.6%)	4.21 (3.8%)	35.3 (5%)	140 (2%)
capture rate ($\times 10^{15}$)	---	11.9 (2%)	---	---
n,2n rate ($\times 10^{15}$)	---	0.207 (3%)	---	---
$Q^{\dagger}$ -factors determined in BIG-10 irradiation:				
^{89}Sr	.954	---	---	0.338
^{111}Ag	1.44	4.12	4.70	16.4
^{115}Cd	1.46	3.07	2.54	3.97
^{136}Cs	1.52	---	10.7	18.1
^{156}Eu	1.21	---	---	9.33

$^{\dagger}Q (= Y_i/Y_i^*)$ denotes a fission product (i) yield ratio for the generalized fission situation (Y_i) to its yield for ^{235}U thermal fission (Y_i^*), our reference fission type.

Comments:

- (1) The energy-insensitive fission products ^{95}Zr , ^{99}Mo , ^{137}Cs , ^{140}Ba , ^{147}Nd were used to determine the quoted fission rates and associated precision estimates. All analyses were accomplished with radio-chemical separation procedures. The LASL-determined Q-factors for the above fission products appropriate to these fissioning species at the neutron energies involved are listed in Table 1.
- (2) In addition, Q-values for the energy-sensitive fission products ^{89}Sr , ^{111}Ag , ^{115}Cd , ^{136}Cs , ^{156}Eu were determined by radiochemical separation procedures. Those Q-value results are also listed in Table 1 and are characteristic of a significantly degraded fission spectrum similar to the CFRMF results obtained previously (3/14/74 Memo - Barr to ILRR).
- (3) For ^{238}U , the capture rate $^{238}\text{U}(n,\gamma)^{239}\text{Np}$ and $n,2n$ reaction rate $^{238}\text{U}(n,2n)^{237}\text{U}$ were determined with radiochemical separation procedures and the results are given in Table 1.
- (4) The accuracy of the fission rate determinations is difficult to estimate but is thought to be ~4% for ^{235}U ; ~5% for ^{238}U and ^{239}Pu ; unknown for ^{237}Np since that nuclide is not one for which we have historical K-factor data - 10% is perhaps reasonable.

Irradiation: Test E - LASL

total exposure: 220 minutes in BIG-10 @ ~1.3 kw

time out (End of Bombardment): 10:15:51 MDT on 9/17/74

materials: The LASL-designated non-fission foil set (LASL 74-21) detailed in the 5/30/74 memo "Documentation for BIG-10 Fission and Non-fission Foil Set Irradiations."

Results: LASL results obtained for the non-fission foil set are summarized in Table 2. They are listed in the order of disassembly of the HEDL-packaged foil set (Fig. 1 in the above documentation).

TABLE 2
LASL RESULTS FOR NON-FISSION FOILS - TEST E

	Reaction Rate ($\times 10^{15}$)	Reaction Rate Ratio [to $^{235}\text{U}(n,f)$]	Reaction Rate Ratio [to $^{58}\text{Ni}(n,p)$]
$^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$	0.595	0.00531	0.465
$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	0.00761	0.0000679	0.00595
$^{27}\text{Al}(n,p)^{27}\text{Mg}$	0.0430	0.0000384	0.0336
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	18.8	0.168	14.7
$^{115}\text{In}(n,n')^{115\text{m}}\text{In}$	2.71	0.0242	2.12
$^{115}\text{In}(n,\gamma)^{116\text{m}}\text{In}$	16.2	0.145	12.7
$^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$	1.92	0.0171	1.50
$^{59}\text{Co}(n,\gamma)^{60}\text{Co}$	1.03	0.00920	0.805
$^{58}\text{Ni}(n,p)^{58}\text{Co}$	1.28	0.0114	1.00
$^{54}\text{Fe}(n,p)^{54}\text{Mn}$	0.935	0.00835	0.730
$^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$	0.322	0.00288	0.252
$^{56}\text{Fe}(n,p)^{56}\text{Mn}$	0.0101	0.0000902	0.00789
$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	0.00746	0.0000666	0.00583
$^{27}\text{Al}(n,p)^{27}\text{Mg}$	0.0429	0.000383	0.0335
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	19.0	0.170	14.8
$^{115}\text{In}(n,n')^{115\text{m}}\text{In}$	2.73	0.0244	2.13
$^{115}\text{In}(n,\gamma)^{116\text{m}}\text{In}$	16.2	0.145	12.7
$^{46}\text{Ti}(n,p)^{46}\text{Sc}$	0.139	0.00124	0.109
$^{47}\text{Ti}(n,p)^{47}\text{Sc}$	0.224	0.00200	0.175
$^{48}\text{Ti}(n,p)^{48}\text{Sc}$	0.00374	0.0000334	0.00292
$^{45}\text{Sc}(n,\gamma)^{46}\text{Sc}$	1.41	0.0126	1.10
$^{235}\text{U}(n,f)\text{F.P.}$	112 (1.1%)	1.00	87.5
$^{238}\text{U}(n,f)\text{F.P.}$	4.19 (2.6%)	0.0374	3.27
$^{238}\text{U}(n,\gamma)^{239}\text{Np}$	11.8 (2%)	0.105	9.22
$^{238}\text{U}(n,2n)^{237}\text{U}$	0.193 (3.5%)	0.00172	0.151
$^{115}\text{In}(n,n')^{115\text{m}}\text{In}$	2.73	0.0244	2.13
$^{115}\text{In}(n,\gamma)^{116\text{m}}\text{In}$	15.9	0.142	12.4
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	18.8	0.168	14.7
$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	0.00730	0.0000652	0.00570
$^{27}\text{Al}(n,p)^{27}\text{Mg}$	0.0422	0.000377	0.0330

Comments:

- (1) The measurements common to the LASL Tests C and E [$^{235}\text{U}(\text{n},\text{f})$, $^{238}\text{U}(\text{n},\text{f})$, n,γ , $\text{n},2\text{n}$] indicate the same fluence levels for these irradiations to within 1%. Confirmation by independent monitors is reported by Hansen (P-5-456, 9/23/74).
- (2) All reaction rate measurements were accomplished on NaI and Ge(Li) systems with radiochemical separations in some instances and direct foil counting where applicable. The measurements are believed to be 3-5% (1σ) accurate. This subjective conclusion is based on long-term experience with our counting systems and the decay schemes involved.
- (3) There is no indication in our raw data of flux gradient, self-shielding, or perturbation effects consistent with the estimates of Hansen (P-5-456). The flux-depression factors given by Hansen (P-5-456) for the Test E non-fissile foil pack have not been applied to the data of Table 2 nor have any corrections for secondary fissions in the fissile foil pack (P-5-214).
- (4) Table 2 lists the "standard" ILRR reaction rate products. Other reaction products were observed in the course of making these measurements. Data are available for the following reaction rates:
 $^{59}\text{Co}(\text{n},2\text{n})^{58}\text{Co}$
 $^{59}\text{Co}(\text{n},\text{p})^{59}\text{Fe}$
 $^{113}\text{In}(\text{n},\gamma)^{114\text{m}}\text{In}$
 $^{197}\text{Au}(\text{n},2\text{n})^{196}\text{Au}$

Also worthy of note is that contaminants were observed in the irradiated Fe foil.

(5) Reaction rate measurement ratios relative to $^{235}\text{U}(\text{n},\text{f})$ F. P. and to $^{58}\text{Ni}(\text{n},\text{p})^{58}\text{Co}$ are listed in Table 2 to facilitate intercomparison with published ILRR results for other Benchmark neutron fields (CFRMF, $\Sigma\Sigma$, Godiva). Such comparisons illustrate the degraded fission spectrum expected in BIG-10 without the very soft neutron component seen in CFRMF and $\Sigma\Sigma$. Quantitative unfolding of the reaction rate data by codes like SAND-II and SPECTRA will provide a check of the measured BIG-10 neutron spectrum.

Expected Achievements

Completion of LASL measurements in the BIG-10-ILRR program tests marks the end of LASL Radiochemistry participation in ILRR on a non-funded basis.

NBS

NATIONAL BUREAU OF STANDARDS

A. SPECTRAL INDICES FOR BIG-10: SPECTRUM-AVERAGED FISSION
CROSS SECTION RATIOS FOR ^{235}U , ^{238}U , ^{239}Pu , ^{237}Np

D. M. Gilliam and J. A. Grundl, NBS

G. E. Hansen and H. H. Helmik, LASL

Objective

- To measure the spectrum-averaged fission cross section ratios for ^{235}U , ^{238}U , ^{239}Pu , and ^{237}Np in the central region of the BIG-10 facility and to provide corrections for perturbations introduced by the measuring instrument.

Summary

The NBS double fission chamber was placed in a close-fitting cavity (Test Region I) at the center of the BIG-10 facility, and fission ratios were observed for back-to-back deposits of the isotopes of interest (^{235}U , ^{238}U , ^{239}Pu , and ^{237}Np). The fission counts were recorded by means of a dual triple-scaler counting system as described in Ref. 1. The triple-scaler counting system was compared with a PDP-8 computer-based analyzer system using LASL-built live-timing circuitry. In nine comparisons, an rms discrepancy of 0.2% was obtained between the two independent systems. In addition, a check of the NBS dead-time corrections was made by the variance-to-mean method using LASL multiscaling hardware. The NBS dead-time corrections were found to agree with the variance-to-mean results within $\pm 0.2\%$ over the count rate range used in the present measurements. Fourteen deposits from the NBS set of reference and working fissionable deposits were employed in the measurements. NBS personnel spent two and one-half man-weeks at the LASL Pajarito site, making sixteen spectral index runs and some subsidiary deposit mass assay comparisons in the BIG-10 system. The fission cross section ratios measured in situ (at the deposit positions within the fission chamber) were as follows: 1.000: $0.0366 \pm 1.8\%$: $1.198 \pm 1.5\%$: $0.314 \pm 2.1\%$ for ^{235}U , ^{238}U , ^{239}Pu , ^{237}Np , respectively. By application of calculated perturbation corrections, the following fission cross section ratios were obtained for the unperturbed BIG-10: 1.000: $0.0372 \pm 1.9\%$: $1.198 \pm 1.5\%$: $0.317 \pm 2.2\%$

for ^{235}U , ^{238}U , ^{239}Pu , ^{237}Np , respectively. For comparison, these cross section ratios were calculated from ENDF/B-IV cross section data using an 87 group computed spectrum for BIG-10. The ratios of the observed spectral indices to the calculated spectral indices were 1.106, 1.021, and 0.985 for the cross section ratios of $^{238}\text{U}/^{235}\text{U}$, $^{239}\text{Pu}/^{235}\text{U}$, and $^{237}\text{Np}/^{235}\text{U}$, respectively.

Accomplishments and Status

The ratios of spectrum-averaged fission cross sections of the isotopes ^{235}U , ^{238}U , ^{239}Pu , and ^{237}Np were measured in the central region of the BIG-10 Critical Assembly by comparison of fission rates obtained with rigorously assayed deposits of these isotopes. These deposits (on thin platinum disks) were mounted back-to-back inside of an NBS double fission chamber located in a close-fitting cavity in the central region of BIG-10. A detailed description of fission chamber construction and operation, and a discussion of the mass assay of the reference deposits can be found in Ref. 1.

Three identical double fission chambers (designated PP 1, PP 2, and PP 5) were used in the measurements, and fourteen deposits from the NBS set of reference and working fissionable deposits were employed. Table NBS-1 lists these deposits and the mass assay data compiled for each of them. Four different batches of material are represented in the ^{238}U deposits, including a natural uranium sample and depleted uranium samples with ^{235}U content ranging from 0.06% down to 2 ppm. A complete listing of the isotopic composition of the four ^{238}U batches and the various batches of all the other deposits is given in Table NBS-2, along with the correction factors appropriate to the BIG-10 spectrum for fissions in isotopes other than the principal isotope of each batch.

The loading of the fission chambers and operation of the counting system were carried out by NBS personnel at the LASL Pajarito site. A complete triple-scaler counting system (as described in Ref. 1) was employed to monitor the fission rate from each side of the double fission chamber during the major series of experimental runs (5/7-10/74); a few additional runs were made later (6/19-20/74) using only a PDP-8 computer-based pulse height analysis system for the data collection. Table NBS-3 lists the basic fission rate data and some of the indicators of data quality which are computed by the data processing program FCDATA.

A brief summary of the triple-scaler nomenclature is given here; a more detailed description may be found in Ref. 1, Section III. The quantities S_U and S_L are the count rates above discriminator levels V_U and $V_L (=V_U/2)$ in the valley of the p.h.d. (pulse height distribution); this "valley" is a low, flat region in the p.h.d. between the single, broad peak on the one side and the noise-alpha tail near zero on the other side. The count rate S_U is regarded as the primary fission rate datum while the difference $(S_L - S_U)$ is used in extrapolating the S_U count rate to zero pulse height. Under normal operation, the quantity $(S_L/S_U - 1)$ is a very regular and reproducible function of the deposit mass thickness (see Fig. 4, Ref. 1), so that departures from the usual value can indicate even very small defects in the chamber or counting system performance. In particular, electronic noise, marring or covering of the fissionable deposit, and the presence of fissionable contamination can give rise to anomalous $(S_L/S_U - 1)$ values. The third scaler in the triple scaler system records the count rate S_{GC} which serves as a check on gain variations. The S_{GC} count rate is taken from a single channel analyzer window beginning at V_U and ending somewhat above, but near the peak of the p.h.d. so that S_{GC} changes markedly with gain shifts. A 1% gain change would cause a 2% change in S_{GC} (while S_L and S_U would change by less than 0.02% for deposits up to $200 \mu\text{g}/\text{cm}^2$).

In Table NBS-3 the sixteen runs are listed in chronological order. The highest S_U count rate used was 4054 s^{-1} , with many runs having count rates in the range 2000 s^{-1} to 3000 s^{-1} . However, at least one run with count rates less than 1600 s^{-1} was made for each fission cross section ratio reported. For even the highest count rates, the dead-time correction never exceeded 0.9%, and the estimated uncertainty in the dead-time corrected count rates never exceeded 0.2%. The statistical uncertainty listed for the S_U ratio was always taken as the larger of the observed standard deviation (s) and the calculated standard deviation (σ). The quantity s/σ is listed for each S_U ratio as an indicator of proper statistical dispersion. As a footnote to Table NBS-3, a list of the normal s/σ ranges is given. No significant departure from proper statistical variation is observed in any of the runs. Next, the experimental $(S_L/S_U - 1)$ data are compared with the nominal values. (The quantity $(S_L/S_U - 1)$ is called the ETZ for Extrapolation to Zero.) Again the greater of s and σ

is listed as the statistical error, and the s/σ comparison is given. No significant statistical departures occurred, but the ETZ's for the T chamber of PP 5 consistently ran higher than the nominal value. The average discrepancy between nominal and observed ETZ's for the six loadings of PP 5 was 0.12%. This discrepancy is insufficient to warrant much discussion. Suffice it to say that for these runs an average of the nominal and observed ETZ values was employed in making the extrapolation to zero correction, and the estimated error was increased to cover the spread of nominal and observed values (see Table NBS-4). The S_{GC} count rate along with s and σ values was also computed for each run (not shown in Table NBS-3). No gain change in excess of 1% was indicated for any of the thirteen triple-scaler runs by the observed standard deviations of the S_{GC} data.

In addition to the routine data quality checks described above, some experimental checks of the triple-scaler system results were made by comparisons with other well developed counting techniques in use at LASL. A PDP-8 pulse height analysis system with LASL-built live-timing electronics was compared with the triple-scaler system, and it was found that these two pulse counting systems were consistent within $\pm 0.2\%$ rms discrepancy (0.37% max. discrepancy) in nine comparisons with count rates as high as 6,700 cps. Dead-time losses for the SCA's in the triple-scaler system were checked by two methods: (i.) comparison of count rate ratios for light and heavy deposits at different power levels (see Table IV of Ref. 1) and (ii.) the variance-to-mean method (Ref. 2) using LASL multi-scaling hardware. The dead-time corrections obtained from the separate methods differed by no more than 0.2% for the highest count rates employed in the fission cross section ratio measurements.

Table NBS-4 lists the observed fission cross section ratio from each run and the estimated uncertainty exclusive of errors in the mass assay of the deposits. All corrections to the fission rates for both the top and bottom chambers are given along with the corrections to the S_U ratio. With each correction an estimated uncertainty is listed. The correlations in systematic errors in the corrections for the two separate chambers were taken into account in deriving the estimated errors for the cross section ratio. All the corrections except the correction for fragment

absorption have already been discussed. The correction for fragment absorption is approximately 0.69% per 100 $\mu\text{g}/\text{cm}^2$ of deposit thickness with an uncertainty of 0.35% or 25% of the correction, whichever is larger. (Further details are given in Ref. 1.) The non-zero floor (0.35%) on this error estimate is necessary because no correction is made for the effect of surface roughness on fragment absorption. The runs for measuring a particular cross section ratio such as $\sigma_f(^{238}\text{U})/\sigma_f(^{235}\text{U})$ have been grouped together in Table NBS-4 so that the spread of the results for that group is easier to see. For the $^{238}\text{U}/^{235}\text{U}$ ratio measurements, the absolute sum of corrections to the ratio ranges from 5% to 24%. The net correction to the ratio is less than 2%, except in one run with a very thick deposit and the run with the 28N deposit, for which a correction of $(20.0 \pm 0.3)\%$ is made for fission in the natural ^{235}U content.

For the $^{238}\text{U}/^{235}\text{U}$ comparison the maximum discrepancy between any two runs is 2.2%; however, the data are not scattered randomly between these limits. The $^{238}\text{U}/^{235}\text{U}$ data are instead clustered around two discrepant values 0.0363 and 0.0369, 1.7% apart. All of the runs using 28S deposits are in the cluster about the value 0.0363, while the remaining runs are clustered at the other value. The masses for the 28HD, 28G, and 28N deposits are all tied to the reference deposit 28N-2-2, while the assay of the 28S deposits is based on the reference deposit 28S-2-2. A direct comparison of these two reference deposits in BIG-10 showed the same 1.7% discrepancy with respect to the mass values reported in Table II, Ref. 1. In order to resolve this discrepancy, further alpha counting comparisons are currently in progress at NBS, and a comparison of the two reference deposits in a $\text{Cf}(\chi)$ field is planned. The uncertainties quoted in Ref. 1. for these deposit masses were 2.0% for 28S-2-2 and 1.5% for 28N-2-2. For the present, equal weight will be assigned to the two clusters so that an average value of $0.0366 \pm 1.8\%$ is obtained. The 1.8% error estimate comes from a 1.7% allowance for the mass ratio uncertainties and 0.6% error from all other sources.

The spread of the $^{239}\text{Pu}/^{235}\text{U}$ results (three runs) is only 0.25%. An average $^{239}\text{Pu}/^{235}\text{U}$ cross section ratio of $1.198 \pm 1.5\%$ is obtained, allowing $\pm 1.4\%$ for mass ratio uncertainty and $\pm 0.5\%$ for other errors.

The two runs measuring the $^{237}\text{Np}/^{235}\text{U}$ cross section ratio agreed within 0.64%; the average of the two gives $0.314 \pm 2.1\%$ for the cross section ratio with the mass ratio error completely dominating the error estimate. The two runs measuring the $^{239}\text{Pu}/^{237}\text{Np}$ ratio agreed to three significant digits. Again deposit mass ratio uncertainty of $\pm 2.1\%$ dominates the overall error. The discrepancy of the single cross section ratio measurement for $^{239}\text{Pu}/^{235}\text{U}$ with the double ratio product $(^{239}\text{Pu}/^{237}\text{Np}) \times (^{237}\text{Np}/^{235}\text{U})$ is 0.67%, well within the quoted errors.

Table NBS-5 presents the calculated fission rate depression factors due to cavity and fission chamber perturbations in the BIG-10 spectrum during the fission rate measurements. The perturbation calculations accounting for the effects of the cavity have been discussed previously in Ref. 3. The correction factor for the platinum backings was derived from the multiple layer self-shielding formula described in Ref. 4. For BIG-10 1.3 ± 0.6 b was selected as the removal cross section appropriate for ^{238}U deposits and 0.9 ± 0.6 b as the removal cross section appropriate for ^{237}Np deposits. Because the deposits are thin relative to mean free path of fast neutrons, the formula for the platinum backing correction reduces to

$$\Phi/\Phi_0 = \frac{1}{2} [1 + E_2(\Sigma t)],$$

where Σ is the macroscopic removal cross section, t is the total platinum thickness, and $E_2(u)$ the exponential integral.

Table NBS-6 summarizes the cross section ratio results, giving both the cross section ratios observed in situ and the corrected results corresponding to an unperturbed BIG-10. The corrected results are compared to calculated spectral indices based on ENDF/B-IV cross sections and the 87 group central flux spectrum from Ref. 5. The calculation of the BIG-10 spectrum tabulated in Ref. 5 was based on ENDF/B-III cross sections. The large discrepancy of nearly 11% for the spectral index $\bar{\sigma}_f(^{238}\text{U})/\bar{\sigma}_f(^{235}\text{U})$ is comparable to the discrepancy seen in the measurements at CFRMF (Ref. 1., Table XII). It has been a persistent feature in the comparisons of ENDF/B data and integral data tests in fission driven fast neutron spectra.

Expected Achievements

The results given here are regarded as final except for the possibility of readjustments associated with refinement of the mass assay of

the NBS reference deposits. When the present upgrading of the reference deposit mass assay is completed, any necessary adjustments in the spectral index results for BIG-10 and CFRMF will be reported.

References

1. J. A. Grundl, D. M. Gilliam, N. D. Dudey, and R. V. Popek, "Measurement of Absolute Fission Rates," Nuclear Technology, 25 (1975), 237.
2. LMFBR Reaction Rate and Dosimetry 9th Progress Report, p. NBS-18.
3. LASL Office Memorandum 9/23/74, To: The ILRR participants in BIG-10 irradiations, From: Gordon Hansen, P-5, LASL, Subject: Record of Tests A, C, and E to September 23, 1974, Symbol: P-5-456
4. LMFBR Reaction Rate and Dosimetry 9th Progress Report, p. NBS-24
5. LASL Office Memorandum 1/23/74, To: ILRR Participants, From: G. E. Hansen, LASL, Subject: BIG-10, Symbol: P-5-214.

TABLE NBS-1
FISSIONABLE DEPOSITS - MASS ASSAY DATA

Deposit Label	Principal Isotope and Atom Percent	Principal Isotope Mass- μ g	Mass Assay Method [†]
25S-2-1	^{235}U 99.748 $\pm$ 0.003	234.9 $\pm$ 1.3%	TFC(25S-2-2)
25S-2-3	^{235}U 99.748 $\pm$ 0.003	222.0 $\pm$ 1.3% 222.8 223.1 $\pm$ 1.3% 222.5 $\pm$ 1.3%	TFC(25S-2-2) AAC, QD AAC, HL WA
25S-01-1	^{235}U 99.748 $\pm$ 0.003	5.13 $\pm$ 1.3%	TFC(25S-2-2)
25S-01-2	^{235}U 99.748 $\pm$ 0.003	4.95 $\pm$ 1.3% 4.96 $\pm$ 1.3% 4.95 $\pm$ 1.3%	TFC(25S-01-1, 25S-2-2) FFC(25S-01-1, 25S-2-2), BIG-10 WA
25K-1-2	^{235}U 99.89 $\pm$ 0.01	132.7 $\pm$ 1.3% 133.2 $\pm$ 1.4% 132.2 $\pm$ 1.5% 133.0 $\pm$ 1.3%	TFC(25K-1-1, 25S-2-2) TFC(25S-2-2) AAC, HL WA
28S-2-2	^{238}U 99.94 $\pm$ 0.01	252.0 $\pm$ 2.0%	Reference Deposit
28S-2-1	^{238}U 99.94 $\pm$ 0.01	255.1 $\pm$ 1.1% 250.5 $\pm$ 2.0% 250.5 $\pm$ 2.0%	AAC, QD FFC(28S-2-2), BIG-10 WA
28N-2-2	^{238}U 99.275	230.0 $\pm$ 1.5%	Reference Deposit
28G-2-3.4	^{238}U 99.98	228.9 230.5 228.9 $\pm$ 1.7% 228.9 $\pm$ 1.7%	AAC, HL Gravimetric FFC(28S-2-1,2;28N-2-2), BIG-10 WA
28HD-8-1	^{238}U 99.999	864 $\pm$ 2% 868 864 $\pm$ 2%	FFC(28S-2-2, 28N-2-2), BIG-10 AAC, HL WA

TABLE NBS-1 (Con't)

Deposit Label	Principal Isotope and Atom Percent	Principal Isotope Mass- μg	Mass Assay Method [†]
49I-1-2	^{239}Pu 99.10 $\pm$ 0.01	104.8 $\pm$ 1.2%	Reference Deposit
49I-1-3	^{239}Pu 99.10 $\pm$ 0.01	104.0 $\pm$ 1.3%	RAC(49I-1-2)
37S-1-1	^{237}Np 99.98 $\pm$ 0.01	117.9 $\pm$ 1.7%	Reference Deposit
37S-2-1	^{237}Np 99.98 $\pm$ 0.01	254.0 $\pm$ 1.7%	FFC(37S-1-1), BIG-10

[†]Mass Assay Notes

TFC(Reference Deposit)	Thermal neutron fission counting comparison to reference deposit listed. If indirect, the intermediate deposit(s) is (are) also listed.
FFC(Reference Deposit)	Fast neutron fission counting comparison. If indirect, the intermediate deposit(s) is (are) also listed.
RAC(Reference Deposit)	Relative alpha counting comparison to reference deposit listed.
AAC	Absolute alpha counting
QD	Alpha emission rate per gram from quantitative deposition.
HL	Alpha emission rate per gram from mass spectrographic isotopic analysis and published half-lives.
WA	Weighted average

TABLE NBS-2

CORRECTION FOR FISSIONS IN MINOR ISOTOPIC CONSTITUENTS

Batch Label and Isotopic Assay (atom percent)		Total Correction and Relative Fission Rate Contributions in the BIG-10 Spectrum
25S		0.99972 (15) [†]
²³⁵ U	99.748 (3) [†]	1.0
²³⁸ U	0.1261(7)	0.0000465(8)
²³⁴ U	0.0608(12)	0.00019 (9)
²³⁶ U	0.0652(12)	0.000046(23)
25K		0.99985 (7)
²³⁵ U	99.89 (1)	1.0
²³⁸ U	0.053(2)	0.0000195(5)
²³⁴ U	0.035(1)	0.00011 (6)
²³⁶ U	0.025(1)	0.000018 (9)
28S		0.9837(7)
²³⁸ U	99.94 (1)	1.0
²³⁵ U	0.061(2)	0.0166(5)
²³⁴ U	<0.001	<0.0001
²³⁶ U	<0.005	<0.0001
28N		0.834 (3)
²³⁸ U	99.275	1.0
²³⁵ U	0.720(1)	0.199 (4)
²³⁴ U	0.005	0.0004
28G		0.9952(1)
²³⁸ U	99.9823	1.0
²³⁵ U	0.01755	0.00481(10)
²³⁴ U	0.00016	0.00001
²³⁶ U	<0.00001	
28HD		0.99995(5)
²³⁸ U	99.999	1.0
²³⁵ U	0.0002(2)	0.00005(5)
49I		0.9977(10)
²³⁹ Pu	99.10 (1)	1.0
²⁴⁰ Pu	0.883(5)	0.0022(10)
²⁴¹ Pu	0.010(1)	0.00010(5)
²⁴² Pu	0.006(1)	0.00001(1)
²³⁸ Pu	<0.005	---
37S		0.9993 (3)
²³⁷ Np	99.98	1.0
²³⁹ Pu	0.02	0.00075(30)

[†]The number in parentheses is the error in the least significant digit.

TABLE NBS-3

SPECTRAL INDEX DATA AND DATA QUALITY CHECKS

Run Date	Labels for Deposits & Chamber	S _U -T Chamber (cps) S _U Ratio S _U -B Chamber (cps)	s/σ [†] for the S _U Ratio	ETZ (S _L /S _U -1) Nominal (%)	ETZ (S _L /S _U -1) Observed (%)	s/σ for the ETZ	Dead Time Corrections to S _U Ratio (%)	Number of Counting Intervals
5-7-74	28S-2-1 PP5 25S-01-2	467.2 1.789 ± 0.27% ^{††} 261.1	0.14	0.85 0.13	0.92 ± 0.02 ^{††} 0.14 ± 0.01	1. 1.	0.04 ± 0.02	4
5-7-74	25S-2-3 PP2 49I-1-3	995. 1.782 ± 0.15% 558.5	0.53	0.81 0.23	0.81 ± 0.03 0.26 ± 0.02	1. 0.5	0.08 ± 0.04	6
5-7-74	37S-1-1 PP5 25S-2-1	167.5 0.1592 ± 0.17% 1052.	0.93	0.38 0.90	0.45 ± 0.01 0.90 ± 0.02	0.7 1.	-0.20 ± 0.05	6
5-7-74	28S-2-2 PP1 25S-2-1	118.6 0.03903 ± 0.24% 3038.	0.77	0.85 0.90	1.01 ± 0.04 0.86 ± 0.05	1.6 1.	-0.59 ± 0.09	5
5-7-74	28S-2-1 PP5 25S-01-1	1128. 1.724 ± 0.25% 654.	0.22	0.85 0.13	1.05 ± 0.03 0.13 ± 0.01	1. 1.	0.09 ± 0.05	4
5-8-74	28N-2-2 PP1 25S-2-1	173.3 0.0427 ± 0.47% 4054.	1.71	0.86 0.90	0.87 ± 0.03 0.91 ± 0.06	0.6 1.	-0.84 ± 0.20	4
5-9-74	25S-2-1 PP1 28G-2-3.4	3054. 27.95 ± 0.21% 109.3	0.83	0.90 0.81	0.87 ± 0.05 0.84 ± 0.02	1. 1.	0.60 ± 0.09	11
5-9-74	25K-1-2 PP5 49I-1-2	2936. 1.073 ± 0.09% 2737.	1.07	0.41 0.23	0.52 ± 0.04 0.27 ± 0.02	1. 1.	0.00 ± 0.16	5
5-9-74	28HD-8-1 PP1 25S-2-1	385.1 0.1262 ± 0.16% 3052.	0.52	(2.77) ^{†††} 0.90	2.43 ± 0.03 0.88 ± 0.04	1. 2.	-0.58 ± 0.15	6
5-10-74	49I-1-3 PP1 25S-2-1	781. 0.5305 ± 0.22% 1472.	1.21	0.23 0.90	0.29 ± 0.01 0.89 ± 0.03	0.5 1.	-0.16 ± 0.08	6
5-10-74	37S-2-1 PP2 49I-1-2	994. 0.6328 ± 0.23% 1571.	1.49	0.89 0.23	0.89 ± 0.03 0.25 ± 0.01	1. 1.	-0.10 ± 0.07	7
5-10-74	28S-2-1 PP5 25S-01-2	3460. 1.784 ± 0.14% 1940.	1.22	0.85 0.13	1.03 ± 0.05 0.14 ± 0.03	1. 1.	0.28 ± 0.14	6
5-10-74	28S-2-1 PP5 25S-01-2	1747. 1.790 ± 0.13% 976.	0.67	0.85 0.13	1.03 ± 0.03 0.12 ± 0.01	1. 2.	0.14 ± 0.07	10
6-19-74	25S-01-1 PP2 37S-1-1	345. 0.1416 ± 0.14% 2437.	---	0.13 0.38	0.17 ± 0.01 0.40 ± 0.01	---	Live Timer	1
6-20-74	49I-1-2 PP2 37S-1-1	2443. 3.350 ± 0.13% 729.	---	0.23 0.38	0.23 ± 0.01 0.39 ± 0.01	---	Live Timer	1
6-20-74	28S-2-1 PP5 25S-01-2	2506. 1.793 ± 0.14% 1398.	---	0.85 0.13	0.94 ± 0.01 0.16 ± 0.01	---	Live Timer	1

Notes

[†]The s/σ limits which should include 90% of normal runs are listed here for comparison. These limits are derived from the assumption that (N-1) s²/σ² follows a χ² distribution.

N (Counting Intervals)

s/σ Limits

2

[0.063, 1.95]

5

[0.42, 1.54]

10

[0.61, 1.37]

20

[0.73, 1.26]

^{††}The error listed is the larger of the observed standard deviation (s) and the calculated standard deviation (σ).

^{†††}From linear extrapolation of data for lighter deposits.

TABLE NBS-4

OBSERVED FISSION CROSS SECTION RATIOS, CORRECTIONS, AND UNCERTAINTIES

Run date	Chamber and deposit labels T/B	Dead-time correction T/Ratio/B	S_U ratio (dead-time corrected) + statistical and dead-time errors	ETZ correction to S_U counts T/Ratio/B	Fragment absorption correction T/Ratio/B	Fission in other isotopes T/Ratio/B	Net correction T/Ratio/B	Absolute sum of corrections T/Ratio/B	Cross section ratio (in-situ) + combined error (exclusive of deposit mass errors)
5-7-74	28S-2-1	1.0010(2)*	1.789±0.27%	1.0177±0.0043	1.0136±0.0035	0.9837±0.0007	1.0157	4.86%	($^{238}\text{U}/^{235}\text{U}$)
	PP 5	1.0004(2)		1.0149±0.0040	1.0133±0.0033	0.9840±0.0007	1.0123	5.26%	0.0362±0.59%
	25S-01-2	1.0006(1)		1.0028±0.0007	1.0003±0.0035	0.9997±0.0002	1.0034	0.40%	
5-7-74	28S-2-2	1.0003(1)	0.03903±0.26%	1.0202±0.0050	1.0137±0.0035	0.9837±0.0007	1.0176	5.05%	($^{238}\text{U}/^{235}\text{U}$)
	PP 1	0.9941(9)		1.0029±0.0020	1.0009±0.0022	0.9840±0.0007	0.9819	8.70%	0.0364±0.40%
	25S-2-1	1.0062(9)		1.0172±0.0043	1.0128±0.0035	0.9997±0.0002	1.0363	3.65%	
5-7-74	28S-2-1	1.0023(4)	1.724±0.25%	1.0190±0.0051	1.0136±0.0035	0.9837±0.0007	1.0184	5.24%	($^{238}\text{U}/^{235}\text{U}$)
	PP 5	1.0009(5)		1.0164±0.0046	1.0133±0.0033	0.9840±0.0007	1.0144	5.70%	0.0362±0.62%
	25S-01-1	1.0014(3)		1.0026±0.0007	1.0003±0.0035	0.9997±0.0002	1.0040	0.46%	
5-8-74	28N-2-2	1.0004(1)	0.0427±0.51%	1.0174±0.0044	1.0125±0.0035	0.834±0.0030	0.8595	19.63%	($^{238}\text{U}/^{235}\text{U}$)
	PP 1	0.9916(20)		0.9992±0.0010	0.9997±0.0020	0.8343±0.0030	0.8264	23.64%	0.0368±0.63%
	25S-2-1	1.0088(20)		1.0182±0.0046	1.0128±0.0035	0.9997±0.0002	1.0400	4.01%	
5-9-74	25S-2-1	1.0062(9)	27.95±0.23%	1.0174±0.0044	1.0128±0.0035	0.9997±0.0002	1.0365	3.67%	($^{238}\text{U}/^{235}\text{U}$)
	PP 1	1.0060(9)		1.0006±0.0010	1.0003±0.0020	1.0045±0.0002	1.0114	7.10%	0.0370±0.32%
	28G-2-3.4	1.0002(0)		1.0168±0.0042	1.0125±0.0035	0.9952±0.0001	1.0248	3.43%	
5-9-74	28HD-8-1	1.0008(2)	0.1262±0.22%	1.0486±0.0122	1.0471±0.0118	1.0000±0.0001	1.0989	9.65%	($^{238}\text{U}/^{235}\text{U}$)
	PP 1	0.9942(15)		1.0305±0.0076	1.0339±0.0085	1.0003±0.0002	1.0596	13.39%	0.0370±1.16%
	25S-2-1	1.0067(15)		1.0176±0.0044	1.0128±0.0035	0.9997±0.0002	1.0372	3.74%	
5-10-74	28S-2-1	1.0070(11)	1.784±0.20%	1.0188±0.0051	1.0136±0.0035	0.9837±0.0007	1.0229	5.57%	($^{238}\text{U}/^{235}\text{U}$)
	PP 5	1.0028(14)		1.0160±0.0044	1.0133±0.0033	0.9840±0.0007	1.0159	6.33%	0.0362±0.59%
	25S-01-2	1.0042(8)		1.0028±0.0007	1.0003±0.0035	0.9997±0.0002	1.0070	0.76%	
5-10-74	28S-2-1	1.0036(6)	1.790±0.15%	1.0188±0.0051	1.0136±0.0035	0.9837±0.0007	1.0195	5.23%	($^{238}\text{U}/^{235}\text{U}$)
	PP 5	1.0014(7)		1.0164±0.0044	1.0133±0.0033	0.9840±0.0007	1.0149	5.74%	0.0363±0.57%
	25S-01-2	1.0021(4)		1.0024±0.0006	1.0003±0.0035	0.9997±0.0002	1.0045	0.51%	
6-20-74	28S-2-1	+0.2% [†]	1.793±0.24%	1.0179±0.0046	1.0136±0.0035	0.9837±0.0007	1.0149	4.78%	($^{238}\text{U}/^{235}\text{U}$)
	PP 5	+0.2%		1.0147±0.0038	1.0133±0.0033	0.9840±0.0007	1.0117	5.16%	0.0363±0.56%
	25S-01-2	+0.2%		1.0032±0.0008	1.0003±0.0035	0.9997±0.0002	1.0032	0.38%	

*The number in parentheses is the uncertainty in the least significant digit.

[†]LASL-built live timer used in computer based pulse height analyzer.

TABLE NBS-4. (Con't)

Run date	Chamber and deposit labels T/B	Dead-time correction T/Ratio/B	S_U ratio (dead-time corrected) $\pm$ statistical and dead-time errors	ETZ correction to S_U counts T/Ratio/B	Fragment absorption correction T/Ratio/B	Fission in other isotopes T/Ratio/B	Net correction T/Ratio/B	Absolute sum of corrections T/Ratio/B	Cross section ratio (in-situ) $\pm$ combined error (exclusive of deposit mass errors)
5-7-74	25S-2-3	1.0020(4)	1.782 $\pm$ 0.16%	1.0162 $\pm$ 0.0041	1.0121 $\pm$ 0.0035	0.9997 $\pm$ 0.0002	1.0302	3.06%	($^{239}\text{Pu}/^{235}\text{U}$)
	PP 2	1.0008(4)		1.0083 $\pm$ 0.0026	1.0067 $\pm$ 0.0026	1.0020 $\pm$ 0.0010	1.0179	4.73%	1.200 $\pm$ 0.41%
	49I-1-3	1.0012(2)		1.0078 $\pm$ 0.0030	1.0054 $\pm$ 0.0035	0.9977 $\pm$ 0.0010	1.0121	1.67%	
5-9-74	25K-1-2	1.0060(10)	1.073 $\pm$ 0.18%	1.0093 $\pm$ 0.0026	1.0072 $\pm$ 0.0035	0.9999 $\pm$ 0.0001	1.0226	2.26%	($^{239}\text{Pu}/^{235}\text{U}$)
	PP 5	1.0000(16)		1.0012 $\pm$ 0.0015	1.0017 $\pm$ 0.0020	1.0022 $\pm$ 0.0010	1.0051	4.45%	1.197 $\pm$ 0.27%
	49I-1-2	1.0060(13)		1.0081 $\pm$ 0.0030	1.0055 $\pm$ 0.0035	0.9977 $\pm$ 0.0010	1.0174	2.19%	
5-10-74	49I-1-3	1.0016(3)	0.5305 $\pm$ 0.23%	1.0087 $\pm$ 0.0033	1.0054 $\pm$ 0.0035	0.9977 $\pm$ 0.0010	1.0134	1.80%	($^{239}\text{Pu}/^{235}\text{U}$)
	PP 1	0.9984(8)		0.9911 $\pm$ 0.0028	0.9927 $\pm$ 0.0027	0.9980 $\pm$ 0.0010	0.9803	5.21%	1.197 $\pm$ 0.46%
	25S-2-1	1.0032(7)		1.0178 $\pm$ 0.0046	1.0128 $\pm$ 0.0035	0.9997 $\pm$ 0.0002	1.0338	3.41%	
5-7-74	37S-1-1	1.0003(1)	0.1592 $\pm$ 0.18%	1.0083 $\pm$ 0.0025	1.0064 $\pm$ 0.0035	0.9993 $\pm$ 0.0003	1.0143	1.57%	($^{237}\text{Np}/^{235}\text{U}$)
	PP 5	0.9980(5)		0.9905 $\pm$ 0.0028	0.9937 $\pm$ 0.0026	0.9996 $\pm$ 0.0004	0.9819	4.91%	0.315 $\pm$ 0.42%
	25S-2-1	1.0023(5)		1.0180 $\pm$ 0.0045	1.0128 $\pm$ 0.0035	0.9997 $\pm$ 0.0002	1.0331	3.34%	
6-19-74	25S-01-1	$\pm 0.2\%$ [†]	0.1416 $\pm$ 0.24%	1.0034 $\pm$ 0.0009	1.0003 $\pm$ 0.0035	0.9997 $\pm$ 0.0002	1.0034	0.40%	($^{237}\text{Np}/^{235}\text{U}$)
	PP?	$\pm 0.2\%$		0.9954 $\pm$ 0.0012	0.9939 $\pm$ 0.0025	1.0004 $\pm$ 0.0004	0.9897	1.91%	0.313 $\pm$ 0.37%
	37S-1-1	$\pm 0.2\%$		1.0080 $\pm$ 0.0020	1.0064 $\pm$ 0.0035	0.9993 $\pm$ 0.0003	1.0137	1.51%	
5-10-74	37S-2-1	1.0022(4)	0.6328 $\pm$ 0.24%	1.0178 $\pm$ 0.0046	1.0138 $\pm$ 0.0035	0.9993 $\pm$ 0.0003	1.0334	3.45%	($^{239}\text{Pu}/^{237}\text{Np}$)
	PP 2	0.9990(7)		1.0102 $\pm$ 0.0032	1.0083 $\pm$ 0.0029	1.0016 $\pm$ 0.0010	1.0192	5.30%	3.79 $\pm$ 0.50%
	49I-1-2	1.0032(6)		1.0075 $\pm$ 0.0028	1.0055 $\pm$ 0.0035	0.9977 $\pm$ 0.0010	1.0139	1.85%	
6-20-74	49I-1-2	$\pm 0.2\%$ [†]	3.350 $\pm$ 0.24%	1.0069 $\pm$ 0.0026	1.0055 $\pm$ 0.0035	0.9977 $\pm$ 0.0010	1.0101	1.47%	($^{239}\text{Pu}/^{237}\text{Np}$)
	PP 2	$\pm 0.2\%$		0.9991 $\pm$ 0.0010	0.9991 $\pm$ 0.0020	0.9984 $\pm$ 0.0010	0.9966	2.96%	3.79 $\pm$ 0.34%
	37S-1-1	$\pm 0.2\%$		1.0078 $\pm$ 0.0020	1.0064 $\pm$ 0.0035	0.9993 $\pm$ 0.0003	1.0135	1.49%	

* The number in parentheses is the uncertainty in the least significant digit.

[†] LASL-built live timer used in computer based pulse height analyzer.

TABLE NBS-5
CALCULATED SPECTRAL CORRECTIONS

Deposit nuclide	Fission rate depression due to cavity and chamber structure	Fission rate depression due to platinum backings	Total fission rate depression factor	Fission rate depression relative to that for ^{235}U
^{235}U	0.991(2)*	1.000(1)	0.991(2)	1.
^{238}U	0.983(4)	0.993(4)	0.976(6)	0.985(6)
^{239}Pu	0.991(2)	1.000(1)	0.991(2)	1.000(3)
^{237}Np	0.987(3)	0.995(3)	0.982(4)	0.991(5)

*The number in parantheses is the error in the least significant digit.

TABLE NBS-6
BIG-10 SPECTRAL INDICES

Nuclides Compared	Fission Cross Section Ratio-- <u>in situ</u>	Fission Cross Section Ratio--in Unperturbed BIG-10	Observed/ENDF/B-IV for Unperturbed BIG-10
$^{238}\text{U}/^{235}\text{U}$	$0.0366 \pm 1.8\%$	$0.0372 \pm 1.9\%$	1.106
$^{239}\text{Pu}/^{235}\text{U}$	$1.198 \pm 1.5\%$	$1.198 \pm 1.5\%$	1.021
$^{237}\text{Np}/^{235}\text{U}$	$0.314 \pm 2.1\%$	$0.317 \pm 2.2\%$	0.985
$^{239}\text{Pu}/^{237}\text{Np}$	$3.79 \pm 2.1\%$	$3.76 \pm 2.2\%$	1.031

B. FISSION/MILLIGRAM OF ^{235}U IN BIG-10 TESTS A, C, E, AND B

D. M. Gilliam and J. A. Grundl, NBS

G. E. Hansen, LASL

Objective

- To monitor each run of the BIG-10 dosimetry foil test groups A, C, E, and B by continuous operation of the NBS double fission chamber in the irradiation cavity. For each run, to determine the integrated number of fissions per milligram of ^{235}U , to check the spectral index $^{238}\sigma_f/^{235}\sigma_f$, and to observe the power profile.

Summary

The entire series of dosimetry foil tests at BIG-10 (including the preliminary Test A, five fission foil set irradiations - Tests C, five non-fission foil set irradiations - Tests E, and five track-etch detector irradiations - Tests B) were monitored continuously by the NBS double fission chamber PP5 in the central test cavity. The deposits 25S-01-2 and 28S-2-1, which had been used in four of the spectral index runs discussed in Section A, were employed in all the irradiations. The chamber remained closed throughout the irradiations (except for the runs listed in Table NBS-4 on 6-19 and 6-20). The pulse counting system for the bottom chamber with the ^{235}U deposit consisted of a full triple-scaler system plus a PDP-8 pulse height analyzer in parallel, while the fission counting for the ^{238}U deposit was done by the PDP-8 alone. All aspects of the irradiations (including the operation of the NBS fission chamber) were performed by LASL personnel.

Each of the C and E series runs and Test A were monitored by two sets of Au, In, and Al activation foils in Test Region II and by external fission chambers in addition to the NBS double fission chamber. One set of these foils was counted

at LASL and the other at HEDL following each run. The run-to-run normalizations as determined by these several independent systems turned out to be more than satisfactorily consistent. The experimental standard deviation of the least-consistent monitor from the weighted mean was 0.88%, and the consistency of the internal and external fission chambers was better than 0.20% (standard deviation). The uncertainty in the weighted average for the run-to-run normalization is judged to be $\pm 0.3\%$ for Tests A, C, and E. For Tests B, only the NBS fission chamber could be used as a monitor. Furthermore, inconsistencies in the PDP-8 and triple-scaler data for the Test B runs as large as 0.5% occurred in two cases. Because of the lack of redundancy in run-to-run monitoring and the occasional inconsistency between the scalers and PDP-8, a larger error, $\pm 0.6\%$, is estimated as the uncertainty of the run-to-run normalization of the Test B runs. The redundant monitoring in the A, C, and E series proved to be especially valuable in the BIG-10 tests because of a contamination problem with the NBS double fission chamber. The final contamination state was determined by thermal neutron beam tests at NBS subsequent to the BIG-10 irradiations. The run-to-run development of the contamination was traceable through pulse height distribution changes evidenced in both the PDP-8 data and the S_L/S_U counting ratios from the scalers. The maximum contamination corrections to the fission count rate (at the last run) were 0.38% for 28S-2-1 and 0.69% for 25S-01-2. Although these corrections are not very large and are expected to give results accurate to $\pm 0.3\%$, one could not have high confidence in the assumptions made in the contamination analysis if the results had not been confirmed by redundant monitors.

The power profile on the C and E runs was very close to a perfect "square" in every case. The allowable reactor start-up period was sufficiently short that the correction for the start-up ramp from the half-power point was only 14 seconds,

and the scram was effectively instantaneous. There was a small gradual power level increase of 0.9% to 2.0% during each of the C and E series runs, but no other departure of the power profiles from the ideal "square" shape was observed. The measured power increase from start-up to scram is tabulated for each run. During the Test A there was an inadvertant power surge of brief duration for which saturation factor corrections are given. The Test B runs were too short for power profiles to be observed. Both the PDP-8 and scaler counting systems monitored the Test B runs in single counting intervals.

The accuracy of the absolute fission counting data (fissions/milligram of ^{235}U) is estimated to be 1.4% for Tests A, C, and E and 1.5% for Test B. Deposit mass assay uncertainties remain the dominant error.

Accomplishments and Status

1. Chamber Performance

General Remarks. In most respects the performance of the double fission chamber and associated electronics was excellent throughout the series of irradiations in spite of superficial wear and tear. The ^{235}U fission rate was monitored redundantly by a triple-scaler counting system and a PDP-8 pulse height analyzer, and the count rates and integrated counts derived from the two separate systems usually agreed within about 0.2%. The pulse height distributions from both chambers showed little change from run to run except for small increases in the count in the valley of the pulse height distribution (p.h.d.) due to the contamination problem discussed below. No significant electronic noise pick-up was encountered except during the shutdown procedure following one of the Test B runs.

It was noted that during the Tests A, C, and E the peak of the p.h.d. for both sides of the double fission chamber would shift about 10% in the negative

direction on the pulse height scale. It was expected that this effect was due to the temperature rise during the irradiations. Subsequently, temperature effects on the p.h.d. were studied in thermal neutron runs at NBS. It was found that the valley region of the p.h.d. was insensitive to the temperature, but that the peak position varied approximately in inverse proportion to the absolute temperature ($^{\circ}\text{K}$). The temperature effect is believed to be due to the change in the average number of ion pairs created per fission fragment due to density changes in the flowing chamber gas (which remains at constant pressure). The peak region is strongly affected because pulses in the peak region are produced by truncated fragment paths along which less ionization is produced at reduced density; the valley region is not strongly affected because this portion of the p.h.d. arises largely from fragments which expend most of their range within the fission deposit itself and expend all their remaining energy in the gas regardless of the density. The p.h.d. shifts seen in the thermal neutron beam runs at NBS were consistent in every way with the spectral shifts in the BIG-10 runs. The peak shifts seen in the BIG-10 runs correspond to a temperature rise of about 30°C .

Contamination Problem. During the BIG-10 irradiations, small increases in the ETZ (see p. NBS-3 for nomenclature) were observed in both sides of the double fission chamber. Figure NBS-1 is a plot of the ETZ histories. By the end of the BIG-10 tests, the above-normal count between V_L and V_U had grown to 0.19% on the ^{235}U side and 0.54% on the ^{238}U side. It was recognized that these increases could be due to chamber contamination, deposit deterioration, or deterioration of pulse formation. Chamber performance deterioration due to radiation damage to insulators or due to radiolytic damage to electrical contacts was thought to be the most likely cause of the high ETZ's before the chamber was returned to NBS for inspection. The post-irradiation inspection of the chamber at NBS indicated that

FIGURE NBS-1. ETZ Histories in the BIG-10 Irradiations

The separate segments within a given test are taken from separate PDP-8 counts for that test. The error bars indicate the run-to-run uncertainty in the ETZ due to uncertainty in the amplifier gain setting. The value of $(S_L/S_U - 1)$ for 28S-2-1 on Test A was taken from the June 19-20 "reference" runs, because an improper adjustment of the gain for that chamber in Test A made the PDP-8 data unusable.

the problem was instead due to contamination by minute amounts of 10% enriched uranium dust. Fortunately, the final contamination state could be determined and the run-to-run development of the contamination could be deduced from the ETZ histories. Also fortunately, the contamination-corrected run-to-run fission rate data from the NBS chamber turn out to be in excellent agreement with the data from other independent monitors (see Section B.2) so that a high level of confidence in the accuracy of the run-to-run normalization is possible in spite of the contamination problem.

Table NBS-7 shows the data from tests made in a thermal neutron beam at NBS before and after the BIG-10 irradiations. The data as shown have been corrected only for relative absorption and scattering effects at the position of the two deposits, so that the tabulated count rates correspond to equal thermal neutron fluxes at each deposit. The reactor power level was held constant with $\pm 1\%$ during these runs. The bottom line of the table gives nominal count rates for a perfectly contamination-free chamber. These nominal count rates are derived from runs prior to the BIG-10 irradiations. The top line in the table shows the count for the contaminated chamber as received following the BIG-10 irradiations, before any internal inspection of the chamber. After this count was done, the chamber was opened and found to be visibly contaminated with black dust. There was no indication of radiation damage to the teflon insulators. The contamination was disturbed as little as possible while platinum blanks were inserted in place of the fissionable deposits used at BIG-10. The second line of Table NBS-7 gives the results for this configuration. The increase in $^{28}\text{S}_\text{L}$ - $^{28}\text{S}_\text{U}$ (1.13 cps to 1.36 cps) in the top chamber indicates that some inadvertant increase in the contamination of the top chamber occurred during the foil exchange. In the bottom chamber the decrease in $^{25}\text{S}_\text{L}$ - $^{25}\text{S}_\text{U}$ was almost exactly the expected amount based on the nominal

count rates from the bottom line of the table ($0.465 \text{ cps} - 0.27 \text{ cps} \approx 0.22 \text{ cps}$), so that no change in the contamination of the bottom chamber appears to have occurred during the exchange of deposits. Next, the chamber was cleaned as well as possible without disassembly. Moderate pressure and use of a solvent were required to remove the black dust from the gold-plated electrode surfaces and teflon insulators. The run with platinum blanks was repeated; the resulting count rates are given on the third line of the table. The cleaning markedly reduced the counts in valley of the p.h.d., confirming the supposition that the black dust had been the cause of the spurious counts. The fissionable deposits were cleaned superficially by blowing off loose dust with a jet of dry air and replaced in the chamber for another run in the thermal neutron beam (fourth line on the table). Finally, the deposits were compared with reference deposits maintained at NBS. The 25S-01-2 deposit mass showed no change ($\pm 15 \text{ ng}$), while the 28S-2-1 deposit was found to have retained $8 \pm 3 \text{ ng}$ of ^{235}U contamination. The more precise determination of contamination of the 28S deposit was possible because of its low ^{235}U content (See Table NBS-2 in Section A).

One interpretation of these contamination measurements and ETZ histories will be presented and shown to be consistent with all the data. This "progressive contamination" interpretation is as follows: During the handling of the chamber between runs, loose dust from the central region of BIG-10 (10% enriched) somehow got into the top chamber. The gas flow from the upper to lower chamber carried some of this dust into the bottom chamber. The migration of this material was short-lived; the dust rather quickly attached itself to some internal surface or blew out of the active region of the chamber. Thus the changes in the ETZ's due to the contamination tend to occur mostly as monotonic steps between runs rather than as continuous or up-and-down variations during the runs. In Figure NBS-2

there is hardly ever a decrease in the ETZ with time (the high value of the ETZ for Test B-ANL-4 is due to electronic noise during the shutdown). The larger steps in the contamination generally occurred at the same times for both top and bottom chambers, whereas a continuous migration of material might show the upper chamber contamination decreasing while the bottom chamber contamination increases or a time lag in contamination of the bottom chamber.

A quantitative interpretation of the contamination data requires introduction of some new nomenclature: Let

$$S_U = S_{U0} (1 + X + Y)$$

$$= S_{U0} + (\Delta S_U)_X + (\Delta S_U)_Y$$

where S_{U0} is the unperturbed count rate, $(\Delta S_U)_Y$ or YS_{U0} is the increase in S_U due to contamination on the fissionable deposit, and $(\Delta S_U)_X$ or XS_{U0} is the perturbation due to contamination on other interior surfaces of the chamber. The p.h.d. of the Y component is indistinguishable from the normal p.h.d. However, the p.h.d. of the X component is degraded due to poorer pulse formation geometry and path truncation, so that the differential count $(S_L - S_U)$ is increased in proportion to the X component provided that the spatial distribution of the X material remains the same as the amount of material builds up). A proportionality constant β can be defined as

$$\beta = \Delta(S_L - S_U) / (\Delta S_U)_X.$$

Under normal conditions

$$S_{L0}/S_{U0} - 1 = ETZ_0$$

where ETZ_0 is 0.13% for 25S-01-2 and 0.85% for 28S-2-1. It can be shown that

$$\Delta \text{ ETZ} \equiv (S_L/S_U - 1) - (S_{L0}/S_{U0} - 1)$$

$$\approx \beta X$$

plus negligible second order terms in the small quantities X , Y , and ETZ_0 .

Since β is the ratio of contamination counts between V_L and V_U to (X-type) contamination counts above S_U , the same value of β should be observed in a thermal neutron beam as in the BIG-10 spectrum, regardless of the isotopic enrichment of the contaminating material. For the bottom chamber, the value of β is easily obtained from the second row of Table NBS-7, $^{25}\beta = 0.27/0.58 = 0.47$. However, the corresponding $(S_L - S_U)$ datum for the top chamber was invalidated by introduction of additional contamination into the top chamber during the foil exchange. Nevertheless, the proper $(S_L - S_U)$ count can be re-constructed by subtracting the nominal $(S_{L0} - S_{U0})$ from the value shown on the first line: $\Delta(S_L - S_U) = (1.13 - .045)\text{s}^{-1} = 1.085 \text{ s}^{-1}$. The $^{28}S_U$ value on the second line of the table could also have been affected by the introduction of extra contamination during the foil exchange, so that it is also necessary to reconstruct this datum. If one ignores — for the moment — that some contamination material may have been removed from the 28S deposit before the run corresponding to the fourth line of the table, then $^{28}S_U$ in line two should be equal to ($^{28}S_U$ from line one) minus ($^{28}S_U$ from line four) plus ($^{28}S_U$ from line three). The reconstructed $^{28}S_U$ for line two is then 0.53 s^{-1} , in good agreement with the measured value. This agreement argues against the possibility that $^{28}S_U$ was significantly increased by addition of contamination during the exchange of deposits and blanks; however, the possibility of removal of a significant amount of dust from the 28S deposit before the line-four run cannot be dismissed. The only consequence of removal of an unknown amount of Y-type material from the 28S deposit before the line-four run would be to

mislabel a part of the 28S Y-type contamination as X-type contamination. Since it is reasonable to assume that the X and Y contaminations increased in proportion to each other during the BIG-10 runs, the possible mislabeling of the separate types does not change the total correction and need not be considered further. The 8 ng of ^{235}U (plus associated ^{238}U) which adhered to the 28S deposit is taken to be the total Y contamination of the 28S deposit to be consistent with $^{28}\text{S}_{\text{U}}$ value for line two as derived above. Finally, using the reconstructed line-two data, one obtains $^{28}\beta = 1.085/0.53 = 2.05$.

Now that the β 's have been obtained, the X corrections ($= \Delta\text{ETZ}/\beta$) for chamber contamination (exclusive of dust on the deposits) can be obtained from the excess ETZ values for each run. Table NBS-8 lists the average ETZ for each run, and Table NBS-9 gives the corresponding X corrections.

The total Y contamination for the 28S deposit was shown above to be 8 ng of ^{235}U (plus associated ^{238}U). Assuming for the moment that the contaminating material was of 10% enrichment (this point will be verified independently later), and using the fission cross section ratios of Section A, the 8 ng of ^{235}U and associated ^{238}U would make a 0.12% contribution to the fission rate of the 28S-2-1 deposit in the BIG-10 spectrum. Thus the ^{28}Y correction for the last run of the BIG-10 tests would be 0.12%, and the Y correction for earlier runs may be scaled in proportion to the excess ETZ. The ^{28}Y corrections in table NBS-9 were obtained in that way.

The Y corrections for the 25S deposit could not be obtained directly. However, by comparison of the 28S-2-1/25S-01-2 counting ratios from Test C MOL with the earlier Test C runs and by comparison of the run "B-ANL--" (December 10, 1974 - Test Regions II and III empty) with the "Reference" runs in May and June, it is possible to infer that a ^{25}Y correction of about 0.3% is required for the C-MOL run

and the B-ANL-- run. From this value, the other ^{25}Y corrections can be scaled in proportion to the excess ETZ data. These ^{25}Y corrections are listed in Table NBS-9.

The question of the enrichment of the contamination particles will now be taken up. While the central region of BIG-10 has 10% enrichment, the reflector is made of depleted uranium, so that a wide range of contamination enrichment values would be possible. The contamination material present in the chamber at the last BIG-10 runs is presumably the same as that observed in the thermal neutron runs at NBS upon the return of the chamber. The enrichment E (^{235}U fraction) of the contamination material can be derived from the comparison of the excess ETZ's induced in the thermal neutron beam and in the BIG-10 spectrum.

$$\Delta \text{ETZ}_{\text{thermal}} \left\{ 1 + \frac{^{28}\sigma}{^{25}\sigma} \left(\frac{1}{E} - 1 \right) \right\} = \Delta \text{ETZ}_{\text{BIG-10}} \text{ for } 25\text{S-01-2}$$

and

$$\Delta \text{ETZ}_{\text{thermal}}^D \left\{ \left(\frac{1}{E} - 1 \right) + \frac{^{25}\sigma}{^{28}\sigma} \right\} = \Delta \text{ETZ}_{\text{BIG-10}} \text{ for } 28\text{S-2-1}$$

where $D = 0.061\%$ is the ^{235}U fraction in the 28S material and the $\frac{^{28}\sigma}{^{25}\sigma}$ is the cross section ratio for $^{238}\text{U}/^{235}\text{U}$ from Section A. From Table NBS-7, $\Delta \text{ETZ}_{\text{thermal}}$ is $0.27/170.1 = 0.16\%$ for 25S-01-1 and $1.08_5/5.32 = 20. \%$ for 28S-2-1. Table NBS-8 gives $\Delta \text{ETZ}_{\text{BIG-10}}$ as 0.19% for 25S-01-2 and 0.54% for 28S-2-1. The formulas above then give E values of 16% and 5.6% respectively. Certainly, the enrichment is actually the same for the contamination in both chambers, so the difference in the inferred enrichment values is merely indicative of the inaccuracy of this enrichment determination. The enrichment value was required only in deriving the ^{28}Y contamination value of 0.12% . If 16% or 5.6% had been assumed for E instead of 10% , the ^{28}Y correction would have been 0.10% or 0.14% respectively. Thus, the one correction which requires knowledge of the enrichment is not significantly affected by the spread of the derived E values.

One more point is worth stressing — that the total contamination of the 28S-2-1 deposit as returned to NBS is more than adequately known in spite of all the indirect paths that had to be followed to get the separate X and Y values. From Table NBS-7 the total contamination contribution to S_U for the top chamber (expressed as an equivalent ^{235}U mass) is simply $[(6.14-5.32)/5.32] \times 250.5 \mu \times 0.061\%$ or $24 (\pm 2)$ ng of ^{235}U . In the BIG-10 spectrum this contamination with associated ^{238}U (at $E = 10\%$) would give a total contamination correction of 0.35% in good agreement with the sum of the derived X and Y corrections (0.38%) of Table NBS-9. So the end point contamination effect is well understood and is not very large. The constancy of β from run-to-run and the proportionality of the X and Y contaminations are considered reasonable approximations over at least the later runs for which the corrections are most important.

Returning to some of the alternative interpretations mentioned at the beginning of this section, a few remarks should suffice. The possibility that the increasing ETZ's were due to deposit deterioration (with concomitant self-contamination of the chambers) is ruled out by the determination of the contamination enrichment value to be about 10% (very different from that of either deposit) and by comparison of the deposits with reference deposits back at NBS. The possibility that the increasing ETZ's were merely due to deterioration of pulse formation characteristics of the chamber is ruled out by the demonstration (Table NBS-7) that the contaminating material produced a (degraded) fission fragment p.h.d. in a thermal neutron beam and by the restoration of good performance of the chamber after cleaning.

Fission Counting Totals. The pulses from the ^{235}U chamber were counted by both a triple-scaler system and the PDP-8 pulse height analyzer, while the pulses from the ^{238}U chamber were recorded only by the PDP-8. The scalers were turned

on at the very beginning of the start-up period and were kept active about 98% of the time until the scram. The scalers were usually stopped and read eight times during each run (except the shorter Test B runs) with about 40 seconds of counting missed for each reading. The PDP-8 counter was started at the half-power point of the start-up and was active about 75% of the time, with four 40 minute counts plus one short count which ended before the scram. The output time for the PDP-8 was longer than for the scalers because the two accumulated pulse height spectra were printed out on a teletype and integrals corresponding to the triple-scaler counts had to be extracted one-by-one. With corrections applied for the output intervals of the two independent counting systems, the integrated fission counts agreed usually within 0.2% as seen in Table NBS-10. On two of the shorter Test B runs, the discrepancy was 0.5%; and in one of the Test B runs, only the PDP-8 was functioning normally. The generally good agreement of the scaler counts and equivalent PDP-8 integrals is not surprising in view of the consistency of the dead-time corrected scaler counts and live-timer accumulations reported in Section A. (Although the scaler system used in the irradiations was not identical to the system used in spectral index runs of Section A, the dead-time of the system employed in the irradiations was checked by the variance-to-mean method as before.) Furthermore, the nearly flat power profile during the runs (See Table NBS-15) made it possible to make simple and accurate corrections for the counter read-out intervals.

2. Comparisons with Activation Foil Monitors and External Fission Chambers.

In Table NBS-11 the normalized monitor responses from the LASL external fission chamber and the LASL and HEDL activation foil packs are compared with the counts from the NBS double fission chamber. All the monitor responses listed in the table have been corrected by application of LASL calculated

flux-depression factors* to correspond to the response of an infinitesimal detector at the center of a cavity-free BIG-10. Each column of the table is separately normalized such that the average of the first four Test C runs is unity. The HEDL foil counting results were supplied by Lloyd Kellogg. Due to delays in getting the monitor foil packs to HEDL, some of the HEDL results have large decay uncertainties. Table NBS-11 also lists the unweighted averaged of all the monitor responses and a weighted average which depends more heavily on the NBS fission chamber counts. (The weighting factors assigned to each monitor are given in the bottom line of Table NBS-12.) The generally good consistency between the unweighted average and the weighted average shows that there is not much systematic bias between the three fission chamber counts on the one hand and the activation foil results on the other (the activation foils having been assigned much less weight in the weighted average). The activation foil data appears to have a larger random dispersion than the fission counter data. The In and Al foils counted at LASL gave results more consistent with the fission chamber counts than the other foil monitors did. Table NBS-12 shows the comparison of all the monitors with the 28S-2-1 results, the unweighted average, and the weighted average. In this table the maximum positive discrepancy, the maximum negative discrepancy, the spread of the discrepancies (the sum of the positive and negative discrepancy magnitudes), and the standard deviation (the root-mean-square discrepancy $\times \sqrt{11/10}$) are given as comparisons. Five of the nine monitors have standard deviations from the weighted average of less than 0.3%, and the other four monitors also agree within 0.9%. When such good agreement is obtained from several independent systems, the probable error in the average is substantially lower than the error one would quote for the individual systems. For this reason the error

* Calculated Flux Depression Factors supplied by G. Hanson (LASL) to ILRR participants, September 23, 1974.

for the run-to-run normalization of Tests A, C, and E is taken to be $\pm 0.3\%$, which is slightly lower than the estimated precision of any of the separate systems in these tests.

In regard to the contamination problem discussed in the preceding section, it is important that the contamination corrections - based on post-irradiation tests and fission chamber data alone - turn out to give good consistency with the independent monitor data. The counts from 28S-2-1 were given the highest weight in the weighted average because the contamination correction was lower and more directly determined for that side of the chamber, in spite of the fact that with this ^{238}U detector different flux-depression corrections were necessary for normalization of the different tests groups (A, C, E, & B) and that no redundant triple-scaler counting system was used.

3. Results

Table NBS-13 shows the spectral index data for the irradiation sets A, B, C, E, and the "reference" runs of Section A. The in situ spectral index derived from deposits 28S-2-1 and 25S-01-2 have been normalized to the value 0.0366 for the "reference" runs. Run-to-run variations within a given test group were small compared to the differences from group-to-group (but the individual run values may be derived from Table NBS-9 if desired). The observed $^{238}\text{U}/^{225}\text{U}$ count rate ratio was 0.36% higher for the Test C runs with fission foil packs, than for the Test E runs with non-fission foil packs, whereas the calculated flux-depression factors * predict only a 0.2% difference. Table NBS-13 also shows the spectral index values corrected for flux-depression perturbations. (Every number in that column would be the same if the measurements and calculations were perfect.) The perturbation corrections indicate slightly smaller spectral index shifts than the experimental data.

* Calculated Flux Depression Factors supplied by G. Hanson (LASL) to ILRR participants, September 23, 1974.

Table NBS-15 gives the power profile and irradiation timing data for the activation foil tests. The power profiles are adequately characterized by listing the slight power level increase that occurred during the runs, because all the runs except Test A were otherwise ideal "square" irradiations. For Test A, a correction factor is also listed to account for a brief power surge that occurred in the latter half of that run.

Table NBS-16 shows the absolute fission counting results (fission/milligram) and shows the weighted average run-to-run normalization data with uncertainties. The run-to-run normalization uncertainty is $\pm 0.3\%$ for the Tests A, C, and E, but is $\pm 0.6\%$ for Test B due to the poorer scaler-PDP 8 agreement and lack of monitor redundancy in the Test B runs. The absolute fission counting errors are given as 1.4% for Tests A, C, and E and 1.5% for Test B. A summary of the significant error components is given in Table NBS-14.

Expected Achievements

As in Section A, the fissions/milligram results given here are regarded as final except for the possibility of readjustments associated with refinement of the mass assay of the NBS reference deposits.

TABLE NBS-7

THERMAL NEUTRON FISSION RATE DATA
FOR CONTAMINATION ASSESSMENT

Run Description	25S-01-2 Bottom Chamber		28S-2-1 Top Chamber	
	S_U	$(S_L - S_U)$	S_U	$(S_L - S_U)$
	cps	cps	cps	cps
Chamber as received following BIG-10 Tests	170.1	.465	6.14	1.13*
Blanks replacing fissionable deposits before cleaning	.58	.27	.55	1.36*
Blanks replacing fissionable deposits after cleaning	.096	.11	.07	.12
Same deposits used at BIG-10 after cleaning chamber and deposits	171.0	.313	5.68	.18
Nominal count rates for a clean chamber with clean deposits	171.0 [†]	.22	5.32 ^{††}	.045

*The increase in $(S_L - S_U)$ in the run with blanks is believed to be due to introduction of additional contamination into the top chamber during the replacement of the deposits with the blanks.

[†] S_U normalized to previous run.

^{††}The depleted uranium count rate is due to the ^{235}U content of 0.061 atom percent.

TABLE NBS-8

HISTORY OF (S_L/S_U-1) VALUES AND OBSERVED S_U RATIOS

Test	Month/ Day	28S-2-1 ETZ (S_L/S_U-1)	25S-01-2 ETZ (S_L/S_U-1)	S_U Ratio (28S-2-1/25S-01-2)
	(1974)	(%)	(%)	
Nominal	---	0.85	0.13	---
Reference	5/7-10 6/20	0.99	0.14	1.789
A	5/29	0.93	0.12	1.786
C-LASL	7/9	0.97	0.15	1.793
C-ANL	7/16	0.98	0.16	1.792
C-ANC	7/23	1.02	0.18	1.790
C-HEDL	7/31	1.02	0.18	1.791
E-ANL	8/6	1.06	0.19	1.786
E-ANC	8/13	1.19	0.23	1.784
E-HEDL	8/27	1.21	0.25	1.784
E-LASL	9/17	1.22	0.25	1.780
E-MOL	10/9	1.30	0.26	1.781
B-ANL-4	11/12	1.35	0.32*	1.777
C-MOL	11/19	1.33	0.30	1.786
B-ANL--	12/10	1.39	0.31	1.783
B-ANL-5	12/10	1.39	0.31	1.774
B-ANL-6	12/10	1.41	0.32	1.780
B-ANL-7	12/10	1.39	0.32	1.778
B-ANL-8	12/10	1.39	0.32	1.778

* High due to noise pickup during scram and shutdown.

TABLE NBS-9
CONTAMINATION CORRECTIONS AND THE CORRECTED
FISSION RATIO

Test	Month/Day	^{28}X	^{28}Y	^{25}X	$^{25}\text{Y}^*$	$^{28}\text{S}_{\text{Uo}}/^{25}\text{S}_{\text{Uo}}$
	(1974)	(%)	(%)	(%)	(%)	
Reference	5/7-10 6/20	0	0	0	0	$1.789(2)^{\dagger}$
A	5/29	.04	.02	0	0	1.785(2)
C-LASL	7/9	.05	.02	.05	.04	1.793(1)
C-ANL	7/16	.06	.03	.05	.04	1.792(1)
C-ANC	7/23	.08	.04	.11	.08	1.791(1)
C-HEDL	7/31	.08	.04	.10	.08	1.792(1)
E-ANL	8/6	.10	.05	.11	.08	1.787(1)
E-ANC	8/13	.16	.08	.21	.16	1.786(1)
E-HEDL	8/27	.17	.08	.24	.18	1.787(1)
E-LASL	9/17	.18	.08	.25	.19	1.783(1)
E-MOL	10/9	.22	.10	.27	.21	1.784(1)
B-ANL-4	11/12	.24	.11	.39	.24	1.782(3)
C-MOL	11/19	.23	.11	.34	.26	1.791(1)
B-ANL--	12/10	.26	.12	.36	.28	1.788(3)
B-ANL-5	12/10	.26	.12	.37	.28	1.779(3)
B-ANL-6	12/10	.27	.13	.39	.30	1.785(3)
B-ANL-7	12/10	.26	.12	.38	.29	1.783(3)
B-ANL-8	12/10	.26	.12	.39	.30	1.783(3)

* ^{25}Y Correction scaled to bring corrected fission ratio for Test C-MOL and B-ANL--into agreement with earlier corresponding runs—the first four Test C runs and the Reference runs, respectively.

† The number in parentheses is the statistical error in the last digit, based on the reproducibility of separate counts, or for Test B on the expected counting errors for the single count taken.

TABLE NBS-10

INTEGRATED FISSION COUNTS FOR 25S-01-2 SCALERS VS. PDP-8

Test	Integrated S_U Count (Dead-time corrected)		Scalers/PDP-8	Average
	Scaler	PDP-8		
	(10^7 counts)	(10^7 counts)		(10^7 counts)
A	1.872	1.868	1.002	1.870
C-LASL	1.842	1.837	1.003	1.840
C-ANL	1.854	1.850	1.002	1.852
C-ANC	1.862	1.858	1.002	1.860
C-HEDL	1.860	1.856	1.002	1.858
E-ANL	1.861	1.857	1.002	1.859
E-ANC	1.853	1.850	1.002	1.852
E-HEDL	1.859	1.856	1.002	1.857
E-LASL	1.859	1.858	1.001	1.859
E-MOL	1.859	1.857	1.001	1.858
B-ANL-4	0.0802	0.0798	1.005	0.0800
C-MOL	1.856	1.854	1.001	1.855
B-ANL--	0.0879	0.0875	1.005	0.0877
B-ANL-5	--	0.0876	--	0.0876
B-ANL-6	0.0879	0.0877	1.002	0.0878
B-ANL-7	0.0879	0.0877	1.002	0.0878
B-ANL-8	0.0879	0.0877	1.002	0.0878

TABLE NBS-11

NORMALIZED FLUENCES AS MONITORED BY FISSION CHAMBERS AND ACTIVATION FOILS

Test	Monitor [†]	28S-2-1 NBS Fission Chamber	25S-01-2 NBS Fission Chamber	LASL External Fission Chambers	Au LASL-II	In LASL-II	Al LASL-II	Au HEDL-II	In HEDL-II	Al HEDL-II	(Unweighted) Average of nine Monitors	Weighted Average
A		1.009	1.011	1.011	1.014	1.014	1.015	1.017	1.020	1.008	1.013	1.011
C-LASL ^{††}		0.994	0.994	0.996	0.997	0.993	0.994	0.991	—	0.987	.993	0.994
C-ANL		1.000	1.000	1.001	.998	1.006	1.004	0.999	0.992	1.000	1.000	1.001
C-ANC		1.003	1.003	1.002	1.002	1.001	0.998	1.006	1.014	1.000	1.003	1.002
C-HEDL		1.003	1.003	1.002	1.003	1.000	1.004	1.006	0.995	1.012	1.003	1.003
E-ANL		1.002	1.003	1.000	1.002	1.002	1.001	—	0.998	1.003	1.001	1.002
E-ANC		0.996	0.997	0.998	1.001	0.995	0.998	1.000	0.996	0.985	.996	0.997
E-HEDL		0.999	1.000	0.999	1.008	1.003	1.003	1.004	0.991	0.989	1.000	1.000
E-LASL		0.997	1.000	0.999	1.005	1.003	1.003	1.006	0.999	0.983	.999	0.999
E-MOL		0.997	1.000	0.999	1.007	1.003	1.001	1.005	1.002	0.989	1.000	0.999
C-MOL		0.997	0.997	0.998	1.001	1.000	1.002	1.015	1.010	0.998	1.002	0.999

[†]Perturbation adjustments have been applied to adjust all monitor responses to correspond to a flat detector (such as ²³⁵U) in Test Region I.

^{††}Each column of this table is normalized separately such that the average of the first four Test C runs is unity.

TABLE NBS-12
CONSISTENCY OF RUN-TO-RUN NORMALIZATION

		NBS 28S-2-1	NBS 25S-01-2	LASL External Fission Chambers	Au LASL-II	In LASL-II	Al LASL-II	Au HEDL-II	In HEDL-II	Al HEDL-II
Discrepancy versus 28S-2-1 (%)	+	0	0	.2	.2	.3	.5	.3	.8	1.4
	-	0	.3	.2	1.0	.6	.6	1.8	1.3	1.2
	Spread	0	.3	.4	1.2	.9	1.1	2.1	2.1	2.6
	Std. Dev.	0	.16	.16	.55	.42	.42	.80	.77	.80
Discrepancy versus Unweighted Average of Nine Monitors (%)	+	.5	.5	.4	.2	.3	.5	.2	.8	1.6
	-	.1	.2	.3	.8	.6	.4	1.3	1.1	.9
	Spread	.6	.7	.7	1.0	.9	.9	1.5	1.9	2.5
	Std. Dev.	.24	.20	.20	.44	.30	.28	.59	.62	.88
Discrepancy versus Weighted Average (%)	+	.2	.2	.2	.3	.3	.4	.3	.8	1.6
	-	.1	.1	.2	.8	.5	.4	1.6	1.1	.9
	Spread	.3	.3	.4	1.1	.8	.8	1.9	1.9	2.5
	Std. Dev.	.14	.10	.11	.46	.30	.29	.70	.76	.88
Assigned Weighting	—	10	4	4	1	2	2	.5	.5	.5

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TABLE NBS-13
SPECTRAL INDEX VARIATIONS FOR TEST A, B, C, E, & REF.

Test	$^{238}\text{U}/^{235}\text{U}$ Fission Cross Section Ratio <u>in situ</u>	$^{238}\text{U}/^{235}\text{U}$ Fission Cross Section Ratio Corrected for Perturbations
Ref., B-ANL--	0.0366	0.0372
A	0.0365	0.0371
B	0.0365	0.0371
C	0.0367	0.0372
E	0.0365	0.0371

TABLE NBS-14

SUMMARY OF ERRORS IN THE FISSIONS/MILLIGRAM RESULTS

Error Factor	Estimated Error Magnitude
	(%)
²³⁵ U deposit mass (255-01-2)	1.3
Run-to-run normalization (contamination correction)	0.3
Fragment absorption	0.4
ETZ	0.1
Fission in isotopic impurities	0.0
Dead-time correction, live- timer accuracy	0.2
Integration of counts over complete power profile	{ 0.2 (Tests A,C,E) 0.5 (Test B)
Flux perturbations	0.2
Combined errors	{ 1.4 Tests A,C,E 1.5 Test B

TABLE NBS-15
IRRADIATION POWER PROFILES AND TIMING DATA

	Count Rate Increase During Run	Irradiation Time*	Time at End of Run
	(%)	(sec)	
A	1.8 [†]	13214	8:51:09 AM MDT 29 May 74
C-LASL	2.0	13214	11:41:40 AM MDT 9 July 74
C-ANL	1.1	13214	10:17:08 AM MDT 16 July 74
C-ANC	0.9	13214	11:34:59 AM MDT 23 July 74
C-HEDL	1.2	13214	10:06:00 AM MDT 31 July 74
E-ANL	1.5	13214	10:18:40 AM MDT 6 Aug 74
E-ANC	1.3	13214	10:08:00 AM MDT 13 Aug 74
E-HEDL	1.7	13214	10:07:58 AM MDT 27 Aug 74
E-LASL	1.1	13214	10:15:51 AM MDT 17 Sept 74
E-MOL	1.1	13214	06:12:28 AM MDT 9 Oct 74
C-MOL	1.1	13214	06:14:35 AM MDT 19 Nov 74

* A correction of 13.8 seconds has been included to account for the finite start-up period.

† A delta-function like power surge occurred during the latter half of Test A so that (especially) for shorter-lived activations, higher relative count rates were produced than would have been in a "square irradiation" with the same fluence. This perturbation depends on the activation decay constant as follows:

$$\begin{array}{llll}
 \text{Decay constant, } \lambda (\text{min}^{-1}) & = & 0.1 & 0.01 & \leq 0.001 \\
 \text{Test A/"square" irradiation} & = & 1.0042 & 1.0026 & 1 + (0.33 \text{ min})\lambda
 \end{array}$$

TABLE NBS-16

FISSIONS PER MILLIGRAM IN BIG-10 IRRADIATIONS
(CORRECTED FOR CAVITY AND INSTRUMENT PERTURBATIONS*)

Test Name	Fission Probability* per nucleus, P_f (10^{-9})	Fissions* per mg of ^{235}U (10^9)	Run-to-Run Normalization
A	1.492 (21) [†]	3.82 (5) [†]	1.011 (3) [†]
C-LASL	1.467 (21)	3.76 (5)	0.994 (3)
C-ANL	1.477 (21)	3.78 (5)	1.001 (3)
C-ANC	1.478 (21)	3.79 (5)	1.002 (3)
C-HEDL	1.480 (21)	3.79 (5)	1.003 (3)
C-MOL	1.474 (21)	3.78 (5)	0.999 (3)
E-ANL	1.478 (21)	3.79 (5)	1.002 (3)
E-ANC	1.471 (21)	3.77 (5)	0.997 (3)
E-HEDL	1.475 (21)	3.78 (5)	1.000 (3)
E-LASL	1.474 (21)	3.78 (5)	0.999 (3)
E-MOL	1.474 (21)	3.78 (5)	0.999 (3)
B-ANL-4	0.0634(10)	0.162(3)	0.0430(3)
B-ANL-5	0.0695(11)	0.178(3)	0.0471(3)
B-ANL-6	0.0695(11)	0.178(3)	0.0471(3)
B-ANL-7	0.0695(11)	0.178(3)	0.0471(3)
B-ANL-8	0.0695(11)	0.178(3)	0.0471(3)

*A correction of 1/0.991 has been applied to derive the unperturbed values listed in this table from the values measured in situ. See Section A for further details.

[†]The number in parentheses is the error in the least significant digit.

C. ABSOLUTE FISSION RATES MEASUREMENTS IN MOL-ΣΣ

A. Fabry, F. Cops, CEN-SCK
J. Grundl, D. Gilliam, NBS

Objective

. Establish absolute fission rates per nucleus for ILRR irradiations in the Mol-ΣΣ Secondary Intermediate-Energy Standard Neutron Field at the CEN/SCK Laboratories in Mol, Belgium. Four fissionable materials, ^{235}U , ^{239}Pu , ^{238}U , and ^{237}Np and the NBS double fission chamber are to be employed. The fission rates are to be reported along with results from permanent, run-to-run flux level monitoring systems operated by CEN/SCK and to be used for ILRR activation foil irradiations. Fission rates are to be specified at the deposit with only nominal estimates of field perturbations due to the fission chamber included. Fission cross section ratios - spectral indices - are to be specified with evaluated errors.

Summary

Absolute fission rate measurements were performed in the Mol-ΣΣ Secondary Standard Neutron Facility in order to prepare for subsequent high-fluence ILRR activation foil irradiations. Twelve separate chamber exposures were carried out from mid-1974 thru early 1975. Fission cross section ratios based on these runs are (1.000: 1.173 ± 0.025 : 0.0564 ± 0.0014 : 0.381 ± 0.011) for ^{235}U : ^{239}Pu : ^{238}U : ^{237}Np respectively. The run-to-run fluence monitoring system - $\text{In}(n,n')$ activation detectors and peripheral fission chambers - were established to a precision of $\pm 0.4\%$.

Accomplishments and Status

The Mol-ΣΣ facility designed for intercomparisons and standardization of experimental techniques applied in zero-power fast critical assemblies is described in Reference 1. Early and very preliminary NBS fission chamber measurements were begun in early 1972. More intensive efforts began in mid-1974 with a full complement of NBS fissionable deposits and improved experimental arrangements. In particular, the treatment of the re-entrant hole for placing active instruments at the center of ΣΣ was extensively investigated,

and external, permanent fission chamber monitor installed-see Ref. 3. As a result of the hole perturbation investigation a correction of $(0.0 \pm 0.2)\%$ has been established for the ^{235}U response obtained with an NBS double fission chamber. The procedures for operating the NBS fission chamber and analyzing fission pulse rate data in the Mol- $\Sigma\Sigma$ are consistent with those employed elsewhere for ILRR measurements (see previous ILRR Reports and Reference 2.) Fission chambers were assembled and sealed at NBS and then sent to CEN/SCK where they were operated by staff members familiar with NBS fission chamber techniques.

Chamber Performance and Operation Chamber performance during the major $\Sigma\Sigma$ exposures was generally satisfied with only one important surprise, a difficulty which made it necessary to discard one long series of chamber exposures. For these runs, the important chamber performance check ratio, S_L/S_U (S_L and S_U are count rates above two bias levels set a factor 1.5 apart in the valley of the fission pulse height distribution) deviated significantly from expectation. After a difficult and time consuming experimental investigation it was ascertained at CEN/SCK that the fission chamber was responding to gamma ray pile up in the chamber manifold. The chamber was returned to NBS where additional checks were performed in high-intensity gamma ray beams. The problem was reduced to negligible proportion by covering all exposed pulse conducting surfaces (e.g. feed thru connector pins), and filling the manifold with a plastic foam packing material. This was the first application of the NBS fission chamber to a neutron field where high gamma-to-neutron ratios extended out to the chamber manifold.

Fission chamber data obtained with significant gamma ray response was not used for the final determination of the $\Sigma\Sigma$ central fission rates. Some useful supplementary information, however, was obtained from these early irradiations:

- (1) Assessment of fission chamber sensitivity to weak spatial gradients at the center of $\Sigma\Sigma$. Along the thermal column axis (direction of maximum gradient), the variation was 0.8% per cm for ^{235}U and 1.6% per cm for ^{238}U .

- (2) Check of response invariance with respect to chamber rotation about the stem. Preliminary measurements in NISUS the sister facility to $\Sigma\Sigma$ in Great Britain, indicated in the existence of such variation. It is suggested that this variation was associated with improper control of access hole streaming.
- (3) Demonstration that any inadvertent small gap in the stepped closure of the inner boron carbide hemispheres of $\Sigma\Sigma$ do not influence fission rates observed at the center of the assembly.
- (4) Establish the neutron transmission equivalence of several different boron carbide shells used in $\Sigma\Sigma$.

Certain operational characteristics of the fission chamber may differ slightly from application to application, or must be verified as meeting expectation in each application. These are noted below.

Extrapolation-to-Zero (ETZ) The flat extrapolation of the fission pulse-height-distribution (phd) from V_L , the voltage setting of the lower discriminator for the S_L pulse scaler, to zero pulse height is termed the ETZ. The ETZ under normal operation depends nearly linearly on fissionable deposit thickness - see Ref. 2, Fig. 4. Pulse rate data for the $\Sigma\Sigma$ runs were reduced with an alternate definition of ETZ, namely, a flat extrapolation of the phd from V_U , the voltage setting of the upper discriminator for the S_U pulse scaler, rather than V_L ; and in almost all of these runs, V_L was set at $(2/3) V_U$ - departing slightly from the usual procedure of Reference 2. Since flat extrapolations are everywhere assumed at the present stage of NBS fission chamber development and interpretation, both definitions of the ETZ lead to an equivalent extrapolated fission pulse rate. Fractional uncertainties must be judged differently, of course since the pulses registered between V_L and V_U (i.e. S_L/S_U-1) are valid fission pulses although they are entered as an ETZ correction in the alternative definition employed here. The $\Sigma\Sigma$ data has not been adjusted to conform to the conventional ETZ definition adopted in Reference 2 but remains as originally analyzed. Henceforth, this report will label the alternative ETZ as ETZ-A. For almost all data reported here $ETA-A = 1.5$ (ETZ); nominal values for ETZ-A are listed in Table

NBS-17. The recipe applied to pulse rate data recorded by the S_U scaler in order to establish absolute fission rates was as follows:

$$(ETA-A)_{APPLIED} = (ETZ-A)_{NOMINAL} + 1/2 (ETZ-A)_{NOMINAL} - 3 \left(\frac{S_L}{S_U} - 1 \right) ,$$

where $3(S_L/S_U - 1)$ is $(ETZ-A)_{OBSERVED}$. This recipe for averaging nominal and observed extrapolations to zero is consistent with the recommendations of reference 2.

Effective Dead Time (τ) Pulse collection dead times depend upon specific electronic modules and on discriminator settings. On-line measurement with the chamber are usually advisable. Observations were undertaken for all three NBS fission chambers in $\Sigma\Sigma$ without its boron shell. Count rates up to 15000 per second were obtained and count rate ratios varied by more than a factor of 40. Supplementary measurements were also carried out in the critical assembly BR02 also at CEN/SCK. A value of $\tau = 4.0 \pm 0.4 \mu\text{sec}$ was derived from these measurements. This value is somewhat larger than the $3.1 \pm 0.5 \mu\text{sec}$ applied to CFRMF and reported in Reference 2. Count rates in $\Sigma\Sigma$ never exceeded 500 cps so that this difference is of little consequence.

Operational Reproducibility Between six and twenty individual counts were taken during each of the eleven irradiations and results generally were within the expectations of statistics. Final S_U pulse rates are quoted with a std. dev. of the mean of between $\pm 0.1\%$ and $\pm 0.5\%$ for all isotopes. Interchange of electronic outputs, carried out for some of the longer runs, gave reproducible results. Pulse height distributions obtained during the course of the irradiations were in agreement with the S_L , S_U , and S_{gc} scaler results when compared.

Irradiation Flux Level and Fluence Monitors The irradiations documented in this report were all performed at a nominal reactor power of 400 kW. Long term precision fluence monitors for $\Sigma\Sigma$ have been in service since 1970 and involve the response of properly placed gold and indium activation foils. For the $In(n,n')$ monitor, up to four separate foils are employed to achieve desired redundancy. Monitoring with activation detectors is satisfactory for

activation detector irradiations in $\Sigma\Sigma$ but it is less so for absolute fission chamber calibrations. For the 1974 irradiations, a permanent fission chamber monitor was installed as a supplementary flux level monitor. This system performed well initially and, judging from count rate ratios relative to the NBS fission chamber, the short term reproducibility during irradiations was better than $\pm 0.2\%$. However, comparison of the fission chamber monitor with the $\text{In}(n,n')$ activation fluence monitors did reveal a slow, long term drift amounting to a few percent. This time-dependence of response was described with a linear least squares fit which was used to establish a proper $\text{In}(n,n')$ to fission monitor ratio for each of the NBS fission chamber irradiations.

The primary irradiation level monitor for $\Sigma\Sigma$ is the $\text{In}(n,n')$ activation response, and all final absolute fission rate results are reported relative to this monitor reaction. The flux-fluence relationship between the fission chamber and the $\text{In}(n,n')$ monitor has been established in measurement analyses separate from this report. For two NBS fission chamber irradiations three weeks apart where both Indium and fission chamber monitors were employed, the agreement of monitor ratios is 0.9%.

Fission Rate Ratios in $\Sigma\Sigma$. The various fissionable deposits used during the course of these preparatory irradiations in $\Sigma\Sigma$ are listed in Table-17 along with the mass of the principle isotope. The masses are based on the latest comparisons of these deposits with the NBS reference deposits. Reference deposit masses remain as reported in Reference 2. Table NBS-18 lists the corrections applied to the S_U fission pulse rates in order to obtain isotopic fission rates. All but the extrapolation-to-zero are fixed corrections; the ETZ-A correction varies slightly from run-to-run since the recipe described earlier for establishing ETZ-A includes the observed S_L/S_U ratio for each irradiation.

A complete summary of the $\Sigma\Sigma$ fission rate measurements appears in Table NBS-19. The results are listed chronologically providing (1) S_U -uncorrected pulse rate per mg of principle isotope; (2) observed ETZ-A with statistical error; (3) fission rate ratio for each deposit pair accompanied by the standard deviation of the array of counts taken and the standard deviation of the mean; (4) nominal cps for the $\Sigma\Sigma$ fission chamber flux level monitor; and (5)

S_U -corrected fission rate relative to the $\Sigma\Sigma$ fission chamber flux monitor with standard deviations of the data and of the mean. Reproducibility of fission rate ratios may be judged from two sets of ratio measurements in which the same pair of fissionable deposits were employed:

$\frac{28N-5-2}{49I-1-1}$:	0.0480, 0.480, 0.474, 0.0476 std. dev. = 0.55%	1.3% max. spread May to Nov. 1974
$\frac{28HD-5-1}{25S-2-3}$:	0.0553, 0.0558, 0.0556, 0.0561 std. dev. = 0.52%	1.4% max. spread Sept. 1974 to Jan. 1975

The precision in terms of the internal standard deviation of $\pm 0.5\%$ over a period of up to six months is quite satisfactory. Final errors will be dominated by other issues.

Final cross section ratios are given in Table NBS-20. The relatively large errors (cf. Ref. 2, Table VII) reflect the use of thick ^{238}U deposits and the attendant large fragment absorption correction. Some complementary fission comparisons of thick and thin ^{238}U deposits could bring this error nearer to that for CFRMF as reported in Ref. 2. Errors for the other cross section ratios are only slightly higher than the CFRMF values. A substantial improvement in accuracy will come about, without any additional measurements, when the entire mass assay error scale at NBS is reduced.

Absolute Fission Rates in $\Sigma\Sigma$ The specification of absolute fission rates for driven and self-sustaining neutron fields always presents unique problems since there is no universal neutron flux level monitor. In ILRR program calibration irradiations flux levels have been monitored differently in each neutron field. The various monitoring schemes employed to date are as follows:

CFRMF. Conventional current reading reactor power level instrumentation was the primary flux monitor. The neutron detector for this instrumentation was a commercial fission chamber operated at the periphery of reactor core entirely decoupled from the spectrum converter arrangement at the center. Gold and Indium foils placed near the calibration irradiation position were auxiliary fluence monitors. Activation counting was done at ANC and HEDL.

Big Ten. The NBS fission chamber operated simultaneously at the calibration position during activation foil irradiations was the primary flux monitor. Commercial fission chambers employed as pulse rate devices were operated in remote positions away from the reactor. Gold and indium foils

at the activation foil position were auxiliary fluence monitors. Activation counting was done at LASL and HEDL.

NBS Thermal Column. The NBS fission chamber operated with the foils inside during irradiation was the primary flux monitor. A commercial fission chamber positioned in the thermal column leakage flux provided a second flux monitor. The latter was given equal weight in establishing run-to-run normalization.

$\Sigma\Sigma$. The primary flux monitor is $\text{In}(n,n')$ activation. Foils are placed within the $\Sigma\Sigma$ cavity and are counted at CEN/SCK after irradiation. In preparation for the ILRR irradiations a commercial fission chamber was placed in the outer region of the thermal column as a secondary flux monitor.

The most significant factor in this outline is that flux monitoring schemes are not uniform and that responsibility for them is largely spread about. It is not easy therefore to investigate these schemes systematically. Each field was subject to tests of precision and consistency, of course, but with each the problems were different. As such, the ILRR program does not really accumulate a great deal of flux monitoring confidence as it moves from one neutron field to the next.

The $\Sigma\Sigma$ irradiations reported here were undertaken to prepare for high-power ILRR activation detector calibration irradiations at a later date. Flux monitoring systematics were being investigated, therefore, and in particular the performance of the thermal column fission chamber was established. As mentioned above a long term drift of the fission chamber response relative to the $\text{In}(n,n')$ monitors was observed. The drift also may be observed relative to the NBS fission chamber. In Table NBS-21 response ratios in chronological order show the magnitude of the changes. In an analysis separate from this report the long-term drift of the thermal column fission chamber was linearized and the fission chamber made to agree with the $\text{In}(n,n')$ monitors. On this basis an $\text{In}(n,n')$ reaction rate has been assigned to each irradiation given in Table NBS-19.

Specific fission rates per nucleus for each irradiation may be obtained by applying to the pulse rate per mg in column 1 of Table NBS-19 an atoms/gm conversion, and corrections from Table NBS-18. Dividing each specific fission rate so obtained by the assigned $\text{In}(n,n')$ reaction rate per nucleus gives the necessary fission rate calibration factor for scheduled ILRR foils irradiations.

These calibration factors are given in Table NBS-22. External and internal precisions included in the Table are consistent providing a welcome demonstration of the reproducibility of fission rate measurements and the $\Sigma\Sigma$ monitoring capability over extended periods of time. Estimates of total systematic errors associated with absolute fission chamber efficiency, field perturbations, and fissionable deposit masses are also included in the Table. The final error in the last column of Table NBS-22 is the rms sum of the errors listed; they do not include estimate of the internal precision for the $\text{In}(n,n')$ monitoring system. The "external precision" of $\sim 0.4\%$ in column 4 of the table, derived from the eleven irradiations covering a period of exactly one year, see Table NBS-21, are probably satisfactory for this purpose.

Expected Achievements No further absolute fission rate measurements in $\Sigma\Sigma$ are planned although some real improvement could be expected if thenner fissionable deposits could be run at some future date. The NBS chamber will be used as a redundant flux monitor during the high power exposures of ILRR activation foils.

References

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2. J. Grundl, D. Gilliam, N. Dudey, R. Popek, "Measurement of Absolute Fission Rates," Nuclear Technology 25, 237(1975).
3. A. Fabry-This Progress Report, CEN/SCK contribution. Also, joint GfK-RCN-CEN-NBS report in preparation.

TABLE NBS-17

FISSIONABLE DEPOSITS EXPOSED AT THE CENTER
OF THE $\Sigma\Sigma$ STANDARD NEUTRON FIELD

DEPOSIT IDENTIFICATION AND PRINCIPAL ISOTOPE		^a NOMINAL ETZ-A	MASS OF PRINCIPAL ISOTOPE (μg)
28N-5-1	^{238}U (natural)	$1.040 \pm .01$	$692 \pm 1.4\%$
28N-5-2	^{238}U (natural)	$1.040 \pm .01$	$686 \pm 1.4\%$
28HD-5-1	^{238}U (99.999%)	$1.037 \pm .01$	$651 \pm 2.2\%$
25S-2-1	^{235}U (99.7%)	$1.016 \pm .005$	$235 \pm 1.5\%$
25S-2-3	^{235}U (99.7%)	$1.015 \pm .005$	$222 \pm 1.4\%$
49I-1-1	^{239}Pu (99.11%)	$1.006 \pm .004$	$105.1 \pm 1.4\%$
37 -5-1	^{237}Np (99.3%)	$1.035 \pm .01$	$630 \pm 1.8\%$

All deposits are 12.7 mm diameter fissionable oxides prepared by vacuum evaporation. Deposit backings are 19.0 mm diameter polished platinum 0.13 mm thick.

- (a) Extrapolation-to-zero from upper discriminator level derived from ETZ values given in Figure 4 of Ref. 2.

TABLE NBS-18
TYPICAL CORRECTIONS FOR DERIVING UNPERTURBED ISOTOPIC FISSION RATES FROM CHAMBER PULSE RATES

CORRECTION	Isotope Deposit	^{238}U		^{235}U	^{239}Pu	^{237}Np
		28N-5-1 28N-5-2	28HD-5-1	25S-2-3	49I-1-1	37-5-1
Dead Time Loss ($\tau = 4.0 \pm 0.4 \mu\text{sec}$)		1.000 ± 0.000	1.000 ± 0.000	1.0009 ± 0.0001	1.0005 ± 0.0001	1.000 ± 0.000
<u>Fragments Undetected</u>						
- Extrapolation-to-zero ^(a) (ETZ-A) _{nominal}		1.050 ± 0.015	1.036 ± 0.013	1.015 ± 0.005	1.008 ± 0.004	1.037 ± 0.013
- Absorption in Deposit		1.037 ± 0.01	1.035 ± 0.01	1.012 ± 0.003	1.006 ± 0.003	1.034 ± 0.01
Fission In Other Isotopes		$0.885 \pm 0.005^{(b)}$	1.000 ± 0.000	0.9995 ± 0.0002	0.997 ± 0.001	0.980 ± 0.008
Neutron Scattering and Absorption		1.006 ± 0.003	1.006 ± 0.003	1.000 ± 0.001	1.000 ± 0.001	1.006 ± 0.003
Access Hole Perturbation		1.000 ± 0.000	1.000 ± 0.000	0.995 ± 0.005	0.995 ± 0.005	1.000 ± 0.000
Net Correction		$0.970 \pm 1.9\%$	$1.079 \pm 1.7\%$	$1.023 \pm 0.8\%$	$1.0059 \pm 0.7\%$	$1.057 \pm 1.9\%$
Absolute Sum of Correction		1.238	1.079	1.033	1.071	1.100

(a) Typical correction for the fission count rate, S_U , above highest discriminator level V_U . Note the difference in definition compared to Reference 2 - see text. When necessary, errors have been chosen conservatively to account for the differences between expected and observed S_L/S_U ratios.

(b) This correction is derived from direct experimental observations combined with isotopic analysis.

TABLE NBS-19
SUMMARY OF FISSION RATE MEASUREMENTS AT $\Sigma\Sigma$ CENTER
WITH NBS DOUBLE FISSION CHAMBER

IRRADIATION DATE AND FISSIONABLE DEPOSIT IDENTIFICATION		S_U -UNCORRECTED FISSION PULSE-RATE PER MG. (cps/mg)	OBSERVED ETZ-A $3(S_L/S_U-1)$	aS_U -CORRECTED FISSION RATE RATIO (I/II)	$\Sigma\Sigma$ FISSION CHAMBER MONITOR (cps)	S_U -CORRECTED FISSION RATE FISSION CHAMBER MONITOR
May 16, 1974						
28N-5-2 (I)		63.3	0.080 +14%	0.0480 +1.1% (+0.3%)	—	—
49I-1-1 (II)		1271	0.014 +10%			—
May 24, 1974						
28N-5-2 (I)		60.9	0.073 + 5%	0.0480 +0.6% (+0.2%)	—	—
49I-1-1 (II)		1223	0.008 + 7%			—
June 21, 1974						
28N-5-1 (I)		62.0	0.052 + 8%	0.0557 +1.0% (+0.2%)	555	0.1084 +0.9% (+0.2%)
25S-2-3 (II)		1055	0.019 + 5%			1.945 +0.5% (+0.1%)
Sept. 30, 1974						
28HD-5-1 (I)		49.7	0.036 +10%	0.0553 +1.2% (+0.4%)	498	0.1077 +1.0% (+0.4%)
25S-2-3 (II)		947	0.018 + 3%			1.945 +0.4% (+0.14%)
Oct 29, 1974						
28HD-5-1 (I)		49.0	0.038 + 2%	0.0558 +0.3% (+0.15%)	494	0.1070 +0.3%
25S-2-3 (II)		926	0.020 + 3%			1.918 +0.2%
Oct. 30, 1974						
28N-5-2 (I)		52.7	0.071 +12%	0.0474 +0.8% (+0.3%)	483	0.1058 +0.8% (+0.3%)
49I-1-1 (II)		1071	0.009 + 6%			2.231 +0.5% (+0.3%)
Nov. 12, 1974						
28N-5-2 (I)		52.1	0.064 ± 3%	0.0476 ±1.0% (±0.4%)	484	0.1044 ±0.8% (+0.3%)
49I-1-1 (II)		1055	0.012 +10%			2.192 +0.3% (+0.1%)
Nov 18, 1974						
28HD-5-1 (I)		47.7	0.041 + 9%	0.0556 +1.2% (+0.3%)	489	0.1050 +1.3% (+0.3%)
25S-2-3 (II)		904	0.021 + 6%			1.887 +0.5% (+0.1%)
Jan 15, 1975						
28HD-5-1 (I)		48.9	0.038 +12%	0.0561 +1.3% (+0.5%)	491	0.1073 +1.3% (+0.5%)
25S-2-3 (II)		919	0.022 + 3%			1.913 +0.4% (+0.15%)
Feb. 18, 1975						
28HD-5-1 (I)		47.4	0.047 +13%	0.1475 +1.0% (+0.25%)	481	0.1064 +1.1% (+0.25%)
37 -5-1 (II)		328	0.039 + 3%			0.7204 +0.5% (+0.1%)
May 7, 1975						
37 -5-1 (I)		340	0.035 + 4%	0.3788 +0.7% (+0.3%)	491	0.7303 +0.4% (+0.15%)
25S-2-3 (II)		926				1.928 +0.5% (+0.2%)

All errors shown are one standard deviation based on the array of S_L , S_U and $\Sigma\Sigma$ monitor count rate data obtained during the irradiation. Std dev of the mean is given in parenthesis. No errors associated with corrections to absolute fission rate or with the $\Sigma\Sigma$ monitoring system are included in this table.

^a" S_U -corrected" refers to the product: S_U -Uncorrected x [net correction from Table NBS-18] - [deposit mass Table NBS-17]

TABLE NBS-20
FINAL CROSS SECTION RATIOS FOR $\Sigma\Sigma$

	Cross Section Ratio	Error Components (1σ)		
		Precision	Correction to Absolute Fission Rate	Mass Ratio
$\frac{\bar{\sigma}_f(^{238}\text{U})}{\bar{\sigma}_f(^{235}\text{U})}$ (a)	0.0564 $\pm 2.5\%$	$\pm 0.3\%$	$\pm 1.4\%$	$\pm 2.0\%$
$\frac{\bar{\sigma}_f(^{239}\text{Pu})}{\bar{\sigma}_f(^{235}\text{U})}$	1.173 $\pm 2.1\%$	$\pm 0.4\%$	$\pm 1.5\%$	$\pm 1.4\%$
$\frac{\bar{\sigma}_f(^{237}\text{Np})}{\bar{\sigma}_f(^{235}\text{U})}$	0.381 $\pm 2.8\%$	$\pm 0.5\%$	$\pm 1.6\%$	$\pm 2.2\%$

TABLE NBS-21
FISSION CHAMBER MONITOR FOR $\Sigma\Sigma$

DATE	NBS Fission Chamber Relative to Thermal Column Fission Chamber Monitor					
	25S-2-3 rel.		28HD-5-1 rel.		28N-5-2 rel.	
June 21, 1974	1.945	1.011				
Sept. 30, 1974	1.945	1.011	0.1077	1.009		
Oct. 29, 1974	1.918	0.997	0.1070	1.003		
Oct. 30, 1974					0.1058	1.007
Nov. 12, 1974					0.1044	0.993
Nov. 18, 1974	1.887	0.981	0.1050	0.984		
Jan. 15, 1975	1.913	0.995	0.1073	1.006		
Feb. 18, 1975			0.1063	0.996		
May 7, 1975	1.928	1.003				
Average	1.923 $\pm$		0.1067 $\pm$		0.1051 $\pm$	

TABLE NBS-22
FISSION RATE CALIBRATION FACTOR FOR $\Sigma\Sigma$ INDIUM MONITOR

Isotope	Nominal Fiss. Rate During Calibration $\times 10^{16}$	Calibration Factor: [(Fiss./sec., nucleus) ÷ (In reaction/sec., nucleus)]					
		Value	(a) External Precision	(b) Internal Precision (Fiss. Chbr. Only)	(c) Systematic Errors		Total Error
					Corr. to Fiss. Rate	Deposit Mass	
^{235}U	4	27.01	$\pm 0.4\%$ ($\pm 0.14\%$)	$\pm 0.2\%$	$\pm 0.8\%$	$\pm 1.4\%$	$\pm 1.7\%$
^{239}Pu	4	31.8	$\pm 0.5\%$ ($\pm 0.25\%$)	$\pm 0.4\%$	$\pm 0.7\%$	$\pm 1.4\%$	$\pm 1.7\%$
^{238}U	0.2	1.524	$\pm 0.4\%$ ($\pm 0.2\%$)	$\pm 1\%$	$\pm 1.8\%$	$\pm 1.4\%$	$\pm 2.5\%$
^{237}Np	1.4	10.27	$\pm 0.4\%$ ($\pm 0.3\%$)	$\pm 0.5\%$	$\pm 1.9\%$	$\pm 1.8\%$	$\pm 2.7\%$

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All errors are one standard deviation.

- (a) Precision for array of (Fiss. Rate/In) values for all irradiations. Std. dev. of the mean is given in parenthesis.
- (b) Average of statistical precision for each irradiation as listed in the last column of Table NBS-19.
- (c) Values listed in Tables NBS-18, and NBS-17.

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J. W. Rogers

Argonne National Laboratory: R. J. Armani, R. R. Heinrich, R. J. Papek

Atomics International: Harry Farrar IV

Brookhaven National Laboratory: B. A. Magurno, S. Pearlstein

Center D'Etude De L'Energie Nucleaire (CEN/SCK): A. Fabray (currently at HEDL)

Exxon: N. D. Dudey

Los Alamos Scientific Laboratory: D. Barr, G. E. Hansen

National Bureau of Standards: D. M. Gilliam, J. A. Grundl

OTHER ILRR PROGRAM PARTICIPANTS

Atlantic Richfield Hanford Company: W. H. Zimmer

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