LMFBR REACTION RATE AND
DOSIMETRY 8th PROGRESS REPORT

FOR PERIOD MARCH
THROUGH SEPTEMBER 1973

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ABSTRACT

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

ACKNOWLEDGEMENTS

The following organizations are presently participating in the Interlaboratory LMFBR Reaction Rate (ILRR) Program.

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

*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.

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)

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SUMMARY

AEROJET NUCLEAR COMPANY

Descriptions of the ILRR-CFRMF tests conducted during this reporting period are documented. Measurements and analyses are presented for the reaction rates from the power level normalization foils of these tests. The reaction rates are reported from the self-shielding test foils counted at ANC. Additional fission product activities from the high purity fission foils have been analyzed and are reported.

ARGONNE NATIONAL LABORATORY

An analysis of the energy sensitivity of the fission products ^{140}Ba , ^{95}Zr , and ^{103}Ru to changes in neutron energy during fission of ^{235}U and ^{239}Pu is presented. It is demonstrated that only the yield of ^{103}Ru in fission of ^{235}U shows a statistically significant energy dependence. Finalized fission-product production-rates from the CFRMF Tests γ and VI and the FTR-EMC Test are presented together with their evaluated yields. Preliminary fission rate measurements in FTR-EMC by both fission chambers and track recorders are discussed. A thorough discussion of the errors associated with track recorder fission-rate measurements is presented. It is demonstrated that the track recorder method is capable of measuring fission rates to an absolute accuracy of $\pm 2\%$ at the 95% confidence level. Tentative results of SSTR fission rate measurements in the second CFRMF series of measurements are presented and discussed.

ATLANTIC RICHFIELD HANFORD COMPANY

Measurement constants, activity analyses, and reaction rates are reported for the HEDL-V, HEDL-VI, the self-shielding foil sets, and other FY-73 ILRR program irradiations. Gamma self-absorption corrections and mutually agreed upon isotopic half-life and gamma branching ratios have been applied. A previous ARHCO contribution on counting of the T 913-5 Ba-140 standard is presented. This contribution was inadvertently omitted from the 3rd Quarterly Report.

ATOMICS INTERNATIONAL

Recent measurements of the helium generated in small specimens of enriched ^{10}B that were encapsulated in vanadium and irradiated in EBR-II have given results that are consistently $\sim 16\%$ higher than calculated values. By comparison, specimens that were not enclosed in miniature vanadium capsules showed evidence of having lost by diffusion $\sim 30\%$ of the helium generated. This would explain the relatively low helium values from the unencapsulated specimens irradiated in CFRMF. A new CFRMF irradiation experiment with encapsulated specimens of ^{10}B and ^6Li is now in progress.

BROOKHAVEN NATIONAL LABORATORY

To complete the comparison of all isotopes presently common to both the ENDF/B and the SAND-II Dosimetry Libraries, the infinitely dilute resonance integrals for nine isotopes (ten reactions) were calculated.

The evaluations for a number of isotopic reactions, unavailable in either of the Dosimetry libraries, were recommended by the Normalization and Standards Subcommittee of CSEWG. One such recommendation was for ^{55}Mn ($n,2n$) ^{54}Mn . A preliminary version of the evaluation is included in this report.

HANFORD ENGINEERING DEVELOPMENT LABORATORY

Quality Assurance work has been initiated at HEDL on high purity foils of ^{235}U , ^{239}Pu , ^{237}Np , and ^{238}U from the HEDL-V foil set. Two additional high purity ^{237}Np foils were sent to ORNL for QA work. The results of this QA work will provide a check and confirmation of earlier ORNL results reported in the ORNL chapter of this report.

Documentation is provided on test identification and radiochemistry foil set selection, fabrication, and assembly for the scheduled FY-74 CFRMF, NBS-Thermal Cavity, NBS-Fission Cavity, BIG-10, and $\Sigma\Sigma$ irradiations. Results of FY-73 ILRR tests related to run-to-run power level normalization, fission product generation rates, measured foil perturbation effects, and neutron self-shielding in non-threshold detectors are documented and discussed.

Run-to-run power level normalization factors are now being obtained by NBS fission chamber and Au and In normalization foil measurements that are

accurate to within ~ 0.5 to 1% and 1 to 2%, respectively.

The experimental results obtained in CFRMF and $\Sigma\Sigma$ indicate that neutron self-shielding in relatively thin foils (less than ~ 10 mils thick) for broad spectrum reactions is significant and must be obtained experimentally for each ILRR test environment. Self-shielding correction factors are provided for correcting previously reported finite foil reaction rates to infinitely dilute values.

Calculated fission rates for correcting for fission of isotopic impurities in the ORNL high purity fission foils are presented for review, recommended changes, and acceptance by participating laboratories.

NATIONAL BUREAU OF STANDARDS

Additional fission rate results are reported for the CFRMF irradiations performed in March 1973. Based on the double fission chamber results, relative fluxes at the center of CFRMF corresponding to reactor power levels of 0.6KW: 6KW:10.4KW are found to be $(1.000):(9.66 \pm 0.03):(17.07 \pm 0.07)$. The central flux itself was observed to be $(1.1 \pm 0.3)\%$ higher in March 1973, than in November 1972.

OAK RIDGE NATIONAL LABORATORY

Archive samples of all batches of ILRR fissile dosimeter isotopes have been prepared, analyzed, and stored. Redundant analyses of dosimeter contents are being made to determine the maximum error involved in reporting the weight of isotope contained. Some results with ^{235}U (oxide) loadings would indicate a maximum error of 2%, but this error can be further reduced. Thin film deposits of $^{235}\text{U}(\text{O}_2)$ and $^{239}\text{Pu}(\text{O}_2)$ were prepared for NBS fission rate experiments and were carefully characterized to determine the content of nuclides and the uniformity of the film deposits.

AEROJET NUCLEAR COMPANY

A. CFRMF ILRR PROGRAM IRRADIATIONS SCHEDULE

J. W. Rogers and Y. D. Harker

Objective

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

Summary

The schedules for all previous irradiations have been summarized in the previous progress reports. Five irradiations were completed during this reporting period and others are planned. The irradiations to investigate self-shielding effects have been made in addition to more solid-state track-recorder irradiations.

Accomplishments and Status

Irradiations completed during this reporting period are summarized in Table ANC-1.

TABLE ANC-1
ILRR-CFRMF IRRADIATIONS

<u>Run Designation</u>	<u>Date</u>	<u>Power</u>	<u>Time</u>
ANL-SSTR Packet 7	3/7/73	~0.6 kW	1 hr.
ANL-SSTR Packet 8	3/7/73	~0.6 kW	1 hr.
ANC-HEDL-ARHCO SS-I	3/13&14/73	~10 kW	21.61 hrs.
HEDL-SS-II	3/14/73	~10 kW	1 hr.
HEDL-SS-III	3/14/73	~10 kW	0.9264 hr.

Approximately 195 hours of CFRMF operating time have been used in support of the ILRR program.

Expected Achievements

Continuing schedules of CFRMF irradiations will be made in support of the ILRR program.

B. CFRMF-ILRR IRRADIATIONS DESCRIPTIONS

J. W. Rogers

Objective

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

Summary

Descriptions of all previous CFRMF-ILRR program tests have been presented in the previous progress reports. Descriptions of the five irradiations conducted during this reporting period are presented.

Accomplishments and Status

Prior to the tests conducted during this reporting period, the CFRMF was operated at ~10 kW for 30 days. Also, the CFRMF was used for reactivity measurements which involved the removal of the servo chamber positioning device. The servo chamber and its positioning device were returned to their respective positions as accurately as possible.

The following is a test by test description of the conditions of those irradiations performed during this reporting period.

1. ANL-SSTR-Packet 7 (3/7/73)

Power Level Setpoint - 522 x 100 (0.6 kW)

Shim Position - 03333

Time In - 05:39:09 MST

Time Out - 06:39:09 MST

Total Exposure - 1 hour

Power Level #1 - 5.33

Power Level #2 - 5.04

Auxiliary Fission Chamber - 0.1335

Immediately before this test, NBS fission chamber measurements were made. During the accumulation of the fission chamber data of this and the following tests, intermittent noise was observed from sources which could not be located. To minimize this noise being included in the data, the pulses were observed with an oscilloscope and when a burst of noise was seen in a set of data, the data were rejected. The foil containment and positioning

arrangement were identical to the ANL-SSTR-Packet 6 (11/8/72) test except that the chamber contained only the ANL-SSTR-Packet 7. No power level normalization foils were irradiated during this test. The ANL-SSTR-Packet 7 was sent to ANL.

2. ANL-SSTR-Packet 8 (3/7/73)

Power Level Setpoint - 522 x 100 (0.6 kW)

Shim Position - 03333

Time In - 06:55:40 MST

Time Out - 07:55:40 MST

Total Exposure - 1 hour

Power Level #1 - 5.33

Power Level #2 - 5.04

Auxiliary Fission Chamber - 0.1335

The foil containment and positioning arrangement were identical to the ANL-SSTR-Packet 7 (3/7/73) test except that the chamber contained only the ANL-SSTR-Packet 8. Immediately after this test, NBS fission chamber measurements were made. Following this irradiation, two of the power level normalization foils (Au-CG and In-2#27) were retained at ANC and the other two were sent to HEDL. The ANL-SSTR-Packet 8 was sent to ANL and the fission chamber data were sent to NBS.

3. ANC-SS-I, HEDL-SS-I and ARHCO-SS-I (3/13 & 14/73)

Power Level Setpoint - 904 x 1000 (~10 kW)

Shim Position - 03333 (250) and 03670 (281)

Time In - 03:08:16 MST on 3/13/73

Time Out - 00:44:55 MST on 3/14/73

Total Exposure - 21 hours 36 min. 39 sec.

Power Level #1 - 94.5

Power Level #2 - 89.5

Auxiliary Fission Chamber - 2.07 → 2.10

Prior to this test the auxiliary fission chamber was moved when some required maintenance was performed and the chamber was replaced in its previous location. As this was the first time the CFRMF had been operated at the ~10 kW level since installing the servo chamber positioning device, the auxiliary fission chamber was used to establish the power level setpoint.

Assuming the auxiliary fission chamber was properly repositioned and functioning normally, the setpoint was raised (to 904 x 1000) until the output of the auxiliary fission chamber was the same as it was in the previous ~10 kW tests (~2.08). NBS fission chamber measurements were made prior to this test. The foil containment and positioning arrangement were identical to the HEDL-VI (11/7/72) test except that the chamber contained the HEDL-SS-I foil set, the ANC-SS-I set was attached to the top of the chamber and the ARHCO-SS-I foil set was attached to the bottom of the chamber. (See Figure HEDL-1, this report.) The foil sets were attached to the outside of the chamber with 15 mil aluminum wire supplied by HEDL.

During this test, the servo controlled regulating rod reached its limit at 16:25:00 MST causing a slight power reduction for a period of 11.5 minutes. This was recorded by the power level #1 monitor system. Assuming the steady state power level to be 94.5 power level-units, there would have been 9.352×10^6 power level-unit-seconds had the power reduction not occurred. By integrating over the reduction a loss of 5.744×10^3 power level-unit-seconds is obtained. This is a net power loss of 0.0781 per cent. To bring the CFRMF back to full power it was necessary to withdraw the shim rod from the 03333 position to a new position, 03670, which corresponds to a 0.558 degree-of-rotation movement or a reactivity of $6 \times 10^{-4} \Delta k/k$. This test ended prematurely due to an inadvertent scram and was not restarted because adequate irradiation was obtained. Following this irradiation, two of the power level normalization foils (Au-CI and In-2#29) and the ANC-SS-I foil set were retained at ANC. The other two power level normalization foils, the ARHCO-SS-I foil set and the HEDL-SS-I foil set were sent to HEDL. The fission chamber data were sent to NBS.

4. HEDL-SS-II (3/14/73)

Power Level Setpoint - 904 x 1000 (~10 kW)

Shim Position - 03670 (281)

Time In - 01:40:18 MST on 3/14/73

Time Out - 02:40:18

Total Exposure - 1 hour

Power Level #1 - 94.0

Power Level #2 - 91.8

Auxiliary Fission Chamber - 2.10

The foil containment and positioning arrangement were identical to the ANC-SS-I, HEDL-SS-I and ARHCO-SS-I (3/13 & 14/73) test except that the chamber contained only the HEDL-SS-I foil set. Following this irradiation, two of the power level normalization foils (Au-DA and In-2#31) were retained at ANC and the other two were sent to HEDL with the HEDL-SS-II foil set.

5. HEDL-SS-III (3/14/73)

Power Level Setpoint - 904 x 1000 (~10 kW)

Shim Position - 03670 (281)

Time In - 03:27:10 MST on 3/14/73

Time Out - 04:22:45

Total Exposure - 55 minutes 35 seconds

Power Level #1 - 94.0

Power Level #2 - 91.8

Auxiliary Fission Chamber - 2.10

The foil containment and positioning arrangement were identical to the HEDL-SS-II (3/14/73) test except that the chamber contained only the HEDL-SS-III foil set. This test ended prematurely due to an inadvertent scram and was not restarted because adequate irradiation had been obtained. Following this irradiation, two of the power level normalization foils (Au-DC and In-2#33) were retained at ANC and the other two were sent to HEDL with the HEDL-SS-III foil set. The CFRMF was restarted and some additional NBS fission chamber measurements were taken and these data were sent to NBS.

Expected Achievements

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

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

D. A. Millsap, Y. D. Harker and J. W. Rogers

Objective

•To meet ILRR program objectives, an accurate characterization of the neutron environment in the CFRMF test region must be available. Important parameters are the neutron energy spectrum and flux gradient around the central test region. The best possible information on the parameters will be obtained and disseminated.

Summary

Previously determined CFRMF neutron environment characterization information has been disseminated in the previous progress reports. More recently, a transport calculation of CFRMF central fluxes has been completed and is presented here.

Accomplishments and Status

The central neutron energy spectrum of the CFRMF has been calculated using a one dimensional transport theory code in P1-S6 approximation where the reactor configuration was represented as a twenty region cell. These fluxes are listed in Table ANC-2. The cross-sections used in this calculation were obtained from the ENDF-B Version III evaluated set.

Expected Achievements

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

TABLE ANC-2

CFRMF Central Fluxes as Calculated by SCAMP

One Dimensional Transport, P1-S6 Approximation, Twenty Region Cell Calculation

SCAMP 73/008

<u>Group Number</u>	<u>Lower Lethargy</u>	<u>Lower Energy (ev)</u>	<u>Real Flux $\phi(u)\Delta u$</u>
1	0.25	7.79 x 10 ⁶	0.015915
2	0.50	6.07	0.047192
3	0.75	4.72	0.099282
4	1.00	3.68	0.15826
5	1.25	2.87	0.22956
6	1.50	2.23	0.30044
7	1.75	1.74	0.32838
8	2.00	1.35	0.37165
9	2.25	1.05	0.48196
10	2.50	8.21 x 10 ⁵	0.65123
11	2.75	6.39	0.87768
12	3.00	4.98	1.00
13	3.25	3.88	0.96504
14	3.50	3.02	0.97404
15	3.75	2.35	0.87086
16	4.00	1.83	0.73469
17	4.25	1.43	0.59642
18	4.50	1.11	0.53875
19	4.75	8.65 x 10 ⁴	0.41965
20	5.00	6.74	0.39890
21	5.25	5.25	0.27323
22	5.50	4.09	0.22890
23	5.75	3.18	0.15576
24	6.00	2.48	0.15655
25	6.25	1.93	0.11410
26	6.50	1.50	0.080494
27	6.75	1.17	0.077870
28	7.00	9.12 x 10 ³	0.065567
29	7.25	7.10	0.051764
30	7.50	5.53	0.044402
31	7.75	4.31	0.042918
32	8.00	3.36	0.043315
33	8.25	2.61	0.034658
34	8.50	2.04	0.031480
35	8.75	1.59	0.033850

TABLE ANC-2 (Continued)

<u>Group Number</u>	<u>Lower Lethargy</u>	<u>Lower Energy (ev)</u>	<u>Real Flux $\phi(u)\Delta u$</u>
36	9.00	1.23×10^3	0.027442
37	9.25	961.1	0.020435
38	9.50	748.5	0.020955
39	9.75	582.9	0.015951
40	10.00	454.0	0.015354
41	10.25	353.6	0.010261
42	10.50	275.4	8.7379×10^{-3}
43	10.75	214.5	6.5036×10^{-3}
44	11.00	167.0	4.9708×10^{-3}
45	11.25	130.1	4.9375×10^{-3}
46	11.50	101.3	1.7187×10^{-3}
47	11.75	78.9	2.6564×10^{-3}
48	12.00	61.4	1.1628×10^{-3}
49	12.25	47.9	1.3074×10^{-3}
50	12.50	37.3	5.1720×10^{-4}
51	12.75	29.0	3.3158×10^{-4}
52	13.00	22.6	2.8618×10^{-4}
53	13.25	17.6	4.8196×10^{-5}
54	13.50	13.7	1.1940×10^{-4}
55	13.75	10.68	5.6173×10^{-5}
56	14.00	8.32	1.6846×10^{-5}
57	14.25	6.48	1.3368×10^{-6}
58	14.50	5.04	8.9927×10^{-7}
59	14.75	3.93	9.4233×10^{-7}
60	15.00	3.06	2.9062×10^{-7}
61	15.25	2.38	8.4601×10^{-8}
62	15.50	1.86	1.6601×10^{-8}
63	15.75	1.44	2.8681×10^{-9}
64	16.00	1.125	3.7852×10^{-10}
65	16.25	0.876	7.1949×10^{-11}
66	16.50	0.683	3.5829×10^{-11}
67	16.75	0.532	2.1580×10^{-12}
68	17.00	0.414	1.3690×10^{-13}
69	∞	0.0	8.2748×10^{-15}

D. CFRMF REACTION RATES FROM POWER LEVEL MONITOR FOILS

J. W. Rogers

Objective

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

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

Summary

The activities of the power level monitor foils from the most recent ILRR-CFRMF program tests have been measured using NaI(Tl) gamma-ray counting techniques.

Accomplishments and Status

The techniques for obtaining the monitor foil activities from which the saturated reaction rates are computed have been described in the previous progress reports. The same techniques were followed in these measurements and the same decay scheme data were used in the computations. Foils from the 0.6 kW test and three ~10 kW tests were counted and the results are tabulated in Table ANC-3.

These results from the ANL SSTR Packet 8 test show an inconsistency between the Au and In reactions of about 2% which is somewhat more than expected from previous experience.

The results from the self-shielding tests are consistent and indicate that all three of these tests were at the same power level to within $\pm 0.5\%$. This power level is 17.06 times the reference 0.6 kW level which is significantly higher than the previous high power runs (15.47 times the 0.6 kW level). This indicates there is some discrepancy between the CFRMF power level control system and the auxiliary fission chamber for the conditions of these tests since by using the output of the auxiliary fission chamber as a reference (see section ANC-B this report), a different reaction rate was obtained.

Expected Achievements

Foils from all future tests will be measured and analyzed.

TABLE ANC-3

CFRMP Power Level Monitor Foil Reaction Rates

<u>Test Identity</u>	<u>Foil</u>	<u>Mass^a</u>	<u>Saturated Reaction^b Rate by NaI(Tl)</u>	<u>Normalization^c Factor</u>	<u>Spectral Index^d</u>
ANL SSTR Packet 8	Au-CG	0.1201	$3.188 \pm 0.017 \times 10^{-15}$	1.020	8.00
	In-2# 27	0.04672	$3.984 \pm 0.024 \times 10^{-16}$	1.004	
ANC-HEDL-ARHCO SS-I	Au-CI	0.1211	$5.329 \pm 0.008 \times 10^{-14}$	17.05	7.95
	In-2# 29	0.04880	$6.705 \pm 0.051 \times 10^{-15}$	16.89	
HEDL-SS-II	Au-DA	0.1236	$5.333 \pm 0.011 \times 10^{-14}$	17.06	7.85
	In-2# 31	0.04887	$6.793 \pm 0.028 \times 10^{-15}$	17.11	
HEDL-SS-III	Au-DC	0.1218	$5.358 \pm 0.014 \times 10^{-14}$	17.14	7.89
	In-2# 33	0.04988	$6.787 \pm 0.024 \times 10^{-15}$	17.10	

^a Determined by weighing at ANC.

^b These uncertainties are the estimated standard deviation (1σ) on span of measurements (does not include uncertainties on efficiencies).

^c Relative to the previous 0.6 kW test values of 3.125×10^{-15} for $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ and 3.970×10^{-16} for $^{115}\text{In}(n,n')^{115\text{m}}\text{In}$.

^d Defined as $^{197}\text{Au}(n,\gamma)^{198}\text{Au}/^{115}\text{In}(n,n')^{115\text{m}}\text{In}$.

E. CFRMF POWER LEVELS

R. G. Helmer and R. C. Greenwood

Objective

•To determine the relative power levels for several CFRMF irradiations by means of the saturated activities of the gold and indium monitor foils.

Summary

Gold and indium monitor foils for the CFRMF experiments ANL-SSTR-8, ANC-SS-I (also known as HEDL-SS-I and ARHCO-SS-I), HEDL-SS-II and HEDL-SS-III were counted using the OR-5 Ge(Li) detector. The power levels of the irradiations ANL-SSTR-4, 5, 6 and 8 are found to have a spread of about 1.2% with uncertainties in the ratios of about 0.6%. The total range of power levels for the neutron self-shielding experiments is also about 1.0%.

Accomplishments and Status

The γ -ray spectra from the gold and indium monitor foils irradiated in the CFRMF experiment ANL-SSTR-8 were measured on the OR-5 Ge(Li) detector. Saturated activities for the ^{198}Au and $^{115\text{m}}\text{In}$ were determined by the methods described in section ANC-D of HEDL-TME-72-129 and the uncertainties estimated as discussed in section ANC-G of HEDL-TME-72-129. The results are given in Table ANC-4 along with those of three previous solid-state track recorder irradiations. As shown, the relative power level shows a spread of only 1.2% with an uncertainty of about 0.6%.

Similar measurements were made for the monitor foils of the first three neutron self-shielding experiments. These results are also given in Table ANC-4. Here the power variation is about 1%.

Expected Achievements

The gold and indium monitor foils from future CFRMF irradiations will be counted.

TABLE ANC-4

SATURATED ACTIVITIES FOR MONITOR FOILS*

Experiment	Foil	Mass (gm)	Number of Measurements	Saturated Activity (10^{-16} dis/sec-atom)	Relative Saturated Activity		Relative Power Level
					Au	In	
ANL-SSTR-4	Au-CA	0.1252	2	31.74 $\pm$ 0.51	1.008 $\pm$ 0.005		1.008 $\pm$ 0.004
	In-2 #21	0.04700	1	4.000 $\pm$ 0.182		1.009 $\pm$ 0.008	
ANL-SSTR-5	Au-CC	0.1223	2	31.66 $\pm$ 0.51	1.006 $\pm$ 0.005		1.000 $\pm$ 0.004
	In-2 #23	0.04945	1	3.935 $\pm$ 0.182		0.992 $\pm$ 0.008	
ANL-SSTR-6	Au-CE	0.1237	2	31.48 $\pm$ 0.51	1.000 $\pm$ 0.005		1.000 $\pm$ 0.004
	In-2 #25	0.04814	1	3.965 $\pm$ 0.182		1.000 $\pm$ 0.008	
ANL-SSTR-8	Au-CG	0.1201	1	31.72 $\pm$ 0.51	1.008 $\pm$ 0.006		1.012 $\pm$ 0.005
	In-2 #27	0.04672	1	4.041 $\pm$ 0.19		1.019 $\pm$ 0.009	
ANC-SS-I	Au-CI	0.1211	3	537.4 $\pm$ 8.4	0.997 $\pm$ 0.005		0.995 $\pm$ 0.004
HEDL-SS-I	In-2 #29	0.04880	2	68.14 $\pm$ 3.3		0.993 $\pm$ 0.006	
ARHCO-SS-I							
HEDL-SS-II	Au-DA	0.1236	2	539.1 $\pm$ 8.4	1.000 $\pm$ 0.004		1.000 $\pm$ 0.003
	In-2 #31	0.04887	1	68.62 $\pm$ 3.2		1.000 $\pm$ 0.006	
HEDL-SS-III	Au-DC	0.1218	2	544.2 $\pm$ 8.5	1.009 $\pm$ 0.004		1.006 $\pm$ 0.003
	In-2 #33	0.04988	2	68.86 $\pm$ 3.2		1.003 $\pm$ 0.006	

* Table includes data from HEDL-TME 73-31 for previous ANS-SSTR experiments.

F. SATURATED ACTIVITIES FOR NEUTRON
SELF-SHIELDING EXPERIMENT ANC-SS-I

R. G. Helmer and R. C. Greenwood

Objective

•To provide saturated activity levels for thin foils irradiated in the CFRMF.

Summary

Thin foils (0.00005") of Cu, Au, Sc, Co, ^{235}U and ^{238}U were irradiated in the CFRMF as experiment ANC-SS-I. Similar packets of foils designated HEDL-SS-I and ARHCO-SS-I were irradiated at the same time. These data will be combined to give base information to determine the neutron self-shielding effect in thicker foils of the same material. The saturated activities have been determined generally with counting uncertainties of <2%. The uncertainties are given without the branching ratio contribution since that does not enter in the neutron self-shielding determination. (Within one laboratory, the detector efficiency also does not contribute.)

Accomplishments and Status

The γ -ray spectra from the foils in the experiment ANC-SS-I were counted with the OR-5 Ge(Li) detector. These foils are 0.00005" thick and along with others will provide base information for the neutron self shielding in thicker foils of the same material. The results of these measurements are given in Table ANC-5. Due to the low activity level the counting durations were long (i.e., ~64 hr for Sc and ~80 hr for the Co). Also the Co foil was counted at 5 cm, instead of the usual 10 cm, to enhance its counting rate relative to the ^{60}Co in the background. A separate measurement of the relative efficiency at 5 and 10 cm for the 1173- and 1332-keV lines was made.

The total uncertainty in the saturated activities is given (see section ANC-G of HEDL-TME-72-129 for contributions included) for general use. However, for neutron self-shielding measurements, the error in the branching ratio should not be included; so this value (in %) is also given. Also for sets of results within one laboratory the uncertainty in the detector efficiency can also be excluded.

Expected Achievements

The data for the ^{235}U and Sc foils in packets HEDL-SS-I, II and III will be analyzed.

TABLE ANC-5

SATURATED ACTIVITIES FROM FOILS IN EXPERIMENT ANC-SS-I

All measurements were made with the OR-5 Ge(Li) detector at 10 cm except where noted and with no absorbers.

Foil	Mass (gm)	Activity	Number of Measure- ments	Gamma-Ray Energy (keV)	Correction Factors			Saturated Activity ^b (10 ⁻¹⁵ dis/sec-atom)		Errors (%)	
					γ -Ray ^a Absorption	Coincidence Summing	Random Summing	Average for γ -Ray	Average for foil	total	without branching
Cu-1	0.001490	⁶⁴ Cu	2	511	0.9814	1.000	0.9995-0.9996		5.77±0.26	4.6	1.6
Au-1	0.006918	¹⁹⁸ Au	2	411	0.9997	1.000	0.9927-0.9973		60.3 ±0.9	1.55	1.5
Sc-1	0.000480	⁴⁶ Sc	2	889	0.9997	0.993	0.9998		3.20±0.05	1.64	1.6
Co-1	0.001645	⁶⁰ Co	1 ^c	1173	0.99997	0.9826	0.9999	12.99	13.02	2.0	2.0
				1332	0.99997	0.9826	0.9999	13.06			
U8-1	0.002585	²³⁹ Np	2	277	0.9988	0.9997	0.9986-0.9994		23.6 ±2.5	10.7	2.9
U5-1	0.003100	¹³² Te	2	228	0.9983	0.9974	0.9954-0.9990		10.1 ±0.7	7.0	1.7
		¹⁰³ Ru	2	497	0.9995	1.000	0.9990-0.9996		6.94±0.26	3.7	1.6
		⁹⁵ Zr	2	756	0.9996	1.000	0.9990-0.9996		14.04±0.26	1.9	1.6
		¹⁴⁰ Ba	2	1596	0.9997	0.9934	0.9990-0.9996		13.08±0.21	1.6	1.6

^aThese values include the absorption in the aluminum covers on the Sc, U8 and U5 foils and the foil used to stop the positrons from the ⁶⁴Cu activity.

^bThe calculation of the U8 and U5 activities assumes the same enrichment as in the high purity 5-mil foils.

^cCounted at 5 cm.

G. SELF-SHIELDING OF REACTIONS IN CFRMF

J. W. Rogers

Objective

• To establish self-shielding effects of certain reactions in individual foils irradiated in the neutron field of CFRMF for ILRR tests. These effects will be used to form necessary self-shielding correction factors in the ILRR program.

Summary

Reaction rates have been obtained from the ANC-SS-I, HEDL-SS-I, II and III foil sets irradiated in the CFRMF.

Accomplishments and Status

The activities of the foils in the ANC-SS-I foil set have been measured using NaI(Tl) gamma-ray counting techniques where possible. The counting techniques used have been described in previous progress reports. Using these activities and the decay scheme data found in section ANC-J of HEDL-TME-72-90, the reaction rates of interest were computed. These results are summarized in Table ANC-6.

TABLE ANC-6

ANC-SS-I FOIL REACTION RATES

<u>Foil</u>	<u>Target Mass (mg)</u>	<u>Reaction</u>	<u>Saturated Reaction^a Rate by NaI(Tl)</u>
Au #1	6.918	$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	$5.979 \pm 0.020 \times 10^{-14}$
Co #1	1.645	$^{59}\text{Co}(n,\gamma)^{60}\text{Co}$	$1.408 \pm 0.012 \times 10^{-14}$
Cu #1	1.02	$^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$	$5.655 \pm 0.058 \times 10^{-15}$
^{235}U #1	3.100	$^{235}\text{U}(n,f)^{140}\text{Ba}$	$2.325 \pm 0.010 \times 10^{-13}{}^b$
Sc #1	0.480	$^{45}\text{Sc}(n,\gamma)^{46}\text{Sc}$	$3.171 \pm 0.034 \times 10^{-15}$

^aThese uncertainties are the estimated standard deviation (1 σ) on span of measurements (does not include uncertainties on efficiencies).

^bAssuming a 5.8% yield.

These foils are all 0.05 mil thick and 0.5" diameter. The Au and Co were counted bare and the Sc and ^{235}U were counted through the ~8 mil Al wrapping. The Cu foil was sandwiched between 10 mils of unirradiated pure Cu to annihilate all the positrons. ORNL material weights were used to compute the reaction rates for all materials except the Au where the HEDL micro-balance weight was used. The ^{238}U was not counted on the NaI(Tl) system because of the resolution and spectrum complexity. All corrections for gamma-ray attenuation in the foils, cladding and absorbers have been made.

The 0.05, 5 and 10 mil foils of Sc and ^{235}U from HEDL-SS-I, II and III tests were also counted at ANC. These results and comparable ANC foil results are tabulated in Table ANC-7. These results show no self-shielding effects in the $^{235}\text{U}(n,f)^{140}\text{Ba}$ reaction and good agreement is obtained between the 0.05 mil foils of the ANC and HEDL foils. For the $^{45}\text{Sc}(n,\gamma)^{46}\text{Sc}$ reaction there may be about 2% self-shielding in the 10 mil foil. There is disagreement between the ANC and HEDL 0.05 mil Sc foils in excess of the uncertainties (1σ) assuming the masses of these foils are correct.

TABLE ANC-7

REACTION RATES OF HEDL AND ANC SELF-SHIELDING FOILS

<u>Thickness</u>	<u>$^{235}\text{U}(n,f)^{140}\text{Ba}(5.8\%)$ Reaction</u>	<u>$^{45}\text{Sc}(n,\gamma)^{46}\text{Sc}$ Reaction</u>
	HEDL	HEDL
0.05 mil	$2.312 \pm 0.04 \times 10^{-13}$	$3.307 \pm 0.04 \times 10^{-15}$
5 mils	$2.346 \pm 0.02 \times 10^{-13}$	$3.275 \pm 0.026 \times 10^{-15}$
10 mils	$2.337 \pm 0.02 \times 10^{-13}$	$3.246 \pm 0.044 \times 10^{-15}$
	ANC	ANC
0.05 mil	$2.325 \pm 0.01 \times 10^{-13}$	$3.171 \pm 0.034 \times 10^{-15}$

Expected Achievements

Any further measurements or analysis deemed necessary after comparison of results from other laboratories will be made and reported.

H. FISSION PRODUCT ACTIVITIES MEASURED FOR IRRADIATIONS IN CFRMF

R. C. Greenwood and R. G. Helmer

Objective

- Obtain fission product decay rates, as a means of determining fission rates, from sets of high purity fission foils irradiated in CFRMF.

Summary

Analysis of the data obtained with the ^{237}Np foil in the HEDL-V experiment was completed and the saturation activity levels for ^{140}Ba , ^{95}Zr and ^{103}Ru were determined. (See section ANC-A of HEDL-TME 73-31 for other results obtained with HEDL-V and HEDL-VI.)

Accomplishments and Status

The analysis of the fission product data for the ^{237}Np foil irradiated in CFRMF as a part of the HEDL V experiment (together with ^{235}U , ^{238}U and ^{238}Pu foils) has been completed and the saturated activities which were determined for several of the resultant fission products are summarized in Table ANC-8. The relative yields of the ^{140}Ba , ^{95}Zr and ^{103}Ru fission products resulting from this irradiation are shown in Table ANC-9 .

Expected Achievements

Fission foils from future tests will be measured and analyzed.

TABLE ANC-8
SATURATED ACTIVITIES FROM ^{237}Np FISSION FOILS IRRADIATED IN CFRMF
(All measurements were made with the OR-5 detector at 25 cm
and with no absorber. Two measurements were made for foil.)

Foil	Activity	Gamma Ray Energy (keV)	Correction Factors			Saturated Activity (10^{-15} dis/sec-atom)	
			γ Self Absorption ^a	Coincidence Summing	Random Summing	Average for Foil	Error in %
$^{237}\text{Np}^b$	^{140}Ba	1596	0.9885	0.9988	0.937-0.941	2.445 ± 0.045	1.9
N7-21	^{95}Zr	756	0.9779	1.000	0.937-0.941	2.61 ± 0.11	4.1
	^{103}Ru	496	0.9628	1.000	0.937-0.941	2.46 ± 0.10	4.0

^aThe γ self absorption term also contains the attenuation factors for the aluminum container.

^bHEDL-V irradiation.

TABLE ANC-9
RELATIVE FISSION PRODUCT YIELDS FOR ^{237}Np IRRADIATED IN CFRMF
IN THE HEDL-V EXPERIMENT

Target	Fission Product Activity	Relative Fission Product Yields
^{237}Np	^{140}Ba	1.000 ± 0.007
	^{95}Zr	1.067 ± 0.037
	^{103}Ru	1.006 ± 0.036

A. FISSION YIELDS AND THEIR RELATIONSHIP TO THE ILRR PROGRAM

N. D. Dudey and R. P. Larsen

OBJECTIVE

•To evaluate and maintain sets of recommended fission yields to be associated with the ILRR program. In particular, to ascertain a set of fission yields whose energy sensitivity (to LMFBR neutron spectra) is sufficiently low that they may be used for K-factor applications.

SUMMARY

An analysis of the energy sensitivity of the fission products ^{140}Ba , ^{95}Zr , and ^{103}Ru to changes in neutron energy during fission of ^{235}U and ^{239}Pu has been made. The analysis is based on regression and correlation techniques. The conclusion drawn is that within the accuracies of the available data only the yield of ^{103}Ru in fission of ^{235}U shows a statistical significant energy dependence. It is further concluded that fission yield data measured in reactor spectra are not capable of establishing energy sensitivities to the required accuracies of $\pm 2\%$ at the 95% confidence levels. Measurements in tailored neutron spectra which can be accurately measured, such as accelerator produced neutron spectra, may be capable of achieving the required accuracy.

ACCOMPLISHMENTS AND STATUS

In the last Quarterly Report (p. HEDL-17 of 7th Quarterly Progress Report, McElroy, Kellogg, and Matsumoto presented a discussion of neutron energy dependencies of fission yields. In the same report, we presented our recommended fission yields for ILRR use; however, we deferred the discussion of neutron energy dependence of the various yields. Because of the significance of this question, (to the K-factor technique, fuel burnup, fission rate measurement) and some of the implications raised by the HEDL discussion, we have decided to present our preliminary assessment of the situation. Our interpretation is somewhat different than the HEDL view.

Selection of Data. The fission yield data we have chosen to examine are limited to fission products of ^{235}U and ^{239}Pu . Much of the data for ^{235}U is the same as presented by HEDL with the following exceptions: We have excluded all reported fission yield data whose quoted errors are in excess of $\pm 8-10\%(1\sigma)$. We have excluded Meek and Rider's results because

they have compiled fission yield data from many different spectra and any energy dependence would be folded into their yields. We have included the ANL measurements that were systematically measured in ZPR-3 (differing EBR-II type spectra) specifically for the purpose of examining the question of energy dependence.

Despite the fact that some questions have been raised concerning the possibility of biases in the Lisman et al. data (see 7th Quarterly Report, p. ANL-1), their data are really the pivot data for several reasons: (1) They were measured in the hardest neutron spectra ($\bar{E} = 1.2$ MeV). (2) They have the smallest quoted errors. (3) They are the most comprehensive (complete) set of fission yields. Although we have chosen for the reasons stated in the 7th Quarterly to use the revised Lisman data for this particular analysis, it must be pointed out that in assessing energy dependence our conclusions would not be altered had we used the yield values published by Lisman.

In our evaluations we have included the CFRMF and EMC results of the ILRR program but, at present, these fission yields must be considered preliminary. Four laboratories have reported ^{140}Ba fission product rates from the HEDL-5 and 6 irradiations. We have converted the reaction rate data to fission yields via the NBS fission chamber data. Similarly, with the EMC results, both ANL and ANC have reported fission product data, and from the fission chamber measurements of the fission rates we can extract yields. Thus, the fission yield values used for CFRMF are an average of four different laboratory measurements, and the EMC yields are an average of two laboratory measurements.

The fission yield results used in this study are summarized in Table ANL-1; the ^{140}Ba , ^{95}Zr , and ^{103}Ru yields are tabulated for ^{235}U and ^{239}Pu fission. These yield values are both preliminary and not necessarily complete with respect to published results. The intent is to provide an alternative approach to the assessment of the energy dependence of these fission yields. The average neutron energies shown in Table-ANL-1 are, for the most part, those reported by HEDL. The average energies for the ZPR-3, Assemblies 60 and 61, spectra are based on neutronics calculations.

TABLE ANL-1

Summary of Fission Yield Data Used on Energy Dependence Analysis

Experiment	Neutron Field	Ref.	$\bar{E}$ (MeV)	Fission Yields					
				^{235}U			^{239}Pu		
				^{140}Ba	^{95}Zr	^{103}Ru	^{140}Ba	^{95}Zr	^{103}Ru
1. ILRR - ANC	CFRMF			5.95	6.40	3.24	4.94	4.43	6.41
ANL	"			5.86	6.30	3.16	4.94	4.50	6.50
HEDL	"			5.97			5.12		
ARHCO	"			6.07			5.03		
Average	CFRMF	1	0.72	5.96	6.35	3.20	5.01	4.46	6.45
2. EMC - ANC	ZPR-9			6.00	6.45	3.16	5.28	4.94	6.99
ANL				5.73	6.31	3.00	5.19	-	6.91
Average		1	0.5	5.84	6.38	3.08	5.24	4.94	6.93
3. ZPR-3	60 C	2	0.85	5.75	6.49	3.36	5.19	-	7.03
Mockup of	61 C	2	0.81	5.61	6.46	3.28	5.31	4.78	7.11
EBR-II	61 I	2	0.43	5.65	6.27	3.22	5.26	4.76	7.01
4. Lisman et al.	EBR-I	3		6.21	6.47	3.50	5.59	4.78	6.87
Lisman (revised)		1	1.2	6.00	6.25	3.3	5.28	4.52	6.6
5. Ass. 57	ZPR-3	4	0.7	5.83			5.00	-	-
6. W. Schultyssek	SNEAK	5	0.54	5.84	6.19	3.11			
	7B		0.44	5.88	-	3.05			
7. Karlsruhe	STARK-5	6	1.0	6.1					
8. Cunnigham et al.	DFR	7	0.79	5.91			5.45		
9. Lisman et al.	Thermal	3	0.0	6.31	6.45		5.61	4.86	

ANL-3

Analysis. On the basis of the fission yields presented in Table ANL-1 we can perform two statistical tests to establish the significance of an energy dependence. The first test is a correlation analysis in which the correlation coefficient R is compared to a critical value R_C . If R_C is computed on the basis of a 95% confidence level, then unless $R > R_C$ the data are not statistically significant, i.e., no energy correlation. Perhaps a better test is to examine R^2 which is equivalent to the percent of the data variance which is accounted for by a correlation between yield and energy. In both correlation tests we find no indication of an energy dependence that is statistically significant for ^{95}Zr and ^{140}Ba . For ^{103}Ru there is strong evidence for energy dependence in ^{235}U fission.

The second statistical test is a regression analysis, in which the equation $Y = a + bE$ is fit to the data. The results of this analysis are presented in Table ANL-2. The t statistic indicates the significance of the slope b (%-yield per MeV) from zero, namely, if $|t|$ exceeds ~ 2 , then the probability that the true slope, β , is not zero is large. An alternate approach is to compute the 95% confidence interval of β , where β is the true energy dependent coefficient which is estimated by b . The 95% confidence interval β for each of the fission yields is also given in Table ANL-2.

TABLE ANL-2
Summary of Statistical Parameters

Parameter	^{235}U		^{239}Pu	
	^{140}Ba	^{95}Zr	^{140}Ba	^{95}Zr
n	12	8	9	6
$\langle \text{FY} \rangle$	$5.89 \pm .19$	$6.36 \pm .12$	$5.26 \pm .19$	$4.72 \pm .19$
a	$5.97 \pm .14$	$6.41 \pm .092$	$5.41 \pm .142$	$4.91 \pm .127$
b	$-0.118 \pm .187$	$-0.0779 \pm .130$	$-.230 \pm .193$	$-.308 \pm .178$
t	-0.634	-0.601	-1.19	-2.39
t_0	1.812	1.943	1.895	1.943
R	.000	.000	.224	.534
R_C	.497	.621	.582	.729
β (95% interval)	-0.46, +0.22	-.33, +.18	-.60, -.14	-.62, +.00

Conclusions. From this analysis we conclude that no energy dependence of the ^{140}Ba and ^{95}Zr fission yields in fission of ^{235}U is statistically discernible within the available data. This analysis has covered the energy range of 0 to 1.2 MeV. For ^{239}Pu , there is also no statistical basis for concluding that there is a dependence of fission yield on neutron energy. It should be pointed out that there are less ^{239}Pu data for making this evaluation than there are for ^{235}U .

Having demonstrated a lack of energy dependence we can assess the accuracy of the fission yield data. At the 1σ confidence level we determine the fission yields to be accurate to 3.2% for ^{140}Ba and 1.9% for ^{95}Zr in ^{235}U fission, 3.6% for ^{140}Ba and 3.8% for ^{95}Zr in ^{239}Pu fission. This analysis also substantiates the yields we recommended in the last Quarterly Progress Report, and our previous conclusion that of the fission products available for dosimetry purposes, ^{95}Zr is the most reliable.

On physical grounds one can refute the possibility of a 10% per MeV increase in fission yields with increasing energy by the following argument: If the yields of ^{140}Ba , ^{137}Cs , and ^{148}Nd increase by 10% in going from say 0.2 $\bar{\text{E}}$ to 1.2 $\bar{\text{E}}$, then one of two possibilities exist. All of the mass yields on the heavy mass peak (133-148) increase with energy or the mass yield curve will have pronounced structure. The change in the relative yields of any two nuclides is known to be quite low ($\leq 3\%$) (Lisman data, etc.); hence, all peak mass yields would have to increase proportionately. Since these masses constitute approximately 80% of the heavy mass peak, the remaining 20% of the heavy mass peak would have to decrease from 20% to 12% of the peak. Since the remaining yields on the shoulders of the mass peak are small, a significant narrowing of the heavy mass peak would have to occur with increasing energy. The opposite behavior is a well established physical fact, namely, with increasing energy the valley fills in and the wings spread out. Hence, with increasing energy the peak yields can only decrease, not increase.

The objective of the ILRR program must be to establish the magnitude of this change for the fission products ^{95}Zr , ^{137}Cs , ^{140}Ba , and ^{148}Nd . If these yields are invariant within $\pm 1-2\%$ over the LMFBR neutron spectra,

then the K-factor approach is valid and furthermore, we can apply the CFRMF measured yields to EBR-II and FFTF. On the basis of existing data we can state that there is no justification to suspect an energy dependence for these fission products, but we cannot state with great certainty that the yields are constant to within $\pm 1-2\%$.

An additional complication to energy sensitivity analysis for these data resides in the energy parameter. Firstly, there is this uncertainty in all of these neutron spectra and secondly, it is not obvious that the average neutron energy is the appropriate parameter with which to study the correlation. Perhaps mean fission energy is a better parameter. Because of the magnitude of the uncertainties in both the yield and energy parameters, it is our opinion that energy dependencies cannot be established to the required accuracies of $\pm 2\%$ (at the 95% confidence level) using reactor spectra. We submit that accelerator produced neutron spectra are the only hope of achieving the required accuracy. This is because the neutron spectra can be measured both instrumentally (time-of-flight, proton-recoil) and by foil techniques and because the spectra can be tailored to maximize the sensitivity, e.g., average neutron energies of 0.3, 0.7, and 1.2 MeV could be utilized.

Once it is established that one fission yield is invariant with neutron energy, then other fission yields can be easily examined. The principal difficulty in determining absolute fission yields resides in establishing the absolute number of fissions. However, fission product (yield) ratios can avoid this problem. Hence, given that the ^{95}Zr yield is constant, then yield ratios relative to ^{95}Zr , e.g., $^{103}\text{Ru}/^{95}\text{Zr}$ can be very accurately measured and the energy dependence of the ^{103}Ru yield could be accurately defined. The ^{103}Ru yield data for ^{235}U in Table ANL-1 show a 10% change in yield per MeV. As more accurate yield ratios are established by the ILRR program, this energy dependence can be more accurately defined. Such analytical efforts should wait until much of the preliminary data are finalized.

EXPECTED ACHIEVEMENTS

Analysis and data testing of this type will be updated as additional data becomes available and as the preliminary data become finalized.

B. REACTION RATES BY FOIL ACTIVATION (EMC and CFRMF)

R. J. Popek and N. D. Dudey

OBJECTIVE

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

SUMMARY

Finalized fission-product production rates from the CFRMF Tests 5 and 6, and the EMC Tests are reported, together with their evaluated errors. Errors associated with fission-product measurements are discussed. Gamma-ray interferences in the EMC Test are presented and found to require as much as 2-4% corrections. Errors quoted by ILRR participants are considered to be optimistic.

ACCOMPLISHMENTS AND STATUS

Measurements of the fission products from foils of ^{235}U , ^{238}U , and ^{239}Pu irradiated in both EMC and CFRMF (Tests 5 and 6) have been completed. Some of these results were reported previously as preliminary data but have now been finalized. The major modifications include: (1) an analysis of the uncertainties associated with the data and (2) corrections for background interferences which were present in unirradiated EMC foils. The results are tabulated in Tables ANL 3-6.

The %-errors quoted in these tables include only the following sources: (1) precision of replicate counts, (2) efficiency uncertainty estimates, (3) errors associated with geometry and self-absorption corrections, and (4) uncertainties associated with the nuclear data, e.g., half-life and branching ratios. The quoted errors are those associated with the accuracy of the fission-product reaction rate not the fission rate. Errors associated with fission-yields will, in general, dominate the uncertainty in the fission rate results.

In the last (7th Quarterly Report) Helmer and Greenwood recommended that the γ -spectra of unirradiated heavy-element foils must be made prior to irradiation so that interference effects might be anticipated. We agree with this suggestion, and can offer some quantitative data to support the suggestion. We have counted archive (unirradiated) samples of the EMC

TABLE ANL-3

Fission Rate Results from CFRMF Tests 5 and 6

Reaction	Sample	Fission Product	DPS/mg	Reaction Rates		% - Error ^a (2σ)	
				Product (10 ⁻¹⁵ $\frac{\text{atom}}{\text{atom-sec}}$)	Fission (10 ⁻¹⁴ $\frac{\text{f}}{\text{atom-sec}}$)	Rel.	Abs.
²³⁵ U(n,f)	U5-20	⁹⁵ Zr	53.4	7.77	12.2	2.0	5.0
		¹⁰³ Ru	43.5	3.89	11.8	1.0	4.6
		¹³¹ I	213	3.90	11.8	1.0	4.4
		¹⁴⁰ Ba	248	7.20	12.4	1.2	4.6
	Mean Fission Rate = 12.1						±.3
	U5-21	⁹⁵ Zr	61.2	7.64	12.0	1.0	4.7
		¹⁰³ Ru	49.6	3.80	11.6	1.0	4.5
		¹³¹ I	248	3.91	11.8	1.6	4.7
		¹⁴⁰ Ba	287	7.13	12.3	1.2	4.6
	Mean Fission Rate = 11.9						±.3
²³⁸ U(n,f)	U8-21	⁹⁵ Zr	2.39	.303	.557	1.0	4.4
		¹⁰³ Ru	4.72	.366	.582	2.2	5.0
		¹³¹ I	11.5	.183	.572	2.5	5.0
		¹⁴⁰ Ba	13.6	.341	.576	1.2	4.6
	Mean Fission Rate = .572						±.011
²³⁹ Pu(n,f)	P9-22	⁹⁵ Zr	50.9	6.44	13.5	2.0	5.2
		¹⁰³ Ru	119	9.29	13.2	1.0	5.6
		¹³¹ I	341	5.45	12.7	1.0	5.6
		¹⁴⁰ Ba	279	7.06	13.4	1.2	4.6
	Mean Fission Rate = 13.2						±.4

^aError in the reaction rate of the fission product. Relative error is the precision of replicate counts; absolute error includes all known sources of counting error in the fission product reaction rate.

TABLE ANL-4

 ^{235}U Fission Rate Results from the EMC Test

Reaction	Sample	Fission Product	DPS/mg	Reaction Rates		% - Error ^a (2 σ)	
				Product ($10^{-16} \frac{\text{atom}}{\text{atom-sec}}$)	Fission ($10^{-14} \frac{\text{fissions}}{\text{atom-sec}}$)	Rel.	Abs.
$^{235}\text{U}(n,f)$	EMC5-1	^{95}Zr	.740	6.33	.992	1.0	4.5
		^{97}Zr	58.8	5.57	1.01	1.0	4.5
		^{99}Mo	14.7	5.44	.959	3.0	6.4
		^{103}Ru	.593	3.11	.945	1.8	4.8
		^{131}I	3.01	3.20	.97	1.4	4.7
		^{132}Te	11.4	4.94	1.04	1.0	5.6
		^{133}I	55.5	6.52		1.0	4.7
		^{140}Ba	3.47	5.87	1.01	1.6	4.7
		^{143}Ce	29.8	5.47		1.0	4.5
	EMC5-8	^{95}Zr	.735	6.28	.984	1.0	4.5
		^{97}Zr	58.4	5.53	1.01	1.0	4.5
		^{99}Mo	14.3	5.29	.933	3.0	6.4
		^{103}Ru	.581	3.04	.924	3.2	5.5
		^{131}I	2.88	3.07	.93	4.2	6.2
		^{132}Te	11.4	4.92	1.04	1.0	5.6
		^{133}I	55.7	6.55		1.0	4.7
		^{140}Ba	3.48	5.90	1.02	1.6	4.7
		^{143}Ce	29.9	5.50		1.0	4.5

^aError in the reaction rate of the fission product. Relative error is the precision of replicate counts; absolute error includes all known sources of counting error in the fission product reaction rate.

TABLE ANL-5

 ^{239}Pu Fission Rate Results from the EMC Test

Reaction Sample	Fission Product	DPS/mg	Reaction Rate		% - Error ^a (2 σ)	
			Product (10^{-16} atom atom-sec)	Fission (10^{-14} fissions atom-sec)	Rel.	Abs.
$^{239}\text{Pu}(n,f)$ EMC N6Q	^{97}Zr	574	28.4	5.80	1.8	6.0
	^{103}Ru	14.4	38.6	5.48	1.2	5.8
	^{132}Te	142	31.5	5.94	4.0	6.8
	^{133}I	627	38.4		3.8	6.7
	^{140}Ba	33.4	29.0	5.52	1.0	5.6
	^{143}Ce	261	24.8		1.2	5.8
EMC N7J	^{97}Zr	561	27.8	5.67	3.0	6.4
	^{103}Ru	14.2	38.1	5.40	1.6	6.0
	^{132}Te	137	30.5	5.75	2.8	6.2
	^{133}I	614	37.6		2.0	6.0
	^{140}Ba	33.1	28.7	5.47	1.4	5.8
	^{143}Ce	243	23.1		3.6	6.7

^aError in the reaction rate of the fission product. Relative error is the precision of replicate counts; absolute error includes all known sources of counting error in the fission product reaction rate.

TABLE ANL-6

 ^{238}U Fission Rate Results from the EMC Test

Reaction	Sample	Fission Product	DPS/mg	Reaction Rate		% - Error ^a (2 σ)	
				Product ($10^{-16} \frac{\text{atom}}{\text{atom-sec}}$)	Fission ($10^{-14} \frac{\text{fissions}}{\text{atom-sec}}$)	Rel.	Abs.
$^{238}\text{U}(n,f)$	EMC8-1	^{95}Zr	.179	.776	.143	2.0	5.0
		^{97}Zr	15.6	.767	.130	1.0	4.4
		^{103}Ru	.339	.904	.144	1.6	4.8
		^{131}I	.861	.469	.147	3.8	5.8
		^{132}Te	3.65	.806	.149	2.6	5.2
		^{140}Ba	.978	.845	.143	4.0	6.0
		^{143}Ce	6.37	.603		4.2	6.2
	EMC8-7	^{95}Zr	.169	.738	.136	3.0	5.4
		^{97}Zr	15.5	.763	.129	1.0	4.4
		^{103}Ru	.335	.896	.142	1.6	4.8
		^{131}I	.842	.458	.143	3.6	5.9
		^{132}Te	3.60	.796	.147	2.3	5.0
		^{140}Ba	.996	.860	.145	4.0	6.0
		^{143}Ce	6.46	.612		3.5	5.7
$^{238}\text{U}(n,\gamma)$	EMC8-1	^{239}Np	549	88.2		1.2	4.6
	8-7	^{239}Np	529	85.1		1.2	4.6

^aError in the reaction rate of the fission product. Relative error is the precision of replicate counts; absolute error includes all known sources of counting error in the fission product reaction rate.

foils and attempted to identify the magnitudes of the interfering gamma rays. We find that for the 497 keV line in ^{103}Ru and the 364 keV line in ^{131}I no interferences are found within the measurement accuracies. For ^{140}Ba , if the counting were done within 30 days of the irradiation, about a 1% correction is required for ^{238}U and 0.5% for ^{235}U ; however, if one counts the ^{140}Ba in ^{235}U as late as 60 days after the irradiation the correction is about 3%. No correction was found for ^{140}Ba in the ^{239}Pu samples. In the case of ^{95}Zr , no correction was detected for the ^{235}U samples, but a 2.6% correction was found for ^{238}U . The data we acquired to assess the corrections to the ^{95}Zr activity in the ^{239}Pu samples is incomplete, but the corrections appear to be as large as 4-5%. For this reason we have not reported ^{95}Zr data for the ^{239}Pu , EMC samples.

These corrections account, in part, for the differences in the ANC and ANL data shown in Table ANL-1. Since ANC did not receive the archive samples, they were not able to make the corrections, whereas the ANL data presented in Table ANL-1 includes the correction for γ -ray interferences. We suggest the recommendations of ANC be followed; namely, a minimum value of fissions/atom of sample be established for all heavy-element fission rate measurements in order to minimize the significance of these corrections.

We wish to raise an additional caution. In reporting of fission-product reaction-rates and fission-rates in the ILRR quarterlies, particular attention should be given to quoted errors. For example, fission rates have been reported to less than 2% accuracy, whereas we are completely unaware of any fission-yield that is known to that accuracy. Furthermore, we suspect that the quoted errors are generally on the optimistic side. Typical examples of this are absolute gamma standards used for efficiency calibrations. Firstly, most standards are quoted at $\pm 1\%$ or less, whereas we have had numerous experiments where two standards did not agree within their quoted accuracies. Furthermore, most efficiencies are obtained by interpolation. Are these interpolation errors included?

Bias errors and random errors are often confused. For example, if all four ILRR labs reported a ^{140}Ba production rate, the accuracy is not obtained from the standard error of the mean value. Standard error represents the precision, not the accuracy. Since all four labs calibrated with the HEDL ^{140}Ba standard, any uncertainty in that standard

represents a bias to the mean of the four laboratory measurements and is not reduced simply because four labs rather than one used it. In the computation of standard errors an implicit assumption is that the errors associated with each data point are the same and that the expected value of the error for each point is zero. The first assumption is rarely, if ever, strictly true although it is often a negligible effect. The second assumption is severely violated if bias are present. In summary, we feel a more cautious use (or abuse) of statistics is warranted.

Finally, we mention a potential source of error neglected by all, but which may be dominate in fission product gamma-ray spectra. That is the uncertainty associated with establishing the net counts under a gamma-ray peak. This uncertainty depends upon numerous factors and is probably time dependent even for a given sample and gamma-ray. Perhaps the magnitude of this uncertainty could be examined by a "round-robin" test among ILRR participants.

EXPECTED ACCOMPLISHMENTS

Foils from future tests will be measured and analyzed. Errors associated with Ge(Li) measurements will be examined in greater detail.

C. Absolute Fission Rates of Pu-239, U-235, and U-238 in the FTR-EMC Test

R. J. Armani

OBJECTIVE

• To provide absolute fission rates in Pu-239, U-235, and U-238 encountered in the FTR-EMC test by absolute fission counter and solid-state track recorder (SSTR) methods.

SUMMARY

Sample packages containing 5-mil thick foils of Pu-239, U-235, and U-238, interspersed with mica solid-state track recorders and fission sources were irradiated in ZPR-9. Fission counters containing each of these isotopes were installed at symmetrical (or nearly so) locations in the matrix with normalizer foils attached to them. Fission rates would be provided by both the fission counters and the mica track recorders.

ACCOMPLISHMENTS AND STATUS

Table ANL-7 summarizes preliminary results of fission measurements by both methods. The Pu-239 and U-238 samples were counted with the automatic track scanner, while the U-235 was manually counted. Reasonable agreement is obtained between the fission counter and SSTR for Pu-239, but discrepancies occur in the U-235 and especially in the U-238 measurements. The large discrepancies in the uranium measurements are attributed to uncertainties in the fission source masses and possible contamination during assembly of the packages. The U-238 measurement was marginal at best because of intensity problems, and a slight U-235 contamination would have a large impact on the measurement.

TABLE ANL-7

Fissions per ng of Fissionable Isotope in EMC Test

	Pu-239	U-238	U-235
Fission counter	1030 + 25	25.96 ± .78	96.8 ± 2.9
SSTR	1068 ± 32	28.91 ± 1.45	104.7 ± 3.1

In each case, the largest portion of the error associated with the measurement is due to uncertainties in the specific alpha activity of the fission source material. Errors in the fission counter data have been revised since last reported to account for this. In addition, a correction of approximately 20% is made to the U-238 fission track measurement to account for the 3% U-234 spike, which contributes an error of approximately equal magnitude as the uncertainty in the mass.

The data in Table ANL-7 indicates that the SSTR data is biased on the high side as compared to fission counter data. Comparative measurements made in the Argonne Thermal Source Reactor in the past did not reveal this bias. Nevertheless, comparative measurements have been undertaken again using a set of 6 U-235 fission sources. Measurements are made in a back-to-back configuration with mica SSTR looking at one source and a fission counter looking at the other. The process is reversed and repeated in the same flux so that each sample is used at least once in the fission counter and with the SSTR.

EXPECTED ACCOMPLISHMENTS

Comparative measurements in the Argonne Thermal Source Reactor will determine if a bias exists between the fission counter and the SSTR methods. The FTR-EMC measurements used materials in the fission counters which were different from the SSTR material for both methods.

The specific alpha activity will be carefully determined by preparation of a large number of samples and intercalibrating them by fission and alpha counting. Then all but about 3 of each isotope will be destroyed for mass and isotopic analysis. The large number of samples will reduce the uncertainty in this measurement.

D. Absolute Fission Rate Measurements by the Track Recorder Technique

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

OBJECTIVE

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

SUMMARY

A discussion of the errors associated with SSTR fission rate measurements is presented. It is concluded that typical fission rate measurements are being made with an accuracy of $\pm 2.6\%$ (at the 95% confidence level) and ultimate accuracies under ideal conditions are about $\pm 2.0\%$. Preliminary results of fission rate measurements in the second CFRMF studies are presented. Tentative indications are that track recorders and fission chamber results agree to within $\sim 2\%$ for ^{238}U and ^{239}Pu , about 4% for ^{237}Np and 5-6% for ^{235}U .

ACCOMPLISHMENTS AND STATUS

1. Error Analysis of SSTR Stock Solutions and Track Recorder Plates

A primary source of uncertainty associated with both SSTR and fission chamber measurements of fission rates resides in the uncertainty in the mass determination of the foils used by the respective techniques. The procedures used for preparing SSTR plates has been described in previous reports; the intent here is to analyze the accuracy of the technique. The procedure may be briefly outlined as follows:

Materials of ^{235}U , ^{238}U , ^{237}Np , and ^{239}Pu of well characterized isotope purity have been reserved for ILRR purposes. We have obtained gram quantities of each of these materials, dissolved them, and very accurately diluted these solutions to obtain what we refer to as stock solutions of each of these materials. The concentrations of ng of material/gram of solution have been carefully calibrated; the accuracies of these calibrations are discussed below. The approach is to transfer known quantities of these stock solutions onto alpha counting plates and to calibrate an alpha counter in terms of cpm/ng of heavy element; again, the accuracy of this calibration will be discussed. Given an accurately

calibrated detector, foils of unknown masses, prepared from the same isotopic material as the stock solutions, may be accurately determined by alpha counting in the calibrated detector. For example, SSTR plates are prepared by electrodeposition (which is not 100% quantitative), and the masses are determined via alpha counting. Similarly, J. Grundl has had fission foils of ^{239}Pu prepared by vacuum deposition of ILRR material. We have sent him quantitative plates of our ^{239}Pu stock solutions so he can determine the masses of his foils in an analogous manner. This technique is essentially the standard procedures used in mass spectrometric isotope dilution techniques. We are simply using alpha spectrometric calibration techniques rather than isotopic dilution and mass spectrometric analysis. The questions we next address are: (1) How accurately are the stock solutions known in terms of ng of material/gram of solution? (2) How accurately are the alpha counters calibrated in terms of cpm/ng? (3) How accurately are the SSTR plates determined in terms of total mass?

In discussing the accuracy of the stock solutions and their associated accuracy, it is perhaps most clear if we follow through an actual example. Good radiochemical procedures were used such as calibrated balances, redistilled and repurified solvents, use of isotopic clean rooms to avoid cross contamination, etc. A piece of ^{239}Pu metal was etched to remove oxides, cleaned in water and alcohol, dried and weighed. The weight was $1.031 \pm .0005$ grams. The sample was dissolved in 6N HCl, plus small amounts of HNO_3 , and a drop of HF. The solution was centrifuged and the undissolved residue collected, dried, and weighed. The residue weight was <1-mg so the initial sample weight was determined to be $1.031 \pm 0.1\%$ grams. The dissolution solution was diluted to a total solution weight of $129.5192 \pm .0002$ grams. All transfers were carried out gravimetrically and in such a manner that two weight measurements were made (before and after weights of the receiving vessel and before and after weights of the transfer vessel). Two separate portions of the primary solution were taken in order to provide two independent series of dilutions. For the first dilution, $0.9560 \pm .0008$ grams of the primary solution was taken and diluted to a weight of $115.636 \pm .0002$ grams. This solution was designated dilution 1-2 and had a concentration of $65.809 \pm .13\%$ ng Pu/gram of solution. A portion of this solution, weighing $1.2384 \pm .0002$ grams was transferred and diluted to a weight of 25.4967

$\pm .0002$ grams. The concentration of the final stock solution (labeled solution 3) has a concentration of $3196.2 \pm .15\%$ ng Pu per gram of solution. The second Pu dilution series was prepared in a similar fashion except that 3 dilutions instead of 2 were carried out. This stock solution (labeled solution 4) had a concentration of $336.34 \pm .18\%$ ng Pu per gram of solution.

Four separate weighed transfers of each stock solution were directly transferred onto plates for alpha counting. The amounts of Pu transferred to these alpha plates varied between 497.16 and 35.01 ng of Pu and were known in absolute accuracy to about $\pm 0.2\%$. The accuracy of the individual plates was partially dependent upon the weight of solution transferred because the balance used in this work was accurate to ± 0.1 -mg.

Each of the eight alpha plates (4 from each stock solution) were alpha counted in two 2π alpha counters. The counting statistics were nominally $\pm 0.3\%$. The counting data provided two pieces of statistical information. Firstly, the standard deviation of a single measurement was a test of the precision of the alpha plating procedure. The precisions were $\pm 0.07\%$ for stock solution 3 and $\pm 0.82\%$ for stock solution 4. Secondly, the combined data provide a means for absolutely calibrating the alpha counter. The mean value of the cpm/ng of Pu was 71.75 cpm/ng with a standard error of $\pm 0.3\%$. The absolute accuracy of the alpha calibration is thus $\pm 0.43\%$ for stock solution 3 and $\pm 0.85\%$ for stock solution number 4.

An independent verification of this methodology can be made by calibrating the 2π alpha detector efficiency with an NBS alpha standard using the published half-lives of ^{239}Pu , ^{240}Pu , ^{241}Pu , and ^{241}Am , and measuring the dpm of the ^{239}Pu in the alpha plates. This theoretical value for the isotopic composition of the Pu is 139.4 d/m/mg, and we determined a value of 140 d/m/mg for series 3, and 141 d/m/mg for series 4. This agreement is well within the accuracies of the half-lives and the NBS alpha standards.

The procedure for preparing calibrations of ^{237}Np was identical to the plutonium. For the ^{238}U and ^{235}U one additional step was required. In order to achieve practical alpha counting rates, the stock solutions were spiked with ^{234}U . The ^{234}U used for this purpose was isotopically purified in a

mass separator, then chemically purified. The ^{234}U was added at the stage at which the two portions of the primary dissolution solution were taken. In this manner two different alpha activity levels could be obtained for the two separate stock solutions of both ^{235}U and ^{238}U . About 0.298% and 0.237% ^{234}U is present in the two ^{238}U solutions and 5.29% and 6.75% ^{234}U is present in the two ^{235}U stock solutions. Because the stock solutions contain a known concentration of either ^{235}U or ^{238}U , the amount of ^{234}U added is not critical; the critical value is cpm (alpha) per gram of solution. This was determined identically to the ^{239}Pu solutions. Summarized below are the nominal errors on the respective quantities.

Material	Stock Solution $\left[\frac{\text{ng}(\text{material})}{\text{gram}(\text{solution})} \right]$	Alpha Calibration $\left[\frac{\text{cpm}(\text{alpha})}{\text{ng}(\text{material})} \right]$	Foil Calibration $[\text{ng}(\text{material})]$
^{239}Pu	$< \pm .2\%$	$\pm 0.85\%$	$\pm 0.9\%$
^{237}Np	$< \pm .2\%$	$\pm 0.3\%$	$\pm 0.5\%$
^{235}U	$\pm .2\%$	$\pm 0.36\%$	$\pm 0.55\%$
^{238}U	$< \pm .2\%$	$\pm 0.5\%$	$\pm 0.6\%$

This table actually summarizes the worst case uncertainties, for example, if all work is carried out with the Pu stock solution number 3, the masses of the SSTRs could be determined to an accuracy of $\pm 0.5\%$. Also the final SSTR mass accuracy is partially dependent upon the total alpha activity present in the samples. Large samples are counted to $< 0.1\%$ counting statistics, whereas very small samples may be only counted to 0.8% counting statistics.

Two stippled alpha plates were sent to J. Grundl to be used for fission foil calibration. Plate number 2-9 Pu 3-3 has a total Pu weight of $0.47655\text{-}\mu\text{g}$ and a ^{239}Pu weight of $0.4724\text{-}\mu\text{g}$ with an assigned uncertainty of $\pm 0.43\%$. Plate number 2-7NP 44-2 has a ^{237}Np weight of $3.1168\text{-}\mu\text{g}$ and an assigned mass uncertainty of $\pm 0.3\%$.

The absolute accuracy of fission rates, when measured by SSTR plates prepared and calibrated by this technique will depend upon (1) the uncertainty in the mass, (2) the number of fission tracks (counting statistics), (3) the accuracy of the optical efficiency used in converting tracks to fissions, and (4) all other unknown sources. These

uncertainties typically are $\pm 0.5\%$, $\pm 1.0\%$, $\pm 0.5\%$, respectively for items 1, 2, and 3, respectively. The primary unknown source is probably cross contamination, especially for the ^{238}U measurements. For this reason, we alpha count all SSTR plates before and after the irradiation. This is admittedly not a sensitive test; however, if a major contamination were to occur we could pick it up. Our experience has been that the after count is generally within $<\pm 1\%$ of the before count. If the after counts were 1-2% lower we use the before count to define the weight because some of the material might have dislodged while the plate was in contact with the mica during irradiation.

In the cases of the ^{238}U and ^{235}U plates which are spiked with ^{234}U , corrections for the fissions due to ^{234}U fission must be made. We always include a pure ^{234}U track recorder so the correction is made on the basis of measured fissions/ng of ^{234}U . In CFRMF, the ^{234}U spikes contribute about 2% of the fissions in both the ^{238}U and ^{235}U foils. Thus, the error introduced by corrections for ^{234}U is negligible. The overall error in fission rates measured in a routine SSTR measurement is typically about 2.6% (at the 95% confidence level). In optimal measurements in which the best stock solutions are used and the number of tracks exceeds 30,000, the uncertainty can be reduced to the level of about $\pm 2\%$ (95% level).

2. Preliminary Results of SSTR Measurements in CFRMF

A second complete series of SSTR fission rate measurements was conducted in CFRMF in November 1972, and April 1973. These track recorders were prepared from the high isotopic purity ILRR materials by the procedures described above. This series of ^{234}U , ^{235}U , ^{238}U , ^{237}Np , and ^{239}Pu track recorders is designated CFRMF-II, as distinguished from the former series of CFRMF-I recorders, which was irradiated in February 1972. The first series was prepared from less well characterized and calibrated fissionable material.

At the present time the series II recorders have been counted on the automatic track scanner at ANL. In order to finalize these counts they must each be manually counted at least once to verify the scanner results even though the two methods have been consistently in agreement to within $\pm 1\%$. For purposes of this preliminary comparison we have utilized the following normalization factors: the fission counter data indicate a 1.02% lower flux level (for the nominal 0.6-kW reactor level) for the

November 1972 irradiation than for the February 1972 series. No ^{239}Pu or ^{237}Np fission rates were measured by fission chambers in the November test so we have used the February fission chamber data reduced by the 1.02% different flux levels as our reference. This may not be valid because our ^{239}Pu and ^{237}Np data were irradiated in April 1973, and as yet we do not know what the fission chamber rates were. We are essentially assuming that the April 1973 test was conducted at exactly the same flux level as the November 1972 test. With these qualifications in mind, the most recent SSTR fission rate data are tabulated in Table ANL-8 and compared to both the fission chamber results and the CFRMF series I track recorder results.

TABLE ANL-8
Preliminary Fission Rate Results from
the Most Recent SSTR Measurements

Isotope	Fission Rate $\times 10^{15}$ (fiss/atom-sec)			Fission Rate Ratio	
	SSTR (Series II)	SSTR (Series I)	F.C.	$\frac{\text{SSTR-II}}{\text{SSTR-I}}$	$\frac{\text{SSTR-II}}{\text{F.C.}}$
^{235}U	13.4	13.2	12.7	1.02	1.06
^{238}U	0.594	-	0.604	-	0.98
^{239}Pu	14.6	15.4	14.6	0.95	1.00
^{237}Np	4.70	4.86	4.50	0.97	1.04

The new track recorder data is in excellent agreement with the fission chamber results for ^{238}U and ^{239}Pu . The recent data has improved the comparison to fission chambers for ^{237}Np , but the ^{235}U comparison is worse. Further discussion is unwarranted until the track counting results have been finalized and absolute errors are assigned.

EXPECTED ACCOMPLISHMENTS

The CFRMF-II Track Recorder data will be finalized and evaluated against the NBS fission chamber results. Further studies of the differences between track recorders and fission chambers will be investigated.

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ATLANTIC RICHFIELD HANFORD COMPANY

- A. ARHCO - CFRMF AI-II REACTION RATES
ARHCO - 1973 SELF-SHIELDING REACTION RATES
ARHCO - HEDL-5 AND HEDL-6 REACTION RATES

W. H. Zimmer

Objective

- To report the measurements of the reaction rates of the fission and non-fission foil sets above.

Summary

The measurement constants, activity analyses, reaction rates, and estimated accuracies are reported for the ARHCO-CFRMF AI-II foils, the 1973 Self-Shielding foils, and the HEDL-5 and HEDL-6 fission foils. All calculation constants not listed such as isotopic half-life, gamma branching ratio, irradiation time, etc., are those mutually agreed upon and previously published.

Commentary

All gamma analytical identification codes listed consist of an alpha character to designate the detector/analyzer system and numeric characters representing the time of analysis in Hanford days in units of thousandths days minus 10000 days. All time values are normalized to Pacific Standard Time.

The CFRMF AI-II Reaction Rate tabulation includes the peaked background found in the nine nonirradiated foils listed. All fission product peaks currently being analyzed in irradiated fission foils were monitored in spectra with average acquisition times of 10 hours. If contributions are not listed they were not found. The average backgrounds found were subtracted from the activity measurements indicated by asterisks.

All reaction rate measurements are listed, regardless of their apparent quality, in the hope that they will be useful now or in the future. As a consequence, the HEDL-5 and -6 reaction rates measured approximately 164 days after TOR contain analyses for short-lived isotopes such as ^{140}Ba which have decayed through more than 12 half-lives. The quality of such a result is of necessity poor as is indicated in the estimated accuracy.

TABLE ARHCO-1
CFRME AI-II REACTION RATES

Foil Identification	TOR (PST)	Milligram Target Isotope	Reaction	keV	Absorption Correction Factor	d/s/mg @ TOR	10 ¹⁵ x Measured Reaction Rate	% Fission Yield	% Estimated Accuracy (2σ)
NI-TJ C531540	10468.269	296.06	⁵⁸ Ni(n,p) ⁵⁸ Co	810.757	0.992057	124.544	1.9534		2.5
U5-17 C531777		289.511	²³⁵ U(n,f)	¹⁴⁴ Ce 133.53 ¹³¹ I 364.49 ¹⁰³ Ru 497.1 ¹⁰⁶ Ru 622.6 ¹⁴⁰ Ba 537.38 1595.4	0.693810 0.954187 0.973479 0.980098 0.975904 0.991417	40.782 832.780 168.616 5.699 930.741 985.065	6.9296 117.67 121.54 1.2607 125.43 132.75	100 3.53 3.3 100 5.8 5.8	4.3 3.0 2.3 12.1 2.6 2.3
				¹³⁷ Cs 661.638 ⁹⁵ Zr 724.184 756.74	0.981208 0.982997 0.983929	1.261* 198.311 202.557	133.74 124.43 127.10	6.2 6.25 6.25	5.4 2.3 2.2
P9-20 C531578		257.924	²³⁹ Pu(n,f)	¹⁴⁴ Ce 133.53 ¹⁰³ Ru 497.1 ¹⁰⁶ Ru 622.6 ¹⁴⁰ Ba 537.38 1595.4	0.717801 0.974434 0.980835 0.976772 0.991816	56.493 398.141 28.899 923.378 939.948	9.8398 137.70 6.5530 142.28 144.83	100 7.05 100 5.2 5.2	6.7 2.4 11.4 6.1 3.5
				¹³⁷ Cs 661.638 ⁹⁵ Zr 724.184 756.74	0.981918 0.983661 0.984569	1.486* 165.094 166.289	151.82 139.13 140.14	6.6 4.77 4.77	16.6 2.8 2.7
P9-17	C10516.999	260.570	Bkg	¹³⁷ Cs 661.638		0.183			22.
U5-12	C10523.999	288.950	Bkg	¹³⁷ Cs 661.638		0.0167			22.
U8-17	C10523.000	311.186	Bkg	¹⁰⁶ Ru 622.6		0.0300			25.
				¹³⁷ Cs 661.638		0.0157			22.
P9-15	C10522.000	255.184	Bkg	¹³⁷ Cs 661.638		0.142			22.
U5-14	C10519.500	280.120	Bkg	¹³⁷ Cs 661.638		0.0168			17.
N7-17	C10518.000	212.275	Bkg	¹³⁷ Cs 661.638		0.0225			22.
N7-20	C10430.999	385.453	Bkg	¹³⁷ Cs 661.638		0.0101			31.
U5-9	C10422.000	286.768	Bkg	¹³⁷ Cs 661.638		0.0152			18.
P9-21	D10433.000	256.945	Bkg	¹³⁷ Cs 661.638		0.141			22.

* Background subtracted.

ARHCO-2

TABLE ARHCO-2
1973 SELF-SHIELDING REACTION RATES

Foil Identification		TOR (PST)	mg Target Isotope	Reaction	keV	Absorption Correction Factor	d/s/mg @ TOR	10 ¹⁵ x Measured Reaction Rate	% Fission Yield	% Estimated Accuracy (2σ)
HEDL-1SS C432571	Co-3	10396.9895	1.538	⁵⁹ Co (n, γ) ⁶⁰ Co	1173.208	1.00	45.571	13.76		4.2
					1332.56	1.00	45.233	13.66		4.4
				⁵⁹ Co (n, p) ⁵⁹ Fe	1099.27	1.00	8.654	0.0610		22.
					1291.56	1.00	7.566	0.0533		20.
HEDL-2SS C432655	Co-1 (1-2)	10397.0697	14.761	⁵⁹ Co (n, γ) ⁶⁰ Co	1173.208	0.99970	1.6522	10.79		8.9
					1332.56	0.99972	1.6105	10.50		9.4
D432644	Co-1 (1-3)		14.070	⁵⁹ Co (n, γ) ⁶⁰ Co	1173.208	0.99970	1.7309	11.28		6.4
					1332.56	0.99972	1.8263	11.91		6.5
				⁵⁹ Co (n, p) ⁵⁹ Fe	1099.27	0.99968	0.4923	0.0744		27.
					1291.56	0.99971	0.4310	0.0651		26.
D433441	Co-1 (1-4)		14.784	⁵⁹ Co (n, γ) ⁶⁰ Co	1173.208	0.99970	1.5198	9.908		6.5
					1332.56	0.99972	1.5337	9.998		6.8
C434522	Co-1 (all)		43.615	⁵⁹ Co (n, γ) ⁶⁰ Co	1173.208	0.99904	1.6054	10.47		12.
					1332.56	0.99910	1.7744	11.57		12.
HEDL-3SS C432576	Co-1	10397.1408	57.411	⁵⁹ Co (n, γ) ⁶⁰ Co	1173.208	0.99876	1.3629	9.591		4.9
					1332.56	0.99883	1.3563	9.545		5.1
				⁵⁹ Co (n, γ) ⁵⁹ Fe	1099.27	0.99872	0.3368	0.0550		23.
					1291.56	0.99881	0.3258	0.0532		19.
HEDL-1SS C432464	U-5#3	10396.9895	3.1984	²³⁵ U (n, f) ¹³¹ I	364.49	0.99901	1350.7	200.1	3.53	5.5
				¹⁰³ Ru	497.1	0.99933	303.58	229.8	3.3	3.9
				¹⁴⁰ Ba	537.3	0.99935	1600.0	226.3	5.8	6.8
				¹⁴⁰ Ba	1595.4	0.99971	1723.2	243.7	5.8	4.7
				⁹⁵ Zr	724.2	0.99951	356.07	234.7	6.25	4.9
				⁹⁵ Zr	756.7	0.99952	347.33	228.9	6.25	4.6
HEDL-2SS C431666	U-5#1	10397.0697	303.281	²³⁵ U (n, f) ¹³¹ I	364.49	0.95649	68.311	210.6	3.53	4.2
				¹⁰³ Ru	497.1	0.97618	13.511	219.2	3.3	3.4
				¹⁴⁰ Ba	537.3	0.97860	76.616	228.6	5.8	5.2
				¹⁴⁰ Ba	1595.4	0.99332	80.801	241.0	5.8	3.7
				⁹⁵ Zr	756.7	0.98648	15.710	176.3	6.25	3.9
				⁹⁵ Zr	756.7	0.98648	15.710	176.3	6.25	3.9
HEDL-3SS D431669	U-5#1	10397.1408	599.565	²³⁵ U (n, f) ¹³¹ I	364.49	0.91686	62.515	208.1	3.53	2.8
				¹⁰³ Ru	497.1	0.95405	12.040	210.8	3.3	2.5
				¹⁴⁰ Ba	537.3	0.95868	69.824	224.8	5.8	3.0
				¹⁴⁰ Ba	1595.4	0.98710	72.897	234.7	5.8	2.5
				⁹⁵ Zr	724.2	0.97212	14.591	223.1	6.25	2.6
				⁹⁵ Zr	756.7	0.97387	14.728	225.2	6.25	2.6

ARHCO-3

TABLE ARHCO-3

HEDL-5 AND -6 REACTION RATES MEASURED APPROXIMATELY 20 DAYS AFTER TOR

Foil Identification	TOR (PST)	mg Target Isotope	Reaction	KeV	Absorption Correction Factor	d/s/mg @ TOR	$10^{14} \times$ Measured Reaction Rate	% Fission Yield	% Estimated Accuracy (2 σ)
HEDL-5 U-5 #21 A290918	10269.2870	285.239	$^{235}\text{U}(n,f)$	^{141}Ce 145.44	0.73593	135.7	8.528	100	2.3
				^{131}I 364.49	0.95494	255.4	113.7	3.53	2.3
				^{103}Ru 497.1	0.97398	51.86	120.4	3.3	2.3
				^{140}Ba 537.3	0.97637	294.0	126.2	5.8	2.3
				1595.4	0.99163	300.5	128.9	5.8	1.5
				^{95}Zr 756.7	0.98427	62.68	127.0	6.25	2.3
HEDL-5 N-7 #21 C290480		306.4353	$^{237}\text{Np}(n,f)$	^{103}Ru 497.1	0.97599	32.769	47.79	5.3	7.8
				^{95}Zr 756.7	0.98545	20.153	46.80	5.5	8.5
				^{140}Ba 537.3	0.97820	102.4	47.60	5.4	9.2
				1595.4	0.99224	99.3	46.16	5.4	5.8
HEDL-5 U-8 #21 A290527		309.793	$^{238}\text{U}(n,f)$	^{141}Ce 145.44	0.71909	4.933	0.3139	100	2.6
				^{131}I 364.49	0.95175	12.51	6.223	3.2	2.7
				^{103}Ru 497.1	0.97222	4.882	6.024	6.29	2.5
				^{140}Ba 537.3	0.97479	14.81	6.221	6.0	3.2
				1595.4	0.99114	14.64	6.150	6.0	2.1
				^{95}Zr 756.7	0.98328	2.537	5.981	5.44	3.3
HEDL-5 P-9 #22 C291405		256.786	$^{239}\text{Pu}(n,f)$	^{141}Ce 145.44	0.74759	139.1	8.890	100	2.4
				^{131}I 364.49	0.95622	360.6	129.5	4.45	2.5
				^{103}Ru 497.1	0.97475	117.0	129.4	7.05	2.4
				^{140}Ba 537.3	0.97707	274.5	133.6	5.2	3.1
				1595.4	0.99197	292.6	142.4	5.2	2.0
				^{95}Zr 756.7	0.98481	50.00	142.5	4.52	3.1
HEDL-6 U-5 #20 C290949	10270.2919	280.821	$^{235}\text{U}(n,f)$	^{141}Ce 145.44	0.74005	116.7	8.552	100	2.3
				^{131}I 364.49	0.95572	211.0	109.4	3.53	2.3
				^{103}Ru 497.1	0.97441	42.72	115.7	3.3	2.3
				^{140}Ba 537.3	0.97675	244.0	122.0	5.8	2.4
				1595.4	0.99175	254.5	127.3	5.8	1.5
				^{95}Zr 756.7	0.98452	51.87	122.6	6.25	2.4

ARHCO-4

TABLE ARHCO-4

HEDL-5 AND -6 REACTION RATES MEASURED APPROXIMATELY 164 DAYS AFTER TOR

Foil Identification	TOR (PST)	Milligram Target Isotope	Reaction	keV	Absorption Correction Factor	d/s/mg @ TOK	10 ¹⁵ x Measured Reaction Rate	% Fission Yield	% Estimated Accuracy (2σ)					
HEDL-5 U-5 #21 D433537	10269.2870	285.239	²³⁵ U(n, f)	¹⁰³ Ru	497.1	0.97398	36.289	84.3	3.3	13.				
				¹⁰⁶ Rh	622.6	0.98050	1.1541	0.8268	100.	40.				
				¹³⁷ Cs	661.6	0.98160	0.35349*	121.5	6.2	5.7				
				¹⁴⁴ Ce	133.5	0.69694	11.240	6.184	100.	4.8				
				¹⁴⁴ Pr	696.4	0.98258	11.143	6.131	100.	12.				
				⁹⁵ Zr	724.2	0.98336	48.676	98.63	6.25	2.4				
				⁹⁵ Zr	756.7	0.98427	49.768	100.8	6.25	2.4				
				¹⁴⁰ Ba	1595.4	0.99163	129.16	55.42	5.8	50.				
				HEDL-5 N-7 #21 C433630		306.4353	²³⁷ Np(n, f)	⁹⁵ Zr	724.2	0.98462	28.675	66.59	5.5	17.
⁹⁵ Zr	756.6	0.98545	18.084					41.99	5.5	21.				
HEDL-5 U-8 #21 D433977	10269.2870	309.793	²³⁸ U(n, f)	¹⁴⁴ Ce	133.5	0.67854	0.50999	0.2842	100.	4.6				
				¹⁴¹ Ce	145.44	0.71909	4.5430	0.2891	100.	4.3				
				¹⁰³ Ru	497.1	0.97222	4.6650	5.757	6.29	2.3				
				¹⁰⁶ Rh	622.6	0.97923	0.17140*	0.1244	100.	6.1				
				¹³⁷ Cs	661.6	0.98041	0.003612*	1.238	6.3	4.9				
				¹⁴⁴ Pr	696.4	0.98146	0.48950	0.2728	100.	13.				
				⁹⁵ Zr	724.2	0.98230	2.3225	5.476	5.44	2.4				
				⁹⁵ Zr	756.7	0.98328	2.3726	5.594	5.44	2.4				
				¹⁴⁰ Ba	1595.4	0.99114	15.522	6.520	6.0	25.				
				HEDL-5 P-9 #22 C433442		256.786	²³⁹ Pu(n, f)	¹⁰³ Ru	497.1	0.97475	120.40	133.1	7.05	5.0
								⁹⁵ Zr	724.2	0.98391	50.706	144.5	4.52	6.7
⁹⁵ Zr	756.7	0.98481	53.084					151.3	4.52	6.2				
HEDL-6 U-5 #20 D433633	10270.2919	280.821	²³⁵ U(n, f)	¹⁴⁴ Ce	133.5	0.70149	10.587	6.796	100.	5.0				
				¹⁰³ Ru	497.1	0.97441	42.041	113.9	3.3	2.6				
				¹⁰⁶ Rh	622.6	0.98081	1.1451	0.9571	100.	20.				
				¹³⁷ Cs	661.6	0.98189	0.31164*	125.0	6.2	6.1				
				¹⁴⁴ Pr	696.4	0.98285	10.711	6.875	100.	12.				
				⁹⁵ Zr	724.2	0.98362	50.404	119.1	6.25	2.5				
				⁹⁵ Zr	756.7	0.98452	51.897	122.7	6.25	2.5				
				¹⁴⁰ Be	1595.4	0.99175	237.36	118.7	5.8	15.				

* Background subtracted.

ARHCO-5

B. Ba-140 STANDARD FOR FISSION RATES

W. H. Zimmer

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 or acquire the necessary reference standard calibrated to within plus or minus two percent (95% confidence level) as required for Fast Reactor Dosimetry and the ILRR program.

Summary

Measurement constants and activity analysis of T913-5 are reported.

Accomplishments and Status

All results are reported in units of d/s/gram T913-5 at a time zero of 12:00PST March 13, 1972 or a decimal day time of 31500 (thousandths of days). Three mounts of 1000.92 microliters were prepared at a hood temperature of 23 degrees C for a density conversion factor of 0.99756 g/ml. Serial numbers identify the two systems used (A&C) and the decimal day time of count (thousandths of days). Peak centers are in units of channels with approximately two channels per keV.

TABLE ARHCO-5 MEASUREMENT CONSTANTS

<u>Isotope</u>	<u>Energy</u> <u>in keV</u>	<u>Gamma per</u> <u>Disinteg.</u>	<u>Half Life</u> <u>in Days</u>	<u>%P</u>
Ba-140	537.3	0.24	12.80	2
La-140	1595.4	0.9619	1.676	1

The La-140 peak activity has been converted to Ba-140 activity assuming equilibrium. Accuracy for individual isotopic activities is the square root of the sum of the squares of the precision of calibration (%P from Table of Measurement Constants) and of the counting statistics precision posted in the Activity Analysis Table.

TABLE ARHCO-6 ACTIVITY ANALYSIS

ISOTOPE	PEAK CENTER	PLUS OR MINUS % OF 95% CL	1/3/CHAN OUT OF COUNT	1/3/CHAN OUT ZERO
T913-5 A28510	#1	BALA140	1000.92 UL	
BA140	1274	+0.9638170E+20	+0.1284344E+07	+0.1292096E+07
BA140	3189	+0.7476361E+00	+0.1326334E+07	+0.1110970E+07
T913-5 C28565	#1	BALA140	1000.92 UL	
BA140	1274	+0.9519774E+20	+0.1279480E+07	+0.1291465E+07
BA140	3192	+0.6549406E+00	+0.1277749E+07	+0.1289985E+07
T913-5 A28625	#2	BALA140	1000.92 UL	
BA140	1274	+0.9521588E+20	+0.1320291E+07	+0.1111618E+07
BA140	3189	+0.7427811E+00	+0.1322397E+07	+0.1132517E+07
T913-5 C28625	#2	BALA140	1000.92 UL	
BA140	1274	+0.8324283E+20	+0.1314677E+07	+0.1125137E+07
BA140	3191	+0.6431243E+00	+0.1321075E+07	+0.1131297E+07
T913-5 A28560	#3	BALA140	1000.92 UL	
BA140	1274	+0.9564882E+20	+0.1302662E+07	+0.1110936E+07
BA140	3189	+0.7429631E+00	+0.1324824E+07	+0.1129836E+07
T913-5 C28592	#3	BALA140	1000.92 UL	
BA140	1274	+0.9519244E+20	+0.1309694E+07	+0.1118861E+07
BA140	3191	+0.6516932E+00	+0.1324098E+07	+0.1131176E+07

ATOMICS INTERNATIONAL

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

Harry Farrar IV

Objective

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

Summary

Recent measurements of the helium generated in small specimens of enriched ${}^{10}\text{B}$ that were encapsulated in vanadium and irradiated in EBR-II have given results that are consistently $\sim 16\%$ higher than calculated values. By comparison, specimens that were not enclosed in miniature vanadium capsules showed evidence of having lost by diffusion $\sim 30\%$ of the helium generated. This would explain the relatively low helium values from the unencapsulated specimens irradiated in CFRMF. A new CFRMF irradiation experiment with encapsulated specimens of ${}^{10}\text{B}$ and ${}^6\text{Li}$ is now in progress.

Accomplishments and Status

a. Helium Measured in Irradiated Boron Specimens. Measurements have been made on numerous small specimens of enriched ${}^{10}\text{B}$, ${}^{11}\text{B}$ and natural boron irradiated at several locations in the core and blanket regions of EBR-II and in the central core location of CFRMF. Some of the EBR-II boron specimens were sealed in miniature vanadium capsules to retain any helium that might have diffused or recoiled out of the boron crystals during or after the irradiation.

Although the EBR-II irradiations were not conducted in connection with the ILRR program, the data obtained with boron specimens from EBR-II and CFRMF are directly comparable and seemed, until recently to be inconsistent with each other. Inclusion of the EBR-II findings in this report makes possible a complete discussion of the subject.

The results of the mass spectrometric helium analyses of unencapsulated "bare" specimens of natural boron (19.8% ^{10}B), enriched ^{10}B (95.58% ^{10}B) and enriched ^{11}B (1.599% ^{10}B) that were irradiated in CFRMF and EBR-II are shown in Table AI-1. The CFRMF irradiation lasted 16.00 hours at a power level of 9.75 kW. The EBR-II irradiations all took place during the 8-day 62.5 MW dosimetry test (Run 50H). The helium measurements were made with a high sensitivity gas mass spectrometer system operated in the "static mode." A new high temperature furnace with resistance-heated graphite crucibles was used to vaporize the boron specimens and release the ^4He . Known quantities of ^3He were added to the furnace before the heating to accurately normalize the ^4He measurement. The concentration values shown in Table AI-1 have an estimated absolute 1 σ uncertainty of 3%.

From the fifth and sixth columns it can be seen that the ^{10}B specimens accumulated over 50 times more helium than the enriched ^{11}B specimens. When the helium concentrations are recalculated with respect to the amount of ^{10}B present, however, in column 6, the results become fairly consistent. This indicates that the isotope ^{11}B contributes very little (if any) helium when compared to the isotope ^{10}B . The variations in the helium concentrations in column 6 are not due to uncertainties in measurement of the amount of helium released. It is possible that some of the helium was not always released by the high temperature furnace, but this is unlikely since the crucible of the furnace is heated until much of the graphite vaporizes; well above the boiling-point-temperature of boron. On several occasions, moreover, unvaporized sections of crucible have been reheated, with no additional release of helium. Low values are therefore presumed to be the result of helium diffusion or recoil.

Also shown in Table AI-1 are values of $\bar{E}$, the average energy of the neutron spectrum; and values of helium concentration calculated by HEDL. The calculations used ENDF/B III energy dependent total helium pro-

TABLE AI-1
HELIUM CONCENTRATIONS IN BARE SPECIMENS OF BORON

Capsule & Location (cm)	$\bar{E}$ (MeV)	Percent ^{10}B	Specimen Mass (mg)	Helium Concentration (appm)			
				Measured per total B	Measured per ^{10}B	Calculated per ^{10}B	Ratio Meas/Calc
B3 (CFRMF Core Center)	0.72	95.58	4.437	0.0121	0.0127	0.0140	0.91
		95.58	2.989	0.0120	0.0125	0.0140	0.89
		95.58	5.333	0.0122	0.0128	0.0140	0.91
WK1 (EBR-II R=15.3 Z=4.3)	0.81	19.8	0.4428	396	2000	2136	0.94
		19.8	0.4235	395	1995	2136	0.93
		19.8	0.3139	353	1780	2136	0.83
		95.58	0.4385	1935	2025	2136	0.95
		95.58	0.1568	1980	2070	2136	0.97
		95.58	0.2278	1595	1670	2136	0.78
		1.599	0.9273	29.4	1840	2136	0.86
		1.599	1.2607	26.8	1680	2136	0.79
WK2 (EBR-II R=15.3 Z=37.8)	0.34	19.8	0.7990	411	2075	3148	0.66
		19.8	1.2254	414	2090	3148	0.66
		1.599	0.8090	50.2	3140	3148	1.00
		1.599	1.8099	50.4	3150	3148	1.00
WK3 (EBR-II R=15.3 Z=69.8)	0.15	19.8	0.6535	257	1300	2063	0.63
		19.8	0.6335	321	1620	2063	0.79
		19.8	1.1872	323	1630	2063	0.79
		1.599	1.3550	29.9	1870	2063	0.91
		1.599	0.9790	30.1	1880	2063	0.91
WK4 (EBR-II R=19.0 Z=4.3)	0.81	19.8	0.9393	338	1710	2102	0.81
		95.58	0.4160	1350	1410	2102	0.67
		1.599	0.7634	17.3	1080	2102	0.51
		1.599	0.9476	11.7	730	2102	0.35
		1.599	0.6617	15.4	965	2102	0.46
WK6 (EBR-II R=33.5 Z=4.3)	0.52	19.8	0.6238	263	1330	1403	0.95
		19.8	0.5829	256	1290	1403	0.92
		1.599	0.4336	19.2	1200	1403	0.86
		1.599	0.4546	18.7	1170	1403	0.83
WK7 (EBR-II R=38.2 Z=4.3)	0.43	19.8	0.7263	225	1135	1153	0.98

duction cross sections, modified by Lippincott (HEDL) to include recent data obtained from M. P. Fricke (GRT). The modifications had the effect of increasing the calculated values by $\sim 13\%$. Total neutron fluence was determined in all cases by HEDL using multiple foil dosimetry sets adjacent to the AI irradiation capsules.

The last column in Table AI-1 gives the ratio of measured-to-calculated helium concentration. This ratio fluctuates between 0.35 and 1.00 and has an average value of 0.82. As mentioned earlier, the low values in any capsule are assumed to be the result of helium diffusion or recoil. The fact that this is occurring at all casts doubt even on the high measured values which could also be suffering from some undetermined loss of helium before the measurement.

Table AI-2 gives the helium concentrations for specimens of ^{10}B that were sealed in miniature vanadium capsules in a manner similar to that described in Section B. During the mass spectrometric analyses, the entire capsules and their contents were analyzed for helium content. The contribution to the total helium from the vanadium itself was found by irradiating a number of empty vanadium capsules along with the others in the X129 subassembly. For the X140 and X140A specimens, no separate pieces of vanadium were available for helium analysis. The contributions for these capsules were estimated from the X129 data simply by assuming the helium generation in vanadium is proportional to the neutron fluence > 5 MeV. In actual fact, this assumption introduces no additional uncertainty to the results since the vanadium capsules contributed negligible amounts of helium, ranging from less than 0.01% to only 0.34% of the total produced. The contribution of helium from the gold foil has not been measured but empirical predictions suggest this should be negligible.

The measured helium concentrations in Table AI-2 have been corrected for vanadium contribution and are presented with respect to the number of ^{10}B atoms present in the sample. The absolute 1σ uncertainty of the measurements averages 2.7 % of the concentration value. The calculated values were determined by HEDL with the same

TABLE AI-2
HELIUM CONCENTRATIONS IN ENCAPSULATED BORON-10

Sample Number *	EBR-II Location			$\bar{E}$ (MeV)	Mass (mg)			Helium Concentration (appm) per ^{10}B		
	Subas- sembly	R (cm)	Z (cm)		B	V	Au	Meas.	Calc.	Meas/ Calc
M1-10B3	X129	21.3	+ 2.65	0.819	0.6542	17.15	0	2053	1862	1.103
M2-10B4	X129	21.3	+10.35	0.755	0.6908	17.16	0	2077	1821	1.140
M3-10B8	X129	21.3	+16.05	0.679	0.4795	17.07	0	2086	1817	1.148
1-6A3	X140	14.3	- 2.5	0.870	0.1490	28.21	1.60	14570	12740	1.144
1-7A3	X140	16.3	- 2.5	0.863	0.1082	28.72	2.04	14810	12094	1.224
1-7B3	X140	16.3	-10.8	0.850	0.1103	28.35	1.91	14010	11235	1.247
1-7C3	X140	16.3	-16.1	0.776	0.1323	31.05	2.05	13980	11860	1.179
1-3A3	X140	18.3	- 2.5	0.853	0.1885	28.00	2.91	13890	12314	1.128
1-9A3	X140A	16.3	- 2.5	0.860	0.1063	28.05	1.59	4856	4285	1.133

* All the capsules were irradiated at 62.5 MW; the X129 specimens for 7.551×10^5 sec, the X140 specimens for 5.543×10^6 sec, and the X140A specimen for 1.890×10^6 sec.

procedures used for Table AI-1. This time, however, the measured helium concentrations are all systematically higher than the calculated values by amounts averaging 16%. This surprising development and the importance to FFTF programs of knowing the $^{10}\text{B}(n, \alpha)^7\text{Li}$ cross section for fast reactor neutron spectra, made it desirable to conduct a completely independent verification of the calibration of the mass spectrometer system for large helium gas samples. Precisely calibrated glass vials were filled with exactly known quantities of helium and were treated as unknown samples. The mass spectrometric helium measurements differed from the true values by less than 1%, confirming the calibration and linearity of the measuring system, and therefore confirming the high values found for the encapsulated ^{10}B specimens.

As mentioned earlier, the calculated values of helium concentration shown in Tables AI-1 and AI-2 were obtained using the flux-spectral data derived from the HEDL dosimetry foils. The cross section used was the ENDF/B III total helium production cross section, which includes a $\sim 1.5\%$ contribution from the $^{10}\text{B}(n, t)2\alpha$ reaction, modified by Lippincott to include the energy-dependent data from Fricke. If the ENDF/B total helium production cross section is used without the modification, the discrepancy between measured and calculated values increases from ~ 16 to $\sim 32\%$.

The importance of all this, as recognized ever since the program began, is that helium measurements provide a completely independent and absolute normalization base for other (n, α) cross section data. Since ^{10}B is an important standard cross section, these results have important consequences, aside from the effects on B_4C control rod design. The disagreement between calculated and measured helium production rates in boron could come from an incorrect energy dependent cross section, an error in the flux value or neutron energy spectrum, or an error in the helium measurement. The possibility of the measurement being in error by more than 2-3% has been essentially ruled out by the calibration verifications of the mass spectrometer system for large helium samples. At a recent AI-HEDL meeting, however, it was noted that better agreement between calculated and measured helium production rates for ^{10}B specimens was obtained if higher flux values derived from certain selected dosimetry foils were used. In particular, the low energy

flux values obtained from the $^{235}\text{U}(n,f)$ measured reaction rate, which is among the most reliable, along with certain high energy reaction rates obtained from subassembly X140, improved the agreement significantly. This suggests that several other important low energy reaction cross sections [e.g. $^{58}\text{Fe}(n,\gamma)$, $^{59}\text{Co}(n,\gamma)$, $^{45}\text{Sc}(n,\gamma)$] used widely for dosimetry applications, have integral values that, as a group, are systematically in error by as much as 10%. These possibilities are now under investigation at the Fast Reactor Materials Dosimetry Center (FRMDC) at HEDL.

The data in the last column of Table AI-2 were examined for possible correlations or trends. It can quickly be seen that in general, the smaller the specimen of ^{10}B , the more the measured helium exceeds the calculated value. This is the sort of thing that happens when resonance neutron self-shielding occurs. Recent calculations at HEDL indicate, however, that self-shielding phenomena for small samples should be no more than 2-5%. This subject must be pursued.

Comparison of the helium concentration measurements in Tables AI-1 and AI-2 reveals that an average value of 30% of the helium generated in the unencapsulated bare boron specimens is somehow escaping. Surprisingly, a similar fraction seems to be lost whether the helium concentration is 12 ppb or 2000 ppm, whether the boron is natural, enriched ^{11}B or ^{10}B , and whether the boron was irradiated at 100°C or 600°C. This helium loss has been verified by the following additional and more complete experiment. Three additional vanadium encapsulated ^{10}B specimens (not shown in Table AI-2) were irradiated at the three locations in subassembly X129. The only difference between these and the others was that the caps were not sealed by welding, and therefore any helium diffusing out of the ^{10}B crystals was able to escape. The mass spectrometric analyses of the open capsules and contents revealed that only 70, 74 and 78% of the helium was still retained when compared with their respective adjacent samples M1-10B3, M2-10B4 and M3-10B8. When capsule M1-10B3 was being analyzed, moreover, the seal of the

vanadium capsule was broken before the boron was heated. At that time, 29% of the helium was released, leaving 71% retained in the boron crystal for a subsequent analysis, in close agreement with the unwelded capsule datum.

As a result of these comparisons, it was decided during the AI-HEDL meeting that the AI-measured rates of helium production for boron will now be used at HEDL in preference to calculated values for correlations of helium released from B_4C control assemblies. Furthermore, it was decided (subject to RRD concurrence) that all future boron specimens would have to be sealed in miniature capsules before irradiation, most probably in vanadium (because the technology is now on hand, and vanadium has a particularly low (n, α) cross section). As a result, it was decided that the highest priority action would be to irradiate some encapsulated boron specimens as soon as possible in CFRMF. It is anticipated that the sealed ^{10}B capsules in CFRMF will produce more helium than calculations predict.

b. Second AI-CFRMF Irradiation Experiment. A second AI irradiation took place in CFRMF on May 23-24, 1973. The irradiation was similar to the first AI irradiation in April 1972; the main difference being that most of the specimens this time were encapsulated in vanadium to make sure no helium escaped by diffusion. The opportunity was taken to also include along with the ^{10}B , some specimens of enriched 6Li . This is another isotope whose (n, α) cross section is of considerable interest as a basic standard to which others are compared.

1) Isotope Procurement. To prepare for this irradiation, new batches of ^{10}B and 6Li were purchased from ORNL for encapsulation in vanadium. Twenty grams of 92.36% enriched ^{10}B and ten grams of 98.72% enriched 6Li were procured to assure enough material is on hand for all anticipated future needs. These particular isotopic enrichments were selected from those available because the batches from which they come have been recently characterized, and therefore the contents of impurities are better defined. It is anticipated that further measurements of the isotopic enrichments and total impurity contents will later be undertaken using small amounts of the specific materials purchased.

2) Encapsulation Techniques. Vanadium encapsulation of the enriched boron and lithium was accomplished at the Isotope Target Laboratory at ORNL, using techniques developed for containing monitor foils. Miniature capsules (1.27 mm OD, 0.76 mm ID) were manufactured from vanadium wire with a purity of better than 99.9%. The outside lengths of the capsules were 6.4 mm for boron specimens and 8.6 mm for the lithium. No helium was used in the preparation of the capsules.

For the earlier AI-CFRMF irradiation in 1972, relatively large unencapsulated crystals of boron weighing between 2 and 6 mg were included in an aluminum disk capsule. The crystals were irregularly shaped, which has made neutron self-shielding calculations difficult. For this irradiation, it was decided to use ^{10}B powder, as supplied by ORNL's Isotope Sales Section. This was necessary, since it was not possible to fit the desired ~ 3 mg of boron into a ~ 2 cubic millimeter capsule interior when the boron was in the form of small crystals. Filling the vanadium capsule with packed powder has the additional advantage of presenting the boron as a cylinder of known dimensions, which will facilitate self-shielding calculations. At ORNL, 6 capsules were loaded under argon with ^{10}B powder to within an estimated 0.5 ± 0.1 mm of the tops of the capsules. A conical plug "lid" was then tapped in the hole, flush with the top. Finally, the whole end was Tungsten Inert Gas (TIG) welded under argon while the capsule was secured in a copper chill-block.

Loading of three vanadium capsules with enriched ^6Li was accomplished in a somewhat different manner. First a technique was devised at ORNL to convert lithium metal with a die to 0.6 mm wire, using another batch of ^6Li . When the right temperature was reached, the lithium extruded easily. The surface of the wire remained shiny in argon throughout the cutting, weighing and loading operations. Short pieces of 0.6 mm lithium wire approximately 7 mm long were cut and weighed (~ 0.9 mg). Following this, the ^6Li was placed into the capsule, a vanadium spacer was inserted, and then pressure was applied to the spacer to press the wire as far away from the weld zone (top) as possible. The spacer was also intended to reflect heat and otherwise protect the lithium from being heated by the welding. The estimated distance between the top vanadium spacer and the top of the capsule was 1.3 mm. A

conical lid was then inserted and TIG welded. During the development of the technique for encapsulating the lithium, several welded capsules were destructively examined. It was found that as long as an inner vanadium spacer was used, the lithium did not melt. Additional lengths of ^6Li wire were drawn and have been shipped to AI for later use as mass spectrometric "blanks."

3) Mass Determinations. Details of ORNL's multiple weighings of the capsules and specimens are summarized in Table AI-3. For the boron samples, the empty capsule and lid, and the full capsule and lid were weighed 5 times independently. After welding, the capsule was weighed an additional three times. In the case of lithium, the ^6Li slug was itself weighed three times, before being inserted in the vanadium capsule. Following this three independent weighings were made of the empty capsule with spacer and lid, the filled capsule with spacer and lid in place, and the final welded capsule. The individual weighings were estimated by ORNL to have a 1σ absolute uncertainty of 2.5 μg .

Examination of columns 4-6 in Table AI-3 shows that the weighings had an average 1σ relative standard deviation of 1.5 μg . This was sufficiently low to make it possible to get adequate accuracy for the weights of the sample materials, even though they resulted from the subtraction of two large quantities. For the boron, the ~ 3 mg specimens were determined with an average 1σ standard deviation of only 0.09%. The lithium specimens were determined both by subtraction (to within 0.5%), and by the direct measurement of the ^6Li slugs before loading (to within 0.2%). The results by the two methods differed by only $\sim 0.5\%$.

Comparison of columns 5 and 6 in Table AI-3 shows that there were small but real changes in mass resulting from the TIG welding operation. On the average, the vanadium capsules containing boron lost 9 μg , which is exactly what two empty vanadium capsules (not shown in Table AI-3) lost during welding. Two of the three capsules containing lithium gained 5 μg during the welding, the other lost 3 μg . The exact reason for these mass changes is not presently known. Kobisk (ORNL) suggests that two processes are probably occurring. Physical transport of the vanadium away from the weld zone could account for the losses. Oxidation of the weld while it is hot, and acting as a getter, could account

TABLE AI-3
MASS DETAILS OF VANADIUM-ENCAPSULATED ^{10}B AND ^6Li SPECIMENS

Sample No.		Sample Material *	Mass (mg) and 1 σ Standard Deviation					
AI	ORNL		Capsule + Lid + Spacers Empty	Capsule + Lid + Spacers + Sample		Sample Only		
				Unwelded	Welded	From Difference	Direct Measure	Weighted Average
RV10B1	3	¹⁰ B	34.1956 ± 0.0010	36.9630 ± 0.0019	36.9477 ± 0.0010	2.7674 ± 0.0029	-	2.7674 ± 0.0029
RV10B2	5	¹⁰ B	34.0908 0.0010	37.1576 ± 0.0015	37.1507 ± 0.0010	3.0668 ± 0.0025	-	3.0668 ± 0.0025
RV10B3	6	¹⁰ B	34.2040 ± 0.0010	37.1260 ± 0.0014	37.1157 ± 0.0010	2.9220 ± 0.0024	-	2.9220 ± 0.0024
RV10B4	7	¹⁰ B	34.5230 ± 0.0010	37.4232 ± 0.0016	37.4183 ± 0.0021	2.9002 ± 0.0026	-	2.9002 ± 0.0026
RV10B5	8	¹⁰ B	35.0574 0.0010	37.9540 ± 0.0016	37.9503 ± 0.0010	2.8966 ± 0.0026	-	2.8966 ± 0.0026
RV10B6	2	¹⁰ B	34.0298 ± 0.0010	36.4944 ± 0.0019	36.4803 ± 0.0010	2.4646 ± 0.0029	-	2.4646 ± 0.0029
RV6LI1	1	⁶ Li	38.9457 ± 0.0011	39.8407 ± 0.0011	39.8456 ± 0.0021	0.8950 ± 0.0022	0.9003 ± 0.0015	0.8986 ± 0.0032
RV6LI2	3	⁶ Li	39.1583 ± 0.0021	40.1187 ± 0.0012	40.1157 ± 0.0011	0.9604 ± 0.0033	0.9677 ± 0.0025	0.9649 ± 0.0049
RV6LI3	4	⁶ Li	38.5640 ± 0.0036	39.4990 ± 0.0036	39.5047 ± 0.0031	0.9350 ± 0.0072	0.9347 ± 0.0010	0.9347 ± 0.0010

* The ^{10}B and ^6Li are enriched to 92.36% and 98.72% respectively. The mass values, however, are for total boron and lithium in the samples.

for the gains. As long as these gains and losses do not affect the mass content of boron or lithium, however, they are of minor importance to the helium measurement studies. Even if the mass changes were due to the sample materials alone, this would only amount to a 0.4% effect. Since it is much more likely the effect is connected with the vanadium, the problem appears to be unimportant to this work. The conclusion that can be drawn from the encapsulation-welding procedures is that the absolute amounts of boron and lithium are known to better than 0.4%, which is well below the uncertainty levels of other aspects of the total experiment.

After welding, all the vanadium capsules were heated in air for an hour at 300°C. Each capsule was examined under a microscope for cracks and checked for leaks (bubbles) under vacuum in a glycol solution. Following this the capsules were shipped from ORNL to HEDL for loading into the irradiation assembly described below.

4) The AI Irradiation Capsule B3. A single thin aluminum capsule shown in Figure AI-1 was made to hold the vanadium encapsulated specimens for irradiation in CFRMF. A central disk had 4 slots and 3 circular holes, whose identification numbers were determined from the figure. The disk was 2.7 mm thick and had a 0.25 mm foil secured to the base with two 0.56 mm aluminum rivets. A 0.25 mm foil lid was secured to the top with a single rivet which therefore acted as a hinge, allowing the lid to swing aside for sample loading. The whole assembly had a diameter of 13.2 mm. The aluminum capsule was designed to hold the various specimens in a known geometrical configuration during the CFRMF irradiation, and was not intended to be vacuum-tight. Arrows scribed on the outside base and lid were used to orient the capsule toward the zenith during the irradiation. By placing the vanadium capsules parallel to the vertical axis of CFRMF, the samples presented a minimum cross sectional area to neutrons coming down the chamber stem.

5) The Complete Irradiation Package. Details on the loading of samples in AI Capsule B3 and the relative locations of the HEDL dosimetry foils are given in Table AI-4. In addition to the vanadium-encapsulated specimens of ^{10}B and ^6Li , three bare irregular-shaped crystals of 96.50% enriched ^{10}B with nominal masses of 2, 3 and 4 mg, were included in

Figure AI-1: AI Irradiation Capsule B3

TABLE AI-4
FOIL SET AND SAMPLE LOADING DETAILS FOR AI CAPSULE B3**

Sample Material	Sample Number *	Sample Mass (mg)	Description of Miniature Capsule					Relative Location in the Assembly ‡
			Capsule Material	Mass † (mg)		Dimensions (inches)		
				Capsule	Total	Diam.	Length	
²³⁹ Pu	P9-20	264.875	Al	252.225	517.10	0.75	0.0218	Immed. above Ni foil TJ
Ni	TJ	296.10	none	-	-	0.50	0.010	Immed. above foil U5-17
²³⁵ U	U5-17	291.462	Al	251.448	542.91	0.75	0.0208	Immed. above capsule B3
¹⁰ B	RV10B2	3.0668	V	34.084	37.151	0.050	0.25	B3 position 1 (top)
¹⁰ B	RV10B1	2.7674	V	34.180	36.948	0.050	0.25	B3 position 1 (bottom)
⁶ Li	RV6LI2	0.9649	V	39.155	40.116	0.050	0.34	B3 position 2 (top)
⁶ Li	RV6LI1	0.8986	V	38.946	39.846	0.050	0.34	B3 position 2 (bottom)
¹⁰ B	RV10B5	2.8966	V	35.054	37.950	0.050	0.25	B3 position 3 (top)
V	RV1	8.09	none	-	-	0.020	0.09	B3 position 3 (top)
⁶ Li	RV6LI3	0.9347	V	38.564	39.505	0.050	0.34	B3 position 3 (bottom)
¹⁰ B	RV10B4	2.9002	V	34.518	37.418	0.050	0.25	B3 position 4 (top)
¹⁰ B	RV10B3	2.9220	V	34.194	37.116	0.050	0.25	B3 position 4 (bottom)
¹⁰ B	R10B16	2.156	none	-	-	irreg	irreg	B3 position 5
¹⁰ B	R10B12	3.171	none	-	-	irreg	irreg	B3 position 6
¹⁰ B	R10B10	4.662	none	-	-	irreg	irreg	B3 position 7

* Encapsulation was performed at ORNL

† Mass of capsule is value determined before sample was loaded, less the amount lost during welding. The total mass is the measured value (including contents) after welding.

‡ See Figures AI-1 and AI-2

** The total weight of the assembled capsule, including the aluminum foil wrapping, was 2.5461 grams.

Capsule B3. These three specimens were taken from the same batch of ^{10}B used in the first AI-CFRMF irradiation and will be used for comparison of results from the two irradiations, and verification that diffusion of helium from the boron does indeed occur even at the relatively low irradiation temperature of $\sim 100^\circ\text{C}$. The different masses were chosen for possible measurement of neutron self-shielding. A single 8 mg piece of 0.9 mm diameter vanadium wire, from the same lot used to fabricate the capsules, was included for possible helium determinations. The sixth vanadium-encapsulated ^{10}B specimen (RV10B6) was not included and will be analyzed for helium as an unirradiated blank sample.

After it was loaded and the lid secured, Capsule B3 was itself placed with other foils in an aluminum "hat" supplied by HEDL. As shown in Figure AI-2, a nickel foil and single aluminum-encapsulated foils of ^{235}U and ^{239}Pu were placed immediately above Capsule B3. It was felt that these three radiochemical foils would provide the most useful comparison with the ^{10}B helium generation measurements. A single sheet of 0.025 mm aluminum foil was used to hold the capsule and foils together. This "hat" assembly was then inserted into HEDL's duplicate NBS fission chamber, which is shown in detail on page NBS-4 of the 1st ILRR Quarterly (HEDL-TME 71-131). Figure AI-3 shows an X-ray of the hat assembly located in the fission chamber.

6) CFRMF Irradiation Details. The test packet "hat" was placed into the NBS fission chamber assembly with the orientation arrow pointing up, and Au and In power level normalization foils were mounted in their normal positions on the assembly. The CFRMF was then brought to the 6 kW power level for pre-irradiation NBS fission chamber measurements. While at power, the test packet and power normalization foils were inserted into the CFRMF at 09:27:26 MDT on May 23, 1973. At 19:16:25 MDT, the reactor scrammed, due to an electrical power dip. The reactor was returned to the 6 kW power level on a stable period of 23.9 ± 0.1 sec at 19:33:50 MDT, while the foil package remained in the core. This stable period started 4 decades below the 6 kW power level. The test packet and normalization foils were removed at 08:27:26 on

Figure AI-2: Helium Generation Capsule and Dosimetry Foils

AI-17

Figure AI-3: X-Ray of Helium Generation Capsule and Assembly

May 24, 1973, whereupon post-irradiation NBS fission chamber measurements were made.

Following the irradiations, the "hat" was disassembled at ANC and the ^{235}U , ^{239}Pu , Ni, and power normalizing foils were removed for radiochemical counting at ANC. Capsule B3 was returned to AI, and two of the power normalization foils were shipped to HEDL.

Expected Achievements

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

1. Helium measurements will be made on encapsulated specimens of ^{10}B and ^6Li irradiated in CFRMF in May 1973.
2. The results will be compared with earlier measurements made with unencapsulated ^{10}B specimens from the first AI-CFRMF irradiation.
3. The CFRMF data will then be reconciled with ^{10}B helium generation data obtained using EBR-II.
4. The effects of neutron self-shielding will be studied.
5. The results will be presented to the ANS winter meeting, San Francisco in November 1973.
6. The results from the measurements in boron will provide an independent absolute normalization that will help improve the reliability of the high energy portion of the $^{10}\text{B}(n, \alpha)^7\text{Li}$ cross section.
7. Helium measurements in ^{10}B will be used with radiochemical integral data from fission and non-fission reactions, to provide benchmarks for data testing of other evaluated energy-dependent cross sections being developed for the ENDF/B dosimetry file.

BROOKHAVEN NATIONAL LABORATORY

A. Evaluated Cross Section Libraries

B. A. Magurno

NATIONAL NEUTRON CROSS SECTION CENTER

Objective

- Continuation of the comparison of ENDF/B Dosimetry and SAND-II Libraries.

Summary

To complete the comparison of all isotopes presently common to both the ENDF/B and the SAND-II Dosimetry Libraries, the infinitely dilute resonance integrals for nine isotopes (ten reactions) were calculated.

The evaluations for a number of isotopic reactions, unavailable in either of the Dosimetry libraries, were recommended by the Normalization and Standards Subcommittee of CSEWG. One such recommendation was for $^{55}_{25}\text{Mn}((n,2n)^{54}_{25}\text{Mn})$. A preliminary version of the evaluation is included in this report.

Accomplishments and Status

Previous BNL contributions to this Quarterly Report have compared all the fission spectrum averaged cross sections of the isotopes presently common to both the ENDF/B and SAND-II Dosimetry Libraries. Some infinitely dilute resonance integrals were similarly compared. This report will complete the comparison of the resonance integrals of the common isotopes. Table I lists the comparisons for $^6_3\text{Li}(n,\text{total He})$, $^{10}_5\text{B}(n,\text{total He})$, $^{27}_{13}\text{Al}(n,p)^{27}_{12}\text{Mg}$, $^{27}_{13}\text{Al}(n,\alpha)^{24}_{11}\text{Na}$, $^{46}_{22}\text{Ti}(n,p)^{46}_{21}\text{Sc}$, $^{59}_{27}\text{Co}(n,\alpha)^{56}_{25}\text{Mn}$, $^{197}_{79}\text{Au}(n,\alpha)^{198}_{79}\text{Au}$, $^{235}_{92}\text{U}(n,f)\text{F.P.}$, $^{238}_{92}\text{U}(n,f)\text{F.P.}$, and $^{239}_{94}\text{Pu}(n,f)\text{F.P.}$. The second column lists the resonance integrals calculated from the ENDF/B Dosimetry Library.* The third column

*The differences in the last four materials above, as calculated here, and those appearing in ENDF-200 are due to the fact that the resonance integrals in ENDF-200 were calculated from the pointwise data and the integrals calculated here are from the data collapsed into 620 groups.

contains those calculated from the SAND-II Library. Both were calculated using INTER.⁽¹⁾ The fourth column contains some integrals from the SAND-II Library, calculated by Simons et al.⁽²⁾ As mentioned in the previous report⁽³⁾ the ENDF/B Library currently extends to 15 MeV, (the library will be continued to 20 MeV in the near future) while the SAND-II Library terminates at 18 MeV. There will be small differences in the comparisons but these differences are presently ignored for the sake of completeness.

A preliminary version of $^{55}_{25}\text{Mn}(n,2n)^{54}_{25}\text{Mn}$ is being submitted to the appropriate subcommittees of CSEWG for inclusion in the ENDF/B Dosimetry Library. This isotope, recommended for evaluation by the Normalization and Standards Subcommittee of CSEWG is not presently in either the ENDF/B or the SAND-II Dosimetry Libraries. The evaluated curve is shown in Figure 1. The calculated threshold is 10.41 MeV. A cross section at 11 MeV was calculated by H. Takahashi using GROGI-III.⁽⁴⁾ A series of cross sections from 12 to 18 MeV were calculated using thresh,^{(5)*} and a single cross section at 20 MeV was averaged from the data. All the above cross sections were inputted as node points for a spline fit in TIGER. ** The $^{55}_{25}\text{Mn}(n,2n)^{54}_{25}\text{Mn}$, if acceptable, will also be included in the ENDF/B Version IV manganese evaluation currently being worked on by H. Takahashi of Brookhaven National Laboratory.

*No renormalization of the points calculated by THRESH to experimental data was necessary since the evaluated experimental cross section and the cross section calculated by THRESH at 14 MeV were extremely close.

**TIGER - "The Interactive Graphics Evaluation Routine," was developed by C. Rindfleisch, D. I. Garber, and R. R. Kinsey, (BNL).

References:

1. O. Ozer, INTER, A Code to Calculate Selected Averaged Cross Sections, Thermal g-Factors, and Infinitely Dilute Resonance Integrals, BNL-17408.
2. R. L. Simons and W. N. McElroy, BNWL-1312, May 1970.
3. LMFBR Reaction Rate and Dosimetry Seventh Quarterly Progress Report, December 1972 - February 1973.
4. GROGI-III - H. Takahashi, March 1973. A modified version of GROGI-II "A Nuclear Evaporation Computer Code." J. Gilat, BNL-50246, June 1970.
5. S. Pearlstein, "Neutron-Induced Reactions in Medium Mass Nuclei," J. Nucl. Energy 27, 81, 1973.

TABLE BNL-1

Infinitely Dilute Resonance Integral

<u>Isotope</u>	<u>ENDF/B to 15 MeV)</u>		<u>SAND-II (to 18 MeV)^a</u>		<u>SAND-II (to 18 MeV)^b</u>
	<u>MAT#</u>	<u>Barns</u>	<u>Barns</u>		<u>Barns</u>
⁶ Li(n,total He)	6401	425.60	435.91		
¹⁰ B (n,total He)	6402	1722.4	1844.4		
²⁷ Al(n, α) ²⁴ Na	6403	.00834	.00795		
²⁷ Al(n,p) ²⁷ Mg	6403	.00991	.00937		
⁴⁶ Ti(n,p) ⁴⁶ Sc	6404	.36377	.27917		
⁵⁹ Co(n, α) ⁵⁶ Mn	6405	.00176	.00175		
¹⁹⁷ Au(n, γ) ¹⁹⁸ Au	6166	1603.1	1561.6		1585.0
²³⁵ U(n,f)F.P.	6157	282.91	301.57		389.0
²³⁸ U(n,f)F.P.	6158	1.6621	1.8444		
²³⁹ Pu(n,f)F.P.	6159	304.45	325.95		345.6

^aCalculated with INTER.⁽¹⁾

^bCalculated by Simons.⁽²⁾

Figure BNL-1 $^{55}\text{Mn}(n,2n)^{54}\text{Mn}$ Evaluated Cross Section

HANFORD ENGINEERING DEVELOPMENT LABORATORY

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

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

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

Specifications have been established for the fabrication by ORNL of different thicknesses of thin foils for use in neutron self-shielding and thermal yield studies in ILRR program neutron fields. Acquisition of the foils has been initiated and ORNL, NBS, and HEDL will cooperate in establishing and performing the necessary quality assurance tests for these foils. QA work has been initiated at HEDL on ^{235}U , ^{239}Pu , ^{237}Np , and ^{238}U foils in the HEDL-V irradiated ORNL encapsulated high purity fission foil set. Two additional high purity ^{237}Np foils were sent to ORNL for QA work.

Accomplishments and Status

Experimental results (see the HEDL chapter of Ref. 4 and Section C of this report) obtained in CFRMF and $\Sigma\Sigma$ indicate that neutron self-shielding in relatively thin foils (less than ~ 10 mils thick) for non-threshold type reactions is significant. A theoretical capability to properly assess corrections for neutron self-shielding within these foils for the different ILRR test environments has not been developed. Consequently, neutron self-shielding

corrections for individual foils must be obtained experimentally, for each ILRR test environment.

Specifications have been established for the fabrication by ORNL of foils of ~ 0.05 (non-fission foils), ~ 0.1 , ~ 0.5 , ~ 1 , ~ 2 , ~ 5 , and ~ 10 mil thicknesses, depending on the foil material. In addition to Au, Co, Cu, Sc, ^{238}U and ^{235}U , foils of Mn and ^{239}Pu are required for future neutron self-shielding studies. Thin (~ 0.1 mils thick) ^{239}Pu foils are also needed for thermal yield measurements. An inventory of up to 10 to 15 foils of each material of the required thicknesses will be needed for planned tests in CFRMF, EMC, $\Sigma\Sigma$, and BIG-10 and for QA work. ORNL, NBS, and HEDL are cooperating in establishing and performing the necessary quality assurance tests for these foils.

QA work has been started by HEDL on the HEDL-V irradiated high purity fission foils encapsulated by ORNL, as all participants have concurred on the destruction of these foils. The HEDL-V foil set is identified in Figures HEDL-4 and 5 and Table HEDL-5 of reference 6 and contains a ^{235}U , ^{238}U , ^{237}Np , and ^{239}Pu foil. The HEDL results are intended to provide a check and confirmation of the ORNL results reported in this Quarterly report.

Questions concerning ORNL QA of the fission foil materials as listed in the ORNL contribution of this Quarterly resulted in additional review of both the isotopic and elemental data reported. ORNL has confirmed that these data should be used for all the high purity fission foil samples with one exception. Elemental data for the Np material is given for the oxide and not the encapsulated metal foil. Initial analysis by ORNL of the metal indicated an elemental composition of 96.17%. As this result could not be confirmed by additional analysis and was somewhat suspect, it was not documented. This value had been used by some participants, pending additional analysis on two of the high purity ^{237}Np foils. A recent communication from ORNL indicates an average elemental composition of 97.50% for the two high purity ^{237}Np foils N7-2 and N7-6. This value should be used pending documentation by ORNL.

Expected Achievements

Depending on available funding, HEDL will initiate the acquisition from ORNL of sets of thin foils of different thicknesses and different materials

for neutron self-shielding studies in CFRMF, EMC, $\Sigma\Sigma$, and BIG-10 and for thermal yield measurements.

Preliminary QA work on the HEDL-V irradiated (ORNL encapsulated) high purity fission foils and two ^{237}Np foils should be complete during the next reporting period.

B. REACTION RATE MEASUREMENTS

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

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 radio-chemically determined reaction rates (and concurrently fission yields) for EBR-II, future FTR, and other fuels and materials irradiation tests. [The accuracy goals are $\pm 5\%$ (95% confidence level) for ^{235}U , ^{239}Pu , and ^{238}U fission rates and $\pm 5\%$ 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 re-adjustment of evaluated energy dependent cross sections in current use by the FRMDC.

Summary

Documentation is provided on foil set selection, fabrication, and assembly for the scheduled FY-74 ILRR program irradiations. Results of FY-73 ILRR tests related to run-to-run power level normalization, fission product generation rates for ^{235}U , ^{238}U , ^{239}Pu , and ^{237}Np fission foils, measured foil perturbation effects, and neutron self-shielding in non-threshold detectors are documented and discussed. Analysis of available test data indicates that the NBS fission chamber results for ^{235}U and ^{238}U can be used to establish run-to-run power level normalization factors that are accurate to within about 0.5 to 1%. The Au and In power level normalization foils provide back-up results that are somewhat less accurate, in the 1 to 2% range.

Accomplishments and Status

1) Documentation for Scheduled FY-74 ILRR Program Irradiations

Based on the ILRR program meeting of May 1973, planned FY-74 test irradiations involve irradiations in: (1) a thermal and a fission cavity at NBS, (2) CFRMF at ANC, (3) BIG-10 at LASL, and (4) $\Sigma\Sigma$ at Mol. Necessary information for tests 74-1 through 74-10 on foil selection, sample numbers, foil size, weights, and covers for the FY-74 thermal, fission and CFRMF irradiations was provided to program participants. The results of preliminary planning meetings for the FY-74 BIG-10 and $\Sigma\Sigma$ tests were provided to program participants; complete details will be provided later.

A summary listing of all planned FY-74 tests is given in Table 1 for completeness, further test planning, and future reference.

To minimize perturbation effects, the two aluminum encapsulated high purity fission foils designated for each tests 74-4 through 74-4B for CFRMF shall be assembled in a single, thin-walled (~ 1 mil) preformed outer aluminum holder (like that shown for Test HEDL-V in Figure HEDL-4 of reference 6). This holder has been formed so that it will fit in the NBS duplicate fission chamber in the same position as an NBS fission foil. A ~ 43 mil wide by ~ 5 mil thick aluminum spacer ring will be used to provide additional separation for the fission foils within the holder. The ring has a diameter of $\sim 1/2$ inches so that it will be held in place by fitting in one of the groves in the fission foil aluminum cover. Documentation of foil placement, thicknesses, weights, etc., for each test package as well as a radiograph which can be used to determine actual as-built foil placement will be provided. This same type holder will be used for the BIG-10 tests. Similar documentation will be provided in subsequent reports for the other tests.

2) Fission, Neutron Self-Shielding, and Power Level Normalization Foils-Measured Reaction Rates

Results of HEDL's radiometric measurements of reaction rates for power level normalization foils and for fission and neutron self-shielding foil sets irradiated in CFRMF and/or EMC in FY-73 are documented and briefly discussed here.

TABLE HEDL-1
PLANNED FY-74 ILRR PROGRAM TEST IRRADIATIONS

Test Number(a) Laboratory, and Time of Irradiation, If Noted	Test Designation and/or Sample Identification	Irradiations Completed by
<u>UNIRRADIATED FOILS</u>		
74-0 (all laboratories)	Background Count of aluminum encapsulated 5 mil thick fission foils: [^{239}Pu (P9-21), ^{237}Np (N7-20), ^{238}U (U8-19), ^{235}U (U5-9), and one 0.05 mil thick ^{235}U (#5) and one 10 mil thick ^{235}U (#3) self-shielding foil].	Un-irradiated reference foil set for periodic counting
<u>NBS-THERMAL NEUTRON FIELD</u>		
74-1(235) (HEDL)	^{140}Ba , ^{95}Zr , ^{97}Zr , ^{103}Ru , ^{132}Te , ^{137}Cs , etc. fission product generation rates. ~0.05 mil ^{235}U(#3)(b)	Nov '73
74-2(235) (ANC)	" " ~0.1 mil ^{235}U(#1)	"
74-3(235) (ANL)	" " ~0.1 mil ^{235}U(#3)	"
74-MOL-1(235)	" " ~0.1 mil ^{235}U(#2)	after Jan '74
74-1(239)(c) (HEDL)	" " ~0.1 mil ^{239}Pu foil	April '74
74-2(239)(c) (ANC)	" " ~0.1 mil ^{239}Pu foil	"
74-3(239)(c) (ANL)	" " ~0.1 mil ^{239}Pu foil	"
<u>ANC-CFRMF NEUTRON FIELD</u>		
74-4 (ANC) (14 hrs)	^{140}Ba , ^{95}Zr , ^{97}Zr , ^{103}Ru , ^{132}Te , ^{137}Cs , etc. fission product generation rates 5 mil ^{235}U(U5-11) ^{239}Pu(P9-14)	Feb. '74
74-MOL-2(d)	" " 2 mil ^{235}U(#1)	"
74-HEDL-1(d)	" " 2 mil ^{235}U(#3)	"
74-5 (ANC) (14 hrs)	" " 5 mil ^{238}U(U8-16) ^{237}Np(N7-16)	"
74-LASL-1(d)	" " 2 mil ^{235}U(#2)	"
74-ANC-1(d)	" " 2 mil ^{235}U(#4)	"
74-6A (HEDL-ARHCO) (7 hrs)	" " 5 mil ^{235}U(5-12) ^{239}Pu(9-15)	"
74-MOL-3(d)	" " 10 mil ^{238}U(#5)	"
74-LASL-2(d)	" " 10 mil ^{238}U(#2)	"

TABLE HEDL-1 (Cont'd)

Test Number(a) Laboratory, and Time of Irradiation, If Noted	Test Designation and/or Sample Identification	Irradiations Completed by
ANC CFRMF NEUTRON FIELD (Continued)		
74-6B (HEDL-ARHCO) (7 hrs)	^{140}Ba , ^{95}Zr , ^{97}Zr , ^{103}Ru , ^{132}Te , ^{137}Cs , etc. fission product gener- ation rates.	5 mil $^{238}\text{U}(\text{U8-17})$ $^{237}\text{Np}(\text{N7-17})$
74-MOL-4(d)	^{198}Au generation rate	2 mil $^{197}\text{Au}(\text{LD})$
74-LASL-3(d)	"	2 mil $^{197}\text{Au}(\text{LE})$
74-7A (ANL) (7 hrs)	^{140}Ba , ^{95}Zr , ^{97}Zr , ^{103}Ru , ^{132}Te , ^{137}Cs , etc. fission product gener- ation rates	5 mil $^{235}\text{U}(\text{U5-13})$ $^{239}\text{Pu}(\text{P9-16})$
74-MOL-5(d)	^{58}Co generation rate	10 mil $^{58}\text{Ni}(\text{MK})$
74-LASL-4(d)	" "	10 mil $^{58}\text{Ni}(\text{ML})$
74-7B (ANL) (7 hrs)	^{140}Ba , ^{95}Zr , ^{97}Zr , ^{103}Ru , ^{132}Te , ^{137}Cs , etc. fission product gener- ation rates.	5 mil $^{238}\text{U}(\text{U8-18})$ $^{237}\text{Np}(\text{N7-18})$
74-AI (~2 days)	AI helium specimen test - to be specified.	[Boron & Lithium] July '74
74-ANL	ANL Track recorder test - to be specified.	[Fission foils] "
74-HEDL-ANL	HEDL-ANL Neutron self shielding measurements - to be specified.	[^{239}Pu foils-0.1, 5, and 10 mils] "

- (a) NBS fission chamber (and as necessary ANL track recorder measurements) will be made to provide unperturbed values of absolute fission rates in each neutron field. Though not explicitly identified, these measurements are an integral part of each test.
- (b) Foil used in HEDL self-shielding set for CFRMF.
- (c) This set of tests may be accomplished instead of tests 74-8A through 74-12.
- (d) These foils will be placed on the normalization foil holder for subsequent counting by Mol (A. Fabry) and LASL (D. Barr) to provide an initial interlaboratory calibration check prior to the BIG-10 irradiations. The two 2 mil ^{235}U foils (74-MOL-2 and 74-HEDL-1, for test 74-4, and 74-LASL-1 and 74-ANC-1, for test 74-5) will be placed together so that one 2 mil ^{235}U foil can also be counted by HEDL and one by ANC for a direct comparison without the necessity of gradient corrections. For test 74-6B, the 2 mil gold foils for MOL and LASL will be placed on opposite sides of the normalization foil holder on top of 2 mil indium foils in the two empty foil positions. Before shipment of the Au foils to MOL and LASL, these extra foils will be counted by ANC together with the other two Au and In run-to-run power level normalization foils to provide data for a check on any flux asymmetry.

TABLE HEDL-1 (Cont'd)

Test Number(a) Laboratory, and Time of Irradiation, If Noted	Test Designation and/or Sample Identification	Irradiations Completed by
<u>NBS-FISSIION NEUTRON FIELD</u>		
74-8A (HEDL-ARHCO)	^{140}Ba , ^{95}Zr , ^{97}Zr , ^{103}Ru , ^{132}Te , etc. fission product generation rates. $\left[\begin{matrix} 5 \text{ mil} \\ ^{235}\text{U}(U5-14) \end{matrix} \right]$	Aug '74
74-8B (HEDL-ARHCO)	" " $\left[\begin{matrix} 5 \text{ mil} \\ ^{239}\text{Pu}(P9-17) \end{matrix} \right]$	"
74-9A (ANL)	" " $\left[\begin{matrix} 5 \text{ mil} \\ ^{235}\text{U}(U5-15) \end{matrix} \right]$	"
74-9B (ANL)	" " $\left[\begin{matrix} 5 \text{ mil} \\ ^{239}\text{Pu}(P9-18) \end{matrix} \right]$	"
74-10A (ANC)	" " $\left[\begin{matrix} 5 \text{ mil} \\ ^{235}\text{U}(U5-16) \end{matrix} \right]$	"
74-10B (ANC)	" " $\left[\begin{matrix} 5 \text{ mil} \\ ^{239}\text{Pu}(P9-19) \end{matrix} \right]$	"
74-11 (ANL)	ANL Track Recorder Fission Rates $[^{235}\text{U-SSTR}]$	"
74-12 (ANL)	" " $[^{239}\text{Pu-SSTR}]$	"
<u>LASL - BIG-10 NEUTRON FIELD</u>		
74-13 (HEDL-ARHCO)	^{140}Ba , ^{95}Zr , ^{103}Ru , ^{132}Te , etc. fission product generation rates. $\left[\begin{matrix} 5 \text{ mil} \\ ^{235}\text{U}(U5-6) \\ ^{239}\text{Pu}(P9-8) \\ ^{238}\text{U}(U8-9) \\ ^{237}\text{Np}(N7-10) \end{matrix} \right] (e)$	May '74
74-14 (LASL)	" " $\left[\begin{matrix} 5 \text{ mil} \\ ^{235}\text{U}(U5-7) \\ ^{239}\text{Pu}(P9-10) \\ ^{238}\text{U}(U8-10) \\ ^{237}\text{Np}(N7-12) \end{matrix} \right] (e)$	"
74-15 (ANC)	" " $\left[\begin{matrix} 5 \text{ mil} \\ ^{235}\text{U}(U5-8) \\ ^{239}\text{Pu}(P9-11) \\ ^{238}\text{U}(U8-11) \\ ^{237}\text{Np}(N7-13) \end{matrix} \right] (e)$	"
74-16 (ANL)	^{140}Ba , ^{95}Zr , ^{103}Ru , ^{132}Te , etc. fission product generation rates. $\left[\begin{matrix} 5 \text{ mil} \\ ^{235}\text{U}(U5-10) \\ ^{239}\text{Pu}(P9-12) \\ ^{238}\text{U}(U8-13) \\ ^{237}\text{Np}(N7-14) \end{matrix} \right]$	May '74
74-17 (MOL)	" " $\left[\begin{matrix} 5 \text{ mil} \\ ^{235}\text{U}(U5-5) \\ ^{239}\text{Pu}(P9-7) \\ ^{238}\text{U}(U8-14) \\ ^{237}\text{Np}(N7-15) \end{matrix} \right]$	"
74-18 (MOL)	Mol Track Recorder - reaction rates $\left[\begin{matrix} ^{235}\text{U}, ^{238}\text{U}, \\ ^{239}\text{Pu}, ^6\text{Li}, \\ \text{and } ^{10}\text{B} \end{matrix} \right]$	"

(e) The grouping of these foils has not been defined as yet, but may be the same as for the CFRMF tests 74-4 and 74-5, 74-6 and 74-6B, etc; i.e., two foils per test.

TABLE HEDL-1 (Cont'd)

Test Number(a) Laboratory, and Time of Irradiation, If Noted	Test Designation and/or Sample Identification	Irradiations Completed by
<u>LASL - BIG-10 NEUTRON FIELD (Cont'd)</u>		
74-19 (AI)	Boron and Lithium helium generation [^{10}B and ^6Li] rates.	May '74
74-20 (HEDL-ARHCO)	Non-Fission Foil Set - reaction rates. [**]	July '74
74-21 (LASL)	" " "	"
74-22 (ANC)	" " " (f)	"
74-23 (ANL)	" " "	"
74-24 (MOL)	" " "	"
74-25 (LASL-ANC)	Self-shielding foil set reaction rates [^{235}U , ^{239}Pu , ^{238}U , ^{197}Au , ^{55}Mn , ^{59}Co , ^{63}Cu , ^{45}Sc]	"
<u>MOL - $\Sigma\Sigma$ NEUTRON FIELD</u>		
73- $\Sigma\Sigma$ -1 (ANL)	ANL Track Recorder fission rates [fission foils]	Completed
74- $\Sigma\Sigma$ -1 (MOL)	^{140}Ba , ^{95}Zr , ^{97}Zr , ^{103}Ru , ^{132}Te , etc. fission product generation rates. [5 mil ^{235}U (U5-4) ^{239}Pu (P9-6) ^{238}U (U8-15) ^{237}Np (N7-11)]	May '74
74- $\Sigma\Sigma$ -2 (ANC, ANL, HEDL)	" " [5 mil ^{235}U (U5-3) ^{239}Pu (P9-5) ^{238}U (U8-8) ^{237}Np (N7-9)]	Aug '74
74- $\Sigma\Sigma$ -3 (MOL)	Self-shielding foil set reaction rates [^{235}U , ^{197}Au , ^{63}Cu , etc.]	July '74 (Cont'g.)
74- $\Sigma\Sigma$ -4 (NBS-MOL)	NBS fission chamber fission rates [^{235}U , ^{239}Pu , ^{238}U , ^{237}Np]	July '74 (Cont'g.)
74- $\Sigma\Sigma$ -5 (AI)	AI helium specimen test - to be specified [Boron & Lithium]	Dec '74

** ^{197}Au (n, γ) ^{198}Au , ^{45}Sc (n, γ) ^{46}Sc , ^{63}Cu (n, γ) ^{64}Cu , ^{23}Na (n, γ) ^{24}Na , ^{58}Fe (n, γ) ^{59}Fe , ^{238}U (n, γ) ^{239}U , ^{238}U (n,f), ^{55}Mn (n, γ) ^{56}Mn *, ^{59}Co (n, γ) ^{60}Co †, ^{115}In (n,n') $^{115\text{m}}\text{In}$, ^{58}Ni (n,p) ^{58}Co , ^{54}Fe (n,p) ^{54}Mn †, ^{46}Ti (n,p) ^{46}Sc , ^{47}Ti (n,p) ^{47}Sc , ^{48}Ti (n,p) ^{48}Sc , ^{56}Fe (n,p) ^{56}Mn *, ^{27}Al (n,p) ^{27}Mg *, ^{27}Al (n, α) ^{24}Na , ^{63}Cu (n, α) ^{60}Co †, ^{127}I (n,2n) ^{126}I , and a single ^{235}U foil to be used with the ^{238}U foil for run-to-run fission rate normalization. (*Because of short half life, all counting must be accomplished at LASL). (†A single set of long half life reaction foils might be used and placed at a specific site adjacent to the power level normalization foils for a long cumulative exposure during the entire series of irradiations. All participating laboratories could count this one set of foils).

(f) Thin, essentially infinitely dilute, non-threshold foils are to be used in the ANC foil package.

Further reactor operations information and power level normalization foil results on these tests are provided in the ANC chapters of these reports⁽¹⁻⁷⁾. The NBS chapters provide information on NBS run-to-run fission chamber power level measurements. The combined results of all available run-to-run power level normalization measurements for the FY-73 high power CFRMF tests are analyzed in Section B-3 in order to establish a consistent set of run-to-run relative power level normalization factors.

The results of HEDL's radiochemistry measurements for the CFRMF irradiated HEDL self-shielding sets, the HEDL-V and HEDL-VI fission foil sets, and AI-II foil set, corrected for elemental and isotopic composition, and the EMC foils irradiated at Argonne are documented in Table 2. A comparative tabulation of all participants ^{235}U and ^{239}Pu results, (after correction of the fission foil data by HEDL) as well as some non-fission foil results, can be found in Table 8A. Some participant data is tentative in that final documentation of their results is not available at this time. Blank spaces are also left in Table 8 for the future addition of un-reported results. As stated in the 5th Quarterly report, all HEDL results are based on the use of nuclear parameters submitted by ANC and adopted by the ILRR participants^(4,5). With the exception of ^{64}Cu , no results have been changed to conform with the nuclear parameters listed in a recent review made by R. G. Helmer of ANC as the effect of such changes on the reported HEDL data appears to be less than $\sim 0.1\%$. Gamma ray self absorption corrections have been made using the correction factors given in Table 3. Table 4 contains the self-shielding ratio data from all participants with relative precision estimates, if specified. Again some data is tentative pending final documentation by the participants.

3) Run-To-Run Power Level Normalization Factors for FY-73 High Power CFRMF Tests

Results of NBS fission chamber and ANC and HEDL gold and indium normalization foil measurements for CFRMF tests HEDL-V, HEDL-VI, AI-II, ANC-ARHCO-HEDL-SS-I, HEDL-SS-II, and HEDL-SS-III are gathered in Table 5. Reference is made to the NBS, ANC, and HEDL chapters of this and previous reports⁽³⁻⁷⁾ for necessary documentation on the individual measurements.

TABLE HEDL-2
MEASURED GENERATION RATES WITH ABSOLUTE ERROR ESTIMATES

Date	Test	Reaction	Foil No.	Foil Thickness (mil)	Foil (a) Weight (mg)	Δt to First Count (days)	No. of Measure-ments	Measured Generation Rate x 10 ¹⁵ observed(b)	Corrected (c)	Error Est. (2σ)
<u>NORMALIZATION FOILS</u>										
3/13-14/73	HEDL-SS-I	¹¹⁵ In(n,n') ^{115m} In	30	2	46.818	0.686	3	6.975	6.992	4.4
3/14/73	HEDL-SS-II	"	32	2	48.194	0.572	4	6.953	6.971	4.5
3/14/73	HEDL-SS-III	"	34	2	47.170	0.524	3	6.981	6.999	4.4
5/24/73	AI-II	"	36	2	48.159	0.672	3	4.043	4.054	4.2
3/13-14/73	HEDL-SS-I	¹⁹⁷ Au(n,γ) ¹⁹⁸ Au	CJ	2	124.097	13.43	4	53.51	54.05	2.1
3/14/73	HEDL-SS-II	"	DB	2	123.566	1.48	3	53.60	54.13	2.3
3/14/73	HEDL-SS-III	"	DD	2	122.119	1.40	4	53.82	54.36	2.0
5/24/73	AI-II	"	DF	2	122.278	7.02	7	30.35	30.66	2.2
<u>SELF-SHIELDING FOIL SETS</u>										
3/13-14/73	HEDL-SS-I	¹⁹⁷ Au(n,γ) ¹⁹⁸ Au	#3	0.05	4.306	1.51	3	60.85	60.86	2.0
		⁵⁹ Co(n,γ) ⁶⁰ Co	#3	0.05	1.529	22.06	2	13.03	13.03	6.2
		⁶³ Cu(n,γ) ⁶⁴ Cu	#3	0.05	1.561	0.849	2	5.798	5.798	3.1
		⁴⁵ Sc(n,γ) ⁴⁶ Sc	#3	0.05	0.430	34.91	2	----	3.278	4.3
		²³⁸ U(n,γ) ²³⁹ Np	#3	0.05	2.990	1.59	4	----	23.42	5.9
					(2.9744)					
		²³⁵ U(n,f) ¹⁴⁰ Ba	#3	0.05	3.220	6.79	3	13.554	13.565	3.5
					(3.1983)					
		¹⁰³ Ru				6.79	3	7.295	7.301	5.0
		⁹⁵ Zr				42.1	2	14.405	14.414	4.8

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TABLE HEDL-2 (Cont'd)

Date	Test	Reaction	Foil No.	Foil Thickness (mil)	Foil (a) Weight (mg)	Δt to First Count (days)	No. of Measurements	Measured Rate $\times 10^{15}$ observed (b)	Generation Corrected (c)	Error Est. (2 σ)
3/14/73	HEDL-SS-II	$^{197}\text{Au}(n,\gamma)$	#1	0.5	36.832	1.44	3	59.25	59.40	2.0
		$^{59}\text{Co}(n,\gamma)$	#1	1.5	44.278	29.36	2	10.06	10.07	6.0
		$^{63}\text{Cu}(n,\gamma)$	#1	0.5	16.363	0.794	3	5.610	5.610	3.0
		$^{45}\text{Sc}(n,\gamma)$	#1	5	47.70	33.70	1	-----	3.246	2.8
		$^{238}\text{U}(n,\gamma)$	#1	1	63.020	1.55	4	-----	22.62	4.8
					(62.6914)					
		$^{235}\text{U}(n,f)^{140}\text{Ba}$	#1	5	305.325	10.99	3	13.340	13.426	2.1
		^{103}Ru			(303.2702)	10.99	3	7.025	7.223	5.1
		^{95}Zr				42.4	2	14.057	14.255	5.8
		$^{197}\text{Au}(n,\gamma)$	JJ	2	123.333	5.48	2	55.64	56.20	2.4
		$^{59}\text{Co}(n,\gamma)$	#1	2	57.395	30.41	2	9.128	9.208	6.0
		$^{63}\text{Cu}(n,\gamma)$	#1	1	31.337	0.710	3	5.439	5.439	3.1
3/14/73	HEDL-SS-III	$^{45}\text{Sc}(n,\gamma)$	#1	10	97.75	34.31	2	-----	3.188	2.3
		$^{238}\text{U}(n,\gamma)$	#1	5	310.090	5.78	3	-----	22.22	4.2
					(308.4730)					
		$^{235}\text{U}(n,\gamma)^{140}\text{Ba}$	#1	10	603.605	11.77	3	13.169	13.336	2.1
					(599.5428)					
		^{103}Ru				11.77	3	6.718	7.069	5.8
		^{95}Zr				41.6	2	13.750	14.133	4.0

TABLE HEDL-2 (Cont'd)

Date	Reaction	Foil No.	Foil Thickness (mil)	Foil (a) Weight (mg)	Δt to First Count (days)	No. of Measurements	Measured Rate x 10 ¹⁵ observed (b)	Generation corrected (c)	Error Est. (2σ)						
HIGH PURITY ORNL ENCAPSULATED FISSION FOILS															
11/6/73	HEDL-V	²³⁵ U(n,f) ¹⁴⁰ Ba	21	5.2	287.161	15.08	2	7.276	7.337	2.2					
					¹⁰³ Ru	(285.2284)	23.24	4	3.80	3.90	5.9				
						⁹⁵ Zr		136.5	3	7.660	7.787	3.7			
		²³⁸ U(n,f) ¹⁴⁰ Ba		21	5.2	311.475	14.63	3	0.355	0.358	2.3				
						¹⁰³ Ru	(309.8507)	22.69	4	0.375	0.386	8.2			
							⁹⁵ Zr		137.5	3	0.303	0.308	5.3		
		²³⁹ Pu(n,f) ¹⁴⁰ Ba		22	5.7	263.707	15.56	2	7.360	7.419	2.1				
						¹⁰³ Ru	(256.7584)	138.3	2	9.158	9.392	6.2			
							⁹⁵ Zr		138.3	2	6.541	6.635	4.4		
		²³⁷ Np(n,f) ¹⁴⁰ Ba		21	6.5-6.8	318.671	18.49	2	2.562[2.527]*	2.591[2.556]*	3.2				
						¹⁰³ Ru	(306.4352)	32.18	2	2.403[2.370]*	2.496[2.462]*	8.8			
							⁹⁵ Zr	[310.6732]*	32.18	2	2.682[2.645]*	2.741[2.704]*	9.0		
						²³⁵ U(n,f) ¹⁴⁰ Ba	20	5.1	282.713	19.63	2	7.313	7.373	2.3	
									¹⁰³ Ru	(280.8104)	23.08	4	3.861	3.963	7.4
										⁹⁵ Zr		134.5	2	7.712	7.839
ENGINEERING MOCKUP CRITICAL (EMC)															
11/27/72	EMC	²³⁵ U(n,f) ¹⁴⁰ Ba	5-5	~5	515.82	24.35	1	0.615	0.619	3.8					
						¹⁰³ Ru	24.35		0.310	0.319	4.9				
		²³⁸ U(n,f) ¹⁴⁰ Ba			8-5	~5	557.35	16.29	1	0.0863	0.0870	4.6			
								¹⁰³ Ru	16.29		0.0899	0.0925	6.5		
		²³⁸ U(n,γ) ²³⁹ Np				16.29		7.031	7.62	10.4					

*Based on elemental composition of 97.50% instead of 96.16%.

TABLE HEDL-2 (Cont'd)

Date	Reaction	Foil No.	Foil Thickness (mil)	Foil (a) Weight (mg)	Δt to First Count (days)	No. of Measurements	Measured Rate x 10 ¹⁵ observed(b)	Generation corrected(c)	Error Est. (2σ)	
AI--BORON-10 AND LITHIUM-6										
5/23-24/73	AI-II	²³⁵ U(n,f) ¹⁴⁰ Ba	17	5.2	291.462	54.13	3	7.480	7.544	3.0
		¹⁰³ Ru			(289.5005)	54.13	3	3.952	4.052	7.2
		⁹⁵ Zr				54.13	3	8.067	8.198	5.6
		²³⁹ Pu(n,f) ¹⁴⁰ Ba	20	5.6	264.875	54.53	3	7.512	7.573	3.3
		¹⁰³ Ru			(257.8956)	54.53	3	9.419	9.647	7.9
		⁹⁵ Zr				54.53	3	6.717	6.817	5.8
		⁵⁸ Ni(n,p) ⁵⁸ Co	TJ	10	296.065	54.47	3	1.995	2.009	3.5

- (a) All samples were reweighed prior to counting with the exception of the encapsulated foils. For the fission foils, the initial weight listed is that documented by ORNL. The second weight (xxxx) is the actual target isotope weight after correcting for elemental and isotopic composition using the data presented by ORNL at the May 1973 program review meeting and in this progress report.
- (b) Data uncorrected for gamma ray shielding or self-absorption. No corrections have been made for neutron scattering, absorption, self-shielding effects or fission yields. All other corrections for count rate loss as discussed in previous quarterly reports, as well as possible incomplete ^{239}Np ingrowth and ^{239}Np equilibrium have been made.
- (c) Corrected for gamma-ray shielding and self-absorption--correction factors are given in Table HEDL-3.

TABLE HEDL-3
 γ SELF ABSORPTION

Irradiation	Reaction	E ₀ (kev)	Foil Thickness (mil)						
			0.05	0.5	1	1.5	2	5	10
Self Shielding (SS)	⁴⁵ Sc(n,γ) ⁴⁶ Sc	889	0.9995*	0.9999	0.9998	0.9996	0.9995	0.9988	0.9976
		1121	0.9996*	0.9999	0.9998	0.9997	0.9996	0.9989	0.9979
	⁵⁹ Co(n,γ) ⁶⁰ Co	1173	0.9999	0.9997	0.9994	0.9991	0.9988	0.9969	0.9939
		1332	0.9999	0.9997	0.9994	0.9991	0.9989	0.9971	0.9943
	¹⁹⁷ Au(n,γ) ¹⁹⁸ Au	411	0.9998	0.9975	0.9950	0.9925	0.9901**	0.9754	0.9516
	²³⁵ U(n,f) ⁹⁵ Zr	724	0.9994*	--	--	--	--	0.9861*	0.9729*
		756	0.9994*	--	--	--	--	0.9868*	0.9744*
	⁹⁵ Nb	765	0.9994*	--	--	--	--	0.9870*	0.9748*
	¹⁰³ Ru	496	0.9992*	--	--	--	--	0.9727*	0.9503*
	¹⁴⁰ Ba	1596	0.9992*	--	--	--	--	0.9936*	0.9875*
	²³⁸ U(n,γ) ²³⁹ Np	104 }	0.9969*	0.9606*	0.8259*	--	0.9268*	--	--
		106 }							
		277	0.9985*	--	0.9842*	--	--	0.9268*	--
	¹¹⁵ In(n,n') ^{115m} In	336	--	--	--	--	0.9974**	--	--
	HEDL-V, VI (***)	Isotope	E ₀	²³⁵ U(5-20)	²³⁵ U(5-21)	²³⁸ U(8-21)	²³⁹ Pu(9-22)	²³⁷ Np(7-21)	
		⁹⁵ Zr	724	0.9839	0.9837	--	0.9850	0.9774	
			756	0.9846	0.9844	--	0.9857	0.9785	
⁹⁵ Nb		766	0.9848	0.9846	--	0.9858	0.9773		
¹⁰³ Ru		496	0.9741	0.9738	--	0.9751	0.9629		
¹⁴⁰ Ba		1596	0.9917	0.9917	--	0.9920	0.9887		
²³⁹ Np		104 }	--	--	0.8179	--	--		
		106 }	--	--	--	--	--		
		277	--	--	0.9219	--	--		
EMC (****)		Isotope	E ₀	²³⁵ U(5-5)	²³⁸ U(8-5)	²³⁹ Pu(N7A)			
	⁹⁵ Zr	724	0.9830	0.9828	0.9855				
	¹⁰³ Ru	496	0.9727	0.9724	0.9758				
	¹⁴⁰ Ba	1596	0.9913	0.9913	0.9922				
	²³⁹ Np	277	--	0.9234	--				
	Isotope	E ₀	Ni-TJ	²³⁵ U(5-17)	²³⁹ Pu(9-20)				
	⁵⁸ Co	813	0.9928	---	--				
	⁹⁵ Zr	724	--	0.9835	0.9849				
	¹⁰³ Ru	496	--	0.9754	0.9764				
	¹⁴⁰ Ba	1596	--	0.9916	0.9919				
AI-II (***)									

* These corrections contain both self shielding for the respective foil thickness and an external absorption correction for those foils with a 1 mil Al wrap.

** Correction for normalization foils.

*** The foil effective thickness was determined using the actual weight and foil diameter, with the exception of ²³⁷Np, as discussed in HEDL-TME 72-154. These corrections include that for 7.4 mil Al encapsulation material.

**** These corrections are based on a nominal 5 mil foil thickness given by ANL and an assumed 7.4 mil Al encapsulation.

TABLE HEDL-4
HEDL FOIL SETS SELF SHIELDING FOIL RATIOS (FOIL/REFERENCE 0.05 MIL FOIL)

Foil Reaction	Thickness (mil)	HEDL		ARHCO		ANC (Greenwood and Helmer)		ANC (Rogers)	
		Ratio	Relative % (1 σ Precision)	Ratio	Relative % (1 σ Precision)	Ratio	Relative % (1 σ Precision)	Ratio	Relative % (1 σ Precision)
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	0.05	1.0000	----	-----	-----	-----	-----	-----	-----
	0.5	0.9759	0.20	-----	-----	-----	-----	-----	-----
	2.0	0.9233	0.25	-----	-----	-----	-----	-----	-----
$^{45}\text{Sc}(n,\gamma)^{46}\text{Sc}$	0.05	1.000*	----	-----	-----	1.000	-----	1.000	-----
	5	0.986	1.4	-----	-----	0.990	-----	0.990	-----
	10	0.975	1.3	-----	-----	0.982	-----	0.970	-----
$^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$	0.05	1.000	----	-----	-----	-----	-----	-----	-----
	1	0.967	0.6	-----	-----	-----	-----	-----	-----
	5	0.940	0.8	-----	-----	-----	-----	-----	-----
$^{59}\text{Co}(n,\gamma)^{60}\text{Co}$	0.05	1.000	----	1.000	-----	-----	-----	-----	-----
	1.5	0.773	1.7	0.804	-----	-----	-----	-----	-----
	2	0.706	1.5	0.698	-----	-----	-----	-----	-----
$^{238}\text{U}(n,\gamma)^{239}\text{Np}$	0.05	1.000	----	-----	-----	-----	-----	-----	-----
	5	0.966	1.7	-----	-----	-----	-----	-----	-----
	10	0.949	1.1	-----	-----	-----	-----	-----	-----
$^{235}\text{U}(n,f)^{140}\text{Ba}$	0.05	1.000	----	1.000	-----	1.000	-----	1.000**	-----
	5	0.990	1.5	0.999	2.6	0.996	-----	1.015**†	-----
	10	0.983	1.6	0.978	2.3	0.974	-----	1.011**†	-----
$^{235}\text{U}(n,f)^{103}\text{Ru}$	0.05	1.000	----	1.000	-----	1.000	-----	-----	-----
	5	0.989	2.4	0.954†	2.6	0.991	-----	-----	-----
	10	0.968	2.3	0.917†	2.4	0.986	-----	-----	-----
$^{235}\text{U}(n,f)^{95}\text{Zr}$	0.05	1.000	----	1.000	-----	1.000	-----	-----	-----
	5	0.989	3.2	-----	-----	0.980	-----	-----	-----
	10	0.980	2.3	0.967	1.9	0.976	-----	-----	-----

*This ratio was determined using a counting system not calibrated or checked for ILRR usage, due to the low count rates. Only relative measurements are used from this system.

**Based on results given in Table ANC-8, this report.

†Not used to obtain average values for Table 7 and Figure 2.

TABLE HEDL-5
CFRMF RUN-TO-RUN FISSION CHAMBER AND Au AND In POWER LEVEL NORMALIZATION FOIL MEASUREMENTS

Test (a)	NBS Fission Chamber		Average Value ^{235}U and ^{238}U	$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$			Average Value ^{197}Au	$^{115}\text{In}(n,n')^{115m}\text{In}$			Average Value ^{115}In	Reactor Power Instr.	
	^{235}U	^{238}U		HEDL	ANC-I ^(b)	ANC-II ^(c)		HEDL	ANC-I ^(b)	ANC-II ^(c)			
	(Fissions/second in foil)			$10^{16} \times$ (reactions/second/nucleus)				$10^{16} \times$ (reactions/second/nucleus)					
	Values			Values				Values					
HEDL-VI	1599±0.20% ^(e)	3762±0.17% ^(e)	-----	303.7	299.2	299.4	-----	39.13	37.82	38.12	-----	6.0 kW	
HEDL-V	1597±0.14% ^(e)	3760±0.10% ^(e)	-----	300.0	298.9	298.4	-----	38.76	37.89	37.97	-----	6.0 kW	
AI-II	1638±0.27% ^(e,f)	3850±0.1% ^(e,f)	-----	306.6	306.0 ^(d)	302.5 ^(d)	-----	40.5	39.3 ^(d)	38.4 ^(d)	-----	6.0 kW	
	Ratio to HEDL VI			Ratio to HEDL VI				Ratio to HEDL VI					
HEDL-VI	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0	
HEDL-V	0.9987	0.9995	0.9991	0.9878	0.9990	0.9967	0.9945	0.9905	1.0019	0.9961	0.9962	1.0	
AI-II	1.024	1.023	1.024	1.0095	1.0227	1.0104	1.0142	1.0350	1.0391	1.0073	1.0271	1.0	
	Values			Values				Values					
ANC/ARHCO/HEDL SS-I	2861±0.23% ^(g)	6720±0.44% ^(g)	-----	540.5	537.4	532.9	-----	69.92	68.14	67.05	-----	10.4	
HEDL-SS-II	2871±0.23% ^(h)	6712±0.44% ^(h)	-----	541.3	539.1	533.3	-----	69.71	68.62	67.93	-----	10.4	
HEDL-SS-III	2871±0.23% ^(h)	6712±0.44% ^(h)	-----	543.6	544.2	535.8	-----	69.99	68.66	67.87	-----	10.4	
	Ratio to SS-I			Ratio to SS-I				Ratio to SS-I					
ANC/ARHCO/HEDL SS-I	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0	
HEDL-SS-II	1.0035	0.9988	1.0012	1.0015	1.0032	1.0008	1.0018	0.9970	1.0070	1.0131	1.0057	1.0	
HEDL-SS-III	1.0035	0.9988	1.0012	1.0057	1.0127	1.0054	1.0079	1.0010	1.0106	1.0122	1.0079	1.0	

(a) HEDL-VI is a single ^{235}U foil, HEDL-V is a set of four fission foils (^{235}U , ^{237}U , ^{238}U , and ^{239}Pu), AI-II is a set of small ^{10}B and ^6Li foils with a single ^{235}U , ^{239}Pu , and Ni foil. The SS-I, SS-II and SS-III are self shielding foil sets with increasing foil thicknesses from ~ 0.05 mils to ~ 10 mils, depending on the foil material.

(b) Counting by R. Greenwood and R. Helmer (ANC).

(c) Counting by J. W. Rogers (ANC).

(d) R. Helmer (ANC) August 1973.

(e) Average of pre- and postirradiation measurement.

(f) J. Grundl (NBS) August 1973.

(g) Preirradiation value.

(h) Postirradiation value.

The estimated 1σ relative uncertainties associated with previous and present NBS fission chamber measurements and ANC and HEDL $^{197}\text{Au} (n,\gamma)^{198}\text{Au}$ and $^{115}\text{In}(n,n')^{115\text{m}}\text{In}$ normalization foil measurements are typically in the 0.1 to 0.5%, 0.2 to 0.5%, and 0.5 to 1.5% range, respectively (see the ANC and NBS chapters). Inconsistencies have been observed between fission chamber and Au and In normalization foil measurements for individual CFRMF irradiations which are outside of expected uncertainties. This has resulted in the suggestion that there may be some asymmetry in the central core region flux distribution that is affecting the 1/2 in. diameter Au and In normalization foil results at the 1 to 2% level.*

At present, it is concluded that 1) the NBS fission chamber results for ^{235}U and ^{238}U can be used to establish run-to-run power level normalization factors that are accurate to within about 0.5 to 1% and 2) the Au and In power level normalization foils provide back-up results that are somewhat less accurate, in the 1 to 2% range.

Relative run-to-run power level normalization factors that are inferred from the NBS ^{235}U and ^{238}U results presented in Table 5 and that are supported by the Au and In results are:

HEDL-VI/HEDL-V/AI-II: 1.000/0.999/1.024

SS-I/SS-II/SS-III: 1.000/1.001/1.001

These relative run-to-run power or flux level normalization factors have an estimated relative 1σ uncertainty of $\sim 0.5\%$.

With reference to the NBS chapter of this report, the observed central flux ratios between 0.6KW, 6KW and 10.4 KW for the March 1973 SS-I, SS-II, and SS-III tests are [1:000]: $[9.66 \pm 0.3\%]$: $[17.07 \pm 0.4\%]$. Using the latter two values, the central flux ratio between 6 and 10.4 KW for the March, 1973 SS-I, SS-II or SS-III test was $1.767 \pm 0.5\%$.

Using the Table 5 fission chamber results for the November 6 and 7, 1972 HEDL-V and VI 6 KW tests and the March 14, 1973 SS-I, SS-II and SS-III 10.4 KW tests, a central flux ratio of $1.790 \pm 0.3\%$ is obtained (the combined Au and

*To date, the azimuthal angle for placement of the NBS fission chamber in the central core region of CFRMF has not been controlled, but it will be for all future tests.

In normalization foil results, Table 5, gave a ratio of $1.792 \pm 0.3\%$. The ratio 1.013 (1.790 divided by 1.767) indicates that the central flux at 6KW in March 1973 was $\sim 1.3\%$ higher than in November 1972. This result is consistent with a reported value of $(1.1 \pm 0.3)\%$ in the NBS chapter of this report.

From reference 6, Section C-2 of the HEDL chapter, the central flux ratio between the 6 KW HEDL-VI or HEDL-V and previous 9.75KW (non-fission foil set tests ANL-I, HEDL-I, ANC-I and AI-I) is $1.615 (\pm 0.8\%)$. The 9.75 KW ARHCO-I non-fission foil set run was a factor of 1.0337 higher than the 1.615 ratio.

In summary, the following relative run-to-run power or flux level normalization factors are defined:

HEDL-V/HEDL-VI:	$1.000 (\pm 0.5\%)$
SS-I, SS-II or SS-III/HEDL-VI:	$1.790 (\pm 0.5\%)$
AI-II/HEDL-VI:	$1.024 (\pm 0.5\%)$
ANL-I, HEDL-I, ANC-I, or AI-I/HEDL-VI:	$1.615 (\pm 0.8\%)$
ARHCO-I/HEDL-VI:	$1.669 (\pm 0.8\%)$

Expected Achievements

The fabrication, assembly, documentation and shipment to ANC of the foil sets for the FY-74 CFRMF tests will be completed in late November or early December, 1973. Similar work for other ILRR tests will be initiated.

C. INFINITELY DILUTE REACTION RATES

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Objective

- In support of ILRR program experimental studies, develop and apply appropriate theoretical and/or experimental methods for correcting measured small finite sample (dosimetry foils) reaction rates to infinitely dilute values.
- Apply developed methods to EBR-II and FTR fuels and materials dosimetry studies.
- Provide calculated values of fission rates for foil isotopic impurities for the CFRMF spectrum.

Summary

Results of preliminary neutron self-shielding studies for the CFRMF neutron field are reported. The present results support earlier findings for CFRMF and $\Sigma\Sigma$ (see sections F and H of the HEDL Chapter of reference 4) that relatively thin broad spectrum detectors (less than ~ 10 mils thick) are subject to serious neutron self-shielding. Self-shielding correction factors are provided for correcting previously reported fission and non-fission foil reaction rates to infinitely dilute values. Measured results of the effect of adjacent foils on individual foil measured reaction rates are also presented.

Accomplishments and Status

1) Foil Neutron Self-Shielding

For various practical reasons, relatively thick 0.5 to 5 mil foils are being used for reaction rate measurements in ILRR neutron fields. CFRMF tests ANC-ARHCO-HEDL-SS-I, HEDL-SS-II, and HEDL-SS-III were intended to establish necessary individual foil self-shielding correction factors for the reactions $^{238}\text{U}(n,\gamma)^{239}\text{U}$, $^{45}\text{Sc}(n,\gamma)^{46}\text{Sc}$, $^{235}\text{U}(n, f)\text{FP}$, $^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$, $^{59}\text{Co}(n,\gamma)^{60}\text{Co}$, and $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ ⁽⁶⁾.

Details of foil set assembly, contents, and placement for the SS-I, SS-II, and SS-III CFRMF irradiations are provided in Section C of reference 6. Foil sets ANC-SS-I, HEDL-SS-I, and ARHCO-SS-I were positioned on the top, in the

center, and on the bottom of the duplicate NBS fission chamber for the irradiation of the 0.05 mil thick foils. Figure 1 is a schematic diagram which shows the relative positioning of each foil and foil set.

Data by J. W. Rogers (ANC) indicate that no flux gradient can be observed, within experimental uncertainties, in a radial direction within $\sim 0.2''$ of the center line (ζ) of the CFRMF central exposure tube. This is consistent with previously measured radial $^{197}\text{Au}(n,\gamma)$ reaction rate data reported in Table ANC-3, reference 5. A plot of these latter data, however, shows an asymmetrical reaction rate distribution. Using this distribution, but ignoring the asymmetry because the azimuthal orientation of the three SS-I foil sets was not controlled, an estimate can be made of the relative flux gradient (or reaction rate gradient) the individual foils within the ANC-SS-I, ARHCO-SS-I and HEDL-SS-I might have been subjected to during irradiation. These estimates are listed in Table 6 and indicate that the ANC-SS-I and ARHCO-SS-I foil results require a correction of $\sim 0.7\%$ to 1.4% ($\pm 0.5\%$) before comparison with the HEDL-SS-I, HEDL-SS-II, and HEDL-SS-III foil set results.

The individual ratios of the HEDL, ANC, and ARHCO reaction rate measurements for the HEDL-SS-I, HEDL-SS-II and HEDL-SS-III self-shielding foil sets are reported in Table 4. The combined HEDL, ANC, and ARHCO results with estimated uncertainties are gathered in Table 7 and are shown graphically in Figure 2 as the solid curves. The $\sim 1/2$ inch diameter foil results have been edge corrected to equivalent infinite area foil results by using the approximation $(t/\pi r)$ where t is the foil thickness and r is its radius.⁽⁸⁾ This correction reduced the observed measured foil reaction rate because the $1/2$ inch diameter foil is not surrounded by a coplanar sheet of absorbing material which would have removed some neutrons entering the foil edge. The corrected results are given in the 4th column of Table 7 and are shown as dashed curves in Figure 2. The infinite area foil results are presented for future comparison with theoretical estimates.

The Table 7 and Figure 2 self-shielding correction factors can be used to correct previously reported finite foil measured reaction rates [Table S-1, Reference 5] to obtain infinitely dilute values, assuming that an 0.05 mil

FIGURE HEDL-1. Schematic Diagram of Self Shielding Foil Set Placement for ANC-SS-1, ARHCO-SS-1, and HEDL-SS-1.

TABLE HEDL-6

ESTIMATED RELATIVE REACTION RATE GRADIENTS FOR INDIVIDUAL FOILS
WITHIN THE ANC-SS-I, ARHCO-SS-I, AND HEDL-SS-I FOIL SETS.

<u>FOIL</u>	<u>ESTIMATED RELATIVE REACTION RATE GRADIENTS</u> ^(a)		
	<u>ANC-SS-I</u>	<u>ARHCO-SS-I</u>	<u>HEDL-SS-I</u>
AU	0.993	0.993	1.000
CO	0.992	0.992	1.000
CU	0.990	0.991	1.000
²³⁵ U	0.989	0.990	1.000
Sc	0.988	0.989	0.999
²³⁸ U	0.986	0.987	0.998

- (a) The estimated limit of uncertainty for the ANC-SS-I and ARHCO-SS-I values relative to the HEDL-SS-I values is less than about ± 0.005 , depending on the actual azimuthal angle of the NBS fission chamber during irradiation; i.e., less than approximately $\pm 0.5\%$.

TABLE HEDL-7
MEASURED FOIL SELF-SHIELDING FACTORS FOR THE
CFRMF NEUTRON FIELD

Reaction	THICKNESS (mil)	Ratio		Relative % Precision, 1σ
		0.5 in. Dia. (a) Foil	Infinite Area (b) Foil	
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	0.05	1.000	1.000	$\pm 0.5^{(c)}$
	0.5	0.976	0.975	± 0.5
	2.0	0.923	0.921	± 0.6
$^{45}\text{Sc}(n,\gamma)^{46}\text{Sc}$	0.05	1.000	1.000	$\pm 0.5^{(c)}$
	5	0.989	0.983	± 1.5
	10	0.976	0.963	± 1.4
$^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$	0.05	1.000	1.000	$\pm 0.5^{(c)}$
	1	0.967	0.966	± 0.8
	5	0.940	0.934	± 0.9
$^{238}\text{U}(n,\gamma)^{239}\text{Np}$	0.05	1.000	1.000	$\pm 0.5^{(c)}$
	1	0.966	0.965	± 1.8
	5	0.949	0.943	± 1.2
$^{235}\text{U}(n,f)\text{FP}^{(e)}$	0.05	1.000	1.000	$\pm 0.5^{(c)}$
	5	0.991	0.985	$\pm 0.5 [1.1]^{(d)}$
	10	0.977	0.964	$\pm 0.6 [0.9]^{(d)}$
$^{59}\text{Co}(n,\gamma)^{60}\text{Co}$	0.05	1.000	1.000	$\pm 0.5^{(c)}$
	1.5	0.789	0.788	± 1.8
	2	0.702	0.700	± 1.6

(a) Individual HEDL value or average of ANL, ARHCO, and HEDL values, Table 4.

(b) Edge corrected by factor of 0.13% per mil based on approximation $(t/\pi r)$, Reference 8.

(c) This first error is associated with foil assay while the other errors are given relative to the 0.05 mil foils and include this error.

(d) Propagated error $\sigma(A)$ of individual laboratory error assignments, see footnote, Table 8-A.

(e) Averaged results using ^{140}Ba , ^{103}Ru , and ^{95}Zr ratios.

Figure HEDL-2: Measured Foil Self-Shielding Factors for the CFRMF Neutron Field

thick foil is subject to negligible neutron self-shielding in CFRMF.

It is believed useful to again stress, as previously done by Fabry in the 4th Quarterly,⁽⁴⁾ "that for a total self-shielding effect of the order of (say) 15%, the reaction rate difference between the outer and inner foils in an equivalent sandwich of three identical detectors may typically not exceed 2-3%; this remark is intended as a warning against the danger to conclude that no self-shielding corrections are needed in situations where no spatial variation of reaction rate is observed within a stack of foils; often also, the bulk of such variations occurs only within the first outer layer of these foils and is hard to observe when the self-shielding is small, but not negligible."

Another observation of possible importance is to note that the incident CFRMF flux-spectrum for the $^{238}\text{U}(n,\gamma)$, $^{235}\text{U}(n,f)$, and $^{10}\text{B}(n,\alpha)$ foils has been formed by transport in and through the ^{238}U block, ~ 0.25 in. ^{10}B sleeve and ~ 0.035 in. ^{235}U sleeve of CFRMF. This will tend to make the observed self-shielding effect less for a $^{235}\text{U}(n,f)$, $^{238}\text{U}(n,\gamma)$ and $^{10}\text{B}(n,\alpha)$ foil relative to other foil reactions for the CFRMF neutron field. On this basis, it can be anticipated that the neutron self-shielding for a 5 mil thick ^{239}Pu foil irradiated in CFRMF could be greater than that for a comparable ^{235}U foil. Because of this, and the experimental observation of $\sim 1\%$ self-shielding in the 5 mil ^{235}U foil, plans are being made for the acquisition of a number of ^{239}Pu self-shielding foil sets for irradiation in CFRMF and other ILRR neutron fields. In the absence of experimental data, it will be assumed that the $^{235}\text{U}(n,f)$ self-shielding results provide a lower bound for $^{239}\text{Pu}(n,f)$.

2) Estimated Neutron Self-Shielding In The AI-II Boron-10 Capsules

Small capsules containing ^{10}B were irradiated in the CFRMF as described in the AI chapter. Self-shielding calculations were made for these specimens in order to estimate the order of magnitude of the effect to be expected. These calculations used the First Flight Transport Approximation (i.e., scattering in the ^{10}B was neglected) and assumed an isotropic neutron flux at the surface of the ^{10}B . The ^{10}B was assumed to be uniformly distributed in a cylindrical shape with radius $R = .0381$ cm and height $h = 0.58$ cm. This was

then transformed into an infinite cylinder with an effective radius of R ($h/h+R$) or .0357 cm. The self-shielding factor for energy group i is then given by:

$$f_i = \frac{1}{2 \Sigma_i R} \left(1 - \frac{4R^3}{\pi} \int_0^\infty dz \int_0^\pi \rho^{-4} (1-\cos\beta)^2 e^{-\Sigma_i \rho} d\beta \right) \quad (1)$$

where $\rho = \sqrt{Z^2 + 2R^2 (1-\cos\beta)}$ (9). The total self-shielding factor is given by:

$$f = \frac{\Sigma_i \sigma_i \phi_i f_i}{\Sigma_i \sigma_i \phi_i}$$

and the effective spectral average cross section is:

$$\bar{\sigma}_{eff} = \bar{\sigma} f$$

$$\text{where } \bar{\sigma} = \frac{\Sigma_i \sigma_i \phi_i}{\Sigma_i \phi_i}$$

The value calculated for the self-shielding for a 3 mg sample in CFRMF was 2.0%. Grouping of the samples will increase the self-shielding and this effect is difficult to estimate exactly because of the complex geometry of the AI-II foil set - see Figures AI-1, AI-2 and AI-3, this report. However, an estimate was made of this effect that indicates a self-shielding increase of only about 25% giving a total self-shielding of 2.5%. It is expected that this number is accurate to better than 20% giving an error in the measured result from the self-shielding correction of less than 0.5% .

3) Foil Perturbation Effects

Tests HEDL-V, HEDL-VI, and AI-II were intended to obtain initial reaction generation rates for the fission products ^{140}Ba , ^{95}Zr , ^{103}Ru , and ^{132}Te for ^{235}U , ^{239}Pu , ^{237}Np , and ^{238}U foils. The AI-II test had the dual purpose of obtaining helium generation rates for ^{10}B and ^6Li and providing a ^{235}U and ^{239}Pu foil with larger concentrations of fission products for possible measurement of ^{137}Cs as well as the above listed fission products.

HEDL-VI contained a single(ORNL aluminum encapsulated)high purity 5 mil

thick ^{235}U foil⁽⁶⁾. In addition to ^{235}U , the HEDL-V foil set contained ^{239}Pu , ^{237}Np , and ^{238}U foils packaged together in an aluminum holder (Figures HEDL-4 and 5, and Table HEDL-5 of reference 6). The AI-II foil set contained a ^{239}Pu and a ^{235}U foil, separated by a nickel foil, but adjacent to the AI ^{10}B and ^6Li capsules in their aluminum holder, Figures AI-1, AI-2, and AI-3, this report.

The results of these three different tests combined with those from the foil neutron self-shielding studies, Section C-1, and previous non-fission foil set irradiations (ANC-I, ANL-I, ARHCO-I, and HEDL-I) provide some experimental data for studying foil set perturbation effects for the ^{10}B $^{235}\text{U}(n,f)$, $^{239}\text{Pu}(n,f)$, $^{197}\text{Au}(n,\gamma)$, $^{115}\text{In}(n,n')$, $^{58}\text{Ni}(n,p)$, and $^{27}\text{Al}(n,\alpha)$ foil reactions. The pertinent results of all of these tests are gathered in Table 8-A. From Section B-2, a run-to-run power level normalization reduction factor of 1.024 ($\pm 0.5\%$) was used to normalize the AI-II test results for comparison with the HEDL-V and VI results. A value of 1.790 ($\pm 0.5\%$) was used to normalize the ANC-ARHCO-HEDL-SS-I, HEDL-SS-II, and HEDL-SS-III test results and a value of 1.615 ($\pm 0.8\%$) was used for the combined ANC, ANL, ARHCO, and HEDL results, Table S-1, reference 5. This same value was used for the AI-I test. The renormalized values are bracketed in Table 8-A. The $\sim 0.5\%$ to 0.8% estimated uncertainty for the normalization factors are propagated into the assigned re-normalized errors.

Grundl, Eisenhower and Spencer (NBS Chapter, reference 5) have estimated that a threshold detector foil at the center of the ANC-I, ANL-I, ARHCO-I, or HEDL-I non-fission foil set (~ 2.7 gram foil package, Table HEDL-1, reference 3) would suffer a response loss of $\sim 2\%$ due to neutron scattering within the foil package. Similarly, a capture detector would see a response enhancement of $\sim 1\%$ or less. Other positions in the foil package were presumed to involve perturbations which are the same or smaller than those at the center.

All foil packages used to date have had about the same physical properties; i.e., the foils in a package fit within a ~ 0.145 in. thick aluminum holder hat, reference 3, and weigh less than ~ 2.7 grams (the total weight for the HEDL-VI, HEDL-V, AI-I, AI-II, and any one of the ANC-I, ANL-I, ARHCO-I, or HEDL-I foil packages was ~ 0.53 , ~ 2.35 , ~ 1.33 , ~ 2.55 , and ~ 2.7 grams, respectively).

TABLE HEDL-8-A

CFRME MEASURED REACTION AND FISSION PRODUCT GENERATION RATES ($\times 10^{15}$ dps/atom)

HIGH PURITY FISSION FOILS RUN AT 6 KW

	Laboratory	^{140}Ba	^{103}Ru	^{95}Zr	
HEDL-VI $^{235}\text{U}(5-20)$ (5 mil)	ANC	7.341 $\pm$ 1.5%	3.992 $\pm$ 3.5%	7.914 $\pm$ 1.8%	
	ANL	7.241 $\pm$ 2.3%	3.912 $\pm$ 2.3%	7.814 $\pm$ 2.6%	
	ARHCO	7.384 $\pm$ 0.8%	3.818 $\pm$ 1.2%	7.557 $\pm$ 1.8%	
	HEDL	7.373 $\pm$ 1.1%	3.963 $\pm$ 3.7%	7.839 $\pm$ 2.0%	
		7.335 $\pm$ 0.8% (0.4%)	3.921 $\pm$ 1.4% (1.0%)	7.781 $\pm$ 1.0% (1.0%)	Averages*
HEDL-V $^{235}\text{U}(5-21)$ (5 mil)	ANC	7.281 $\pm$ 1.5%	3.962 $\pm$ 3.5%	7.824 $\pm$ 1.8%	
	ANL	7.170 $\pm$ 2.3%	3.821 $\pm$ 2.3%	7.683 $\pm$ 2.4%	
	ARCHO	7.476 $\pm$ 0.8%	3.973 $\pm$ 1.2%	7.937 $\pm$ 1.2%	
	HEDL	7.326 $\pm$ 1.2%	3.900 $\pm$ 3.0%	7.787 $\pm$ 1.9%	
		7.313 $\pm$ 0.8% (0.9%)	3.914 $\pm$ 1.3% (0.9%)	7.808 $\pm$ 0.9% (0.7%)	Averages
$^{239}\text{Pu}(9-22)$ (5 mil)	ANC	7.154 $\pm$ 1.6%	9.296 $\pm$ 3.7%	6.434 $\pm$ 1.9%	
	ANL	7.187 $\pm$ 2.3%	9.456 $\pm$ 2.9%	6.555 $\pm$ 2.7%	
	ARCHO	7.177 $\pm$ 0.9%	9.254 $\pm$ 1.4%	6.686 $\pm$ 2.3%	
	HEDL	7.419 $\pm$ 1.1%	9.392 $\pm$ 3.2%	6.635 $\pm$ 2.2%	
		7.234 $\pm$ 0.8% (0.9%)	9.350 $\pm$ 1.5% (0.5%)	6.578 $\pm$ 1.2% (0.8%)	Averages

AI-BORON-10 AND LITHIUM-6 FOILS WITH TWO HIGH PURITY FISSION FOILS RUN AT 6 KW

AI-II $^{235}\text{U}(5-17)$ (5 mil)	ANC	7.491 $\pm$ 1.5% [7.315 $\pm$ 1.6%]	4.068 $\pm$ 3.7% [3.973 $\pm$ 3.7%]	8.025 $\pm$ 1.9% [7.837 $\pm$ 2.0%]	
	ANL	[%]	[%]	[%]	
	ARCHO	7.487 $\pm$ 1.8% [7.312 $\pm$ 1.8%]	4.011 $\pm$ 1.2% [3.917 $\pm$ 1.3%]	7.861 $\pm$ 0.8% [7.677 $\pm$ 1.0%]	
	HEDL	7.544 $\pm$ 1.6% [7.367 $\pm$ 1.7%] [7.331 $\pm$ 1.0%] (0.2%)	4.052 $\pm$ 2.6% [3.957 $\pm$ 2.6%] [3.949 $\pm$ 1.6%] (0.4%)	8.198 $\pm$ 2.9% [8.006 $\pm$ 2.9%] [7.840 $\pm$ 1.2%] (1.2%)	Averages
$^{239}\text{Pu}(9-20)$ (5 mil)	ANC	7.490 $\pm$ 1.6% [7.314 $\pm$ 1.7%]	9.858 $\pm$ 3.8% [9.627 $\pm$ 3.8%]	6.745 $\pm$ 2.0% [6.587 $\pm$ 2.1%]	
	ANL	[%]	[%]	[%]	
	ARCHO	7.466 $\pm$ 3.6% [7.291 $\pm$ 3.6%]	9.709 $\pm$ 1.2% [9.481 $\pm$ 1.3%]	6.661 $\pm$ 2.0% [6.505 $\pm$ 2.0%]	
	HEDL	7.573 $\pm$ 1.7% [7.396 $\pm$ 1.8%] [7.334 $\pm$ 1.5%] (0.4%)	9.647 $\pm$ 4.0% [9.421 $\pm$ 4.0%] [9.510 $\pm$ 1.9%] (0.6%)	6.817 $\pm$ 2.9% [6.657 $\pm$ 2.9%] [6.583 $\pm$ 1.4%] (0.7%)	Averages
$^{58}\text{Ni}(\text{TJ})$ (10 mil)	ANC	^{58}Co 1.997 $\pm$ 1.6% [1.950 $\pm$ 1.7%]			
	ANL	[%]			
	ARCHO	1.953 $\pm$ 1.3% [1.907 $\pm$ 1.4%]			
	HEDL	2.009 $\pm$ 1.8% [1.962 $\pm$ 1.9%] [1.940 $\pm$ 1.0%] (0.9%)			Average

HEDL-29

TABLE HEDL 8-A (Cont'd)

NEUTRON SELF SHIELDING FOILS RUN AT 10.4 KW

<u>HEDL-SS-I</u>	<u>Laboratory</u>	<u>^{140}Ba</u>	<u>^{103}Ru</u>	<u>^{95}Zr</u>	
$^{235}\text{U}(\#3)$ (0.05 mil)	ANC	13.375± % [7.472± %]	7.059± % [3.944± %]	14.310± % [7.994± %]	
	ANC	13.500± % [7.542± %]	- - - - -	- - - - -	
	ARCHO	13.630±2.1% [7.615±2.2%]	7.584±2.0% [4.237±2.1%]	14.488±1.7% [8.094±1.8%]	
	HEDL	13.565±1.8% [7.578±1.9%] [7.552±1.1%] (0.4%)	7.301±2.5% [4.079±2.5%] [4.087±1.4%] (2.1%)	14.414±2.8% [8.053±2.8%] [8.047±1.5%] (0.4%)	Average
		<u>^{198}Au</u>			
$^{197}\text{Au}(\#3)$ (0.05 mil)	HEDL	60.86±1.0% [34.00±1.3%]			
<u>HEDL-SS-II</u>	<u>Laboratory</u>				
$^{235}\text{U}(\#1)$	ANC	13.315± % [7.439± %]	6.994± % [3.907± %]	14.018± % [7.831± %]	
	ANC	13.699± % [7.653± %]	- - - - -	- - - - -	
	ARCHO	13.619±1.6% [7.608±1.7%]	7.234±1.7% [4.041±1.8%]	11.019±2.0% [6.156±2.1%] †	
	HEDL	13.426±1.1% [7.501±1.1%] [7.550±0.8%] (0.7%)	7.223±2.6% [4.035±2.6%] [3.994±1.4%] (1.1%)	14.225±2.9% [7.964±2.9%] [7.898±2.1%] (0.8%)	Average
		<u>^{198}Au</u>			
$^{197}\text{Au}(\#1)$ (0.5 mil)	HEDL	59.40±1.0% [33.18±1.3%]			
<u>HEDL-SS-III</u>	<u>Laboratory</u>				
$^{235}\text{U}(\#1)$ (10 mil)	ANC	13.033± % [7.281± %]	6.959± % [3.888± %]	13.968± % [7.803± %]	
	ANC	13.646± % [7.623± %]	- - - - -	- - - - -	
	ARCHO	13.326±1.0% [7.445±1.2%]	6.957±1.3% [3.887±1.4%]	14.010±0.94% [7.827±1.1%]	
	HEDL	13.336±1.1% [7.450±1.2%] [7.450±0.6%] (0.9%)	7.069±2.5% [3.949±2.5%] [3.908±1.3%] (0.5%)	14.133±2.4% [7.896±2.5%] [7.842±1.2%] (0.4%)	Average
		<u>^{198}Au</u>			
$^{197}\text{Au}(\text{JJ})$ (2 mil)		56.20±1.2% [31.40±1.4%]			

† Not used for obtaining average value.

TABLE HEDL 8-A (Cont'd)

NEUTRON SELF SHIELDING FOILS RUN AT 10.4 KW

ANC-SS-I	Laboratory	^{140}Ba	^{103}Ru	^{95}Zr	
$^{235}\text{U}(\#1)$ (0.05 mil)	ANC	$13.154 \pm 1.9\%$ [7.349 $\pm$ 2.0%]	$6.979 \pm 3.7\%$ [3.899 $\pm$ 3.7%]	$14.120 \pm 1.9\%$ [7.888 $\pm$ 2.0%]	
	ANC	$13.576 \pm \%$ [7.584 $\pm \%$] [7.467 $\pm$ 1.6%]	- - - - - [3.899 $\pm$ 3.7%]	- - - - - [7.888 $\pm$ 2.0%]	Average

NON-FISSION FOIL SETS WITH ^{235}U NORMALIZATION FOILS RUN AT 9.75 KW

ANC-I/ANL-I/** ARHCO-I/HEDL-I	Laboratory		Average Value
$^{235}\text{U}(n,f)^{140}\text{Ba}$	A11	(4 mils)	$11.90 \pm 1.4\%$ [7.368 $\pm$ 1.6%]
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	A11	(0.5 mil) (0.05 mil) (0.5 mil) (0.135% Au-A1)(20 mil)	[32.89 $\pm$ 0.8%] [33.61 $\pm$ 0.9%] [32.76 $\pm$ 0.7%] [33.75 $\pm$ 1.0%]
$^{115}\text{In}(n,n')^{115m}\text{In}$	A11	(2 mil)	[3.910 $\pm$ 0.2%]
$^{58}\text{Ni}(n,p)^{58}\text{Co}$		(10 mil)	[1.894 $\pm$ 0.5%]
$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	A11	(5 mil)	[0.01271 $\pm$ 0.6%]

AI BORON-10 FOILS WITH AU, IN, AND AL NORMALIZATION FOILS RUN AT 9.75 KW

AI-I	Laboratory		
	HEDL	$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ (0.5 mil) Top Foil	[32.37 $\pm$ 1.1%]
		(0.5 mil) Bottom Foil	[32.46 $\pm$ 1.1%]
	HEDL	$^{115}\text{In}(n,n')^{115m}\text{In}$ (2 mil) Top Foil	[4.027 $\pm$ 2.8%]
		(2 mil) Bottom Foil	[4.020 $\pm$ 2.8%]
	HEDL	$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$ (5 mil) Bottom Foil	[0.01290 $\pm$ 1.8%]

- Note: 1) Where required in this table, data were corrected by HEDL for target composition.
 2) Some data in this table are tentative in that documentation of some ANC counting of HEDL-SS foils is forthcoming.
 3) Numbers in brackets [xxxx $\pm$ xx%] in this table are the reported value normalized to the HEDL-VI run using the normalization factors given in Section B-3.
 4) For some foils, two values are reported for ANC; i.e., by R. Greenwood and R. Helmer, and/or J. W. Rogers.

* The 1 σ absolute error estimates of the individual measurements were propagated to obtain the average value error $\sigma(A)$. $\sigma(A)$ is defined by

$$\sigma(A) = \frac{1}{N} \left[\sum_{i=1}^N \sigma_i^2 \right]^{1/2}$$

which assumes equal weighting for each of the N values of σ_i . When an individual laboratory error is not specified, the largest error in the given set of N values of σ_i was assumed to be a reasonable estimate to use with the other errors for obtaining the average value error. The standard error of the set of values is given in brackets immediately below the propagated error value and the larger of the two errors is taken as the uncertainty in the average value, A.

**From Table S-I, reference 5.

Based on the NBS prediction of less than $\sim 1\%$ response increase for a 2.7 gram foil package and available run-to-run flux level normalization factor error estimates of less than $\sim 0.5\%$ to 0.8% , differences of more than $\sim 1\%$ in test-to-test measured reaction and fission product generation rates should not be observable for the broad spectrum foils $^{235}\text{U}(n,f)$, $^{239}\text{Pu}(n,f)$ and $^{197}\text{Au}(n,\gamma)$. Except for the self-shielding foil set tests SS-I, SS-II, and SS-III and AI-I, Table 8-B, this is indeed the case for foils of the same thickness.* (The enhanced ratios for thinner foils and reduced ratios for thicker foils are indications of the neutron self-shielding discussed in the previous section). Based on the $^{197}\text{Au}(n,\gamma)$ [0.05 mil and 0.5 mil foils] and the $^{235}\text{U}(n,f)\text{FP}$ [~ 5 mil foils] reactions, the series of self-shielding tests show an ~ 1 to 2% higher reaction rate which may be significant and could be associated with the relative difference in individual foil placement; i.e., thin aluminum spacer rings were used in the series of self-shielding tests to provide additional separation between individual foils to reduce foil-to-foil perturbation effects. This separation would tend to reduce the foil-to-foil perturbation effect of neutrons that are incident on a foil's surface at low angles.

For the AI-I test, Table 8-B, the 0.5 mil $^{197}\text{Au}(n,\gamma)$ results suggest the possibility of as much as a $\sim 1.5\%$ reduction in reaction rate while the threshold detector reactions, $^{27}\text{Al}(n,\alpha)$ and $^{115}\text{In}(n,n')$ suggest increases of up to as much as ~ 1.5 and $\sim 3.0\%$, respectively. Considering the lighter weight of the AI-I foil set (~ 1.33 grams), these results appear to be inconsistent, within experimental uncertainties, with the NBS prediction of up to an $\sim 1\%$ response increase for a broad spectrum monitor and up to a $\sim 2\%$ response loss for threshold detectors at the center of a ~ 2.7 gram foil set. These combined differences for the AI-I test could be an indication, therefore, that ^{10}B in the AI-I capsule may have perturbed the incident central flux spectrum sufficiently to cause a slight spectral hardening affect.

The $\sim 1\%$ increase in the $^{239}\text{Pu}(n,f)$ reaction rate for the AI-II over the HEDL-V test does not appear to be significant in view of no detected increase for $^{235}\text{U}(n,f)$, Table 8-B. This coupled with a 2.4% increase for $^{58}\text{Ni}(n,p)$ may have significance, however, relative to the effect of the ^{10}B and ^6Li in the ~ 2.6 gram AI-II capsule on a slight hardening of the central flux spectrum as

*The Table 8-A averaged values were used to obtain the Table 8-B relative reaction and fission product generation rate ratios.

TABLE 8-B

CFRMF TEST-TO-TEST MEASURED REACTION AND FISSION PRODUCT GENERATION RATE RATIOS

^{235}U	Foil/Referenced Foil			
	^{140}Ba	^{103}Ru	^{95}Zr	Average
HEDL-VI ^{235}U (5-20) (5 mils) (One ^{235}U Foil)	1.0000*	1.0000*	1.0000*	1.0000*
HEDL-V ^{235}U (5-21) (5 mils) (Foil set, one ^{235}U , ^{239}Pu , ^{237}Np , and ^{238}U foil)	0.9970	0.9982	1.0035	0.9996
AI-II ^{235}U (5-17) (5 mils) (Foil set, one ^{235}U , Ni, and ^{239}Pu foil with a ^{10}B and ^6Li capsule)	0.9995	1.0071	1.0076	1.0047
HEDL-SS-I ^{235}U (#3) (0.05 mils) (One ^{235}U foil with a ^{238}U , Sc, Co, Cu, and Au foil)	1.0296	1.0423	1.0342	1.0354
HEDL-SS-II ^{235}U (#1) (5 mils) (One ^{235}U foil with a ^{238}U , Sc, Co, Cu, and Au foil)	1.0293	1.0186	1.0150	1.0210
HEDL-SS-III ^{235}U (#1) (10 mils) (One ^{235}U foil with a ^{238}U , Sc, Co, Cu, and Au foil)	1.0157	0.9967	1.0078	1.0067
ANC-SS-I ^{235}U (#1) (0.05 mils) (One ^{235}U foil with a ^{238}U , Sc, Co, Cu, and Au foil)	1.0180 (1.0293)**	0.9944 (1.0055)**	1.0138 (1.0251)**	1.0087 (1.0200)**
ANC-I/ANL-I/ARHCO-I/HEDL-I ^{235}U (4 mils) (One ^{235}U and ^{238}U foil with a non-fission foil set)	1.0045	--	--	1.0045
^{239}Pu	^{140}Ba	^{103}Ru	^{95}Zr	Average
HEDL-V ^{239}Pu (9-22) (5 mils)	1.0000*	1.0000*	1.0000*	1.0000*
AI-II ^{239}Pu (9-20) (5 mils)	1.0138	1.0171	1.0008	1.0106

* Reference foil for ratios

** Corrected for gradient, see Table 6.

TABLE 8-B (Con.t)

<u>$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$</u>		<u>Foil/ Referenced Foil</u>	<u>Renormalized to 0.05 mil foil</u>
ANC-I/ANL-I/ ARHCO-I/HEDL-I	(0.5 mil) (0.05 mil) (0.5 mil)	1.0000* 1.0219 0.9960	0.9786 1.0000 0.9747
(0.135% Au-A1)	(20 mil)	1.0261	1.0004
AI-I	(0.5 mil)	0.9842	0.9631
	(0.5 mil)	0.9869	0.9658
HEDL-SS-I	(0.05 mil)	1.0337	1.0115
HEDL-SS-II	(0.5 mil)	1.0088	0.9872
HEDL-SS-III	(2 mil)	0.9547	0.9342
<u>$^{58}\text{Ni}(n,p)^{58}\text{Co}$</u>			
ANC-I/ANL-I/ ARHCO-I/HEDL-I	(10 mil)	1.0000*	
AI-II	(10 mil)	1.0243	
<u>$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$</u>			
ANC-I/ANL-I/ ARHCO-I/HEDL-I	(5 mil)	1.0000*	
AI-I	(5 mil)	1.0149	
<u>$^{115}\text{In}(n,n')^{115\text{m}}\text{In}$</u>			
ANC-I/ANL-I/ ARHCO-I/HEDL-I	(2 mil)	1.0000*	
AI-I	(2 mil)	1.0300	
	(2 mil)	1.0281	

* Reference foil for ratios

suggested for the AI-I test. In the previous section of this report, an estimate of a $\sim (2.5 \pm 0.5)\%$ neutron self-shielding effect for the ^{10}B in the AI-II capsule was made, to which another $\sim 1\%$ may be required for spectral hardening.

In summary, significant foil flux perturbation effects (up to ~ 2 to 3%)* can be expected for individual foils being used for the ILRR program. The magnitude of the effect will depend on the materials and the physical arrangement of foils within each aluminum foil set holder and the subsequent placement within the NBS duplicate fission chamber. At present, only estimated corrections for these effects can be made.

4) Calculated Fission Rates for Foil Isotopic Impurities

Reaction rates per unit flux (spectral averaged cross sections) for a number of fission reactions are presented in Table 9. These values of fission rates per unit flux are being used by HEDL to make corrections for fissions by minor impurity isotopes in the high purity ^{235}U , ^{238}U , ^{239}Pu , and ^{237}Np fission foils irradiated in CFRMF. The participating laboratories should review these values, recommend changes, and agree to a final set of values that will be accepted and used by all laboratories.

Expected Achievements

Further calculations will be done to analyze self-shielding effects on larger ^{10}B samples in BIG-10 and CFRMF and to better define perturbation effects of samples on each other.

* Neutron self-shielding effects within individual foils are not included, and may be much larger, see Section C-1.

TABLE HEDL-9

<u>FISSION CROSS SECTIONS FOR CFRMF</u>			
<u>ISOTOPE</u>	<u>SPECTRAL AVE. CROSS SECTION (Barns)</u>	<u>RATIO TO ²³⁵U</u>	<u>ESTIMATED UNCERTAINTY</u>
²³³ U	2.454	1.52	10%
²³⁴ U	.541	.34	20%
²³⁵ U	1.612	1.00	5%
²³⁶ U	.162	.10	>20%
²³⁸ U	.007202	.004	10%
²³⁹ U	.132	.08	>20%
²³⁷ Np	.5552	.34	10%
²³⁸ Np	1.81	1.12	>20%
²³⁹ Np	.988	.61	>20%
²³⁸ Pu	1.53	.95	20%
²³⁹ Pu	1.794	1.11	5%
²⁴⁰ Pu	.552	.34	10%
²⁴¹ Pu	2.09	1.30	10%
²⁴² Pu	.403	.25	>20%

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NATIONAL BUREAU OF STANDARDS

A. ABSOLUTE FISSION CHAMBER MEASUREMENTS IN CFRMF

J. A. Grundl and J W Rogers*

Objective

● Measure absolute fission rates per nucleus at the center of the CFRMF Reactor for four fissionable isotopes: ^{235}U , ^{239}Pu , ^{238}U , and ^{237}Np . The NBS double fission chamber, and fission foils from a permanent set of fissionable deposits at NBS, are to be employed. Specific efforts to identify and evaluate error sources will focus on chamber performance, foil assay, and flux perturbations.

Summary

Fission chamber measurements were performed during ILRR irradiations at CFRMF in March 1973. Techniques for obtaining data and the derivation of absolute fission rates were the same as those described in the Seventh ILRR Quarterly. Additional information was obtained on the relationship between central flux and the reactor power instrumentation: (1) for reactor power levels of 0.6 KW:6KW:10.4KW, the observed central flux ratios were $[1:000]:[9.66 \pm 0.03]:[17.07 \pm 0.07]$, respectively; (2) the central flux at both 0.6 KW and 6 KW in March 1973, was $(1.1 \pm 0.3)\%$ higher than in November 1972. The fission rate ratio $^{235}\text{U}/^{238}\text{U}$ does not depend upon the reactor power level and agrees with earlier results to better than 0.5%. Some chamber performance difficulties which arose in the course of these latest CFRMF measurements are thought to signal the onset of radiation damage within the fission chamber. The influence of these difficulties upon the fission rate data is believed to be no worse than a few tenths of a percent.

*Staff of ANC, Idaho

Accomplishments and Status

Fission chamber measurements were performed at CFRMF during the following ILRR irradiations in March 1973: (1) fission track recorders (Pu and NP) exposed at 0.6 KW power level; and (2) three self-shielding foil sets exposed at one and ten KW (see Table ANC-1 in the fifth ILRR Quarterly). The NBS double fission chamber PP1 was operated as in November 1972, with the same ^{235}U and ^{238}U foils and with the same amplifier and scaler arrangement. All fission counting data was reduced to absolute fission rates consistent with the procedures described in the last fission rate summary report (Seventh ILRR Quarterly Progress Report^{*}).

1. Chamber Performance

For the ANL irradiation at 0.6 KW nine pairs of triple-scaler counts were taken before and after the irradiation. The standard deviations of the four sets of fission counts were within the expectation of counting statistics: $\pm 0.3\%$ to $\pm 0.45\%$. Statistics for the average of the nine counts was $< \pm 0.2\%$. The extrapolations-to-zero, $S_1/S_u^{(1)}$, were in the expected range: 1.008 ± 0.0005 for ^{238}U and 1.002 ± 0.0005 for ^{235}U . The gain check ratios, S_{gc}/S_u , showed no significant drift. For the three self-shielding foil irradiations, pre- and post-irradiation fission counts were taken at 0.6 KW, 6 KW, and 10.4 KW the final irradiation power level. Standard deviations for the more than thirty pairs of counts taken were in accordance with counting statistics. The average fission counting rates at 10.4 KW were established to $\approx \pm 0.1\%$ for both ^{235}U and

*erratum: scaler ratio designations, " S_u/S_1 " and S_{gc}/S_1 " on p. NBS-3 of the Seventh ILRR Quarterly should read " S_1/S_u " and " S_{gc}/S_u ."

⁽¹⁾ratio of scaler S_1 biased near the alpha pulse height cut-off to scaler S_u biased at twice the bias voltage of S_1 .

^{238}U , and the step-wise fission count rate checks at 0.6 KW and 6 KW were established to $< \pm 0.3\%$.

Three problems arose in the course of these March fission chamber measurements at CFRMF. They will be outlined separately although it is thought that they have a common origin.

a. Equivalence of the amplifier-scaler networks. In a brief post-irradiation measurement (after the ANL fission track recorder exposure) the double fission chamber outputs were interchanged at the preamps. In this condition the ^{235}U fission count rate was reproduced to within $\pm 0.3\%$, the standard deviation of the counts taken; on the ^{238}U side of the chamber, however, the fission count rate dropped by 0.8%, well outside of the standard deviation of the counts. The extrapolation-to-zero and gain check scaler ratios for ^{238}U were normal. Such amplifier interchanges undertaken during the first CFRMF measurements in early 1972 (Third ILRR Quarterly) did not show any such count rate shift. Further investigation of this effect was not undertaken. All reported fission rate data has been taken with the chamber connected to the same amplifier-scaler line as previously.

b. Gain change in the ^{238}U chamber. In one set of pre-irradiation fission counts (at 10.4 KW before the self-shielding foil irradiation) the gain check scaler ratios fell below the normal value of 0.50. On the ^{238}U side the ratio dropped to as low as 0.26; on the ^{235}U side the minimum value was 0.38. The effect of these variations in fission chamber output on the average of the nine fission counts for ^{238}U is less than 0.1%. Observations two months later of more serious reductions in the ^{238}U pulse level from the chamber suggest that chamber insulators and/or internal mechanical connections may be suffering radiation damage.

c. Intermittent noise. With the fission chamber inside of CFRMF, occasional flashes of noise were observed during the March irradiations. With the chamber outside of the reactor and electrically isolated from metal structures the chamber was quiet. Routine 100 sec counts with the reactor shutdown showed noise associated count rates in the S_u scaler of as much as 1 per sec; the average observed was around 0.1 per sec. Attempts to correlate the noise with the switching of electrical equipment were partially successful. Data taken with the reactor at power was monitored visually with an oscilloscope so that few of the counts accepted for reporting back to NBS are considered to have any noise contribution in the S_u scaler count. There is nothing in the scaler ratios, S_1/S_u , to indicate the presence of spurious counts.

At least some of the difficulties outlined above are believed to be associated with radiation damage. Teflon parts in the dummy fission chamber used for the high-fluence ILRR foil irradiations showed serious radiation damage some time ago. The PP1 fission chamber was returned to NBS for disassembly and inspection in June 1973.

2. Power Level Reproducibility and Linearity

As noted in earlier reports, the power level of the CFRMF Reactor is highly reproducible over the period of time associated with individual ILRR irradiations. We can now make comparisons over longer time intervals. At the 0.6 KW and 6 KW reactor power levels the fission chamber indicates that a small change in central flux occurred between November 1972 and March 1973:

	<u>^{235}U</u>	<u>^{238}U</u>
$\frac{\text{March 1973}}{\text{November 1972}}:$	1.012 ± 0.003	1.016 ± 0.004

There is also more data on the power level normalization problem now. Table NBS-1 shows relative central flux levels corresponding to the reactor power levels established by the out-of-core neutron detectors. The flux level ratio 6 KW to 0.6 KW agrees very well with the earlier measurements in February and November of 1972 (see Seventh Quarterly). The new values at 10.4 KW suggest that the disagreement between central flux and reactor power instrumentation is not monotonic with power level.

3. Absolute Fission Rates for CFRMF

Fission count rate data from the double fission chamber was reduced to absolute fission rates in exact accordance with the procedures described in the Seventh ILRR Quarterly. Table NBS-2 lists the final observed fission

rates for the March 1973 ILRR irradiations at CFRMF. Values given are entirely comparable with those listed in Table NBS-2 of the Seventh Quarterly including assigned errors. Applying foil masses as previously reported, fission rates per nucleus may be obtained; these are given in Table NBS-3 along with comparable results reported earlier. The average cross section ratios in the last column are seen to be in close agreement, an indication that the CFRMF spectrum is unchanging. Errors in foil masses remain at a nominal $\pm 3\%$ pending the outcome of a systematic foil assay investigation.

TABLE NBS-1
POWER LEVEL NORMALIZATION

Reactor Instrumentation:	<u>0.6 kW</u> (1.000)	<u>6 kW</u> (10.000)	<u>10.4 kW</u> (17.32)
NBS Fission Chamber:			
<u>March 1973</u>			
^{235}U :	1.000	$9.65 \pm 0.3\%$	$17.07 \pm 0.4\%$
^{238}U :	1.000	$9.66 \pm 0.5\%$	$17.07 \pm 1.2\%$
<u>*Previous Summary Report</u>			
^{235}U :	1.000	$9.63 \pm 0.3\%$	-
^{238}U :	1 000	$9.66 \pm 0.4\%$	-

*Seventh ILRR Quarterly Progress Report (typographical mistake on error assignments corrected).

TABLE NBS-2

ABSOLUTE FISSION RATES
FOR
ILRR IRRADIATIONS AT CFRMF [MARCH 1973]

	^{235}U <u>fissions/sec</u>	^{238}U <u>fissions/sec</u>	^{235}U <u>^{238}U</u>
ANL-SSTR Irradiation @ 0.6 kW (3-7-73)			
pre-irradiation:	168.1 $\pm$ 0.25%	395.6 $\pm$ 0.25%	0.4249 $\pm$ 0.35%
post-irradiation:	168.0 $\pm$ 0.25%	396.7 $\pm$ 0.25%	0.4235 $\pm$ 0.35%
Self-shielding Foil Irradiations @ 10.4 kW (3-12-73)			
pre-irradiation:	2861 $\pm$ 0.23%	6760 $\pm$ 0.44%	0.4232 $\pm$ 0.5%
post-irradiation:	2871 $\pm$ 0.23%	6752 $\pm$ 0.44%	0.4252 $\pm$ 0.5%

The standard deviation listed is associated with reproducibility of count rate data, and non-systematic errors in the dead time correction and the extrapolation-to-zero of the fission pulse height distribution.

TABLE NBS-3

FISSION RATE PER NUCLEUS
FOR
ILRR IRRADIATIONS AT CFRMF [MARCH 1973]

	<u>^{235}U</u>	<u>^{238}U</u>	<u>$\frac{\sigma_f(^{235}\text{U})}{\sigma_f(^{238}\text{U})}$</u>
ANL-SSTR Irradiation @ 0.6 kW (3-7-73)			
$\frac{\text{fiss./sec.}}{\text{nucleus}}$	1.285 ± 0.003 $\times 10^{-14}$	6.14 ± 0.05 $\times 10^{-16}$	20.9_5
Foil Self-shielding Irradiations @ 10.4 kW (3-12-73)			
$\frac{\text{fiss./sec.}}{\text{nucleus}}$	2.192 ± 0.007 $\times 10^{-13}$	1.047 ± 0.01 $\times 10^{-14}$	20.9_3
*Previous Summary (result @ 0.6 kW)			
$\frac{\text{fiss./sec.}}{\text{nucleus}}$	1.270 ± 0.003 $\times 10^{-14}$	6.04 ± 0.07 $\times 10^{-16}$	21.0_3

Error assignments exclude uncertainties in foil mass.

*Seventh ILRR Quarterly Progress Report.

OAK RIDGE NATIONAL LABORATORY

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

E. H. Kobisk and H. L. Adair

Objective

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

Summary

Archive samples of all batches of ILRR fissile dosimeter isotopes have been prepared, analyzed, and stored. Redundant analyses of dosimeter contents are being made to determine the maximum error involved in reporting the weight of isotope contained. Some results with ^{235}U (oxide) loadings would indicate a maximum error of 2%, but this error can be further reduced. Thin film deposits of $^{235}\text{U}(\text{O}_2)$ and $^{239}\text{Pu}(\text{O}_2)$ were prepared for NBS fission rate experiments and were carefully characterized to determine the content of nuclides and the uniformity of the film deposits.

Accomplishments and Status

Archive Samples. Small quantities of ILRR reserved isotopes (^{235}U , ^{238}U , ^{237}Np , ^{239}Pu) were carefully prepared for long-term storage as archive samples and are now in a repository at ORNL. Isotope batch numbers and sample weights are given in Table ORNL-1. These materials were packaged in two stainless steel containers as shown in Figure ORNL-1.

TABLE ORNL-1
ARCHIVE SAMPLES

<u>Batch No.</u>	<u>Oxide Weight, g (± 0.0001)</u>
U-235-264-C	2.3592
U-238-ES-Z	2.3558
Pu-239-453-B-O	2.2680
Np-237-NP21	2.2700
Np-237-NP24HP	2.2700
	<u>Metal Weight, g (± 0.0001)</u>
U-235-264-CM	2.0214
U-238-ES-ZM	2.6693
Pu-239-453-B-M	2.0000

Uranium metal and oxide samples were placed in their respective inner containers, evacuated to $<10^{-2}$ torr, and then backfilled with pure argon to atmospheric pressure. These samples were then welded inside the outer stainless steel capsules in an argon atmosphere. Plutonium and neptunium samples were loaded and welded under an argon atmosphere. Both inner and outer containers of each archive capsule are labeled with the appropriate batch number of the contained material. Unfortunately, the extreme reactivity of neptunium metal precluded encapsulation of the residual metal (Batch Np-237-NP21M) as corrosion was excessive. Analyses of each batch of material were performed and are recorded in Tables ORNL-2 through ORNL-5.

FIGURE ORNL-1. Archive Sample Capsule Assembly

TABLE ORNL-2
ANALYSES FOR THE MAJOR ELEMENT (THE AMOUNT OF THE MAJOR
ELEMENT PER UNIT WEIGHT OF MATERIAL)

<u>Batch No.</u>	U (mg/g) <u>±0.5%</u>	Np (mg/g) <u>±0.5%</u>	Pu (mg/g) <u>±0.5%</u>
U-235-264-C	846.8		
U-235-264-CM	994.4		
U-238-ES-Z	845.4		
U-238-ES-ZM	994.8		
Pu-239-453-B-O			895.2
Pu-239-453-B-M			982.5
Np-237-NP21		861.0	
Np-237-24HP		874.5	

TABLE ORNL-3
ISOTOPIC COMPOSITION OF THE MAJOR ELEMENTS

Batch U-235-264-C

<u>Isotope</u>	<u>Atom Percent</u>
233	<0.0005
234	0.034 ±0.001
235	99.89 ±0.01
236	0.025 ±0.001
238	0.052 ±0.002

Batch U-235-264-CM

233	<0.0005
234	0.035 ±0.001
235	99.89 ±0.01
236	0.025 ±0.001
238	0.053 ±0.002

Batch U-238-ES-Z

233	<0.0001
234	<0.0001
235	0.0012 ±0.0001
236	<0.0001
238	99.998 ±0.001

TABLE ORNL-3 (Cont'd)

Batch U-238-ES-ZM

<u>Isotope</u>	<u>Atom Percent</u>
233	<0.0001
234	<0.0001
235	0.0015 $\pm$ 0.0001
236	<0.0001
238	99.998 $\pm$ 0.001

Batch Pu-239-453-B-M

238	$\leq$ 0.002
239	99.11 $\pm$ 0.01
240	0.880 $\pm$ 0.005
241	0.010 $\pm$ 0.001
242	0.005 $\pm$ 0.001
244	<0.0005

Batch Pu-239-453-B-O

238	$\leq$ 0.002
239	99.11 $\pm$ 0.01
240	0.875 $\pm$ 0.005
241	0.010 $\pm$ 0.001
242	0.005 $\pm$ 0.001
244	<0.0005

Batch Np-237-NP21

235	<0.0005
236	<0.0005
237	$\geq$ 99.99 $\pm$ 0.01
238	$\leq$ 0.003
239	$\leq$ 0.004

Batch Np-237-24HP

235	<0.0005
236	<0.0005
237	$\geq$ 99.99 $\pm$ 0.01
238	$\leq$ 0.003
239	$\leq$ 0.003

TABLE ORNL-4
IMPURITY ANALYSIS (Wt. ppm)

	²³⁵ U 264-C	²³⁵ U 264-CM	²³⁸ U ES-Z	²³⁸ U ES-ZM	²³⁹ Pu 453-B-M	²³⁹ Pu 453-B-O	²³⁷ Np NP21	²³⁷ Np NP24HP
Al	<1	30	5	30	1,000	15	1	2
As	<1	<1	1	<1		1	1	<1
B	<0.5	<0.5	2	<0.5	0.5	0.2	<0.5	<0.5
Ba	<1	<1	<1	<1	<1	<1	<1	<1
Ca	2	2	15	2	40	8	8	2
Co	<1	<1	<1	<1	<1	<1	<1	<1
Cr	<1	30	1	30	20	<1	<1	<1
Cu	5	10	2	10	40	6	4	2
Fe	8	80	25	200	400	10	2	4
K	<1	<1	<1	<1	20	10	<1	1
Mg	<2	<1	2	1	100	1	<1	<1
Mn	<1	7	<1	7	7	<1	<1	<1
Na	<1	<1	<1	<1	<1	1	<1	<1
Ni	<1	100	2	30	30	1	1	1
P	1	<1	10	<1	1	<1	<1	<1
Pb	1	<1	<1			<1	<1	
Si	<3	20	3	20	20	10	<1	3
Sn			5			1		2

TABLE ORNL-4 (Cont'd)

	²³⁵ U 264-C	²³⁵ U 264-CM	²³⁸ U ES-Z	²³⁸ U ES-ZM	²³⁹ Pu 453-B-M	²³⁹ Pu 453-B-O	²³⁷ Np NP21	²³⁷ Np NP24HP
S	4	1	10	1	10	1,000	4	4
Ti	10	2	10	2	<1	<1	<1	<1
²³⁸ U	100	100	M	M	10	5	1	10
V	100	20	100	200	<1	<1	<1	<1
W	<1	<1	<1	<1				
Zn	<1	<1	<1	<1	10	2	2	<1
Zr	1	<1	<1	2	2	<1		<1
²³⁵ U	M	M	4	3	10	1	<1	<1
²³⁷ Np	<1	<1	<1	<1	1		M	M
²³⁹ Pu	<1	<1	≤1	3	M	M	1	<1
²⁴⁴ Cm	<1	<1	<1	<1	<1	<1	<1	<1

M - Major

TABLE ORNL-7 (Cont'd)

 ^{239}Pu , Batch 277A

<u>Isotope</u>	<u>Atom Percent</u>
238	≤ 0.0002
239	99.978 ± 0.002
240	0.021 ± 0.002
241	0.0005 ± 0.0003
242	0.0005 ± 0.0003
244	< 0.0002

After deposition, each target was α -counted in a low geometry system; the ^{235}U targets were positioned 1.27 cm from the silicon detector while the ^{239}Pu targets were positioned at 15.24 cm or 38.10 cm depending on their thickness. Geometry factors were determined by destructive analysis of additional samples of ^{235}U or ^{241}Am ; analysis was performed to $\pm 0.5\%$. Table ORNL-8 summarizes the analytical data for each target sample prepared and shipped to NBS.

TABLE ORNL-8
ISOTOPE CONTENT OF NBS TARGETS

<u>Date</u>	<u>Target No.</u>	<u>^{235}U (μg)</u>	<u>Estimated Standard Deviation</u>
12-12-72	25K-1-1	126.3 ± 1.9	± 1.7
12-15-72	25K-02-1	28.5 ± 0.6	one measurement
<u>Date</u>	<u>Target No.</u>	<u>^{239}Pu (μg)</u>	<u>Estimated Standard Deviation</u>
12-27-72	49K-01-2	13.3 ± 0.1	± 0.1
1-4-73	49K-4-1	472.2 ± 4.7	± 0.6

Alpha spectra for targets 25K-02-1, 25K-1-1, 49K-01-2, and 49K-4-1 were determined. In each case, instrument calibration was approximately 1.2 keV/channel. The ^{235}U content was determined by integrating the

counts under the ^{235}U peaks and subtracting contributions due to ^{236}U , ^{238}U , and background. Error in the geometry factor amounted to $\pm 1\%$ and remaining error (Table ORNL-8) resulted from counting statistics. It was originally thought that the best method of analysis for ^{235}U was to obtain the amount of ^{234}U and, on the basis of the mass analysis, calculate the ^{235}U content. Unfortunately, the mass analysis for the ^{234}U had an uncertainty of $\pm 2.6\%$.

Target uniformity for sample 49K-01-2 was determined using a 2 mm aperture and scanning the ^{239}Pu deposit. Equidistant positions in the x- and y-direction through the center of the circular targets were counted. Data from these measurements are given in Table ORNL-9. The uniformity was not as good as was expected ($\sim 5\%$ maximum deviation from the average thickness at the edge), but with the exception of the edge (~ 3 mm radius increment) the interior of the targets showed a variation in thickness of only $\sim 3\%$. Probably the less thick target deposit on the outer edge was the result of a mask effect which is not uncommon.

TABLE ORNL-9
 ^{239}Pu TARGET UNIFORMITY MEASUREMENTS

<u>Position</u>	<u>x-Axis, Counts/100 sec</u>	<u>y-Axis, Counts/100 sec</u>
1	12526	12859
2	13227	12810
3	13659	13829
4	13775	13400
5	13459	13168
6	13076	12773

Although the batches of ^{235}U and ^{239}Pu used to prepare these targets are not the same as the ILRR reserved materials, they are well characterized and can be used for reaction rate studies similar to those presented in the

last ILRR Quarterly Report.¹ Indeed the accuracy of the isotope content in these targets should reduce the error level shown in NBS fission chamber rate measurements.¹

¹LMFBR Reaction Rate and Dosimetry 7th Quarterly Progress Report, December 1972, January, February 1973. HEDL-TME 73-31.