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LMFBR Reaction Rate And
Dosimetry 3rd Quarterly
Progress Report
December 1971,
January, February 1972

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PRELIMINARY REPORT

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LMFBR Reaction Rate And
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Progress Report *Rpt*
December 1971,
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3rd Prog Rpt
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Hanford Engineering Development Laboratory

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W. N. McElroy
March, 1972

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ABSTRACT

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

ACKNOWLEDGEMENTS

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

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

FOREWORD

PURPOSES OF ILRR PROGRAM

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

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

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

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

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SUMMARY

PROGRAM HIGHLIGHTS

Comparison of preliminary ANC, ANL, ARHCO, and HEDL ^{235}U and ^{238}U radiochemistry (^{140}Ba - ^{140}La) fission rate measurements with NBS fission chamber measurements provides an initial consistency check on the use of recently revised ^{140}Ba fission yield values of 5.8% for ^{235}U and 6.0% for ^{238}U for fast reactor fuels and materials dosimetry. (These revised values are based on ANL studies of fission track recorder and radiochemistry measurements in EBR-II type fast reactor spectra.) Using these yield values and neglecting expected small corrections for uncertainties in foil assay, size and placement differences, and neutron scattering and absorption effects, the combined ANC, ANL, ARHCO, and HEDL measured CFRMF ^{235}U average fission rate value of 1.286×10^{-14} ($\pm 1.7\%$, 1 standard error) agrees with the NBS fission chamber measurement of 1.283×10^{-14} to well within 1% for the low power tests. For the high power tests, after normalizing for power level differences, the combined ARHCO-I and HEDL-I value of 1.337×10^{-14} ($\pm 0.7\%$, 1 standard error) shows a 4.1% disagreement relative to the low power measurements. At high power, however, there are corrections, not yet applied, for a larger foil irradiation package size; moreover, some uncertainty is introduced from the run-to-run power level normalization. For ^{238}U fission, the low power NBS fission chamber measurements and combined low and high power ARHCO and HEDL radiochemistry measurements agree to within $\sim 4\%$. While preliminary and subject to change, these results are sufficiently encouraging to warrant special notice at this time.

AEROJET NUCLEAR COMPANY

NBS fission chamber measurements and ANL fission track, ANC, ARHCO, ANL and HEDL radiochemistry foil irradiations were made in the CFRMF. Experimental devices and conditions for these measurements and irradiations are presented. The CFRMF power level control and monitor system is described and discussed. Radiochemistry foils (Au, In and ^{235}U) were counted and the results are presented and discussed. A compilation of decay scheme information for use in analysis of the radiochemistry foil measurements is presented. Full-energy peak efficiencies for Ge(Li) detectors are presented and evaluated on relative and absolute bases.

ARGONNE NATIONAL LABORATORY

Methods for preparation of ^{235}U , ^{237}Np , and ^{239}Pu stock solutions are described. SSTR's for these materials were prepared and irradiated in the

CFRMF facility. A very preliminary result for a single ^{235}U SSTR is reported. Stock solution and SSTR preparation of ^{238}U are nearing completion.

The measured fission rate of an exploratory ^{235}U foil irradiated for seven hours in CFRMF at a stated reactor power of 0.6 kW on December 6, 1971, was 1.313×10^{-14} fissions/atom-sec, based upon ^{140}Ba activity. A complete ANL-I non-fissile foil set irradiated in CFRMF on February 15, 1972, for seven hours at a stated reactor power of 9.75 kW is now being counted.

ATLANTIC RICHFIELD HANFORD COMPANY

Measurement constants, activity analyses and reaction rates are reported for the ARHCO-I radiochemistry foil set irradiated for seven hours in CFRMF on February 8, 1972, at a stated reactor power of 9.75 kW. ARHCO low power fission foil reaction rates are reported in combination with the HEDL measurements in the HEDL section.

ATOMICS INTERNATIONAL

Boron specimens have been prepared at Atomics International for irradiation in the CFRMF facility at Aerojet Nuclear. Ten milligram-size crystals of enriched ^{10}B and two crystals each of natural boron and enriched ^{11}B were placed within holes in a 1/2-inch diameter thin aluminum disk. This disk was placed between monitor foils of In, Au and Al in a foil set holder supplied by HEDL. Irradiation of this holder in the CFRMF core center will take place in early April, 1972, within the duplicate NBS fission chamber.

BROOKHAVEN NATIONAL LABORATORY

The SAND-II Library of evaluated energy dependent cross sections was received by the National Neutron Cross Section Center on June 21, 1971, for comparison with ENDF/B-III. The isotopes and reactions of primary importance were defined by HEDL and these were plotted and compared where common to both libraries. Data are given on seven such reactions.

HANFORD ENGINEERING DEVELOPMENT LABORATORY

A meeting at AEC Headquarters in December 1971 resulted in the establishment of a pool of highly enriched fission foil materials for use by participants of the ILRR program and for routine LMFBR dosimetry. This material is now being used to fabricate aluminum encapsulated metal fission foils at ORNL for the CFRMF and other ILRR program tests.

A ^{140}Ba - ^{140}La standard for the CFRMF fission foil radiochemistry measurements has been prepared and will be distributed to participants in early March 1972.

Results of low and high power CFRMF tests related to run-to-run power level normalization, check-out of ^{140}Ba - ^{140}La counting for ^{235}U and ^{238}U fission foils, and results of the high-power HEDL-I Non-Fission foil set irradiation are documented and discussed. The HEDL-I foil set was irradiated for seven hours on February 16, 1972, at a stated reactor power of 9.75 kW.

Comparison of previous SAND-II analysis of the evaluated reaction cross sections for $^{54}\text{Fe}(n,p)^{54}\text{Mn}$ and $^{58}\text{Ni}(n,p)^{58}\text{Co}$ with recent energy dependent activation measurements of Paulsen and Widera show very good agreement, a result which supports the use of these cross sections for fast reactor dosimetry.

SAND-II Monte Carlo error analysis results for the ^{235}U fission spectrum based on Fabry's recent evaluation of available integral reaction rate data are presented. These integral data are intended for use in developing a CSEWG benchmark for the ^{235}U fission spectrum for reactions of current interest for fast reactor dosimetry.

Background information and some available flux-spectral and integral data on the CEN-SCK $\Sigma\Sigma$ facility are provided to assist in the planning and selection of appropriate standard neutron fields for future ILRR program study.

LAWRENCE LIVERMORE LABORATORY

During the past two quarters, the development and debugging of the statistical model codes COMNUC, CASCADE, and UHL have continued. The coupled-channels optical model program FOURPLUS was put into operation and tested. Some preliminary results have been obtained for the neutron induced reactions on ^{197}Au and ^{239}Pu .

LOS ALAMOS SCIENTIFIC LABORATORY

Neutron flux spectra associated with several LASL critical assemblies are presented in order to provide background information for the planning and selection of standard neutron fields for future ILRR program study.

NATIONAL BUREAU OF STANDARDS

Fission rate measurements at the CFRMF were performed during the first week of February 1972. The NBS Fission chamber and associated electronics

was transported to Idaho without incident and performed reliably at the reactor. Fission rates per nucleus are given but without final evaluation or error assignments.

Properties of the NBS ^{252}Cf spontaneous fission sources as they are relevant for activation detector calibration are reviewed in order to provide background information for the planning and selection of standard neutron fields for future ILRR program study.

OAK RIDGE NATIONAL LABORATORY

Batches of oxides and metals containing at least 200 g of each isotope ^{235}U , ^{238}U , ^{237}Np , and ^{239}Pu , have been allocated by the AEC Division of Research for sole use in the various LMFBR associated dosimetry programs. Work is now in progress to form 20 samples of high purity metal from each of the isotopes noted. All oxide reductions to metal and subsequent rolling of 0.005-in. thick sheets of ^{235}U , ^{238}U , and ^{239}Pu are in progress or have been completed. Delivery of the ^{235}U and ^{238}U samples to HEDL is anticipated by March 31, 1972.

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

Preliminary radiochemistry foil irradiations were made during December 1971 and January 1972. NBS fission chamber measurements, ANL fission track irradiations and ARHCO, ANL, HEDL and ANC radiochemistry foil irradiations were made in February 1972. More HEDL radiochemistry foil irradiations and the AI helium production irradiation are planned for March 1972.

Accomplishments and Status

Irradiations so far completed are summarized in Table ANC-I.

Table ANC-I

ILRR-CFRMF Irradiations

<u>Run Designation</u>	<u>Laboratory</u>	<u>Date</u>	<u>Power</u>	<u>Time</u>
<u>Preliminary Radiochemistry Foils</u>				
Preliminary-I	ANC, ANL, ARHCO			
	HEDL	12/6/71	~0.6kW	7 hours
Preliminary-II	HEDL	12/7/71	~0.6kW	7 hours
Preliminary-III	HEDL	1/26/72	~0.6kW	7 hours
<u>Fission Rate Measurements</u>				
Fission Chamber	NBS	2/1-2/4/72	~0.1-6kW	32 hours
Fission Tracks	ANL	2/3-4/72	~0.6kW	5 hour
<u>Radiochemistry Foils</u>				
ARHCO-I	ARHCO	2/8/72	~9.75kW	7 hours
ANL-I	ANL	2/15/72	~9.75kW	7 hours
HEDL-I	HEDL	2/16/72	~9.75kW	7 hours
ANC-I	ANC	2/29/72	~9.75kW	7 hours

Tentative plans for more HEDL radiochemistry foils and AI helium production irradiations have been made. Approximately 105 hours of CFRMF operating time have been used in support of the ILRR program.

Expected Achievements

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

B. CFRMF FLUX SPECTRAL, POWER LEVEL, GRADIENT DETERMINATIONS

J. W. Rogers and I. E. Stepan

Objective

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

Summary

Previously determined neutron environment information has been disseminated and reported in the previous progress reports. Additional flux gradient information has been obtained and is presented. An up-to-date shape of the neutron energy spectrum is presented. The CFRMF power level control and monitor system is described and discussed. Power level reproducibility studies are presented and discussed.

Accomplishments and Status

1. Flux Gradient

During the NBS fission chamber measurements some additional vertical flux gradient information was obtained from ^{239}Pu and ^{237}Np . Fission rates were measured at 4 positions above the centerline of the CFRMF. These results are presented in Table ANC-II along with previous ^{235}U and ^{237}Np results obtained with other fission chambers. These results show that in the region between 0.5" above and 0.5" below the centerline where the foil packages and sensitive portions of the fission chambers are located during irradiation there is apparently a 0.3% gradient observable.

Table ANC-II

CFRMF Vertical Relative Fission Rates

Inches above CFRMF Centerline	^{235}U (a)	^{237}Np (a)	^{237}Np (b)	^{239}Pu (b)
4	0.897	0.934	0.905	0.910
3	0.940	0.964	0.942	0.948
2	0.976	0.988	0.969	0.976
1	0.992	0.999	0.995	0.997
0	1.000	1.000	1.000	1.000

^aANC (Kirm type) fission chambers

^bNBS (back to back type) fission chamber

2. Flux Shape

In the previous ILRR progress reports the figure depicting the shapes of the measured and calculated CFRMF neutron energy spectrum was from a preliminary data analysis. Figure ANC-I is presented here to update these results.

3. Power Level Control

Due to the importance of knowing what the power level of the CFRMF is at each irradiation in the ILRR program it is felt that a description of the power level control system would be helpful for those concerned.

By means of a compensated ion chamber, reference voltage source and appropriate amplifiers, the regulating rod is controlled at a position which maintains the reactor critical and at a constant and preset power. The regulating rod control system is basically a conventional direct-current servo system. The signal applied to the first stage of amplification is the difference between a current proportional to the power level of the reactor as measured by an ion chamber, and a current obtained from a reference voltage source. The power level error signal is then amplified and used to drive the regulating rod to correct for any power level deviation. The amplification between the ion chamber and torque motor is obtained by three amplifier stages of solid state electronics. Three

stages are used, partly for the convenience of breaking up the amplification by function and partly to obtain compatible impedance levels. The first stage is a high quality D.C. amplifier with low drift characteristics. The second stage is used to add in and adjust the gain of the tachometer and limit stop signals. The third stage provides the necessary power amplification to drive the torque motor.

The gamma compensated ion chamber, reference voltage and first stage D.C. amplifier primarily determine the reproducibility and linearity of multiples of the power level. Positioning of the chamber and the gamma to neutron ratio of the radiation incident on the chamber are the primary variables affecting it. At the CFRMF, where reactivity measurements are made at low power level (~ 50 Watts) and irradiations are made at higher power levels (up to 10 kilowatts), the chamber must be at different positions for each application. When the chamber is moved there is a repositioning error that will result in a power level shift. At present adequate data are not available to quote any uncertainty due to positioning of the chamber. When the CFRMF is operated at the different power levels the gamma to neutron ratio at the chamber changes, resulting in a change in the gamma compensation on the chamber. This affects the response of the chamber, resulting in possible shifts in power level. Again adequate data on this effect are not available to quote uncertainties.

The reference voltage setting device, which is used to preset the level at which the power is controlled, consists of a voltage power supply, potentiometer and two switchable precision resistors ($\times 100$ and $\times 1000$). The overall stability of this device over the normal range of ambient temperatures is approximately 0.01%.

The variable resistance of the potentiometer and switchable precision resistors in conjunction with the input impedance of the first stage D.C. amplifier cause the changes in the output of the reference voltage for the power level. The reference voltage output was measured as a function of potentiometer setting at both of the precision resistor settings and the results are tabulated in Table ANC-III. These results provide the necessary information for determining the linearity of the power level in relation to the reference voltage settings.

Table ANC-III

CFRMF Relative Reference Voltage Output

<u>Potentiometer</u>	<u>x100</u>	<u>X1000</u>
100	16.5	16.7
200	33.2	33.3
300	49.9	50.0
400	66.6	66.7
500	83.3	83.3
600	100.	100.
700	117.	117.
800	134.	134.
900	150.	151.
950	159.	160.
975	163.	164.
990	165.	167.
1000	167.	169.

These results are presented relative to the 600 setting on the potentiometer.

In addition to setting and controlling the power level it is necessary to monitor it. The power level of the CFRMF is normally monitored by two power level channels, #1 and #2. These two power level channels are identical except for chamber location. A power level channel consists of a compensated ion chamber, battery power supply, Sigma Pre-Amplifier, Sigma Amplifier, and the two channels share a strip chart recorder. The signal from the chamber is fed to the Sigma Pre-Amp. This Pre-Amplifier is made up of two stages of D.C. amplification and a cathode follower output with associated gain controlling networks and a dual compound regulated power supply. The gain of the Pre-Amplifier can be decreased by a factor of ten and is accomplished when the power reaches ~750 watts. The output of the Pre-Amp is routed to the Sigma Amplifier and then to the strip chart recorder. An additional auxiliary power level monitor was installed at the CFRMF consisting of an auxiliary ^{235}U fission chamber, battery power supply and high stability current amplifier. The linearity of this system over the

range of the CFRMF power level is excellent. The output of this system is read directly on a digital voltmeter.

The actual power of the CFRMF reactor core at the 600 x 100 reference voltage setting was determined by obtaining the ^{235}U fission rate throughout the core. Dilute ^{235}U in Al wires was used to obtain the fission rate distributions and power per gram of ^{235}U in the core fuel elements. The actual power level at this setting was found to be 600 ± 12 watts.

Figure ANC-II is a midplane diagram of the CFRMF reactor core and associated parts. This diagram shows the location of core components, power level distribution and control and monitor chamber positions.

Table ANC-IV is a compilation of results obtained from several monitors of the CFRMF power level at different reference voltage settings. The gold activation monitors were 5-mil thick foils placed in the center of the CFRMF and irradiated for one hour at the different settings. The foils were counted using standard NaI techniques and the saturated reaction rates were computed from the results. The ^{233}U Kirn type fission chamber was also located at the center of the CFRMF and standard pulse counting electronics were used to collect the fission rate data from this chamber. The power control (servo), standard auxiliary fission chamber and level monitors #1 and #2 compensated ion chambers (CIC) have been described previously and are located as shown in Figure ANC-II. The results are presented in percentage of what was obtained relative to the setting of 600 x 100 on the reference voltage of the power level control (servo).

Expected Achievements

More fission rate profiles are planned. More evaluation of the power level control and monitor systems will be made.

TABLE ANC-IV
CFRMF Power Level Control Settings and Monitor Readings

Reference Voltage Setting (% of 600x100)	Au Activation	²³³ U Kirm Fission Chamber	²³⁵ U Auxiliary Fission Chamber	Power Level #1 CIC Chamber	Power Level #2 CIC Chamber
100 x 100 (16.7)	16.8	16.7	16.7	16.2	16.7
340 x 100 (56.8)	- - -	56.8	- - -	57.1	56.8
500 x 100 (83.5)	83.5	83.5	83.1	83.5	78.8
600 x 100 (100.)	100	100	100	100	100
700 x 100 (116.)	- - -	117	- - -	116	116
800 x 100 (133.)	- - -	134	- - -	134	134
900 x 100 (150.)	- - -	150	- - -	190*	189*
1000 x 100 (167.)	164	166	162	165*	157*
100 x 1000 (167.)	165	- - -	162	178*	180*
500 x 1000 (835.)	810	- - -	802	752*	778*
990 x 1000 (1650.)	1640	- - -	1620	1650*	1570*

*These values are what was obtained following the x 0.10 gain change of the Pre-Amp and indicate there is a linearity problem which is being investigated.

Figure ANC-I. Measured and Calculated CFRMF Neutron Energy Spectrum

ANC-8

Figure ANC-II. Midplane Core Diagram of CFRMF Reactor

C. CFRMF-ILRR IRRADIATIONS DESCRIPTIONS

J. W.-Rogers

Objective

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

Summary

Descriptions of all CFRMF-ILRR program tests are presented. Any known differences in any conditions are stated.

Accomplishments and Status

Ten separate tests have been made in the ILRR program using the CFRMF and are tabulated in Table ANC-I (section ANC-A). The following is a test by test description with the main emphasis on those aspects in which ANC was involved.

1. Preliminary I (12-6-71)

Power Level Setpoint - 983 x 1000 (600 watts)

Shim Position - 03030

Time In - 09:00:30 MST

Time Out - 16:00:30 MST

Total Exposure - 7 hours

Power Level #1 - 78.5

Power Level #2 - 66.0

The foils for these tests are described elsewhere (see Figure HEDL-1). The foils were contained in a spool shaped aluminum basket 0.985" long and 1.480" in diameter. The foils were placed on a 20-mil thick aluminum shelf that is 10 mils below the centerline of the basket. This shelf is inside the 20-mil thick 0.600" inside diameter cylinder part of the basket. The basket was then placed on an aluminum stand in the CFRMF sample tube so as to position the shelf 10 mils below the centerline. This allowed the foil package to be located perpendicular to the vertical plane and within 10 mils of the CFRMF centerline.

After irradiation, two foils (Au-B and ^{235}U -4) were retained by ANC, one foil (^{235}U -3) was sent to ANL[†] and all other foils were sent to HEDL.*

2. Preliminary II (12-7-71)

Power Level Setpoint - 983 x 1000 (600 watts)

Shim Position - 03030

Time In - 02:46:37 MST

Time Out - 09:46:37 MST

Total Exposure - 7 hours

Power Level #1 - 79.0

Power Level #2 - 66.0

The foil containment and positioning were identical to Preliminary-I except that the foils were separated by 1-mil paper spacers, see Figure HEDL-1.

After irradiation all foils were sent to HEDL.*

3. Preliminary III (1-26-71)

Power Level Setpoint - 600 x 100 (equal to old 983 x 1000 setting)

This change in the setpoint resulted from the necessity to reposition (prior to this irradiation) the ion chambers in the power level control and monitor systems. This repositioning was necessary to allow CFRMF operation at the 10-kW level.

Shim Position - 03030

Time In-00:16:24 MST

Time Out - 07:16:24 MST

Total Exposure - 7 hours

Power Level #1 - 57.6

Power Level #2 - 53.6

The foil containment and positioning were identical to Preliminary-II except that 0.5-mil thick aluminum foil was used to form foil packets of indium, ^{238}U , and gold. Two other packets of one gold and one indium foil each (power level monitors) were assembled and positioned 1 inch above the other three packets by means of a piece of 5-mil thick stainless steel tubing 0.5 inch in diameter. The gold-indium packets were attached to one end of the tubing with one small piece of tape and the other end of the tube inserted into the cylinder part of the basket and rested on the top foil packet, see Figure HEDL-1.*

[†] See Table ANL-3.

* See Tables HEDL-3 to 7.

4. Fission Chamber NBS (2-2-72)

Power Level Setpoint - 600 x 100 (600 watts)

Shim Position - 03030

Time In - 20:48:46 MST

Time Out - 21:18:46 MST

Total Exposure - 30 minutes

Power Level #1 - 56.0

Power Level #2 - 56.0

Auxiliary Fission Chamber - 0.136

The foil containment and positioning arrangement were completely changed for this and all subsequent tests. This arrangement is described in detail in Section HEDL-C. Figure ANC-III is a photograph of the entire assembly that is inserted into the CFRMF sample tube for each irradiation. At the bottom is the 12.070" aluminum stand on which the foil containing chamber* rests. The upper three inches of the stand are hollow with 20-mil thick wall and end. Attached to the chamber is the stem leading to the manifold on the upper end. Mounted on the stem one inch above the chamber is a positioning and foil holding spacer plate. On the top side of this spacer plate are milled out recessions in which power level monitor foils (Au and In) can be placed external to the foil containing chamber. Only the two recessions 180° apart are used to hold the power level monitor foils. The foils, separated by 0.5-mil aluminum, are placed in the recessions and retained by a piece of 20-mil diameter aluminum wire. At the top of the stem against the bottom of the manifold is mounted another positioning spacer plate to assure complete alignment and positioning of the assembly in the sample entry tube of the CFRMF. In the case of fission chamber measurements gas supply and electrical connections are attached to the manifold and in the case of foil irradiations only a string is attached to the manifold for insertion and removal of the assembly. Figure ANC-IV is a closeup photograph of the foil containing chamber and Au and In power level normalizing foil holder. Figure ANC-V is a photograph of the chamber disassembled down to the foil location. In this test the fission chamber contained two fission foils (^{235}U and ^{238}U). (Note that the foils in the chamber are parallel to the vertical plane of the CFRMF in this arrangement.)

* Duplicate NBS fission chamber, see Figure HEDL-2.

Prior to this test an auxiliary fission chamber power level monitor system (see Section ANC-B) was installed as an additional indicator of the CFRMF power level.

After irradiation, two foils (Au-AA and In-2#1) were retained by ANC and the other two monitor foils were sent to HEDL.*

5. Fission Tracks ANL (2-4-72)

Power Level Setpoint - 600 x 100 (600 watts)

Shim Position - 03030

Time In - 11:05:30 MST

Time Out - 12:05:30 MST

Total Exposure - 1 hour

Power Level #1 - 56.2

Power Level #2 - 56.2

Auxiliary Fission Chamber - 0.136

The foil containment and positioning arrangement were identical to the Fission Chamber NBS (2-2-72) test except that a duplicate chamber-stem-manifold assembly without gas and electrical attachments was used. The chamber contained an ANL fission track packet (described in section ANL-A).

After irradiation, two foils (Au-AC and In-2#3) were retained by ANC, the other monitor foils were sent to HEDL* and the fission track packet was sent to ANL.

6. Fission Chamber NBS (2-4-72)

Power Level Setpoint - 600 x 100 (600 watts)

Shim Position - 03030

Time In - 13:42:50 MST

Time Out - 14:42:50 MST

Total Exposure - 1 hour

Power Level #1 - 56.2

Power Level #2 - 55.8

Auxiliary Fission Chamber - 0.136

* See Tables HEDL-3 and 4.

The foil containment and positioning arrangement were identical to the Fission Chamber NBS (2-2-72) test except that the fission chamber contained two other fission foils (^{239}Pu and ^{237}Np).

After irradiation, two foils (Au-AE and In-2#5) were retained by ANC and the other two monitor foils were sent to HEDL.*

7. ARHCO-I (2-8-72)

Power Level Setpoint - 975 x 1000

Shim Position - 03333

The shim position was changed because the regulating rod was operating too close to the outer limit with the old shim position of 03030.

Time In- 00:49:14 MST

Time Out - 07:49:17 MST

Total Exposure - 7 hours

Power Level #1 - 87.5 - 88.9

Power Level #2 - 98.1 - 99.7

The two values of power level shown represent the minimum and maximum readings observed throughout the irradiations.

Auxiliary Fission Chamber - 2.06 - 2.07

The fission chamber was repositioned prior to this test to assure that no saturation levels would be reached at this power level. At the 500 x 100 setpoint the reading was reduced by 74.55%.

The foil containment and positioning arrangement were identical to the Fission Tracks ANL (2-4-72) test except that the chamber contained the ARHCO-I radiochemistry foil packet (described in the HEDL chapter and reference HEDL-4).

After irradiation, two foils (Au-AG and In-2#7) were retained by ANC, the other two monitor foils and foil packet were sent to HEDL.*

* See Tables HEDL-3 and 4.

8. ANL-I (2-15-72)

Power Level Setpoint - 975 x 1000

Shim Position - 03333

Time In - 01:22:11 MST

Time Out - 08:22:11 MST

Total Exposure - 7 hours

Power Level #1 - 84.5 - 86.8

Power Level #2 - 95.2 - 96.6

Auxiliary Fission Chamber - 2.06

Prior to this test all chambers of the CFRMF were actually moved when new chamber securing devices were installed and reactivity measurements were made. The chambers were returned to their previous positions as closely as possible.

The foil containment and positioning arrangement were identical to the ARHCO-I (2-8-72) test except that the chamber contained the ANL-I radio-chemistry foil packet.

After irradiation two foils (Au-AI and In-2#9) were retained by ANC, the other two monitor foils were sent to HEDL^{*} and the foil packet was sent to ANL.

9. HEDL-I (2-16-72)

Power Level Setpoint - 975 x 1000

Shim Position - 03333

Time In - 00:12:17 MST

Time Out - 07:12:17 MST

Total Exposure - 7 hours

Power Level #1 - 85.5 - 86.0

Power Level #2 - 95.5 - 96.0

Auxiliary Fission Chamber - 2.06

The foil containment and positioning arrangement were identical to the ANL-I (2-16-72) test except that the chamber contained the HEDL-I radiochemistry foil packet.

After irradiation, two foils (Au-AK and In-2#11) were retained by ANC, the other two monitor foils and foil packet were sent to HEDL.*

* See Tables HEDL-3 and 4.

10. ANC-I (2-29-72)

Power Level Setpoint - 975 x 1000

Shim Position - 03333

Time In - 01:04:14 MST

Time Out - 08:04:14 MST

Total Exposure - 7 hours

Power Level #1 - 84.5 - 86.5

Power Level #2 - 96.0 - 97.5

Auxiliary Fission Chamber - 2.065 - 2.085

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

The foil containment and positioning arrangement were identical to the HEDL-I (2-16-72) test except that the chamber contained the ANC-I radiochemistry foil packet. Another foil packet, containing some aluminum samples from ORNL material to be used in encapsulation of the fission foils, was attached to the stem two inches above the chamber.[†]

After irradiation, two foils (Au-AM and In-2#13) and the radiochemistry foil packet were retained by ANC, the other two monitor foils* and the ORNL aluminum foil packet were sent to HEDL.

Expected Achievements

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

[†] See HEDL chapter, Section A.

* See Tables HEDL-3 and 4.

Figure ANC-II. Foil Containment and Positioning Assembly for CFRMF-ILRR Tests.

Figure ANC-IV. Assembled Foil Containing Chamber and Monitor Foil Holding Spacer Plate.

Figure ANC-V. Disassembled Foil Containing Chamber.

D. DECAY-SCHEME INFORMATION FOR ANALYSIS OF RADIOCHEMISTRY FOIL MEASUREMENTS

R. G. Helmer

Objective

- To provide a set of decay scheme information allowing various laboratories to carry out consistent analyses of the radiochemistry foil packets (see description in the HEDL chapter and reference HEDL-4).

Summary

For the radioactivities that are to be observed in the foils in these packets, the available decay scheme literature has been reviewed. Values are presented for the half life and the absolute gamma-ray intensities for the one or two most prominent transitions in each isotope.

Accomplishments and Status

There are five radiochemistry foil packets to be irradiated and analyzed. The first step in this analysis of these foils will be to see if the results from the various laboratories agree. The decay scheme information needed for this comparison is given in Table ANC-V. Although for the interlaboratory comparison, one need only use the same half lives and branching ratios, the accuracy of these values is important for the subsequent analysis of the CFRMF and other neutron spectra.

Two levels of analysis are presented in Table ANC-V. For those values where an error (1σ level) is given, an extensive review of the available literature has been made. We have, where possible, considered the quality of the work, and feel that the values quoted are the best available, and that the errors are reasonable. Where no error is shown only a brief literature review has been made. In these cases, it is felt that uncertainty is in the last digit given for each value.

Expected Accomplishments

If, in the process of the radiochemistry foil experiments, it is found that more complete data are needed, the pertinent information in Table ANC-V will be improved where possible.

Table ANC-V

Half-lives and Branching Ratios for Radiochemistry Foil Measurements

<u>Parent Isotope</u>	<u>Half-life</u>	<u>Gamma-Ray Energy (keV)</u>	<u>Branch Ratio % of decays</u>
²⁷ Mg	9.5m	842	70
		1013	30
			} sum = 100.0
²⁴ Na	15.00h	1368.55	99.99
		2753.9	99.90
¹⁹⁸ Au	2.696d	411.794 ± 0.008	95.47 ± 0.10
^{116m} In	54.0m	1293.3	80
^{115m} In	4.5h	335	47
⁶⁴ Cu	12.78h	511.00	36.8 ± 1.6
⁶⁰ Co	5.261yr	1332.491 ± 0.041	99.985 ± 0.001
		1173.208 ± 0.025	99.86 ± 0.02
⁵⁸ Co	71.3d	810.757 ± 0.021	99.44
⁵⁴ Mn	312.6 ± 0.3d	834.827 ± 0.021	99.978
⁵⁹ Fe	44.6d	1099.224 ± 0.025	56
		1291.564 ± 0.028	44
⁴⁶ Sc	84.0d	889.258 ± 0.018	99.98
		1120.516 ± 0.025	99.99
⁴⁷ Sc	3.39d	160	69.0 ± 2.5
⁴⁸ Sc	1.83d	983.3	99.99
¹⁴⁰ La	40.26 ± 0.02h	1596.18 ± 0.05	95.33 ± 0.16
¹⁴⁰ Ba	12.79d	537.38	23.8
²³⁹ Np	2.35d	277.5	14
		228.2	12

$$\left. \begin{array}{l} 70 \\ 30 \end{array} \right\} \frac{T_{Ba}}{T_{Ba}-T_{La}} = 1.151$$

E. EVALUATION OF DECAY-SCHEME DATA ON FISSION-PRODUCT NUCLIDES

R. C. Greenwood, R. G. Helmer and C. W. Reich

Objective

• To critically evaluate the currently available data on the gamma-ray branching ratios of these nuclides and to provide accurate data for the determination of absolute amounts of the various nuclides produced by the irradiation of fission foils in the ILRR program.

Summary, Accomplishments and Status

The currently available decay-scheme data on the fission-product nuclides ^{95}Zr , ^{95}Nb , ^{97}Zr , ^{97}Nb , ^{103}Ru and ^{140}La have been critically evaluated. From this evaluation we have obtained a set of values for the absolute intensities of the gamma rays emitted in the decay of these nuclides. These data are summarized in Tables ANC-VI-XI.

Expected Accomplishments

The evaluation of data on additional fission-product nuclides will continue, with the emphasis being on those nuclides deemed to be the most important to the ILRR effort.

Table ANC-VI

Summary of gamma-ray branching-ratio data for ^{95}Zr

E_{γ} (keV)	Gamma-Ray Intensity ($\gamma/100$ disintegrations)
235.65 ± 0.03	0.24 ± 0.06
724.18 ± 0.02	44.1 ± 0.5
756.72 ± 0.02	54.7 ± 0.5

Table ANC-VII

Summary of gamma-ray branching-ratio data for ^{95}Nb

E_{γ} (keV)	Gamma-Ray Intensity ($\gamma/100$ disintegrations)
765.79 ± 0.02	99.83 ± 0.04

Table ANC-VIII

Summary of gamma-ray branching-ratio data for $^{97}\text{Zr}^*$

E_γ (keV)	Gamma-ray Intensity ($\gamma/100$ disintegrations)
254.4 $\pm$ 0.1	1.3 $\pm$ 0.2
355.53 $\pm$ 0.05	2.1 $\pm$ 0.3
507.75 $\pm$ 0.05	4.9 $\pm$ 0.5
602.45 $\pm$ 0.05	1.5 $\pm$ 0.2
703.74 $\pm$ 0.05	1.0 $\pm$ 0.1
743.35 $\pm$ 0.05	93.3 $\pm$ 0.4
1021.4 $\pm$ 0.15	1.1 $\pm$ 0.2
1148.0 $\pm$ 0.1	2.5 $\pm$ 0.3
1362.6 $\pm$ 0.1	1.0 $\pm$ 0.1
1750.4 $\pm$ 0.1	1.1 $\pm$ 0.2

* Only the stronger (intensity $>1\%$) gamma rays are included here

Table ANC-IX

Summary of gamma-ray branching-ratio data for ^{97}Nb

E_γ (keV)	Gamma-ray Intensity ($\gamma/100$ disintegrations)
177.97 $\pm$ 0.30	0.05 $\pm$ 0.01
238.37 $\pm$ 0.30	0.05 $\pm$ 0.01
549.25 $\pm$ 0.20	0.05 $\pm$ 0.01
657.92 $\pm$ 0.10	98.1 $\pm$ 0.1
719.47 $\pm$ 0.20	0.09 $\pm$ 0.01
857.50 $\pm$ 0.20	0.05 $\pm$ 0.01
909.57 $\pm$ 0.10	0.04 $\pm$ 0.01
1024.53 $\pm$ 0.30	1.1 $\pm$ 0.1
1117.01 $\pm$ 0.20	0.09 $\pm$ 0.01
1148.56 $\pm$ 0.30	0.05 $\pm$ 0.01
1268.63 $\pm$ 0.10	0.16 $\pm$ 0.02
1515.64 $\pm$ 0.20	0.12 $\pm$ 0.02
1629.13 $\pm$ 0.20	0.03 $\pm$ 0.01

Table ANC-X

Summary of gamma-ray branching-ratio data for ^{103}Ru
 $(T_{1/2} = 39.6 \pm 0.5 \text{ days})$

E_{γ} (keV)	Gamma-ray Intensity ($\gamma/100$ disintegrations)
39.758 $\pm$ 0.014	0.066 $\pm$ 0.005
53.275 $\pm$ 0.010	0.33 $\pm$ 0.03
114.65 $\pm$ 0.05	0.011 $\pm$ 0.005
241.8 $\pm$ 0.1	0.013 $\pm$ 0.004
294.78 $\pm$ 0.08	0.27 $\pm$ 0.03
357.5 $\pm$ 0.3	<0.007
443.70 $\pm$ 0.12	0.38 $\pm$ 0.05
497.08 $\pm$ 0.01	90. $\pm$ 3.
557.11 $\pm$ 0.13	0.81 $\pm$ 0.05
610.40 $\pm$ 0.07	5.7 $\pm$ 0.2
611.53 $\pm$ 0.25	$\approx$ 0.05

Table ANC-XI

Summary of gamma-ray branching-ratio data for ^{140}La
 $(T_{1/2} = 40.26 \pm 0.02 \text{ hours})$

E_{γ} (keV)	Gamma-ray Intensity ($\gamma/100$ disintegrations)	E_{γ} (keV)	Gamma-ray Intensity ($\gamma/100$ disintegrations)
64.132 $\pm$ 0.006	$\approx$ 0.01	892.0 $\pm$ 2.0	0.23 $\pm$ 0.14
68.916 $\pm$ 0.006	0.062 $\pm$ 0.010	898.7 $\pm$ 2.0	0.27 $\pm$ 0.14
109.417 $\pm$ 0.005	0.022 $\pm$ 0.002	919.57 $\pm$ 0.10	2.86 $\pm$ 0.14
131.122 $\pm$ 0.006	0.52 $\pm$ 0.05	925.20 $\pm$ 0.09	7.1 $\pm$ 0.3
173.542 $\pm$ 0.009	0.12 $\pm$ 0.02	936.9 $\pm$ 0.4	0.06 $\pm$ 0.02
241.964 $\pm$ 0.011	0.48 $\pm$ 0.05	951.06 $\pm$ 0.06	0.51 $\pm$ 0.04
266.550 $\pm$ 0.012	0.52 $\pm$ 0.05	1062 $\pm$ 2	0.23 $\pm$ 0.14
306.9 $\pm$ 0.2	0.026 $\pm$ 0.009	1416 $\pm$ 2.5	0.2 $\pm$ 0.1
328.762 $\pm$ 0.010	19.5 $\pm$ 0.8	1521.8 $\pm$ 2.0	0.14 $\pm$ 0.10
397.79 $\pm$ 0.11	0.08 $\pm$ 0.02	1596.18 $\pm$ 0.05	95.33 $\pm$ 0.16
432.505 $\pm$ 0.020	2.86 $\pm$ 0.11	1877.3 $\pm$ 1.5	0.071 $\pm$ 0.024
438.5 $\pm$ 0.4	0.02 $\pm$ 0.01	2347.8 $\pm$ 0.2	0.88 $\pm$ 0.04
446.0 $\pm$ 0.5	$\approx$ 0.024	2465.3 $\pm$ 0.8	0.17 $\pm$ 0.05
487.022 $\pm$ 0.016	45.3 $\pm$ 1.9	2494 $\pm$ 3	0.012 $\pm$ 0.008
615.2 $\pm$ 0.7	0.042 $\pm$ 0.021	2521.45 $\pm$ 0.10	3.46 $\pm$ 0.14
653.2 $\pm$ 2.0	0.041 $\pm$ 0.002	2533.4 $\pm$ 0.7	0.004 $\pm$ 0.002
751.70 $\pm$ 0.04	4.17 $\pm$ 0.19	2547.5 $\pm$ 0.5	0.112 $\pm$ 0.007
798.3 $\pm$ 1.5	0.17 $\pm$ 0.09	2898.8 $\pm$ 0.6	0.064 $\pm$ 0.005
815.74 $\pm$ 0.04	22.7 $\pm$ 0.7	3118.7 $\pm$ 0.6	0.026 $\pm$ 0.003
867.85 $\pm$ 0.05	5.3 $\pm$ 0.2	3319.6 $\pm$ 0.9	0.045 $\pm$ 0.010

F. GE(LI) DETECTOR EFFICIENCY CURVES

R. C. Greenwood and R. G. Helmer

Objective

- To determine the gamma-ray efficiency, as a function of the gamma-ray energy, of two large Ge(Li) detectors. These detectors will then be used for the absolute disintegration rate measurements on the radio-chemistry foils for the ILRR program.

Summary

Values of the full-energy peak efficiency have been determined at ≈ 25 gamma-ray energies for OR-5 (a 35-cm³ open-end coaxial Ge(Li) detector) and ND-3 (a 60-cm³ closed-end coaxial detector). Above 200 keV, the average uncertainty in the measured efficiency values is 1.0%. Methods of interpolation to determine the efficiency at an arbitrary energy have been investigated. Between 300 and 1500 keV, this interpolation should be accurate to better than 1%.

Accomplishments and Status

The measured values of the full-energy peak efficiencies for the OR-5 and ND-3 detectors are given in Tables ANC-XII and XIII. Over half of these values were obtained using standard sources from IAEA or NBS. The gamma-ray emission rates of the remaining sources were determined by measurements on a NaI(Tl) detector. The uncertainties in Tables ANC-XII and XIII contain four contributions which have been combined in quadrature.

- a. The error in the disintegration rate of the sample as quoted by the supplier (or in the NaI(Tl) measurement of the gamma-ray emission rate).
- b. The uncertainty in the gamma-ray branching ratio and from any decay correction. (Note that this contribution is not present for the sources calibrated with a NaI(Tl) spectrometer.)
- c. The counting statistics
- d. The reproducibility of the source position. This contribution is taken to be 0.5% in all cases.

From the data in Tables ANC-XII and XIII, we have some test of the accuracy of the disintegration rates of the standard sources. For two sources (^{137}Cs and ^{60}Co) we have IAEA and NBS standards. The efficiency values determined with these sources are compared in Table ANC-XIV. These results indicate that the two suppliers agree within about 1%; and in all cases there is no indication that the quoted errors are too small.

As noted, the gamma-ray emission rates of several sources were measured with a NaI(Tl) spectrometer. This NaI(Tl) detector is a home-mounted assembly for which the dimensions of the crystal and all of the mounting materials are accurately known. This allows an accurate determination of the source-detector distance as well as the total efficiency at any energy. In a test of the consistency of the NaI(Tl) standardizations with those done elsewhere, several of the IAEA and NBS sources were counted on the NaI(Tl) detector. The results are given in Table ANC-XV. Excluding the four measurements for which there are difficulties in analyzing the NaI(Tl) spectrum, the average ratio of emission rates is 1.002. The largest deviation of the two values is 1.5%. These results indicate that the remaining NaI(Tl) standardizations are probably accurate to 1%.

In the determination of gamma-ray intensities, it is necessary to interpolate efficiency values for energies between those of the measured points. As a first test of the accuracy of such an interpolation, we have compared the values obtained from a hand-drawn curve through the values in Table ANC-XII (on a $\log \epsilon$ vs $\log E$ plot) with those from a fit to the function

$$\log_e \epsilon = \sum_{n=1}^N a_n (\log_e E)^{n-1}$$

Two sets of coefficients a_n were computed; one for $E < 200$ keV and the second for $E > 200$ keV. A comparison of the interpolated values from the hand-drawn curve and the polynomials is given in Table ANC-XVI where $N = 4$ and 5 (i.e., third- and fourth-order polynomials) for the two energy regions, respectively. These results indicate excellent agreement (<1% difference) between the interpolated values from 300 to 1500 keV. The RMS difference in this region is 0.6% which is essentially the interpolation precision on the hand-drawn curve. (As noted in section ANC-D, this energy region includes most of the gamma rays of interest in the radiochemistry foil experiments.)

Outside of this region the differences increase. Below 150 keV this is due in part to the larger errors on the data points. Between 150 and 300 keV it is due to the lack of data points and the large curvature in the function. Above 1500 keV the increased differences are caused by the lack of points above 2600 keV as well as the tendency of the polynomial to go to zero too rapidly. This is illustrated by the divergence of the extrapolated values.

Expected Achievements

The experimental efficiency curves will be fitted with other functions in order to determine the best method of interpolation. From this procedure, estimates of the uncertainty in the interpolated efficiencies will be made.

Table-ANC-XII
Summary OR-5 Efficiencies

	E_{γ}	ϵ	
^{109}Cd	22.10	$(1.510 \pm 0.076) \times 10^{-3}$	$(\pm 5\%)$
^{241}Am	26.345	$(2.188 \pm 0.177) \times 10^{-3}$	$(\pm 8.1\%)$
^{137}Cs	32.06	$(2.973 \pm 0.104) \times 10^{-3}$	$(\pm 3.5\%)$
^{139}Ce	33.30	$(3.311 \pm 0.166) \times 10^{-3}$	$(\pm 5\%)$
^{153}Gd	41.33	$(4.275 \pm 0.214) \times 10^{-3}$	$(\pm 5\%)$
^{241}Am	59.537	$(5.145 \pm 0.093) \times 10^{-3}$	$(\pm 1.8\%)$
^{109}Cd	88.037	$(5.186 \pm 0.156) \times 10^{-3}$	$(\pm 3\%)$
^{153}Gd	100.31	$(4.964 \pm 0.148) \times 10^{-3}$	$(\pm 3\%)$
^{57}Co	122.063	$(5.076 \pm 0.107) \times 10^{-3}$	$(\pm 2.1\%)$
^{57}Co	136.473	$(4.464 \pm 0.509) \times 10^{-3}$	$(\pm 11.4\%)$
^{141}Ce	145.440	$(4.491 \pm 0.072) \times 10^{-3}$	$(\pm 1.6\%)$
^{139}Ce	165.853	$(4.134 \pm 0.124) \times 10^{-3}$	$(\pm 3\%)$
^{203}Hg	279.188	$(2.320 \pm 0.019) \times 10^{-3}$	$(\pm 0.83\%)$
^{51}Cr	320.078	$(1.963 \pm 0.022) \times 10^{-3}$	$(\pm 1.1\%)$
^{113}Sn	391.688	$(1.5154 \pm 0.0167) \times 10^{-3}$	$(\pm 1.1\%)$
^7Be	477.593	$(1.1794 \pm 0.0130) \times 10^{-3}$	$(\pm 1.1\%)$
^{85}Sr	513.996	$(1.0561 \pm 0.0116) \times 10^{-3}$	$(\pm 1.1\%)$
^{228}Th	583.174	$(9.299 \pm 0.121) \times 10^{-4}$	$(\pm 1.3\%)$
^{137}Cs	661.638	$(7.960 \pm 0.065) \times 10^{-4}$	$(\pm 0.82\%)$
^{137}Cs	661.638	$(7.860 \pm 0.054) \times 10^{-4}$	$(\pm 0.69\%)$
^{54}Mn	834.827	$(6.149 \pm 0.038) \times 10^{-4}$	$(\pm 0.62\%)$
^{88}Y	898.021	$(5.790 \pm 0.087) \times 10^{-4}$	$(\pm 1.5\%)$
^{60}Co	1173.208	$(4.336 \pm 0.025) \times 10^{-4}$	$(\pm 0.57\%)$
^{60}Co	1173.208	$(4.382 \pm 0.029) \times 10^{-4}$	$(\pm 0.67\%)$
^{22}Na	1274.511	$(4.033 \pm 0.024) \times 10^{-4}$	$(\pm 0.60\%)$
^{60}Co	1332.491	$(3.779 \pm 0.022) \times 10^{-4}$	$(\pm 0.57\%)$
^{60}Co	1332.491	$(3.824 \pm 0.026) \times 10^{-4}$	$(\pm 0.67\%)$
^{88}Y	1836.074	$(2.660 \pm 0.033) \times 10^{-4}$	$(\pm 1.24\%)$
^{144}Ce	2185.62	$(2.103 \pm 0.036) \times 10^{-4}$	$(\pm 1.7\%)$
^{228}Th	2614.47	$(1.7720 \pm 0.0167) \times 10^{-4}$	$(\pm 0.94\%)$

Table ANC-XIII

Summary ND-3 Efficiencies

	E (keV)	ε		
¹³⁹ Ce	33.30	(4.299 ± 0.258) × 10 ⁻⁵	(± 6%)	
¹⁵³ Gd	41.33	(4.302 ± 0.215) × 10 ⁻⁴	(± 5%)	
²⁴¹ Am	59.537	(2.424 ± 0.044) × 10 ⁻³	(± 1.8%)	
¹⁰⁹ Cd	88.037	(4.447 ± 0.133) × 10 ⁻³	(± 3%)	
¹⁵³ Gd	100.31	(4.348 ± 0.130) × 10 ⁻³	(± 3%)	
⁵⁷ Co	122.063	(4.814 ± 0.101) × 10 ⁻³	(± 2.1%)	
⁵⁷ Co	136.473	(4.369 ± 0.498) × 10 ⁻³	(± 11.4%)	
¹⁴¹ Ce	145.440	(4.514 ± 0.072) × 10 ⁻³	(± 1.6%)	
¹³⁹ Ce	165.853	(4.280 ± 0.128) × 10 ⁻³	(± 3%)	
²⁰³ Hg	279.188	(2.655 ± 0.023) × 10 ⁻³	(± 0.85%)	
⁵¹ Co	320.078	(2.286 ± 0.025) × 10 ⁻³	(± 1.1%)	
¹¹³ Sn	391.688	(1.7872 ± 0.0197) × 10 ⁻³	(± 1.1%)	
⁷ Be	477.593	(1.4444 ± 0.0159) × 10 ⁻³	(± 1.1%)	
⁸⁵ Sr	513.996	(1.3335 ± 0.0147) × 10 ⁻³	(± 1.1%)	
²²⁸ Th	583.174	(1.1578 ± 0.0151) × 10 ⁻³	(± 1.3%)	
¹³⁷ Cs	661.638	(1.0075 ± 0.0083) × 10 ⁻³	(± 0.82%)	
¹³⁷ Cs	661.638	(9.968 ± 0.069) × 10 ⁻⁴	(± 0.69%)	
⁵⁴ Mn	834.827	(8.007 ± 0.050) × 10 ⁻⁴	(± 0.63%)	
⁸⁸ Y	898.021	(7.486 ± 0.120) × 10 ⁻⁴	(± 1.6%)	
⁶⁰ Co	1173.208	(5.731 ± 0.032) × 10 ⁻⁴	(± 0.56%)	
⁶⁰ Co	1173.208	(5.811 ± 0.040) × 10 ⁻⁴	(± 0.68%)	
²² Na	1274.511	(5.387 ± 0.033) × 10 ⁻⁴	(± 0.61%)	
⁶⁰ Co	1332.491	(5.063 ± 0.028) × 10 ⁻⁴	(± 0.56%)	
⁶⁰ Co	1332.491	(5.129 ± 0.035) × 10 ⁻⁴	(± 0.68%)	
⁸⁸ Y	1836.074	(3.663 ± 0.048) × 10 ⁻⁴	(± 1.3%)	
¹⁴⁴ Ce	2185.62	(2.975 ± 0.051) × 10 ⁻⁴	(± 1.7%)	
²²⁸ Th	2614.47	(2.490 ± 0.023) × 10 ⁻⁴	(± 0.93%)	

Table ANC-XIV
Comparison of Efficiency Values from Standard
Sources from Two Suppliers

Source	Gamma-Ray Energy (keV)	Detector	Source Supplier	Efficiency Value (x10 ⁴)	Difference Between Sources	
					Value (%)	Average(%)
¹³⁷ Cs	661	OR5	IAEA	7.960 ± 0.065	1.26 ± 1.07	1.16
			NBS	7.860 ± 0.054		
		ND3	IAEA	10.075 ± 0.083	1.07 ± 1.08	
			NBS	9.968 ± 0.069		
⁶⁰ Co	1173	OR5	IAEA	4.336 ± 0.025	1.06 ± 0.88	1.23
			NBS	4.382 ± 0.029		
		ND3	IAEA	5.731 ± 0.032	1.39 ± 0.89	
			NBS	5.811 ± 0.040		
	1332	OR5	IAEA	3.779 ± 0.022	1.18 ± 0.89	
			NBS	3.824 ± 0.026		
		ND3	IAEA	5.063 ± 0.028	1.30 ± 0.88	
			NBS	5.129 ± 0.035		

Table ANC-XV
Comparison of NaI(Tl) and IAEA or NBS Standardizations

Source	Gamma-Ray Energy (keV)	Relative Gamma-Ray Emission Rate $\frac{\text{NaI}}{\text{std}}$
^{241}Am	59	1.033 ^a
^{57}Co	122,136	1.015
^{203}Hg	279	0.995 ^b
^{137}Cs -IAEA	661	1.003
^{137}Cs -NBS	661	0.988
^{54}Mn	834	1.005
^{22}Na	1274	1.033 ^c
^{60}Co -IAEA	1332	1.042 ^d
^{60}Co -NBS	1332	1.037 ^d
$^{88}\gamma$	1836	1.004

^aIn NaI(Tl) spectrum, there is some ambiguity in the separation of the full-energy and iodine x-ray escape peaks.

^bThis source was 7 half-lives old.

^cSome error in background subtraction due to presence of 511-1274-keV coincidence summing.

^dIn NaI(Tl) spectrum, the 1173 and 1332 keV peaks are not resolved.

Table ANC-XVI

Comparison of Efficiency Values Interpolated from
Hand Drawn Curve and $\log_e \epsilon$ vs $\log_e E$ Polynomial for the OR-5 Detector

Gamma-Ray Energy (keV)	Efficiency ($\times 10^3$)		Difference (%)
	Hand Curve ^a	Polynomial	
30	2.74	2.699	1.5
40	4.09	3.874	5.4
50	4.79	4.675	2.4
60	5.13	5.154	-0.5
70	5.30	5.393	-1.7
80	5.38	5.464	-1.5
90	5.36	5.422	-1.2
110	5.19	5.146	0.9
130	4.85	4.755	2.0
150	4.43	4.339	2.1
170	4.00	3.937	1.6
190	3.61	3.566	1.2
210	3.25	3.240	0.3
250	2.63	2.649	-0.7
290	2.20	2.206	-0.3
330	1.875	1.873	0.1
400	1.475	1.466	0.6
500	1.11	1.109	0.1
600	0.895	0.8915	0.4
700	0.750	0.7471	0.4
800	0.647	0.6449	0.3
900	0.574	0.5688	0.9
1000	0.515	0.5099	1.0
1200	0.427	0.4236	0.8
1400	0.359	0.3624	-0.9
1600	0.310	0.3156	-1.8
1800	0.272	0.2781	-2.2
2000	0.241	0.2467	-2.3
2200	0.216	0.2199	-1.8
2400	0.195	0.1966	-0.8
2600	0.178	0.1761	1.1
3000	0.1515	0.1416	6
3500	0.1265	0.1079	15
4000	0.1085	0.08206	24

^aBelow 200 keV, a third-order polynomial is used and above this energy a distinct fourth-order polynomial is employed. The points above 2800 keV are an extrapolation.

G. COUNTING OF FOILS FROM PRELIMINARY-I RADIOCHEMISTRY FOIL PACKET

R. C. Greenwood and R. G. Helmer

Objective

- To determine activity levels to be expected in, and to test counting procedures for, planned fission foil experiments.

A ^{235}U foil was counted over a period from 1 day to 35 days after irradiation (7 hours at 0.6 kW) in the CFRMF. Although the activity level was very low and the results are, therefore, limited by the counting statistics, excellent agreement is found between measurements on the same detector system and between two different systems. A gold monitor foil was also counted.

Summary, Accomplishments and Status

The gamma-ray spectra from the gold and ^{235}U foils were measured on two large Ge(Li) detectors. Four spectra taken on the ^{235}U foil (#4) were used to test the consistency of the data being obtained. These results are shown in Table ANC-XVII. For each Ge(Li) detector, the data from the second spectrum have been corrected for decay back to the time of the first measurement. Due to the low activity levels, the statistical errors in the peak areas are large (i.e., $\sim 1\%$) in spite of the long counting times (i.e., 11 to 22 hours). In Table ANC-XVII, the errors include the uncertainty in the peak area (i.e., the square root of the total count) and an error of 0.5% for the uncertainty of the source position (10.0 ± 0.025 cm from the detector). The agreement among all the sets of data is quite good.

The data in Table ANC-XVII, along with that for ^{140}La (^{235}U foil) and ^{198}Au (gold foil) were used to compare the results from the two detectors. When corrected to end of irradiation (16:00 MST on 12/6/71), the results are given in Table ANC-XVIII. The differences shown in the last column are all less than the associated errors and are $\sim 1.0\%$.

From these data, the disintegration rates per atom have been computed at the time of the reactor shutdown. (No corrections have been made to the efficiencies for the finite size of these sources.) For the gold foil (of 120.52 mg), the result is

$$2.338 \times 10^{-16} \text{ dis/sec-atom}$$

For the uranium foil (with 48.33 mg of ^{235}U), the result is

$$1.120 \times 10^{-17} \text{ dis/sec-atom.}$$

Table ANC-XVII

Reproducibility of Gamma-Ray Emission Rates

Detector ^a	Time Between Counts (days)	Activity	Gamma Emission Rate (γ /sec)	Difference (%)
OR-5	13.95	^{103}Ru	184.8 ($\pm 1.2\%$)	1.9 ± 1.6
			190.4 ($\pm 1.0\%$)	
		^{95}Zr	182.1 ($\pm 1.7\%$)	0.6 ± 2.1
			186.1 ($\pm 1.3\%$)	
ND-3	7.89	^{103}Ru	182.1 ($\pm 1.0\%$)	2.2 ± 1.3
			186.1 ($\pm 0.9\%$)	
		^{95}Zr	144.6 (1.3%)	1.3 ± 1.8
			142.7 (1.2%)	

^aOR-5 is a 35-cm³ open-end coaxial detector and ND-3 is a 60-cm³ closed-end coaxial.

Table ANC-XVIII

Comparison of Gamma Emission Rates from Two Detectors

Data corrected for decay to 1600 on 12/6/71. The errors include contributions from peak area ($\sqrt{n}$), source position (0.5%) and efficiency curve interpolation (0.5%).

Isotope	Gamma Energy (keV)	Detector	Gamma Emission Rate (γ /sec)	Difference (%)
^{103}Ru	497	OR-5	214.3 ($\pm 1.0\%$)	1.2 ± 1.3
		ND-3	216.8 ($\pm 0.9\%$)	
^{95}Zr	756	OR-5	157.0 ($\pm 1.2\%$)	1.0 ± 1.6
		ND-3	158.5 ($\pm 1.0\%$)	
^{140}La	1596	OR-5	1521 ^a ($\pm 1.2\%$) ^a	0.6 ± 1.6
		ND-3	1530 ^a ($\pm 1.0\%$)	
^{198}Au	411	OR-5	81,790 ($\pm 0.75\%$)	1.1 ± 1.1
		ND-3	82,660 ($\pm 0.76\%$)	

^aThis value is computed from the ^{140}Ba half-life and the 1596-keV gamma rate at more than 16 days after the irradiation. It is not corrected for the difference in ^{140}Ba and ^{140}La decay rates.

H. GAMMA-RAY COUNTING OF MONITOR FOILS

R. G. Helmer

Objective

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

Summary

The gamma-ray spectra of the gold and indium monitor foils from the ARCHO-I, ANL-I and HEDL-I irradiations in the CFRMF were measured with a large Ge(Li) detector. The relative gold and indium gamma-ray activities from these three irradiations have been determined with uncertainties of less than 1%. Subsidiary experiments were carried out to test the reproducibility of the counting system.

Accomplishments and Status

The gamma-ray spectra from the gold and indium monitor foils were measured on a large Ge(Li) detector. As discussed in section ANC-F, the efficiency of this detector, ND3, has been accurately measured. Also, the counting losses in the peak areas from random summing and other effects have been measured as a function of the counting rate.

All of the samples were measured at a source-detector distance of 10 cm. The foils were mounted on source cards in a manner to minimize the variability in position and to allow the foils to be demounted for subsequent weighing. The foils were placed between two slightly cone-shaped pieces of 0.010" mylar and held in the center of a source card. The cards were mounted so that they were 9.05 cm from the detector housing. It is estimated that this position was reproducible to ± 0.01 cm (1 σ level), which results in an uncertainty in the efficiency of $\pm 0.2\%$.

For the indium foils, the 335-keV gamma ray of the 4.5-h ^{115m}In activity was counted. Each foil was counted three times. The first two counts were made at about 9 and 25 hours after the irradiation. The third

count was made to assure the purity of the 335-keV peak. For the three foils, the differences between the counting rates computed from the first and second measurements (corrected for decay) were 0.21%, 0.66% and 0.43%. These differences are all smaller than the statistical counting errors of the second measurements (which are $\approx 0.9\%$) and are much smaller than the uncertainty in the decay corrections. Due to the uncertainty in the half-life, it is felt that the most accurate flux measure would result from using only the first count. These results are given in Table ANC-XIX and indicate a spread of about $3\frac{1}{2}\%$ in the activity levels (per unit mass).

For the gold foils, the 411-keV transition from the decay of ^{198}Au was counted. Each foil was measured three times over a period of one week, with the first spectrum taken about 24 hours after the irradiation. In all cases about 10^6 counts were obtained in the 411-keV peak. The results of these measurements are given in Table ANC-XX. As noted there, four contributions to the errors are taken in account; (1) that from the peak area, taken as the square root of the number of counts; (2) that from the correction for count-rate dependent losses (e.g., random summing); (3) that from the decay correction (where the uncertainty in the half-life is taken to be 0.06%); and (4) that from the source position, 0.2%. In this case, the later measurements are the most accurate due to the large error contribution from (2) in the early runs. (The analyzer dead times varied from 15% down to 3%.) As shown, the results agree with the indium values in that the activity from the ARHCO-1 irradiation is about 3% higher than the others. It is also noteworthy that the various measurements for each foil are consistent.

Since these were among the first measurements made for the ILRR program on this counting system, another set of spectra was also taken as a check on the consistency to be expected. A weak ^{60}Co (2% dead time) source and a strong ^{137}Cs (15% dead time) source were each counted six times over a period of 7 and 14 days. For the ^{60}Co sample, a total of 59,000 counts were collected in the two full-energy peaks (1173 and 1332 keV). The variation in the combined peak areas is consistent with that expected from the counting

statistics. In fact, the "quality of fit" (i.e., the weighted sum of the squares of the residuals divided by the number of degrees of freedom) is 0.68 compared to the expected value of ≈ 1.0 . Therefore, any errors must be small compared to the 0.4% statistical error.

A more sensitive test resulted from the ^{137}Cs spectra since the full-energy peak contained $\approx 10^6$ counts. The measured peak areas are given in Table ANC-XXI. As shown, this set of values has a quality of fit of 2.6. This indicates that actual uncertainties are about 0.16% instead of the 0.10% expected from the counting statistics. Since the same source is used for each run, the position error should be small, but it still may be $\approx 0.1\%$. (This would be one-half of the value used in Table ANC-XX.) This source position error and the counting statistics would account for the observed variation.

Expected Achievements

Similar measurements will be made on the monitor foils from future irradiations. Also the ^{60}Co and ^{137}Cs sources will be counted. As more data are collected a more complete evaluation of the uncertainties may be possible.

Table ANC-XIX

Relative Gamma-Ray Counting Rates from Indium Monitor Foils

Experiment	Peak Count ^a Rate (cts/sec)	mass ^b (mg)	Relative Count Rate (cts/sec-mg)
ARHCO-1	314.5 $\pm$ 1.1	49.76	1.035 $\pm$ 0.004
ANL-1	296.0 $\pm$ 0.7	48.48	≈ 1.000
HEDL-1	298.1 $\pm$ 0.7	48.40	1.009 $\pm$ 0.004

^aCorrected for decay to 8 hours after irradiation. Uncertainty includes counting statistics, 0.2% for source position and contribution from 0.3% error in half-life. At 335 keV, the detector efficiency is 2.16×10^{-3} ($\pm 1.0\%$).

^bANC values determined after irradiation.

Table ANC-XX

Relative Gamma-Ray Counting Rates from Gold Monitor Foils
 Samples were all Counted at 10 cm on ND3 with Live Times of 550 to 2000 seconds.

Experiment	Decay Time (days)	Peak Count ^a Rate (cts/sec)	Error Contributions (%) ^b				Average Count ^c Rate (ct/sec)	Mass (mg)	Relative Count Rate (cts/sec-mg)
			Peak Area	Source Position	Decay Correction	Summing Correction			
ARHCO-1	1	1740.5 ± 17.4	0.10	0.2	0.00	1.0	1726.9 ± 3.4	123.4	1.026 ± 0.003
	7	1725.5 ± 4.8	0.13	0.2	0.10	0.1			
	8	1723.8 ± 4.8	0.13	0.2	0.11	0.1			
ANL-1	1	1652.9 ± 16.5	0.08	0.2	0.00	1.0	1657.8 ± 4.1	121.6	≅1.000
	2	1668.2 ± 12.2	0.07	0.2	0.02	0.7			
	7	1654.3 ± 4.5	0.12	0.2	0.09	0.1			
HEDL-1	1	1651.6 ± 16.5	0.08	0.2	0.00	1.0	1643.3 ± 4.8	122.0	0.988 ± 0.004
	2	1654.1 ± 12.1	0.08	0.2	0.02	0.7			
	6	1635.1 ± 5.1	0.11	0.2	0.08	0.2			

^aCorrected to 25 h after the irradiations.

^bSee text for a discussion of these errors.

^cAt 411 keV, the detector efficiency is 1.71×10^{-3} ($\pm 1.0\%$)

Table ANC-XXI

Reproducibility of Peak Area from ¹³⁷Cs Source. Spectrum Counted for 1000 Seconds Live Time with Source at 10 cm and 15% Dead Time.

Date	Peak Area (counts)	Area-Average
		$\frac{\text{Area-Average}}{\sqrt{\text{Area}}}$
2/15	1,026,078	-0.22
2/16	1,025,507	-0.78
2/16	1,027,650	1.33
2/17	1,024,492	-1.78
2/18	1,028,869	2.53
2/22	1,025,215	-1.07

Average 1,026,302

$$\frac{1}{5} \sum \left[\frac{\text{Area-Average}}{\sqrt{\text{Area}}} \right]^2 = 2.63$$

I. CFRMF REACTION RATES FROM RADIOCHEMISTRY FOILS

J. W. Rogers and Y. D. Harker

Objective

- To present the results of radiochemistry foil measurements at ANC. These results are to provide a monitor of the neutron fluence and spectral index of the ILRR-CFRMF program tests.

Summary

Radiochemistry foils from the various ILRR program tests in the CFRMF have been counted using NaI(Tl) and Ge(Li) gamma-ray counting techniques. The methods of counting are discussed and the saturated reaction rates for the reactions of interest are presented.

Accomplishments and Status

Reaction rates determined from gamma-ray spectra measurements on several foils activated in the ILRR-CFRMF tests have been obtained using standard NaI(Tl) techniques. A 3" x 3" NaI(Tl) crystal detector was used at a source-detector distance of 10 cm. All sources were the 0.5" diameter 2-mil thick gold and indium monitor foils and were mounted as described in section ANC-H. A 1.024 g/cm² polystyrene absorber was used to stop the source beta radiation. All sources were carefully weighed either just before or just after counting. To eliminate the need to make high counting rate corrections, all counts were done at decay times long enough to yield acceptably low rates.

The 411.8-keV gamma ray from the decay of ¹⁹⁸Au was counted from the gold foils. In order to have acceptable counting rates at 10 cm, decay times of 16 to 20 days were allowed for the higher exposure foils. In all measurements more than 10⁵ counts were collected in the peak.

The 335-keV gamma ray from the decay of ^{115m}In was counted from the indium foils. Decay times of between 20 and 28 hours were allowed so that good counting rates and a single gamma-peak spectrum were obtained. In all measurements more than 10⁴ counts were collected in the peak.

Some of the foils that were counted by NaI(Tl) were also counted by Ge(Li) as discussed in sections ANC-G and H. For comparison purposes the Ge(Li) and NaI(Tl) results were converted to saturated reaction rates; i.e., $\Phi\sigma$ (reactions per unit time per atom). The decay-scheme data found

in section ANC-D were used in this analysis. Table ANC-XXII summarizes the results. The values given in this table are considered to be absolute reaction rates. The efficiencies of the Ge(Li) system were determined as described in section ANC-F. The efficiency of the NaI(Tl) system is that computed from the known characteristics of the detector. Therefore, the differences between the results are not surprising since no intercalibration between the systems has been made.

Expected Achievements

Further analysis of these results will be made. Foils from all other tests will be measured and analyzed.

Table ANC-XXII
CFRMF Reaction Rates from Radiochemistry Foils

Test Identity	Foil	Mass ^a (g)	Saturated Reaction ^b Rate by NaI(Tl)	Number of Measurements	Saturated Reaction Rate by Ge(Li)	Number of Measurements
Preliminary-I (12-6-71)	Au-B 235U-4	0.1205 0.04833	---- ----	---- ----	3.238 x 10 ⁻¹⁵ 1.237 x 10 ⁻¹⁴	2(.53%) 3(.57%)
Preliminary-III (1-26-72)	Au-MD In-14	0.1229 0.05034	3.173 x 10 ⁻¹⁵ 3.785 x 10 ⁻¹⁶	2(.64%) ^c 2(.51%)	---- ----	---- ----
Fission Chamber NBS (2-2-72)	Au-AA In-2#1	0.1216 0.04878	3.193 x 10 ⁻¹⁵ 3.788 x 10 ⁻¹⁶	3(.33%) 1	---- ----	---- ----
Fission Tracks ANL (2-4-72)	Au-AC In-2#3	0.1214 0.04802	3.186 x 10 ⁻¹⁵ 3.753 x 10 ⁻¹⁶	2(.25%) 2(.14%)	---- ----	---- ----
Fission Chamber NBS (2-4-72)	Au-AE In-2#5	0.1239 0.04909	3.193 x 10 ⁻¹⁵ 3.775 x 10 ⁻¹⁶	2(.74%) 1	---- ----	---- ----
ARHCO-I (2-8-72)	Au-AG In-2#7	0.1234 0.04976	4.902 x 10 ⁻¹⁴ 5.984 x 10 ⁻¹⁵	3(.26%) 3(1.04%)	5.076 x 10 ⁻¹⁴ 6.173 x 10 ⁻¹⁵	3(.57%) 1
ANL-I (2-15-72)	Au-AI In-2#9	0.1216 0.04848	4.708 x 10 ⁻¹⁴ 5.795 x 10 ⁻¹⁵	5(.97%) 3(.27%)	4.945 x 10 ⁻¹⁴ 5.964 x 10 ⁻¹⁵	3(.54%) 1
HEDL-I (2-16-72)	Au-AK In-2#11	0.1220 0.04840	4.665 x 10 ⁻¹⁴ 5.838 x 10 ⁻¹⁵	6(.53%) 3(.11%)	4.886 x 10 ⁻¹⁴ 6.016 x 10 ⁻¹⁵	3(.68%) 1

a) determined by weighing at ANC.

b) events per atom per second.

c) estimated percent standard deviation on span of measurements (does not include uncertainties in efficiencies and decay corrections).

ARGONNE NATIONAL LABORATORY

A. CFRMF REACTION RATES BY TRACK-RECORDER METHOD

R. J. Popek and N. D. Dudey

OBJECTIVE

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

SUMMARY

Methods for preparation of ^{235}U , ^{237}Np , and ^{239}Pu stock solutions are described. SSTR's for these materials were prepared and irradiated in the CFRMF facility. Stock solution and SSTR preparation of ^{238}U are nearing completion.

ACCOMPLISHMENTS AND STATUS

The principal limitation to the accuracy achievable with solid state track recorders is the accuracy with which the number of atoms exposed to the track recorder is known. These amounts are usually in the nanogram range. This limitation also exists for fission counters; however, SSTR's have one additional requirement, namely, the atoms must be uniformly distributed over the surface of the track recorder. Our approach is to prepare stock solutions of ^{235}U , ^{238}U , ^{239}Pu , and ^{237}Np whose specific alpha activities are absolutely known, electrodeposit the material on 5-mil platinum discs, determine their masses by alpha counting, place the fissile material in intimate contact with the track recorder (mica in this case) and irradiate the package. As fissions occur in the material the fission products recoil into the mica, producing radiation-damage tracks. These tracks are etched in 49% HF for 2-3 hr and become visible under an optical microscope. The number of fission events is then determined by counting all of the individual tracks.

1. Preparation of Stock Solutions

The procedure for preparing the stock solution of ^{235}U is as follows. All new glassware was used for each solution. Since all chemical reagents potentially are contaminated with trace amounts of uranium, they must be purified. Hydrochloric acid was purified by passing through an anion-exchange resin, and nitric acid was prepared by redistillation; extreme

precautions were taken to avoid cross contamination. Approximately 1 g of uranium metal was etched in acid to remove oxide, dried, and quickly weighed. Such a large amount was required to assure that any surface oxidation after cleaning would contribute a negligible error in the weight of metal. The uranium was dissolved in nitric acid and diluted to a convenient volume in a tared weighing bottle.

The atom content of material on the platinum plates must ultimately be verified by alpha counting each plate since we could not assure the electrodeposition was quantitative within 0.5% accuracy. It was therefore necessary to spike the ^{235}U and ^{238}U with a higher specific alpha activity than the material itself. ^{234}U was chosen as the spike material. The isotopic composition of the ^{234}U , as well as those of the primary fissile materials, are given in Table ANL-1.

TABLE ANL-1
ISOTOPIC COMPOSITION OF SSTR STOCK SOLUTIONS

^{239}Pu Solution		^{235}U Solution		^{234}U Solution		^{237}Np Solution	
^{239}Pu	99.99%	^{235}U	93.12%	^{234}U	99.885%	^{237}Np	99.99%
^{240}Pu	8 ppm	^{234}U	0.96%	^{235}U	0.064%		
		^{236}U	0.32%	^{236}U	0.035%		
		^{238}U	5.59%	^{238}U	0.014%		

A stock solution of ^{234}U was prepared by dissolving approximately 1 milligram in nitric acid. Unfortunately the ^{234}U contained trace amounts of ^{232}U together with the alpha decay products of ^{232}U . (Although the ^{232}U itself does not interfere, the decay products would change the specific activity significantly.) The decay products, except lead, were removed by passing the uranium solution through an anion-exchange column in 6N HCl. Thorium and other decay products pass through but lead and uranium stick to the column. The uranium was eluted with 0.1N HCl together with ^{212}Pb

which has a half-life of 10.6 hr. The ^{212}Pb is not an alpha emitter but has alpha-emitting daughter products. Therefore, the ^{212}Pb was allowed to decay for a few days and the uranium plus any ^{228}Th grow-in was again eliminated by ion exchange. Any new ^{212}Pb grow-in at this time is insignificant. This process removes all daughter products of ^{232}U . Although these then continue to build up, their grow-in rate is known. The initial ^{234}U contained about 10% alpha activity due to ^{232}U ; by making the separations the alpha-activity buildup due to daughter products is less than 1% of the alpha activity during the first 25 days after the separation.

After the spike solution and the stock solutions have been prepared, the spike solution is added to the ^{235}U to obtain the desired specific alpha activity level. All transfers are made by means of weight burettes, a procedure that is considerably more accurate than volumetric transfers. The new specific activity was determined by preparing four alpha plates by means of weight-burette transfers and alpha counting the plates on a 2M gas-flow proportional counter. This calibration provided a value for the number of alpha counts per gram of ^{235}U . Thus alpha counting of any subsequent electrodeposited plates provided a direct measure of the number of atoms of ^{235}U on the plate. Electrodeposition of the solutions onto platinum plates involved a modification of the method of Handley and Cooper.¹ The details of the modified procedure are described in Ref. 2. The four calibration plates agreed to better than 0.5%; the accuracy of determining the number of atoms on the electrodeposited plates is better than $\pm 1\%$.

Stock solutions of ^{239}Pu and ^{237}Np were prepared by dissolution in nitric acid. Aliquots of these solutions were electroplated and the mass of fissile material on the plates determined by direct alpha counting. The accuracy of the plutonium and neptunium determinations is limited by counting statistics, alpha contaminants in the solutions, and the accuracy of their half-lives for alpha decay. The estimated accuracy for determining the amount of material on the plates is $\pm 0.8\%$.

2. CRFMF Irradiations

Track recorder measurements in CRFMF for ^{235}U , ^{237}Np , and ^{239}Pu were made on February 3 and 4, 1972. Table ANL-2 summarizes the conditions of

the SSTR irradiations. The purpose of this first test was primarily to compare the SSTR fission rates to those measured by Grundl by means of fission chambers. In addition, we want to examine several variables associated with the measurement. SSTR numbers 4, 5, and 6 were designed to test the precision of the measurements. Numbers 8 and 9 were designed to test uncertainties associated with the time it takes to insert the recorders into the reactor core and to remove the recorders from the core. Test number 7 was designed to provide fission-rate ratios which could be directly compared with the ratios obtained with fission chambers.

TABLE ANL-2. First SSTR Irradiation in CFRMF

Date	SSTR Number	Sample Material	Weight (ng)	Time (min)	Power (watts)
2/3/72	4	^{235}U	99.7	60.0	600
		^{235}U	98.2	"	"
2/3/72	5	^{239}Pu	75.2	73.0	"
		^{239}Pu	87.0	73.0	"
2/3/72	6	^{237}Np	353	59.0	"
		^{237}Np	322	59.0	"
2/3/72	8	^{237}Np	3388	6.0	"
2/3/72	9	^{237}Np	3509	6.1	"
2/4/72	7	^{235}U	85.5	60.0	"
		^{239}Pu	78.4	60.0	"
		^{237}Np	323.0	60.0	"

Counting of the track recorders has only begun; however, we were able to obtain a very preliminary value for the ^{235}U sample in SSTR number 7. Based upon only one count of the tracks, the fission rate was measured to be 1.34×10^{-14} f/atom-sec.

Expected Achievements

The SSTR's exposed in the first CFRMF irradiation will be counted and the fission rates reported. Stock solutions and track recorders for ^{238}U will be prepared. It is anticipated that a second series of track recorders will be irradiated in CFRMF.

We have obtained an additional quantity of ^{234}U which is being isotopically separated on a mass separator. The mass separation will remove virtually all ^{232}U impurity so that a stock solution of ^{234}U can be prepared which will not require such elaborate radiochemical processing to remove alpha-active contaminants. The new solution should be available in March.

REFERENCES

1. T. H. Handley and J. H. Cooper, Anal. Chem. 41, 381 (1969).
2. N. D. Dudey, et al., "Activation-Rate Measurements in the ZPR-3 Mockup Critical Experiments", ANL-7781 (1971).

B. CFRMF REACTION RATES BY FOIL ACTIVATION

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

OBJECTIVE

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

SUMMARY

The measured fission rate of an exploratory ^{235}U foil irradiated for 7 hr in CFRMF at a nominal power of 600 W on December 6, 1971 was 1.313×10^{-14} fissions/atom-sec, based upon ^{140}Ba activity. A complete nonfissile foil set irradiated in CFRMF is now being counted.

ACCOMPLISHMENTS AND STATUS

A preliminary irradiation test in CFRMF was conducted on December 6, 1971. From this irradiation we received a single ^{235}U foil (designated ID-3) from which the fission rate was to be determined; this value has been compared with results from ANC, ARHCO, and HEDL. The irradiation was for 7.0 hr at a CFRMF Power setpoint of 983.0×1000 .

The sample was counted on our calibrated 1600 Ge(Li) detector system at three different geometries (3-10 cm) for a total of 6 times over a period of 26 days. The results, determined from ^{140}Ba , ^{95}Zr , and ^{103}Ru , are tabulated in Table ANL-3. The purpose of comparing fission rates determined by means of various fission products is to obtain a feeling for

the precision of the fission yields. The results indicate better than 2% agreement between ^{95}Zr and ^{140}Ba , although the ^{103}Ru result differs by about 9%. This indicates that the applicable fission yield for ^{103}Ru may be 3.0% rather than the 3.3% which was measured in previous work in spectra similar to EBR-II.¹

TABLE ANL-3. ^{235}U : CFRMF Test (12/6/71)

Fission Product	Fission Yield (%)	Fission Rate ($\times 10^{14}$)
^{140}Ba	5.8	1.313
^{95}Zr	6.4	1.288
^{103}Ru	3.3	1.207

The fission rates based upon ^{140}Ba are of primary interest in this interlaboratory comparison. Based upon the 6 counts, the standard deviation of a single measurement was 0.7%, which is comparable to the counting statistics of each measurement.

A complete nonfissile foil set of fast-neutron dosimeters was irradiated for a nominal 70 kWh in CFRMF on February 15, 1972. These samples are now being counted on both Ge(Li) and NaI(Tl) detectors.

EXPECTED ACHIEVEMENTS

Counting, data processing, and reporting of results from the nonfissile foil set irradiated on February 15, 1972, will be completed. Irradiation and counting of the primary fissile foils are expected in the next quarter.

REFERENCES

1. R. P. Larsen, private communication, 1971.

ATLANTIC RICHFIELD HANFORD COMPANY

A. ARHCO-I CFRMF REACTION RATES

W. H. Zimmer

Objective

- To perform non-destructive measurement of the reaction rates of fission and non-fission foils irradiated in CFRMF.

Summary

Measurement constants, activity analyses and reaction rates are reported for the ARHCO-I radiochemistry foil package and power normalization foils. Detailed information on the foil package design, contents, and placement in CFRMF for irradiation at a stated reactor power of 9.75 KW is given in the HEDL section.

ARHCO low power fission foil reaction rates are reported in combination with the HEDL measurement in the HEDL section.

Accomplishments and Status

Except for reaction rates, all results are reported in units of disintegrations per second per milligram of sample, see Tables 1 and 2. The units for reaction rate (saturated activity) are disintegrations per second per atom. Time zero is Hanford time 9997.2842 PST or 2/8/72 at 07:49:14 MST. The precision reported is in units of percent of the activity at the 95% confidence level. Precision for the average of two isotopic peaks of comparable individual precision may be taken as the square root of the average of the sum of the squares of the individual precisions divided by the square root of two. Likewise, for three peaks, the precision of the average is the square root of the average of the sum of the squares of the individual precisions divided by the square root of three. The accuracy for an individual isotopic activity is the square root of the sum of the squares of the precision of the average activity of an isotope and of the precision of calibration (% P) for that isotope. The latter value is included in Table 1.

Atomic weights for calculation of d/s/atom reaction rates are based on the International Atomic Weights referenced to C-12. Avogadro's number is 6.022169E23. All "E" values represent an exponent base of ten.

TABLE ARHCO-1
REACTION PRODUCT NUCLEAR DATA SUMMARY

Isotope	Energy in keV	Gamma per Disintegration	Half Life in Days	% P
^{113}mIn	391.71	0.64	0.07210	2
^{115}mIn	336.25	0.450	0.1867	2
^{116}mIn	1097.2	0.53	0.0375	2
	1293.3	0.80		2
^{24}Na	1368.53	1.00	0.6250	2
^{46}Sc	889.258	1.00	83.80	2
	1120.516	0.996		2
^{47}Sc	159.39	0.6875	3.422	4
^{48}Sc	983.5	1.00	1.83	2
	1037.6	0.978		2
	1311.8	1.00		2
^{64}Cu	1345.76	0.0048	0.5358	3
^{196}Au	355.72	0.8866	6.170	2
^{198}Au	411.794	0.9549	2.696	2
^{54}Mn	834.78	1.00	312.99	1
^{56}Mn	846.8	0.988	0.1072	2
	1810.9	0.289		2
^{59}Fe	1099.224	0.565	45.1	2
	1291.564	0.432		2
^{58}Co	810.757	0.9950	71.3	2
^{60}Co	1173.208	0.9986	1920.8	2
	1332.56	0.9999		2
^{140}Ba	1595.4	1.1068	12.80	1

No self absorption corrections have been made. The results for foil Au-Al #3 C998528 are low due to geometry error. The initial analysis was made with the foil mashed together rather than coiled.

The isotopic abundance of ^{235}U is taken as 0.93098349 and the ^{140}Ba fission yield as 0.058. The ^{140}Ba fission yield for ^{238}U is taken as 0.060. The equilibrium isotopic ratio of ^{140}La to ^{140}Ba was taken using a ^{140}La half-life of 1.676 days.

In Table 2, the assigned ARHCO foil serial number (such as A997749 for In-2) is in the same units of decimal days as that given in the text for the Hanford time zero. These numbers can be used (as can the D/S/Milligram values @ T of count and @ T zero) to calculate ΔT 's for estimating uncertainties associated with isotope half lives.

Expected Achievements

ARHCO measurements of a limited number of other fission and non-fission foils from HEDL "Non-Fission" foil sets II, III, and IV are not anticipated to start before May 1972.

TABLE ARHCO-2

SUMMARY OF ARHCO MEASURED FOIL D/S/MILLIGRAM VALUES AND
REACTION RATES FOR THE ARHCO-I NON-FISSION FOIL SET

ISOTOPE	PEAK	PLUS OR MINUS	D/S/MILLIGRAM	D/S/MILLIGRAM	REACTION
CENTER	% @ 95% CL	@ T OF COUNT	@ T ZERO	RATE	
IN-2 (a)					
#7	46.642 MILLIGRAMS				
A997749*					
IN115M	670	+0.4737634E+00	+0.3995331E+04	+0.2245464E+05	6.763E-15
IN113M	783	+0.9225832E+01	+0.1130858E+02	+0.9882425E+03	4.686E-15
IN116M	2195	+0.1066173E+02	+0.3432986E+02	+0.1855427E+06	3.733E-14
IN116M	2588	+0.3713258E+01	+0.3471311E+02	+0.1876140E+06	
IN-2 #8					
#8	46.578 MILLIGRAMS				
C997757					
IN115M	672	+0.4183275E+00	+0.3854333E+04	+0.2231523E+05	6.721E-15
IN113M	784	+0.8739576E+01	+0.9802868E+01	+0.9251440E+03	4.387E-15
IN116M	2199	+0.8586046E+01	+0.3529483E+02	+0.2211579E+06	4.175E-14
IN116M	2593	+0.7893669E+01	+0.3129571E+02	+0.1960994E+06	
AL-5 #7					
#7	47.477 MILLIGRAMS				
A997791					
NA24	2738	+0.2927280E+01	+0.7835466E+02	+0.1374866E+03	2.229E-17
AL-5 #8					
#8	47.580 MILLIGRAMS				
C997800					
NA24	2743	+0.2440332E+01	+0.7439455E+02	+0.1318473E+03	2.137E-17
IN-2 #9					
#9	49.900 MILLIGRAMS				
A997811					
IN115M	670	+0.4189850E+00	+0.3175395E+04	+0.2246560E+05	6.766E-15
IN113M	783	+0.1023563E+02	+0.6029547E+01	+0.9563134E+03	4.535E-15
IN116M	2196	+0.1445562E+02	+0.1139068E+02	+0.1936527E+06	3.793E-14
IN116M	2588	+0.1235733E+02	+0.1090641E+02	+0.1854196E+06	
AL-5 #9					
#9	46.564 MILLIGRAMS				
C997820					
NA24	2743	+0.2503404E+01	+0.7238937E+02	+0.1311711E+03	2.127E-17
TI-S					
159.038	MILLIGRAMS				
C997847					
SC47	316	+0.7591564E+00	+0.2579538E+02	+0.2891136E+02	5.476E-16
SC46	1783	+0.1233559E+02	+0.1022554E+01	+0.1027327E+01	4.080E-16
SC46	2247	+0.1283216E+02	+0.9634969E+00	+0.9679939E+00	
SC48	1972	+0.3694145E+01	+0.7791935E+01	+0.9644001E+01	9.660E-18
SC48	2079	+0.3830855E+01	+0.7568323E+01	+0.9367239E+01	
SC48	2630	+0.4144304E+01	+0.7421751E+01	+0.9185827E+01	

* Time = 9997.749

(a) Except for the fission foils, a number after the element symbol indicates the foil thickness in mils. A letter identifies the foil or in the case of Au-Al indicates an alloy.

TABLE ARHCO-2 (Cont'd)

ISOTOPE	PEAK CENTER	PLUS OR MINUS % @ 95% CL	D/S/MILLIGRAM @ T OF COUNT	D/S/MILLIGRAM @ T ZERO	D/S/ATOM @ T ZERO
TI-R 157.073 MILLIGRAMS					
C997874*					
SC47	316	+0.5395213E+00	+0.2450211E+02	+0.2761247E+02	5.230E-16
SC46	1783	+0.9758328E+01	+0.8497122E+00	+0.8538688E+00	3.624E-16
SC46	2247	+0.9247041E+01	+0.8962500E+00	+0.9006342E+00	
SC48	1972	+0.2656498E+01	+0.7301361E+01	+0.9129711E+01	8.815E-18
SC48	2080	+0.2814135E+01	+0.6859705E+01	+0.8577461E+01	
SC48	2630	+0.2988272E+01	+0.6857516E+01	+0.8574723E+01	
CU-0.5 #3 15.231 MILLIGRAMS					
A997831					
CU64	2692	+0.5907179E+01	+0.5969289E+04	+0.1211281E+05	5.886E-15
AU-0.5 #5 31.153 MILLIGRAMS					
C998380					
AU196	711	+0.1970193E+02	+0.1349329E+02	+0.1526128E+02	1.548E-16
AU198	825	+0.2136931E+00	+0.8962278E+04	+0.1187943E+05	5.378E-14
AU-0.5 #6 33.188 MILLIGRAMS					
C998420					
AU196	712	+0.1966896E+02	+0.1303234E+02	+0.1480632E+02	1.502E-16
AU198	824	+0.2073423E+00	+0.8936477E+04	+0.1196768E+05	5.418E-14
FE-C 808.987 MILLIGRAMS					
A998402					
? C/S	277	+0.3734772E+01	+0.2726317E-02	+0.5917356E-02	
AS76	1119	+0.2078989E+01	+0.7433232E+00	+0.1503603E+01	
MN54	1670	+0.1419585E+01	+0.9368705E+00	+0.9391928E+00	2.310E-15
MN56	1695	+0.3194790E+01	+0.2080399E+00	+0.2868268E+03	3.422E-17
MN56	3618	+0.9938757E+01	+0.2081871E+00	+0.2870298E+03	
FE59	2199	+0.7095729E+01	+0.1224910E+00	+0.1246139E+00	7.869E-16
FE59	2583	+0.8548736E+01	+0.1257976E+00	+0.1279778E+00	
CU64	2693	+0.1811509E+02	+0.3283134E+01	+0.1394496E+02	
NA24	2737	+0.2143237E+02	+0.1269551E-01	+0.4386647E-01	
AU-0.05 #3 3.595 MILLIGRAMS					
C998484					
AU198	825	+0.3611796E+00	+0.9052311E+04	+0.1232392E+05	5.579E-14
AU196	712	+0.3059554E+02	+0.1454574E+02	+0.1664496E+02	1.689E-16
SC-5 #3 50.261 MILLIGRAMS					
A998504					
SC44M	540	+0.3309955E+02	+0.5138982E+00	+0.7267618E+00	
SC46	1779	+0.1469766E+01	+0.9542731E+02	+0.9639513E+02	3.038E-15
SC46	2242	+0.1560820E+01	+0.9869464E+02	+0.9969561E+02	
SC44	2315	+0.2100110E+02	+0.7221539E+00	+0.1021279E+01	
K42	3049	+0.1730515E+02	+0.7317297E+01	+0.3779811E+02	

* Time = 9997.874

TABLE ARHCO-2 (Cont'd)

ISOTOPE	PEAK CENTER	PLUS OR MINUS % @ 95% CL	D/S/MILLIGRAM @ T OF COUNT	D/S/MILLIGRAM @ T ZERO	REACTION RATE
AU-AL	#3	102.496 MILLIGRAMS AL (0.1386 MILLIGRAMS AU)			
C998528*					
AU198	825	+0.1972506E+01	+0.8590234E+04	+0.1132790E+05	5.355E-14 ^(b)
NA24	2744	+0.2299789E+01	+0.3237985E+02	+0.1286604E+03	2.086E-17
AU-AL	#3	.1386 MILLIGRAMS AU			
A1007415**					
AU198	822	+0.2099086E+01	+0.9360564E+03	+0.1266220 +05	5.732E-14 ^(b)
NI-TA	297.171	MILLIGRAMS			
A998582*					
CO58	1622	+0.1040157E+01	+0.5999382E+02	+0.6075565E+02	3.082E-15
CO-0.5	#7	14.680 MILLIGRAMS			
A10002278					
CO60	2347	+0.1924154E+01	+0.10463E+02	+0.10482E+02	9.811E-15
CO60	2665	+0.1997680E+01	+0.10601E+02	+0.10621E+02	
CO-0.5	#8	14.505 MILLIGRAMS			
A10003294					
CO60	2347	+0.1998114E+01	+0.9721E+01	+0.9742E+01	8.918E-15
CO60	2666	+0.2123284E+01	+0.9419E+01	+0.9439E+01	
FE59	2199	+0.9074316E+01	+0.1985E+01	+0.2177E+01	4.829E-17
FE59	2584	+0.7773177E+01	+0.2039E+01	+0.2236E+01	
CO-0.5	#8	14.505 MILLIGRAMS			
A10005984					
CO60	2347	+0.2492172E+01	+0.988550E+01	+0.991658E+01	9.120E-15
CO60	2666	+0.2636939E+01	+0.966904E+01	+0.96994E+01	
CO-0.5	#9	14.795 MILLIGRAMS			
A10004000					
CO60	2348	+0.2072965E+01	+0.10429E+02	+0.10454E+02	9.835E-15
CO60	2666	+0.2147729E+01	+0.10674E+02	+0.10700E+02	
FE59	2200	+0.1036354E+02	+0.1947E+01	+0.2159E+01	4.717E-17
FE59	2584	+0.8447612E+01	+0.1941E+01	+0.2152E+01	

* Time = 9998.528

** Time = 10007.415

(b) As stated in the text the first value is low because the foil was counted mashed together, while the second value was counted as a coiled disk-the same as for the HEDL-I foil Sets, Au-Al Table HEDL-8.

TABLE ARHCO-2 (Cont'd)

ISOTOPE	PEAK CENTER	PLUS OR MINUS % @ 95% CL	D/S/MILLIGRAM @ T OF COUNT	D/S/MILLIGRAM @ T ZERO	REACTION RATE
U-235	#3	256.32 MILLIGRAMS†			
C1011438*					
BA140	3192	+0.1254148E+01	+0.1977136E+03	+0.4255100E+03	1.963E-13
U-238	#3	69.84 MILLIGRAMS			
C1011482					
BA140	3192	+0.8395718E+01	+0.1061710E+02	+0.2290414E+02	9.629E-15
U-235	#3	256.32 MILLIGRAMS			
C1017540					
BA140	3191	+0.8432501E+01	+0.1461492E+03	+0.4376997E+03	2.019E-13
U-238	#3	69.84 MILLIGRAMS			
C1017488					
BA140	3191	+0.1000797E+02	+0.7592202E+01	+0.2267384E+02	9.532E-15

* Time = 10011.438

† This original weight was found to be in error by 1.84%, the HEDL reweighing gave a corrected weight of 251.68 mg. The ^{235}U reaction rate should be increased, therefore, by a factor of 1.0184.

ATOMICS INTERNATIONAL

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

H. Farrar IV

Objective

• In order to help improve the absolute accuracy of reaction rates and of flux-fluence measurements, several "standard" cross sections are being examined at different laboratories by various techniques. The objective of Atomics International's preliminary contribution to the Interlaboratory IMFBR Reaction Rate (ILRR) Program is to 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 IMFBR's. The determination by mass spectrometry of the amount of helium generated in milligram-size specimens will provide an additional and independent absolute base for normalization of available energy dependent (n,α) cross section data.

Summary

Boron specimens have been prepared at Atomics International for irradiation in the CFRMF facility at Aerojet Nuclear. Ten milligram-size crystals of enriched ${}^{10}\text{B}$ and two crystals each of natural boron and enriched ${}^{11}\text{B}$ were placed within holes in a $\frac{1}{2}$ -inch diameter thin aluminum disk. This disk was placed between monitor foils of In, Au and Al in a foil set holder supplied by HEDL. Irradiation of this holder in the CFRMF core center will take place within a chamber closely resembling the NBS fission chamber. After the irradiation, the spectrum integrated (n,α) cross sections of the boron isotopes will be determined by mass spectrometric measurement of the concentration of helium generated in the specimens.

Accomplishments and Status

In the ILRR program, the two important (n,α) reactions that lend themselves to study by mass spectrometric measurement of the helium generated are the ${}^6\text{Li}(n,\alpha){}^3\text{H}$ and ${}^{10}\text{B}(n,\alpha){}^7\text{Li}$ reactions. Other elements with lower (n,α) cross

sections do not produce enough helium for precise measurement, when the irradiations are accomplished in the low-flux test reactors to be used in the LLRR program. The low melting point temperature of lithium makes this element unattractive to encapsulate in metallic form, yet there are definite drawbacks in using lithium compounds for the generation of helium. For the first experimental irradiation in CFRMF, it was therefore decided to include only crystals of boron, which could be prepared for irradiation without complication.

Twenty eight crystalline specimens of boron were prepared by dividing large crystals into a suitable size for eventual mass spectrometric analysis. The specimens were first examined under microscope for integrity, then weighed. Masses of the specimens ranged from 2.1 to 5.3 mg. Four of the specimens were enriched ^{11}B (98.55%) and four were natural boron; most of the samples, however were enriched ^{10}B (96.50%).

Two thin aluminum disks, shown in Figure AI-1, were made to contain the boron specimens. Each disk has fourteen holes whose identification numbers are determined from the figure relative to three orientation marks drilled in the perimeter. The bottom and top lids were spot-welded to the center disk before and after sample loading respectively, and an arrow was scratched on the top lid pointing in the direction of the "x" orientation mark. Each $\frac{1}{2}$ -inch diameter disk is approximately 0.12-inch thick, which allows it to fit into the non-fission foil set holder shown in HEDL's chapter of this report, Figure HEDL-3. In each disk, the ten ^{10}B specimens were placed around the perimeter, with the ^{11}B and natural boron specimens in the center. The locations of the specimens, their isotopic enrichments and their masses are given in Table AI-1.

The first CFRMF irradiation of boron specimens will last 16 hours at approximately 9 KW, and this is scheduled to take place on April 10, 1972. For this irradiation, disk B 1 has been loaded in HEDL's non-fission foil set holder along with other $\frac{1}{2}$ -inch diameter foils in the order shown in Table AI-2. Each foil was separated from the adjacent foils by 0.00065-inch thick foils of aluminum. In order to determine possible spatial flux heterogeneities, the AI disk containing the boron specimens was oriented so that the arrow on the top lid will point in the direction of the stem of the assembly. After the

Figure AI-1 Aluminum Disk for Holding Boron Specimens

TABLE AI-1

LOCATION DETAILS OF BORON SPECIMENS

Disk	Location Number	Specimen Number	Mass (mg)	Isotopic Comp. (%)	
				^{10}B	^{11}B
B 1	1	R10B4	5.330	96.50	3.50
	2	R10B6	3.871	96.50	3.50
	3	R10B15	4.531	96.50	3.50
	4	R10B8	2.991	96.50	3.50
	5	R10B2	4.447	96.50	3.50
	6	R10B14	2.258	96.50	3.50
	7	R10B5	3.596	96.50	3.50
	8	R10B17	2.669	96.50	3.50
	9	R10B9	3.442	96.50	3.50
	10	R10B19	2.193	96.50	3.50
	11	R11B4	5.100	1.45	98.55
	12	RNB4	5.043	19.78	80.22
	13	R11B3	4.735	1.45	98.55
	14	RNB1	2.981	19.78	80.22
B 2	1	R10B10	4.662	96.50	3.50
	2	R10B12	3.171	96.50	3.50
	3	R10B7	4.384	96.50	3.50
	4	R10B13	2.414	96.50	3.50
	5	R10B11	3.817	96.50	3.50
	6	R10B18	2.245	96.50	3.50
	7	R10B1	3.630	96.50	3.50
	8	R10B20	2.226	96.50	3.50
	9	R10B3	3.517	96.50	3.50
	10	R10B16	2.156	96.50	3.50
	11	R11B1	4.631	1.45	98.55
	12	RNB2	3.761	19.78	80.22
	13	R11B2	4.353	1.45	98.55
	14	RNB3	2.151	19.78	80.22

TABLE AI-2

DETAILS OF THE ATOMICS INTERNATIONAL FOIL SET

Foil and Relative Position	Foil Number	Thickness (inches)	Mass * (grams)	Reaction Products of Primary Interest
Al (top)	2	0.005	0.04625	^{24}Na
Au	2	0.0005	0.04095	^{198}Au
In	2	0.002	0.04921	$^{115\text{m}}\text{In}$
B set	B 1	0.123	0.7670	^4He from B isotopes
In	1	0.002	0.04879	$^{115\text{m}}\text{In}$
Au	1	0.0005	0.03804	^{198}Au
Al (bottom)	1	0.005	0.04654	^{24}Na

* The mass of the aluminum disk B 1 (with lids) is 0.7138g and the mass of the boron specimens totals 0.05319g. The mass of aluminum in disk B 2 is 0.6558g and it contains 0.04712g of boron.

loading of the disk and foils, the total outer thickness of the foil set holder with its contents including a piece of 0.015-inch Al-0.1% Ta wire to hold it together, was 1.333g. No plans have yet been made to irradiate disk B 2 which was assembled as a spare. The effects of self-shielding of thermal neutrons by the resonances of Au and In are expected to be insignificant, especially since the neutron flux in CFRMF in these low energy regions is expected to be small. The possibility of there being such an effect due to a larger-than-expected thermal flux has to be weighed against the importance of having these gold and indium foils at this location for the proper normalization of the reactor run.

Expected Achievements

After the irradiation, the foil set holder will be shipped to HEDL where the Al, In and Au foils will be removed for radiochemical determinations. The disk will be shipped to Atomics International where the boron specimens will be identified, reweighed and analyzed for helium content by high sensitivity gas mass spectrometer. Separate analyses of the ten ^{10}B specimens, which are expected to contain no more than a few parts per billion (atomic) helium, will

give not only an average value, but also will provide a measure of the uncertainties involved. These measured concentrations, when coupled with the fluence and neutron spectral data obtained using multiple-foil dosimetry, will give spectrum-integrated (n,α) cross sections. These will subsequently be compared with integral cross sections obtained by calculational techniques using energy dependent (n,α) cross section data, providing an independent and absolute normalization for the calculations.

BROOKHAVEN NATIONAL LABORATORY

NATIONAL NEUTRON CROSS SECTION CENTER

A. Evaluated Cross Section Libraries

B. Magurno

Objective

- Comparison of the SAND-II and ENDF/B-III evaluated energy dependent cross sections.

Summary

The SAND-II Library of evaluated energy dependent cross sections was received by the National Neutron Cross Section Center on June 21, 1971, for comparison with ENDF/B-III. The isotopes and reactions of primary importance were defined by McElroy⁽¹⁾ and these were plotted and compared where common to both libraries.

Accomplishments and Status

The isotopes and reactions common to both libraries are $\text{Li}^6(n,\alpha)\text{H}^3$, $\text{B}^{10}(n,\alpha)\text{Li}^7$, $\text{Al}^{27}(n,\alpha)\text{Na}^{24}$, $\text{Au}^{197}(n,\gamma)\text{Au}^{198}$, $\text{U}^{235}(n,f)\text{F.P.}$, $\text{U}^{238}(n,f)\text{F.P.}$, and $\text{Pu}^{239}(n,f)\text{F.P.}$ The curves and comments follow.

$\text{Li}^6(n,\alpha)\text{H}^3$

The cross sections are in good agreement at low energies (Fig. 1). The SAND-II curve starts to diverge from the ENDF/B-III curve (Fig. 2) at one keV and varies to as much as 20% higher than the ENDF/B-III curve near 100 keV. The peak cross section of the resonance around 240 keV also differs by 20% between the two curves. The requested cross section comparison, i.e., that of total helium production, could not be made since the SAND-II data contain the (n,α) cross section, but not the $(n,n'\alpha)$ or the $(n,2n\alpha)$ reactions.

$B^{10}(n,\alpha)Li^7$

As in Li^6 , the SAND-II and ENDF/B-III are in good agreement at low energies (Fig. 3), but the SAND-II curve starts to diverge from the ENDF/B curve (Fig. 4) and from $1/v$ at approximately 10 eV. The SAND-II cross section is as much as 10% higher than the ENDF/B-III cross section at 10 keV. The ENDF/B-III dips slightly below $1/v$ in the region of 10 - 100 keV as recommended by the Normalization and Standards Subcommittee of the Cross Section Evaluation Working Group. The SAND-II cross section curve crosses below the ENDF/B-III curve (Fig. 5) at 100 keV, but more or less follows the shape of the ENDF/B-III curve to about 3.5 MeV where it again increases to a cross section value larger than the ENDF/B-III value and diverges sharply from the ENDF/B-III curve (Fig. 6). The SAND-II cross section curve in the area of 5 MeV is too high to be only the n,α cross section. Adding MT 107 (n,α) and double MT 105 ($n,T2\alpha$) of ENDF/B-III to calculate the total He production gives a value twice that indicated by SAND-II. At 14 MeV, the SAND-II Library is in approximate agreement with ENDF/B-III for the total He production.

$Al^{27}(n,\alpha)Na^{24}$

The SAND-II and ENDF/B-III curves are very close on the low energy side of the curve (Fig. 7). They cross each other on both sides of the peak, finally diverging to a 12% difference at 17 MeV. The behavior of the SAND-II curve at 18 MeV is probably due to keypunch errors. $Q = -3.1316 \text{ MeV}^{(2)}$ and the threshold = 3.25 MeV. Neither curve has a satisfactory shape at threshold (Fig. 8). ENDF/B-III plots from 3.25 MeV and SAND-II starts at 4.6 MeV.

$Au^{197}(n,\gamma)Au^{198}$

The ENDF/B-III and SAND-II curves for $Au^{197}(n,\gamma)$ are in very good agreement to 10 eV (Fig. 9). The SAND-II curve then exhibits a behavior,

between 10 and 100 eV (Fig. 10), that is not consistent with the known capture cross section.⁽³⁾ From 100 eV to 1 keV, no comparison whatever can be made unless group averages are taken of the ENDF/B-III data. From 1 keV to 1 MeV the SAND-II curve (Fig. 9), although higher in cross section than ENDF/B-III, agrees (except for the two mysterious dips) with the general shape. At 1 MeV the SAND-II curve drops below that of the ENDF/B-III curve but swings upward suddenly to a value of 50 mb at 18 MeV.

$U^{235}(n,f)F.P.$

The SAND-II and ENDF/B-III cross section curves for this isotope are in fair agreement over the entire energy range (Fig. 11). Noticeable but minor differences occur between 1 and 3 keV. The resolved energy range could not be meaningfully compared, but there seems to be general agreement between the two curves. A segment (10-25 eV) of the comparison is blown up and presented (Fig. 12) as a demonstration.

$U^{238}(n,f)F.P.$

The SAND-II and ENDF/B-III curves are in good agreement except between 2 and 3 MeV⁽³⁾ where there is an 8% difference in cross section, and between 8 and 9 MeV where there is a 4% difference (Fig. 13). The threshold (Fig. 14) of the SAND-II curve is at 400 keV while the ENDF/B-III curve starts at 50 keV, taking some sub-threshold fission into account.

$Pu^{239}(n,f)F.P.$

The SAND-II cross section curve is higher than, but parallel to, the ENDF/B-III curve up to 10^{-2} eV (Fig. 15, 16), where it drops below the ENDF/B-III curve and shows some slight cross section variations up to 0.1 eV. There is good agreement over the first resonance, but the behavior of the SAND-II curve is not supported by the known structure in the energy range 0.6 - 7 eV. As in

U^{235} the resolved energy region could not be meaningfully compared, but a blow-up of the area between 10 - 100 eV (Fig. 17) shows a general agreement between the two curves. A final comparison between 300 eV and 20 MeV (Fig. 18) shows the SAND-II curve varying around, but generally lower than, the ENDF/B-III curve.

Expected Achievements

Group averages of ENDF/B-III for direct comparison to the SAND-II Library, and a linear plot of the ratio of SAND-II to ENDF/B-III for each of the reactions common to both libraries. Where possible, resonance integrals for $1/e$ and fission spectrums, and Maxwellian and fission spectrum average cross sections will be calculated and compared.

References for BNL Chapter

1. W. N. McElroy, private communication, HEDL (1971).
2. A. H. Wapstra and N. B. Gove, "The 1971 Atomic Mass Evaluation," Nuclear Data Tables 9 (1972).
3. Note added in proof: W. N. McElroy, private communication, HEDL, February 25, 1971. The SAND-II version of this cross section has recently been changed. (To date no new data has been received at NNCSC.)

Figure BNL-1 Lithium-6 (n, α) Neutron Cross Section

Figure BNL-2 Lithium-6 (n, α) Neutron Cross Section

Figure BNL-3 Boron-10 (n,α) Neutron Cross Section

Figure BNL-4 Boron-10 (n,α) Neutron Cross Section

Figure BNL-5 Boron-10 (n,α) Neutron Cross Section

Figure BNL-6 Boron-10 (n,α) Neutron Cross Section

Figure BNL-7 Aluminum-27 (n,α) Neutron Cross Section

Figure BNL-8 Aluminum-27 (n,α) Neutron Cross Section

Figure BNL-9 Gold-197 (n,γ) Neutron Cross Section

Figure BNL-10 Gold-197 (n,γ) Neutron Cross Section

Figure BNL-11 Uranium-235 Fission Neutron Cross Section

Figure BNL-12 Uranium-235 Fission Neutron Cross Section

Figure BNL-13 Uranium-238 Fission Neutron Cross Section

Figure BNL-14 Uranium-238 Fission Neutron Cross Section

Figure BNL-15 Plutonium-239 Fission Neutron Cross Section

Figure BNL-16 Plutonium-239 Fission Neutron Cross Section

Figure BNL-17 Plutonium-239 Fission Neutron Cross Section

Figure BNL-18 Plutonium-239 Fission Neutron Cross Section

HANFORD ENGINEERING DEVELOPMENT LABORATORY

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

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

Objectives

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

A meeting at AEC Headquarters in December 1971 resulted in the establishment of a pool of highly enriched fission foil materials for use by participants of the ILRR program and for routine LMFBR dosimetry.^(1,2) The ORNL contribution to this Quarterly Report provides additional information on the aluminum encapsulated metal fission foils being prepared for the CFRMF and other ILRR tests. Quality Assurance tests on the high purity aluminum encapsulation material have been initiated at HEDL.

Accomplishments and Status

AEC approval for the release of fissionable material for the fabrication of fission foils for the CFRMF test and routine LMFBR dosimetry was obtained at a meeting on December 8, 1971, at AEC Headquarters.^(1,2) The ORNL contribution to this Quarterly Report provides information on the materials assigned to the fissionable materials pool and the status of the work to fabricate the twenty ^{235}U , ^{238}U , ^{237}Np , and ^{239}Pu foils for the CFRMF and other ILRR tests.

Quality Assurance tests on the high purity aluminum material that ORNL will use for encapsulation of the high purity fission foilshave been initiated.

The RT-4 rabbit tube position of the NBS reactor and the CFRMF central exposure tube facility are both being used for activation studies on this material. Neutron characteristics of the RT-4 and the CFRMF facilities are discussed in previous Quarterly Reports.^(1,2) Irradiations in these two facilities will provide information on impurities with different activation response for thermal and fast reactor neutrons. Consequently, possible interference problems from impurities in the aluminum for direct counting of encapsulated fission foils (for the fission products ^{99}Mo , ^{140}Ba , ^{137}Cs , etc.) for both low and high energy neutrons will be assessed.

Appropriate physical and chemical properties of the high purity aluminum will also be studied and documented for future reference.

Expected Achievements

Initial Quality Assurance tests at HEDL on the high purity aluminum encapsulation material for the high purity fission foils will be completed by the middle of April 1972.

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

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

Objectives

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

Summary

The HEDL Radiochemistry Standards Laboratory is scheduled to provide a ^{140}Ba - ^{140}La standard solution in March 1972.^(1,2) ILRR program participants designated to receive one or more standard solutions are: ANC (J. W. Rogers), ANL (R. Armani), ANL (N. Dudey), ANL (E. Ebersole), ARHCO (W. Zimmer), HEDL (L. Kellogg), and NBS (S. Garfinkel).

The standard will consist of approximately 5 grams of solution containing a combined total (^{140}Ba plus ^{140}La) of approximately 2×10^6 dps/g. Actual calibration values to $\pm 2\%$ or better (95% confidence level) will be provided shortly after the standards are shipped.

Accomplishments and Status

Barium-140 (from which the HEDL radiochemistry standards laboratory will prepare calibrated ^{140}Ba - ^{140}La standard solutions for distribution to requesting laboratories in early March 1972) has been received. Radiochemical purification of the ^{140}Ba was made on February 15, 1972.

Arrangements to have a 4π γ ion chamber reference measurement made by the National Bureau of Standards on a suitable aliquot of this standard have been made. A ^{140}Ba standard solution from Amersham/Searle Corporation has been ordered to provide an additional point of reference and should be received about March 10, 1972.

Expected Achievements

Preliminary calibration, for general packaging purposes, will be made March 3, 1972, after allowing the ^{140}La daughter to reach $\sim 99.5\%$ of its "equilibrium" value.

Packaging and shipment of the individual units will be done March 8 and 9, 1972. Special packaging of activities will be made for the National Bureau of Standards.⁽³⁾

Calibration of the standard solution using 4π β - γ coincidence counting techniques will be started March 10, 1972. The calibration reference date time will be March 13, 1972, 12:00 PST. Final certification of the reference standard solution will be made available by April 3, 1972.

C. CFRMF REACTION RATES BY RADIOCHEMISTRY

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

Objective

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

Summary

Results of low and high power CFRMF tests related to run-to-run power level normalization, check-out of ^{140}Ba - ^{140}La counting for ^{235}U and ^{238}U fission foils, and results of the high-power HEDL-I Non-Fission foil set irradiation are documented and discussed. Analysis of these preliminary test data indicates that the Au and In power level normalization foils should permit determination of relative reactor power levels to within $\sim 1\%$. Differences in reaction rates between sandwich foil measurements using Au, Co, In, ^{238}U , and ^{235}U show foil neutron self shielding, scattering, and possible placement angular orientation effects. For reactions sensitive to low energy neutrons, such as ^{235}U and ^{239}Pu , these effects must be given further consideration before a valid comparison can be made among different sets of measurements from these first tests.

Accomplishments and Status

CFRMF Radiochemistry Test Irradiations

Three preliminary CFRMF test irradiations⁽²⁾ (designated as Tests I, II, and III) were completed during December 1971 and January 1972 at a stated reactor power of 600 watts.⁽⁵⁾ The ANC-I, ANL-I, ARHCO-I, and HEDL-I Non-Fission foil set high power irradiations⁽⁴⁾ were completed during February 1972 at a stated reactor power of 9.75 kW,⁽⁵⁾ see ANC Chapter, Section A.

Results of HEDL's radiochemistry measurements for the preliminary tests, for the power level determinations for the low-power NBS fission chamber and ANL fission track measurements, and the high-power ARHCO-I, ANL-I, and HEDL-I Non-Fission foil set irradiations are documented and briefly discussed here. Further reactor operations information on these tests, the ANL-I and ARHCO-I Non-Fission foil set irradiations, and the ANC reactor power level instrumentation and normalization foil activation measurements are given in the ANC chapter of this Quarterly Report. The NBS chapter provides information on some NBS fission chamber power level measurements.

A Test Plan⁽⁴⁾ for the high-power Non-Fission foil radiochemistry measurements and an irradiation schedule for the first NBS fission chamber, first ANL fission track, and first AI helium specimen (mass spectrometry) measurements was reviewed and revised as a result of meetings at ANC the first week in February 1972.

CFRMF Test Data Documentation

The following summary statements and comments pertain to the Test Plan and those tests completed to date:

A. Figure 1 is a schematic representation of foil placement relative to the CFRMF central exposure tube center line (Q) for the foils selected for irradiation in preliminary Tests I, II, and III.⁽⁵⁾ Each foil package was centered in the 1.5" ID central exposure tube. These tests were designed to provide information on (1) the counting of ^{235}U and ^{238}U fission foils, (2) the power instrumentation normalization procedures, and (3) the use of $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ and $^{115}\text{In}(n,n')^{115\text{m}}\text{In}$ irradiated foils as backup flux level and fluence monitors.^(1,2) Based on calculations for the CFRMF spectrum, the Au primarily monitors flux in the energy range between $\sim 10^{-4}$ to 0.5 MeV while the In covers the range ~ 0.5 to 6 MeV.^(1,2) This two-foil set combination, therefore, monitors most of the neutron energy range of interest for the CFRMF tests.

B. After preliminary Tests I, II, and III, all irradiations were performed within a duplicate NBS fission chamber built at NBS according to NBS design drawings.⁽⁶⁾ A schematic representation of this duplicate chamber inserted in the CFRMF central exposure tube is shown in Figure 2. Figures ANC-3 to 5 of this Quarterly Report are photographs of the chamber and its placement.

TEST I: 12/6/71 IRRADIATION FOIL PLACEMENT^a

		Foil Weights (grams)
Foil Package → CFRMF ☒	Top	
	Au-B*	0.1206 (0.1205) ^b
	²³⁵ U-4	0.05198 (0.05186)
	²³⁵ U-3	0.05084 (0.05079)
	²³⁵ U-2	0.04966 (0.04937)
	²³⁵ U-1	0.05174 (0.05158) (0.05098)
	Au-A	0.1220 (0.1220)
Bottom		

TEST II: 12/7/71 IRRADIATION FOIL PLACEMENT^a

		Foil Weights (grams)
Foil Package → CFRMF ☒	Top	
	In-1	0.04806 (0.04790)
	Au-C	0.1255 (0.1255)
	²³⁵ U-5	0.05123 (0.05106) (0.05121)
Bottom		

TEST III: 1/26/72 IRRADIATION FOIL PLACEMENT^a

		Foil Weights (grams)
Normalizing Foil Package →	Top	
	Au-MD	0.1229
	In-14	0.05034
	Au-ME	0.1234
	In-15	0.04778
1.000"		
Foil Package → CFRMF ☒	Au-MA	0.1243
	Au-MB	0.1233
	Au-MC	0.1211
	²³⁸ U-1	0.06734
	²³⁸ U-2	0.07020
	²³⁸ U-3	0.06690
	²³⁸ U-4	0.06975
	²³⁸ U-5	0.06856
	²³⁸ U-6	0.07130
	²³⁸ U-7	0.06868
	²³⁸ U-8	0.06906
	²³⁸ U-9	0.07204
	²³⁸ U-10	0.06841
	In-11	0.04815
	In-12	0.04968
	In-13	0.04843
Bottom		

* Foil reference numbers.

- Seven-hour irradiations at a stated reactor power of 600 watts. All foils are ~0.5" in diameter with the surface normal to each foil oriented parallel with the axis of the ~1.5" ID CFRMF central exposure hole. All Au and In foils are ~2 mils thick and the ²³⁵U and ²³⁸U foils are about 0.8 and 1 mil thick, respectively, see Table HEDL-3 of reference 2.
- First bracketed values are ANC checks on original HEDL weights; second bracketed values are ARHCO checks.

FIGURE HEDL-1 Au, In, ²³⁸U, AND ²³⁵U FOIL PROPERTIES AND PLACEMENT FOR PRELIMINARY CFRMF TESTS

The foils for these preliminary tests had their plane perpendicular to the exposure tube axis, Figure 1, while foils irradiated in the NBS fission chamber, Figure 2, had their planes parallel to that axis.

C. For the Non-Fission foil irradiations, a 3/4" diameter by ~5 mil thick aluminum foil holder with a ~1/2" central diameter and ~145 mil central depth was used for maintaining foil placement. As shown schematically in Figure 3, in the design of this holder, the 3/4" diameter holder lip is the same dimension as that of the NBS fission chamber foils. Using this foil holder, the locations of all foils are fixed to within ~10 mils relative to the NBS fission foil position shown in Figure 2. (Also see Figure NBS-1 of reference 1.) With this arrangement, accurate corrections will be possible for flux gradients, scattering, etc.*

D. Five identical sets of Non-Fission foils for radiochemistry analysis were prepared by HEDL. Two sets were prepared for analysis by HEDL (I and II), one set by ANC (I), one set by ANL (I), and one set by ARHCO (I). Information on these foil sets is provided in Table 1. In Table 1, nominal data on foil thicknesses, weights, reaction products to be counted, and half lives are listed. Additional information and more precise data for each laboratory's assembled foil set(s) are given in Appendix A of reference 4. It is noted that Al, Au, and In foils were placed at the top, bottom, and mid-point of each of these five foil sets to provide information on reproducibility of counting, self-shielding, flux gradients, etc. Each foil set was designed so that the total package inside thickness was ~145 mils with an ~1/2" inside diameter, Figure 3. Some of the foils within the package, such as the ^{235}U and ^{238}U foils which are subject to fission fragment loss, were individually wrapped in thin aluminum. These two foils were included to provide preliminary information for radiochemistry fission foil measurements and also to obtain information on the n, γ reaction for ^{238}U . Table 2 provides isotopic information on the ^{235}U and ^{238}U . A single wrapping of 0.135% Au-Al 20 mil wire around the outside of the holder's hat top, Figure 3, was included to obtain additional data on the infinitely dilute $^{197}\text{Au}(n, \gamma)^{198}\text{Au}$ reaction rate. Information on this alloy is given in reference 1, pages HEDL-3 to HEDL-6.

* The rather large size of the Non-Fission foil package was primarily dictated by a trade-off between minimizing reactor utilization time and obtaining sufficient information on a large enough set of foil reactions.

Figure HEDL-2 Schematic Diagram of NBS Fission Chamber Placement in the CFRMF Central Exposure Tube

Figure HEDL-3 Metal Non-Fission Foil Set Holder for ANC, ANL, ARHCO, and HEDL Radiochemistry Measurements

TABLE HEDL-1
METAL NON-FISSION FOIL SET FOR CFRMF 70 kW-HOUR IRRADIATION FOR
ANC, ANL, ARHCO, AND HEDL RADIOCHEMISTRY MEASUREMENTS

Foil and Position ^a	Approximate Thickness (mils)	Approximate Weight (grams)	Reaction Products of Primary Interest and Half Lives in Current Use at HEDL
Al ^b	5	0.047	²⁷ Mg (9.5 min); ²⁴ Na (15.00 ±0.04 hr)
Au ^b	0.5	0.034	¹⁹⁸ Au (2.697 ±0.005 d)
In ^b	2	0.049	^{116m} In (54 min); ^{115m} In (4.5 hr)
Cu	0.5	0.015	⁶⁴ Cu (12.84 ±.1 hr)
Co	0.5	0.015	} ⁶⁰ Co (5.268 ±0.01 y)
Co	0.5	0.015	
Co	0.5	0.015	
Ni	10	0.30	⁵⁸ Co (71.3 ±0.2 d)
Fe	30	0.81	⁵⁴ Mn (312.5 ±0.5 d); ⁵⁹ Fe (44.5 ±0.4 d)
Al ^b	5	0.047	²⁷ Mg (9.5 min); ²⁴ Na (15.00 ±0.04 hr)
Au ^b	0.05	0.0035	¹⁹⁸ Au (2.697 ±0.005 d)
In ^b	2	0.049	^{116m} In (54 min); ^{115m} In (4.5 hr)
Ti ^c	10	0.16	} ⁴⁶ Sc (84.0 d); ⁴⁷ Sc (3.43 ±0.05 d);
Ti ^c	10	0.16	
Sc	5	0.050	⁴⁶ Sc (84.0 d)
²³⁵ U ^d	4	0.25	¹⁴⁰ La (40.27 ±0.05 hr); ¹⁴⁰ Ba (12.8 ±0.1 d)
²³⁸ U ^d	1	0.067	¹⁴⁰ La (40.27 ±0.05 hr); ¹⁴⁰ Ba (12.8 ±0.1 d); ²³⁹ Np (2.35 d)
In ^b	2	0.049	^{116m} In (54 min); ^{115m} In (4.5 hr)
Au ^b	0.5	0.034	¹⁹⁸ Au (2.697 ±0.005 d)
Al ^b	5	0.047	²⁷ Mg (9.5 min); ²⁴ Na (15.00 ±0.04 hr)
Au-Al ^e	20 Wire	0.10	¹⁹⁸ Au (2.697 ±0.005 d)

- a. All foils are ~0.52" in diameter and are separated by ~0.65 mil thick x 0.5" diameter aluminum spacers. The order of the foils given in the table is the same as that used for placement within the ~5 mil thick aluminum holder (hat) shown in Figure 3. The first foil listed in the table is placed closest to the brim of the hat.
- b. These foils will also provide information on counting reproducibility, gradients, and resonance self absorption.
- c. Two foils used to increase total quantity of material.
- d. These foils are individually wrapped in ~0.65 mil thick aluminum to prevent loss of fission fragments - see Figure 6.
- e. 0.135 ±0.005% Au-Al 20 mil wire wrapped around outside of foil holder hat top, see Figures 3 and 5.

TABLE HEDL-2
FISSION FOIL DATA

^{235}U : Enriched ^{235}U , 5 mils thick,* 3 pieces, 12.5 grams (Density: Calculated to be $\sim 18.8 \text{ g/cm}^3$)

	<u>Isotopic Analysis**</u>	<u>Errors 95% Confidence Level**</u>
^{234}U	0.657%	0.0016%
^{235}U	93.098%	0.014%
^{236}U	0.323%	0.0017%
^{238}U	5.922%	0.014%

^{238}U : ORNL Lot No. 105(C), depleted to 12 ppm ^{235}U (Density: Assumed to be 19.05 g/cm^3)

* Cleaning of the surface oxidation may have removed up to 1 mil of total thickness.

** These results were provided by R. Armani (ANL) and were taken from a computer printout.

E. Each assembled foil set, Table 1, was designed for insertion in the duplicate NBS fission chamber, Figure 2, with the hat top down and the 3/4" brim (lip) occupying the position used for placement of NBS fission foils. These Non-Fission foil sets were sized for an ~ 7 -hour irradiation at $\sim 10 \text{ KW}$.

F. A sixth (HEDL-III) Non-Fission foil radiochemistry set for non-metallic and thick metal foils was prepared for analysis by HEDL, ARHCO, ANL, and/or ANC as appropriate. This foil set is described in reference 4.

G. A seventh (HEDL-IV) Non-Fission resonance self-shielding foil set was prepared for counting at HEDL to provide information on resonance self-shielding for the resonance detectors used in the first six sets. Details on this foil set are provided in reference 4.

H. A photograph of the as-assembled and fabricated seven foil sets is shown in Figure 4A. The NBS duplicate fission chamber housing with the bottom teflon insert in place and the aluminum fission foil holder ring and top teflon insert removed are also shown. Figure 4B is another view of the chamber with one of the Non-Fission foil sets inserted in the aluminum fission foil holder

Figure HEDL 4A NBS Duplicate Fission Chamber Enclosure and Non-Fission Foil Sets

Figure HEDL-4B Non-Fission Foil Set Placement Within the NBS Duplicate Fission Chamber Enclosure

ring and locked in place by the top teflon insert. Other components of the fission chamber (aluminum and teflon) are inserted on top of the teflon insert to complete the assembly for irradiation. Individual foils can be clearly identified and located in Figure 5, a radiograph of one of the assembled Non-Fission foil sets positioned within the NBS duplicate fission chamber. Only a few of the chamber's actual components were used for this simulated assembly.

I. All possible efforts were made to obtain high purity foil materials for all the aforementioned foil sets to minimize the effect of impurities on subsequent foil counting. Counting of foils is being accomplished by both NaI(Tl) and Ge(Li) detectors at the participating laboratories. ANL has provided information on their counting techniques in reference 2, pages ANL-2 to ANL-13. Similar information is expected to be included in other participants' future Quarterly Report contributions.

J. Two-mil thick by 1/2" diameter Au foils and 2-mil by 1/2" diameter In foils were provided to ANC for the three Preliminary Tests I, II, and III, and for placement in the power level normalization foil holder for all subsequent irradiations. One Au and one In foil (sandwiched together and with the In foil on the bottom) were placed in each of the ~0.008" deep holes (depressions) in the "Normalizing Foil Holder," Figure 2. One Au and one In foil were counted by ANC and the other two by HEDL. The HEDL results for In and Au are summarized in Tables 3 and 4, and the ANC results are given in their chapter of this Quarterly Report.

K. Results of HEDL's, ANL's, ANC's, and ARHCO's low power fission foil measurements (corresponding to the low power In and Au results in Tables 3 and 4) are summarized in Tables 5, 6, and 7 and Figure 6. Results of HEDL's high power fission and Non-Fission foil radiochemistry measurements are summarized in Table 8. Single foil flux level estimates based on the high power measurements are given in Table 9 to provide a consistency check on all results.

Discussion of Radiochemistry Reaction Rate Measurements

A. $^{115}\text{In}(n,n')^{115\text{m}}\text{In}$

$^{115}\text{In}(n,n')^{115\text{m}}\text{In}$ measured reaction rates are summarized in Table 3. Additional details on foil placement, counting, etc., and references to the Figures and Tables of the previous section complete the documentation for this reaction for the low and high power tests.

HEDL-13

Figure HEDL-5 X-ray of Non-Fission Foil Set Placement in the NBS Duplicate Fission Chamber

TABLE HEDL-3

SUMMARY OF HEDL MEASURED
 $^{115}\text{In}(n,n')$ ^{115m}In REACTION RATES

Date	Test	Foil No.	Foil Thickness (mils)		10^{+16} x Reactions ^b Per Sec.-Nuclei	Ratio ^c	Reactor Power (kW)	Remarks
			Total ^a	Actual				
1/26/72	III	15	4	2	4.429	0.9980	0.600 ^d	Foil Near Norm. Foil Position. ^e
1/26/72	III	11	6	2	4.437	0.9998	0.600	Sandwich - Top foil. ^e
1/26/72	III	12	6	2	4.435	0.9993	0.600	Sandwich - Middle foil.
1/26/72	III	13	6	2	4.450	1.003	0.600	Sandwich - Bottom foil.
2/8/72	ARHCO-I	8'	2	2	67.95 (4.438) ^e	Norm. Point	9.187	In Norm. Foil Holder. ^f
2/8/72	ARHCO-I	9	6	2*	67.66 (4.419)	0.9957	9.187	In NBS Chamber. ^f
2/8/72	ARHCO-I	8	6	2*	67.21 (4.390)	0.9892	9.187	In NBS Chamber. ^f
2/8/72	ARHCO-I	7	6	2*	67.63 (4.417)	0.9953	9.187	In NBS Chamber. ^f
2/15/72	ANL-I	10'	2	2	66.05 (4.438)	Norm. Point	8.930	In Norm. Foil Holder. ^f
2/16/72	HEDL-I	12'	2	2	66.26 (4.438)	Norm. Point	8.958	In Norm. Foil Holder. ^f
2/16/72	HEDL-I	12	6	2*	66.85 (4.478)	1.009	8.958	In NBS Chamber. ^f
2/16/72	HEDL-I	11	6	2*	67.23 (4.503)	1.015	8.958	In NBS Chamber. ^f
2/16/72	HEDL-I	10	6	2*	66.95 (4.484)	1.010	8.958	In NBS Chamber. ^f

- a. Total effective thickness of In (for a beam geometry) in an assembled foil set for considering scattering effects. An asterisk following the thickness indicates the foil set surface normal was perpendicular to the CFRMF central exposure tube axis as a result of irradiation in the NBS fission chamber enclosure, Figure 2.
- b. Counted at HEDL with a NaI(Tl) detector at the 10 cm position with a counting precision of $\sim 0.4\%$ for each of the 2 mil foils. No correction has as yet been made for the attenuation of the ^{115m}In gamma rays within each foil - corrections are estimated to be less than 0.3% for a 2 mil foil.
- c. All high power results have been re-normalized (bracketed values) to a power level of 0.6 kW based on the measured 2 mil In normalizing foil power level factors of 15.311 for ARHCO-I, 14.883 for ANL-I, and 14.930 for HEDL-I. All re-normalized values are then ratioed to the 0.600 kW average value of 4.438×10^{-16} . Calculations indicate that the correction for flux gradients for most of the foils is less than $\sim 0.2\%$, based on ^{237}Np data, reference 2. The exception is for all foils irradiated in the normalizing foil holder, Figure 2, which still require multiplication by a factor of 1.005 before comparison with foils irradiated at the reactor ζ .
- d. Stated reactor power, reference 5.
- e. The foil placement for Tests I, II, and III is shown in Figure 1. The NBS fission chamber enclosure was not available for these first three tests and the foil sets were placed with the surface normal to the assembled foil set parallel with the central exposure tube axis.
- f. Figure 3 shows the arrangement of foils within the foil set holder. The brim of the hat has the same diameter as the NBS fission chamber foils and is designed so that it is held in the same position as a fission chamber foil.
- g. Counted by ARHCO, reference 7.
- h. This is the average value of the measured 2 mil In normalization foil reaction rate for two irradiation positions, Figure 1. Since the foils were not irradiated in the power level normalization foil holder, Figure 2, the measured values were corrected to the normalization foil holder position by dividing by a factor of 1.0040 for the #15 foil and a factor of 1.0050 for the #11, #12, and #13 foils. These two factors are based on the ^{237}Np gradient data presented in Figure ANC-1, reference 2.

The value of 4.438×10^{-16} ($\pm 0.2\%$) in Table 3 was established as the indium foil 0.600 kW (low power) reaction rate reference point for the determination of subsequent run-to-run reactor power level normalization factors. Using this value and those obtained from the normalization foils for the high power ARHCO-I (#8' foil), the ANL-I (#10' foil), and the HEDL (#12' foil) irradiations yielded power levels of 9.187 kW, 8.930 kW, and 8.958 kW relative to the 0.600 kW level for these three high power tests, respectively.

The measured reaction rates for the three indium foils placed at the top, middle, and bottom of the Non-Fission foil sets (for the ARHCO-I and HEDL-I irradiations in the NBS duplicate fission chamber) are listed below each normalizing foil result in Table 3. In each case, the foil with the lowest identification number was at the bottom. After correcting all values back to the same power level of 0.600 kW, the ratio of each value to the 4.438×10^{-16} reference value is given in the third from last column. Good agreement is found among all of the listed values counted by HEDL as well as those counted by ARHCO.⁽⁷⁾ Within experimental uncertainties and expected flux gradients, these In results do not show any measurable high energy ($> \sim 0.5$ MeV) effect based on foil placement or angular orientation within the central exposure tube.

B. $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$

$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ measured reaction rates are summarized in Table 4. Additional details on foil placement, counting, etc., and references to the figures and tables of the previous section complete the documentation for this reaction for the low and high power tests.

The value of 3.199×10^{-15} ($\pm 0.72\%$) in Table 4 was established as the gold foil 0.600 kW (low power) reaction rate reference point for the determination of subsequent run-to-run reactor power level normalization factors. Using this value, and those obtained from the normalization foils for the high power ARHCO-I (#AH foil), the ANL-I (#AJ foil), and HEDL-I (#AL foil) irradiations yielded power levels of 9.295 kW, 8.952 kW, and 8.823 kW relative to the 0.600 kW level for these three high power tests, respectively. For the HEDL-I test, the value of 8.823 kW based on Au is discrepant with the value of 8.958 kW based on In. This same discrepancy was also observed by ANC for their normalizing foil set.⁽⁵⁾ A comparison of the HEDL-I and ARHCO-I 0.5, 0.05, and 0.003 mil Au foil results (for foils irradiated in the NBS fission chamber) does not show this power level discrepancy. At this time, there is no explanation for it.

TABLE HEDL-4
SUMMARY OF HEDL-MEASURED
 $^{197}\text{Au}(n,\gamma)$ ^{198}Au REACTION RATES

Date	Test	Foil No.	Foil Thickness (mils)		10 ¹⁵ x Reactions ^b Per Sec.-Nuclei	Ratio ^c	Reactor Power (kW)	Remarks	
			Total ^a	Actual					
12/6/71	I	A	4	2	3.108	0.8757	0.600 ^d	Top of ²³⁵ U set. ^e	
12/7/71	II	C	2	2	3.196	0.9005	0.600	Top of ²³⁵ U set. ^e	
1/26/72	III	ME	4	2	3.150	0.8876	0.600	1" Above Sandwich Set. ^e	
1/26/72	III	MA	6	2	3.081	0.8681	0.600	Sandwich - Top Foil. ^e	
1/26/72	III	MB	6	2	3.002	0.8459	0.600	Sandwich - Middle Foil.	
1/26/72	III	MC	6	2	3.084	0.8690	0.600	Sandwich - Bottom Foil.	
2/2/72	NBS-I	AB	2	2	3.173	3.199 ^h (±0.72%)	0.8941	0.600	In Norm. Foil Holder. ^g
2/4/72	NBS-I	AF	2	2	3.218		0.9067	0.600	In Norm. Foil Holder. ^g
2/4/72	ANL-I	AD	2	2	3.205		0.9031	0.600	In Norm. Foil Holder. ^g
2/8/72	ARHCO-I	6	1.05	0.5*	54.18 (3.497) ^c	0.9853	9.295	In NBS Chamber. ^g	
2/8/72	ARHCO-I	3'	1.05	0.05*	54.72 (3.532)	0.9952	9.295	In NBS Chamber. ^g	
2/8/72	ARHCO-I	5	1.05	0.5*	53.83 (3.475)	0.9791	9.295	In NBS Chamber. ^g	
2/8/72	ARHCO-I	3	0.003 ^f	0.003*	54.98 (3.549)	1.000	9.295	0.135% Au-Al Wire in NBS Chamber. ^g	
2/8/72	ARHCO-I	AH	2	2	49.56 (3.199)	Norm. Point	9.295	In Norm. Foil Holder. ^g	
2/15/72	ANL-I	AJ	2	2	47.73 (3.199)	Norm. Point	8.952	In Norm. Foil Holder. ^g	
2/16/72	HEDL-I	8	1.05	0.5*	52.55 (3.574)	1.007	8.823	In NBS Chamber. ^g	
2/16/72	HEDL-I	4'	1.05	0.05*	53.36 (3.629)	1.023	8.823	In NBS Chamber. ^g	
2/16/72	HEDL-I	7	1.05	0.5*	51.78 (3.521)	0.9921	8.823	In NBS Chamber. ^g	
2/16/72	HEDL-I	4	0.003 ^f	0.003*	53.20 (3.618)	1.019	8.823	0.135% Au-Al Wire in NBS Chamber. ^g	
2/16/72	HEDL-I	AL	2	2	47.04 (3.199)	Norm. Point	8.823	In NBS Chamber. ^g	

- a. Total effective thickness of Au (for a beam geometry) in an assembled foil set for considering resonance self shielding. An asterisk following the thickness indicates the foil set surface normal was perpendicular to the CFRMF central exposure tube axis as a result of irradiation in the NBS fission chamber enclosure, Figure 2.
- b. Counted at HEDL with a NaI detector at the 20 cm position with a counting precision of $\sim 0.1\%$ to 0.4% for pure Au samples and 1 to 2% for the alloy. No correction has as yet been made for the attenuation of the ^{198}Au gamma rays within each foil - corrections are estimated to be less than 1%, 0.3%, and 0.6% for the 2 mil, 0.5 mil, and 20 mil 0.135% Au-Al wire, respectively.
- c. All high power results have been re-normalized (bracketed values, adjacent column) to a power level of 0.6 kW based on the measured 2 mil gold normalizing foil power level factors of 15.492 for ARHCO-I, 14.920 for ANL-I, and 14.705 for HEDL-I. All re-normalized values are then ratioed to the ARHCO-I 0.135% Au-Al wire value of 3.549×10^{-15} reactions/sec.-nuclei, assuming it represents the infinitely dilute reaction rate at the core Q_c . Calculations indicate that the correction for flux gradients for most of the foils is less than $\sim 0.1\%$, based on the ^{235}U data, reference 2. The exceptions are foil #ME and foils irradiated in the normalizing foil holder, Figure 2, which still require multiplication by factors of 1.006 and 1.011, respectively, before comparison with foils irradiated at the reactor Q_c .
- d. Stated reactor power for the 0.6 kW level, reference 5. For higher powers, estimated power levels are given based on the 0.6 kW value and the relative difference of the 2 mil Au normalizing foils measured reaction rates.
- e. The foil placement for Tests I, II, and III is shown in Figure 1. The NBS fission chamber enclosure was not available for these first three tests, and the foil sets were placed with the surface normal to the assembled package parallel with the central exposure tube axis.
- f. Approximately 7" of the 20 mil 0.135% Au-Al alloy was wrapped around the "non-fission foil set" holder hat as shown in Figure 3. The effective thickness for this alloy for a beam geometry measurement is given.
- g. Table 1 shows the arrangement of foils within the foil set holder. The brim of the hat has the same diameter as the NBS fission chamber foils and is designed so that it is held in the same position as a fission chamber foil, Figure 2.
- h. This is the average value of the measured 2 mil Au normalization foil reaction rate for a series of three irradiations at the stated 600 watt reactor power level. This is the reference value for re-normalization to the 600 watt level of all other runs at different power levels.

The HEDL measured reaction rates for the Au foils placed at the top (0.5 mils), middle (0.05 mils), bottom (0.5 mils), and on the outside (0.003 mils, effective thickness of the 0.135% Au-Al alloy wire) of the HEDL-I and ARHCO-I Non-Fission foil sets are listed above each normalizing foil result in Table 4. After correcting all values back to the same power level of 0.600 kW, the ratio of each value to the ARHCO-I 3.549×10^{-15} * reference value is given in the third from last column. The overall agreement is good after considering power level differences, the angular orientation of foils, and their total effective beam geometry thicknesses (column 4 of Table 4). The variation from unity suggests that foils with the surface normal parallel with the CFRMF exposure tube axis appear to be subject to rather high neutron self shielding effects. That is an ~10%, 12%, and 15% effect for a 2 mil, 4 mil, and 6 mil Au foil, respectively. On the other hand, Au foils of up to 0.5 mil (~1 mil for a beam geometry) thickness show no apparent effect when the surface normal is perpendicular to the exposure tube axis.** These results suggest that scattering and self shielding effects (as well as possible low energy neutron streaming from both open ends of the central exposure tube) must be studied and better understood before further track record and radiochemistry irradiations are accomplished.

C. $^{235}\text{U}(n,f)^{140}\text{Ba}$

ANC, ANL, ARHCO, and HEDL $^{235}\text{U}(n,f)^{140}\text{Ba}$ measured fission rates for preliminary Tests I and II are summarized in Table 5. The four ~0.8 mil thick foils for Test I were stacked together (bare) and placed in a single outer wrapping of thin aluminum for irradiation at a stated reactor power of 0.600 kW.⁽⁵⁾ A single ~0.8 mil thick foil wrapped in thin aluminum was irradiated for Test II at the same power level. The placement of the fission foils for Tests I and II relative to Au and In power level normalization foils is shown schematically in Figure 1.

* This value was selected because it normally would represent the value for an infinitely dilute foil.

** Based on $^{235}\text{U}(n,f)$ results, however, neutron scattering in the rather large foil package may have enhanced the observed gold reaction rate by as much as 5 to 10% for the Au-Al alloy, 0.5 mil, and 0.05 mil gold foils.

TABLE HEDL-5
PRELIMINARY CFRMF TESTS I AND
II ^{235}U MEASURED FISSION RATES^a

Foil Reference Number ^b	10 ⁺¹⁴ x Measured Fission Rate (Fissions/Sec.-Nuclei) ^c				
	HEDL	ARHCO ⁽⁷⁾	ANC ⁽⁵⁾	ANL ⁽⁸⁾	Average
1	1.245	1.227	-	-	1.236 ^d
2	1.253	1.285	-	-	1.269
3	-	-	-	1.313	1.313
4	-	-	1.237	-	1.237 ^d
5	1.204	1.188	-	-	1.196 ^e

Foils 2 and 3: Average value = $1.291 \pm (\pm 2.4\% 1\sigma)$
 $(\pm 1.7\% 1 \text{ standard error})$

Foils 1 and 4: Average value = $1.237 \pm (\pm 0.06\% 1\sigma)$
 $(\pm 0.04\% 1 \text{ standard error})$

- a. At a stated reactor power of 600 watts, reference 5, with the normal to the foils surface parallel with the CFRMF central exposure tube axis, Figure 1. These results have not been corrected for ^{238}U fissions, an 0.4% correction.
- b. The diameter of the individual foils was measured to be between 0.51 to 0.52" (an average of ~ 0.515 ") with a nominal as punched out and cleaned thickness of ~ 0.8 mils and weight of $\sim 0.0380 \text{ g/cm}^2$. The enrichment was stated to be 93.2% ^{235}U , but has not been verified by QA.
- c. Based on a ^{140}Ba yield of 5.8%, reference 2. No correction has been made for the attenuation of the ^{140}La gamma rays within each foil - corrections are estimated to be less than 0.2% for the 0.8 mil foils used.
- d. These foils show the effect of fission fragment loss from one surface of the foil, see Figure 6.
- e. This foil shows the effect of fission fragment loss from both surfaces, see Figure 6.

After irradiation for seven hours, the five fission foils from Tests I and II were separated from the aluminum covers and were then counted at the participating laboratories. As indicated in the footnotes of Table 5, foils #2 and #3 were not subject to fission fragment loss, foils #1 and #4 were subject to loss from one surface, and foil #5 was subject to loss from both surfaces. These individual foil results were combined to develop the percent fission fragment loss versus foil thickness (g/cm^2) curve (solid line) of Figure 6 for ^{235}U fission.

D. $^{238}\text{U}(n,f)^{140}\text{Ba}$

Results for ^{238}U fission similar to those for ^{235}U are summarized in Table 6 for Preliminary Test III. The placement and packaging of the ten ^{238}U sandwich foils, relative to the Au and In normalizing foils, are shown schematically in Figure 1. This foil set was irradiated at a stated reactor power of 0.600 kW for seven hours.⁽⁵⁾

The results of the counting of the Au and In foils for Tests I, II, and III are summarized in Tables 3 and 4 and, as stated before, indicate that a run-to-run power level normalization to within ~1% is possible using these two foils.

The ^{238}U foils #2 through #9 were not subject to fission fragment loss, but foils #1 and #2 were from one surface. In Figure 6, these two foils yield a result for percent fission fragment loss for a ~2 mil foil. As with ^{235}U , a ^{238}U curve could have been developed from these data for foils ≥ 2 mils thickness.

The difference between the ^{238}U and ^{235}U results of Figure 6 may be primarily due to experimental uncertainties in the relative counting of the fission foils; however, it might also be an indication of the effect of low energy neutron streaming. That is, the two outer foils #1 and #4 of Test I, Table 5, might have been subject to an enhanced fission rate (relative to the two center foils) due to possible streaming and absorption effects of lower energy neutrons. Such an effect could account for the apparent greater loss of fission fragments from a ^{238}U foil, which is not subject to low energy fission.

E. $^{238}\text{U}(n,\gamma)^{239}\text{U}$

Results for $^{238}\text{U}(n,\gamma)^{239}\text{U}$ for the same foils listed in Table 6 are summarized in Table 7. All values are normalized to the average value of foils #1 and #10. These relative numbers can be converted to absolute values of reactions per second per nucleus by multiplying by 1.441×10^{-15} . The poor counting statistics of ~2% are primarily responsible for the scatter in the data; nevertheless, there is a definite trend of an unexpected* reduction in

* Unexpected, because the CFRMF ^{238}U core material would be expected to remove all neutrons at the major resonances of ^{238}U .

Figure HEDL-6 Estimated Percent Loss of Fission Products from Different Thicknesses of ²³⁵U and ²³⁸U Irradiated in the CFRMF Central Exposure Tube

TABLE HEDL-6
PRELIMINARY CFRMF TEST III
 ^{238}U MEASURED FISSION RATES^a

Foil Reference ^b Number	$10^{+16} \times$ Measured Fission Rate (Fissions/Sec.-Nuclei) ^c
1	5.687 ^d
2	5.929
3	5.827
4	5.868
5	5.837
6	5.833
7	5.796
8	5.866
9	5.775
10	5.640 ^d

Foils 2 to 9: Average value = 5.841 ($\pm 0.8\%$ 1σ)
($\pm 0.3\%$ 1 standard error)

Foils 1 and 10: Average value = 5.664 ($\pm 0.6\%$ 1σ)
($\pm 0.4\%$ 1 standard error)

- ^a. At a stated reactor power of 600 watts, reference 5, with the normal to the foils surface parallel with the CFRMF central exposure tube axis, Figure 1. These results require less than $\sim 0.03\%$ correction for ^{235}U fissions.
- ^b. The diameter of the individual foils was measured to be between 0.518 to 0.522" (an average of ~ 0.52 ") with a nominal as punched out and cleaned thickness of ~ 1 mil and weight of ~ 0.0505 g/cm². The depletion was stated to be 12 ppm of ^{235}U , but has not been verified by QA.
- ^c. Based on a ^{140}Ba yield of 6.0%, reference 2. No correction has been made for the attenuation of the ^{140}La gamma rays within each foil - corrections are estimated to be less than 0.2% for a 1 mil foil.
- ^d. These foils show the effect of fission fragment loss from one surface of the foil, see Figure 6.

TABLE HEDL-7
PRELIMINARY CFRMF TEST III
 $^{238}\text{U}(n,\gamma)^{239}\text{U}$ MEASURED REACTION RATES^a

Foil ^b Reference Number	Relative Measured ^c Reaction Rate
1	0.998
2	0.957
3	0.954
4	0.942
5	0.976
6	0.964
7	0.937
8	0.977
9	0.969
10	1.002

- ^a. Irradiated as a ten-foil sandwich set at a stated reactor power of 600 watts, reference 5, with the normal to the foil set surface parallel with the CFRMF central exposure tube axis, Figure 1.
- ^b. See footnote (b) of Table 6.
- ^c. Counted at HEDL with a Ge(Li) detector at ~6 cm position with a counting precision of ~2% for each of the 1 mil foils. All values are normalized to the average value of foils 1 and 10. No correction has been made for the attenuation of the 0.1064 MeV gamma-rays within each foil - the correction is estimated to be 4.07% for a 1 mil foil. To convert these relative numbers to absolute values of reactions per second per nucleus, multiply by 1.441×10^{-15} . This conversion factor is only approximate, and a better value is being determined.

activity of up to possibly 5 to 6%. This reduction is presumably due to neutron self-shielding/scattering as a result of the primary CFRMF spectrum and possibly some low energy neutron streaming from the open ends of the CFRMF central exposure tube.

Assuming that the measured reaction rate (Table 8) for the single 1-mil ^{238}U foil in the HEDL-I foil set (with its normal perpendicular to the exposure tube axis) represents an infinitely dilute foil (and after correcting for

power level difference), it is found that there is an apparent 12 to 13% self-shielding/scattering effect for a 10-mil ^{238}U foil with its surface normal parallel with the CFRMF central exposure tube axis, a result similar to that found for gold.

F. HEDL-I Measured Reaction Rates

Results of the radiochemistry measurements for the HEDL-I Non-Fission foil set are summarized in Table 8. The CFRMF reactor was operated for 7 hours at a stated reactor power of 9.75 kW⁽⁵⁾ for the HEDL-I test. The results of the Au and In power level normalization foil measurements for this test are given in Tables 3 and 4, respectively. As previously stated, the reason for the discrepancy between the Au and In results is not understood; however, the combined analyses of the Au results for the ANL-I, HEDL-I, and ARHCO-I tests, coupled with those for In, indicate that the power levels for the ANL-I and HEDL-I tests (run on successive days) were essentially identical but a factor of 0.969 lower than that for the ARHCO-I test. Based on foil activation measurements, therefore, HEDL's best estimate for the powers of these three tests relative to the 0.600 kW level are 8.95 kW, 8.95 kW, and 9.24 kW, respectively.

Gamma-ray counting of the HEDL-I Non-Fission foil set was accomplished with NaI(Tl) and Ge(Li) detectors. In Table 8, the counting precisions reported (parenthesized values next to the reaction rates) are in units of percent at the 95% confidence level. The bracketed values are the times (in days) from reactor shut down to count time. Table 9 documents reaction product nuclear data used by HEDL in arriving at the measured values of reaction rates, Table 8. Time zero (reactor shut down) for the HEDL-I test is 06:12:17 PST or 07:12:17 MST.⁽⁵⁾ For most of the isotopes, half life values are given with an estimated uncertainty. The estimated accuracy of absolute calibration for each isotope is given (at the 95% confidence level) in the last column of the table. The gamma peak energies and branching ratios used by HEDL are listed in columns 2 and 3.

In Table 8, estimated correction factors for gamma-ray attenuation within individual foils are listed in column 4, but have not been applied in order to simplify the initial laboratory-to-laboratory comparisons for the "Non-Fission" foil sets.

Procedures used by HEDL for propagating errors and assigning final absolute uncertainties will be documented in a future Quarterly Report, as was recently done by ANL.⁽²⁾

TABLE HEDL-8
SUMMARY OF HEDL MEASURED REACTION RATES FOR α
THE HEDL-1 NON-FISSION FOIL SET

Reaction	Foil No.	Thickness (mils)	γ -Ray Attenuation Correction Factors	$10^{15} \times$ Reactions Per Sec.-Nuclei		Average ^d
				NaI(Tl) ^d	Ge(Li) ^d	
$^{235}\text{U}(n,f)^{140}\text{Ba}^b$	4	~ 4	1.0051	202.1 (± 0.94) [19.22] ^e	203.2 (± 1.2) [16.40] ^e	202.7
$^{238}\text{U}(n,f)^{140}\text{Ba}^c$	4	~ 1	1.0012	9.438 (± 1.7) [19.27]	9.495 (± 4.1) [19.41]	9.484
$^{238}\text{U}(n,\gamma)^{239}\text{U}$	4	~ 1	1.0407	-	23.80 (± 1.8) [19.36] ^e	23.80
$^{45}\text{Sc}(n,\gamma)^{46}\text{Sc}$	4	~ 5	1.0011	2.957 (± 0.70) [14.42]	2.871 (± 2.9) [21.56]	2.914
$^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$	G	~ 33	1.0190	0.7494 (± 9.0) [19.11]	0.7477 (± 13.5) [21.80]	0.7486
$^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$	4	~ 0.5	1.0000	5.448 (± 3.7) [0.6146]	-	5.448
$^{59}\text{Co}(n,\gamma)^{60}\text{Co}$	12	~ 0.5	1.0003	9.41 (± 1.1) [2.089]	-	9.232
$^{59}\text{Co}(n,\gamma)^{60}\text{Co}$	11	~ 0.5	1.0003	8.793 (± 1.6) [1.551]	-	
$^{59}\text{Co}(n,\gamma)^{60}\text{Co}$	10	~ 0.5	1.0003	9.493 (± 1.1) [1.634]	-	
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	8	~ 0.5	1.0025	52.55 (± 0.57) [1.395]	-	52.72
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	4	~ 0.05	1.0003	53.36 (± 0.47) [1.361]	-	
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	7	~ 0.5	1.0025	51.78 (± 0.47) [1.385]	-	
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	4	0.135% Alloy	1.0056	53.20 (± 4.1) [12.28]	-	
$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	12	~ 5	1.0008	0.01981 (± 1.7) [0.7174]	-	0.01996
$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	11	~ 5	1.0008	0.01989 (± 1.7) [0.7285]	-	
$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	10	~ 5	1.0008	0.02019 (± 1.7) [0.7063]	-	
$^{46}\text{Ti}(n,p)^{46}\text{Sc}$	U	~ 10	1.0017	0.3313 (± 4.3) [19.2]	-	0.3339
$^{46}\text{Ti}(n,p)^{46}\text{Sc}$	T	~ 10	1.0017	0.3364 (± 4.3) [19.2]	-	
$^{47}\text{Ti}(n,p)^{47}\text{Sc}$	U	~ 10	1.0095	0.5604 (± 1.0) [0.5389]	-	0.5565
$^{47}\text{Ti}(n,p)^{47}\text{Sc}$	T	~ 10	1.0095	0.5525 (± 0.53) [1.176]	-	
$^{48}\text{Ti}(n,p)^{48}\text{Sc}$	U	~ 10	1.0035	0.008390 (± 2.4) [0.5389]	-	0.008425
$^{48}\text{Ti}(n,p)^{48}\text{Sc}$	T	~ 10	1.0035	0.008459 (± 1.7) [1.176]	-	
$^{54}\text{Fe}(n,p)^{54}\text{Mn}$	G	~ 33	1.0217	2.276 (± 1.4) [19.12]	2.299 (± 2.2) [21.79]	2.288
$^{58}\text{Ni}(n,p)^{58}\text{Co}$	TD	~ 10	1.0081	3.018 (± 0.65) [1.590]	3.015 (± 0.35) [23.84]	3.017
$^{115}\text{In}(n,n')^{115\text{m}}\text{In}$	12	~ 2	1.0026	6.685 (± 0.43) [0.6979]	-	6.701
$^{115}\text{In}(n,n')^{115\text{m}}\text{In}$	11	~ 2	1.0026	6.723 (± 0.41) [0.6827]	-	
$^{115}\text{In}(n,n')^{115\text{m}}\text{In}$	10	~ 2	1.0026	6.695 (± 0.43) [0.6905]	-	

- a. At a stated reactor power of 9.75 kW. (5) See Tables 3 and 4 for power inferred by Au and In power level normalization foils.
- b. Based on ^{140}Ba yield of 5.8%, reference 2.
- c. Based on ^{140}Ba yield of 6.0%, reference 2.
- d. Not corrected for neutron resonance self shielding, neutron scattering, or foil counting gamma-ray self shielding (attenuation). Correction factors for this latter effect are given in the 4th column of the table.
- e. The first parenthesized number is the estimated 2σ percent counting precision and the second bracketed number is the time (in days) from reactor shutdown to the time of count.

Corrections for neutron scattering and absorption effects have not been made for any of the foil reactions, Table 8. For most reactions these corrections are expected to be small (<5%). In addition to the $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ and $^{238}\text{U}(n,\gamma)^{239}\text{U}$ reactions, another exception is the $^{59}\text{Co}(n,\gamma)^{60}\text{Co}$ reaction for which the three 0.5 mil sandwiched foils, Tables 1 and 8, show a much larger effect. Preliminary SAND-II calculations based on an assumed isotropic flux, the individual foil thicknesses of Table 1, and using only absorption cross sections show less than ~1% neutron attenuation (self absorption) for all reactions including $^{197}\text{Au}(n,\gamma)$ and $^{238}\text{U}(n,\gamma)$. The exception is $^{59}\text{Co}(n,\gamma)^{60}\text{Co}$, with a calculated value of ~6% for an infinitely dilute foil sandwiched between two 0.5 mil foils. More exact calculations and experimental data on such effects will be required to obtain infinitely dilute reaction rates after the foil activation results of all of the participating laboratories have been documented, compared, and studied.

To provide an overall rough consistency check on the relative accuracy of all measured reaction rates, the average values given in the last column of Table 8 were used with the SAND-II calculated values of the CFRMF spectral averaged cross sections (reaction rates per unit total flux)⁽¹⁾ to obtain single foil total flux level estimates, Table 10. If there were no errors in the assumed shape of the CFRMF spectrum, the energy dependent SAND-II reaction cross sections, and measured reaction rates (after correcting for neutron scattering and absorption effects), then each foil would be expected to give the same value of total flux level. The preliminary results of Table 10 show an overall consistency check to within $\pm 19\%$. At this time, the best HEDL-I estimate of total flux is, perhaps, 1.2×10^{11} n/cm²-sec based on ^{235}U because of its overall flat energy response and more reliable cross section.

Expected Achievements

Because of funding limitations, no further CFRMF radiochemistry foil irradiations are anticipated before May 1972. HEDL foil sets II, III, and IV have not yet been irradiated. Work will continue on the analysis of available test data and the definition of a test plan for the CFRMF radiochemistry foil irradiations of the high purity aluminum encapsulated fission foils being fabricated by ORNL.

TABLE HEDL-9
REACTION PRODUCT NUCLEAR DATA SUMMARY*

<u>Isotope</u>	<u>Energy in keV</u>	<u>Gamma per Disintegration</u>	<u>Half Life in Days</u>	<u>Percent Accuracy of Calibration (95% Confi- dence Level)</u>
^{115m} In	336.25	0.450	0.1850	4
²⁴ Na	1368.53	1.00	0.6250 ±0.0017	2
	2275.41	0.9985		2
⁴⁶ Sc	889.26	1.00	84.0	2
	1120.51	0.996		2
⁴⁷ Sc	159.39	0.6875	3.43 ±0.03	3
⁴⁸ Sc	983.5	1.00	1.83	4
	1037.6	0.978		4
	1311.8	1.00		2
⁶⁴ Cu	1345.76	0.0048	0.5350	3
	511.0 γ [±]	0.37		3
¹⁹⁸ Au	411.79	0.9549	2.697 ±0.005	2
⁵⁴ Mn	834.78	0.9998	312.6 ±0.3	2
⁵⁹ Fe	1099.22	0.565	44.6 ±0.4	3
	1291.58	0.435		2
⁵⁸ Co	810.7	0.9944	71.3 ±0.2	3
⁶⁰ Co	1173.2	0.9986	1921.2 ±3.7	2
	1332.5	0.9999		2
¹⁴⁰ Ba	1595.4 (¹⁴⁰ La)	1.1058	12.80 ±0.05	2
¹⁴⁰ La	1595.4	0.965	1.6779 ±0.0021	2
²³⁹ Np	106.4	0.228	2.35	2
	228.2	0.121		
	277.9	0.14		

* Reference: Primarily, the Nuclear Data Tables. Atomic weights for calculation of reaction rates are based on the International Atomic Weights referenced to C-12. Avogadro's number used was 6.022169×10^{23} . Abundances are primarily taken from Nuclear Data Sheets.

TABLE HEDL-10
SINGLE FOIL TOTAL FLUX LEVEL
ESTIMATES FROM THE HEDL-I FOIL SET

Reaction	Measured ^a Reaction Rate	Spectral ^b Averaged Cross Section $\bar{\sigma}$ (cm ²)	Nominal 5% ^b Activity Limits (MeV)		10 ⁻¹¹ x Total Flux Level (n/cm ² -sec)
			Lower	Upper	
²³⁵ U(n,f) ¹⁴⁰ Ba	2.027 -13 ^e	1.695 -24	3.6 -4	2.4	1.196
²³⁸ U(n,f) ¹⁴⁰ Ba	9.484 -15	6.797 -26	1.4	7.4	1.395
²³⁸ U(n,γ) ²³⁹ U	2.380 -14 ^d	2.428 -25	8.4 -5	1.0	0.980
⁴⁵ Sc(n,γ) ⁴⁶ Sc	2.914 -15	2.360 -26	6.3 -4	0.8	1.235
⁵⁸ Fe(n,γ) ⁵⁹ Fe	7.486 -16	9.283 -27	3.2 -4	0.9	0.806
⁶³ Cu(n,γ) ⁶⁴ Cu	5.448 -15	5.965 -26	5.8 -4	0.6	0.913
⁵⁹ Co(n,γ) ⁶⁰ Co	9.232 -15	1.019 -25	1.2 -4	0.2	0.906
¹⁹⁷ Au(n,γ) ¹⁹⁸ Au	5.272 -14	5.4 -25 ⁱ	1.6 -4	0.5	0.976
²⁷ Al(n,α) ²⁴ Na	1.996 -17	2.484 -28	6.5	12	0.804
⁴⁶ Ti(n,p) ⁴⁶ Sc	3.339 -16	3.170 -27	3.5	9.7	1.053
⁴⁷ Ti(n,p) ⁴⁷ Sc	5.565 -16	4.029 -27	2.1	7.6	1.381
⁴⁸ Ti(n,p) ⁴⁸ Sc	8.425 -18	9.032 -29	6.7	13	0.933
⁵⁴ Fe(n,p) ⁵⁴ Mn	2.288 -15	1.959 -26	2.4	8.3	1.168
⁵⁸ Ni(n,p) ⁵⁸ Co	3.017 -15	2.535 -26	2.2	8.0	1.190
¹¹⁵ In(n,n') ^{115m} In	6.701 -15	4.750 -26	0.8	6.2	1.411

Average Value 1.090(±19% 1σ)
(±4.9%, 1
std. error)

- a. At a stated reactor power of 9.75 kW.⁽⁵⁾ Units of reactions per second per nuclei from Table 8.
- b. Based on the ANC derived flux spectrum, Figure HEDL-1, reference 1, and the SAND-II evaluated energy dependent cross sections,⁽⁹⁾ see Table HEDL-4 of reference 1.
- c. 2.027 - 13 = 2.027 x 10⁻¹³.
- d. No corrections have been made for neutron scattering from adjacent foils in the foil set or for resonance self shielding in this or any of the other (n,γ) reactions. The corrections are estimated to be small, less than ~5%, except for Co -- see Table 8.
- i. From Table HEDL-6, reference 2.

D. REACTION CROSS SECTIONS

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

Objectives

- Establish and maintain an updated set of evaluated energy dependent cross sections for foil reactions used by the FRMDC for routine neutron dosimetry. Establish better consistency and accuracy for these cross sections in order to achieve the long range objective of 5 to 10% accuracy (at the 95% confidence level) for measured integral flux spectra and fluence values for LMFBR fuels and materials development programs.

Summary

Comparison of Paulsen's and Widera's⁽¹⁰⁾ recent energy dependent activation measurements of the $^{58}\text{Ni}(n,p)^{58}\text{Co}$ and $^{54}\text{Fe}(n,p)^{54}\text{Mn}$ cross section in the 1 to 6 MeV region with those derived on the basis of SAND-II analysis of measured reaction rates in a number of fast reactor spectra provides information to help establish the absolute magnitude of these as well as of high energy fission cross sections relative to the hydrogen (n,p) scattering cross section.

Accomplishments and Status

SAND-II determination of flux-spectra and fluence from a set of measured reaction rates requires the use of a consistent set of evaluated energy-dependent cross sections $\sigma(E)$.⁽¹¹⁾

A reasonably consistent set of $\sigma(E)$ values has been developed and is in current use for the SAND-II code,⁽⁹⁾ which is used by HEDL for routine fast reactor dosimetry and will be used to provide multiple foil flux spectral data for the ILRR program.⁽¹¹⁾ Better consistency and accuracy for these cross sections are required. Present accuracy is in the 20 to 40% range at the 95% confidence level.⁽⁹⁻¹³⁾ This accuracy can be improved if the sources of existing inconsistencies between calculated integral averaged cross sections $\bar{\sigma}_c$ using the current evaluated energy dependent cross sections $\sigma(E)$ and measured values $\bar{\sigma}_m$ can be identified and effectively eliminated - see the references listed in the previous Quarterly Reports.^(1,2)

$^{58}\text{Ni}(n,p)^{58}\text{Co}$ and $^{54}\text{Fe}(n,p)^{54}\text{Mn}$ Cross Sections

Study of the $^{58}\text{Ni}(n,p)$ and $^{54}\text{Fe}(n,p)$ evaluated energy dependent cross

sections is aimed at understanding and reducing existing inconsistencies between measured and calculated fission and non-fission reaction rates in fast neutron spectra, such as the Fission, Godiva, and EBR-II spectra. The method of evaluating energy dependent cross sections to establish consistency with integral measurements in different neutron spectra by the SAND-II unfolding technique has been described elsewhere.^(9,12,13)

In this application, the effect of flux-spectral uncertainties was minimized by unfolding (readjusting) the cross sections of interest relative to a selected set of standard cross sections. This was accomplished for $^{58}\text{Ni}(n,p)$ and $^{54}\text{Fe}(n,p)$ by selecting the best estimate (based on spectrometry and calculations) of five spectra [the Fission, APFA-III Godiva, EBR-II Core Center, and ECEL Cores 14 and 16 (similar to the FTR core spectrum)]. The best estimates were then readjusted in magnitude and shape by SAND-II unfolding using four standard reference reaction cross sections [$^{238}\text{U}(n,f)$, $^{235}\text{U}(n,f)$, $^{237}\text{Np}(n,f)$, and $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$] and the corresponding absolute values of measured reaction rates for each reaction in the five spectra. This established an integrally consistent set of five reference flux-spectra relative to the five reference cross sections. These spectra were then used with the measured absolute reaction rates for Ni and Fe (obtained by measurements in the same five spectra) as input to the SAND-II code to unfold (readjust) a best input estimate of the energy dependent Ni and Fe cross sections.

By this procedure, the best input Ni and Fe cross sections were readjusted by the SAND-II procedure relative to the $^{235}\text{U}(n,f)$, $^{238}\text{U}(n,f)$, $^{237}\text{Np}(n,f)$, and $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$ reference set of cross sections - with the effect of uncertainties in the five spectra having only an indirect effect. Further, because this is an integral approach, the effect of fine structure (both known and unknown) in both the reference cross sections and the unfolded Ni and Fe cross sections should have little effect on the setting of the relative magnitudes of these cross sections. For mono-energetic measurements, however, such fine structure could cause scatter between different sets of measurements and could be a more serious problem in fixing overall absolute or relative evaluated cross section scales.

The SAND-II evaluated energy dependent cross sections for $^{54}\text{Fe}(n,p)$ and $^{58}\text{Ni}(n,p)$ are shown in Figures 7 and 8, respectively, as taken from references 14 and 9. For $^{54}\text{Fe}(n,p)$, the solid line (representing the SAND-II derived

Figure HEDL-7 SAND-II Solution for $^{54}\text{Fe}(n,p)^{54}\text{Mn}$ Reaction Cross Section

Figure HEDL-8 SAND-II Solution for $^{58}\text{Ni}(n,p)^{58}\text{Co}$ Reaction Cross Section

group averaged cross sections⁽⁹⁾ is the readjusted $\sigma(E)$. The $\sigma(E)$ used as a best input approximation is specified in the figure.⁽¹⁵⁾ The dashed curve is a solution using a constant input. Fairly unique definition of the cross section is implied by the similarity in the solutions based on the constant and best input forms - which is an important test of the adequacy of the combined neutron energy coverage of the five reference flux-spectra used in the study.

In Figure 8, similar results are presented for $^{58}\text{Ni}(n,p)$, but here the SAND-II derived group averaged values are individually shown so that a more careful and direct comparison can be made with the detailed energy dependent activation measurements of Paulsen and Widera.⁽¹⁰⁾ These measurements are shown for both $^{54}\text{Fe}(n,p)$ and $^{58}\text{Ni}(n,p)$ by the solid points with error bars in Figures 7 and 8,, respectively. Allowing for fine structure in the real cross sections, the agreement between Paulsen and Widera's mono-energetic results and the SAND-II derived cross sections is very good for the $^{54}\text{Fe}(n,p)$ and the $^{58}\text{Ni}(n,p)$ reactions in the region of maximum response (below ~4 to 6 MeV) for the five spectra used in this study. For $^{58}\text{Ni}(n,p)$, the agreement is most striking, and certainly may be somewhat fortuitous considering present uncertainties.

There is a fundamental significance of such a comparison in the study and determination of the absolute magnitude of high energy fission cross sections. This significance is based on the observation that Paulsen and Widera's mono-energetic data are derived from absolute activation measurements (reaction rates) and absolute neutron fluence measurements using a proton recoil telescope counter. Consequently, their mono-energetic data are established relative to the high energy hydrogen scattering cross section, while the SAND-II evaluated $\sigma(E)$'s are established relative to the high energy fission cross sections of ^{235}U , ^{238}U , ^{237}Np , and the (n,α) cross section of ^{27}Al . Comparison of these two sets of data, therefore, should help to determine if the current SAND-II set (or for that matter, any other set) of evaluated fission cross sections for ^{235}U , ^{238}U , and ^{237}Np * are at a proper barn scale level

* Here we note that for the energy range of interest, the current SAND-II fission cross sections are basically those of Davey's.⁽¹⁶⁾

relative to the hydrogen scattering cross section at high energies - that is, if there is no significant laboratory-to-laboratory bias or uncertainty in the absolute measurements of the ^{58}Co and ^{54}Mn activation rates of Paulsen's and Widera's and those used for the SAND-II analysis.*

With existing uncertainties, positive conclusions cannot be reached from the present study, but future studies of integral reaction rates and ratio measurements, particularly from the ILRR program, coupled with results such as those reported by Paulsen and Widera, should provide new information to cross section evaluators to help establish the absolute magnitude of high energy fission cross sections for ^{235}U , ^{238}U , and ^{237}Np . This will be particularly important for fast reactor dosimetry applications involving fuels and materials tests in EBR-II and, later FTR.

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

Expected Achievements

A Task Group is being established through the Normalization and Standards Subcommittee of CSEWG to develop an "ENDF/B or A File for Dosimetry Applications." A meeting is being planned for late March 1972 to consider an initial set of ~20 evaluated cross sections for this data file. This has come about after the status of the HEDL effort to improve the SAND-II evaluated energy dependent cross sections and the comparisons made by the NNCSC of these cross sections with ENDF/B files were discussed at the December 1-2, 1971, CSEWG meeting at Brookhaven National Laboratory.

* Because of previous National and International round robins on the counting of ^{58}Co and ^{54}Mn , the laboratory-to-laboratory uncertainty and bias are expected to be small.

E. MULTIPLE FOIL TECHNIQUE ERROR ANALYSIS

W. N. McElroy

Objective

- Develop, apply, and test appropriate analytical and numerical methods for the assignment of errors for the multiple foil technique of flux-spectral definition and cross section evaluation.

Summary

An error analysis study for the ^{235}U fission spectrum as determined by the SAND-II code using an evaluated set of measured reaction rates (spectral averaged cross sections) provided by A. Fabry⁽¹⁷⁾ was completed. His cross sections are a first attempt to establish an accepted set of integral reaction rates and ratios as a CSEWG benchmark for the ^{235}U fission spectrum.

Accomplishments and Status

A SAND-II Monte Carlo Error Analysis Code has been developed⁽¹⁸⁾ and is being used and tested for the assignment of errors to multiple foil flux-spectra, re-evaluated energy-dependent cross sections, and empirically derived damage functions.⁽¹¹⁾ Error analysis results are provided here for the ^{235}U fission spectrum. Results of an earlier study have been reported.⁽¹⁹⁾ Reference is made to the first Quarterly Report, reference 1,, and reference 18 for detailed information on the Monte Carlo procedure and the SAND-II evaluated cross section error assignments. Table 11 provides an updated listing of the cross section error assignments used for this study. Table 12 lists the values of a recently evaluated set of measured reaction rates per unit flux (spectral averaged cross sections) and the corresponding assigned errors (1σ) prepared by A. Fabry.^{(17)*}

The Cranberg⁽²⁰⁾ form was assumed to be the best current approximation to the true fission spectrum. It together with a selected set of 16 measured reaction rates per unit flux, Table 12, and the listed errors, Table 11, were input

* A. Fabry developed this set of integral data for submission to the next EURATOM WGRD meeting and for submission to the CSEWG Data Testing Subcommittee for consideration as a possible benchmark for the ^{235}U fission spectrum.

TABLE HEDL-11
SAND-II EVALUATED CROSS SECTION ERROR ASSIGNMENT ^a

Reaction	1-162 ^b	162-226	226-361	361-406	406-440	440-455	455-463	463-471	471-481	481-491	491-501	501-521	521-551	551-571	571-621
	1-10-4-7 ^c	4-7-1-5	1-5-1-2	1-2-1-1	1-1-6-1	6-1-1.4	1.4-2.2	2.2-3.0	3.0-4.0	4.0-5.0	5.0-6.0	6.0-8.0	8.0-11.0	11.0-13.0	13.0-18.0
	(%)	(%)	(%)	(%)	(%)	(%)	(%)	(%)	(%)	(%)	(%)	(%)	(%)	(%)	(%)
⁵⁸ Ni(n,p)	0	0	0	0	0	250	30	10	10	5	5	4	6	10	10
⁵⁴ Fe(n,p)	0	0	0	0	1000	500	50	10	10	8	8	7	5	10	10
²⁷ Al(n,α)	0	0	0	0	0	0	0	0	0	30	20	6	6	10	10
⁵⁹ Co(n,α)	0	0	0	0	0	0	0	0	0	1000	15	8	8	10	10
⁶⁰ Ni(n,p)	0	0	0	0	0	0	1000	500	250	50	15	8	8	8	8
⁴⁵ Sc(n,γ)	2	5	5	6	10	10	11	11	11	11	11	11	11	11	11
²³² Th(n,γ)	1.4	5	5	5	5	8	8	8	8	8	8	8	8	8	8
²³⁷ Np(n,f)	16	10	10	10	20	5	3	2	5	5	5	10	10	10	10
²³⁹ Pu(n,f)	0.5	8	8	8	7	6	5	5	6	6	6	6	6	10	10
²³² Th(n,f)	0	0	0	0	0	100	25	20	10	10	10	10	10	10	10
²³⁵ U(n,f)	2	8	8	8	7	6	5	3	6	6	6	6	6	10	10
²³⁸ U(n,γ)	1.5	5	5	5	5	5	5	5	5	5	5	5	5	10	10
²³⁸ U(n,f)	0	0	0	0	1000	30	4	2	3	3	3	4	4	10	10
¹¹⁵ In(n,γ)	2.5	5	5	5	5	10	17	17	17	17	17	17	17	17	17
⁵⁵ Mn(n,γ)	0.8	5	5	5	5	5	5	8	8	8	8	8	10	10	10
⁶⁴ Zn(n,p)	0	0	0	0	1000	750	100	50	15	15	15	15	15	15	15
³² S(n,p)	0	0	0	0	0	0	100	20	5	5	5	5	5	8	8
²⁸ Si(n,p)	0	0	0	0	0	0	0	0	0	250	50	15	15	15	15
³⁴ S(n,α)	0	0	0	0	0	0	0	0	0	1000	50	25	15	10	10
²⁴ Mg(n,p)	0	0	0	0	0	0	0	0	0	100	50	10	10	10	10
³⁵ Cl(n,α)	0	0	0	0	0	0	0	500	20	20	30	30	30	30	30
⁵⁸ Ni(n,2n)	0	0	0	0	0	0	0	0	0	0	0	0	0	30	20
¹²⁷ I(n,2n)	0	0	0	0	0	0	0	0	0	0	0	0	30	30	15
⁹⁰ Zr(n,2n)	0	0	0	0	0	0	0	0	0	0	0	0	0	30	15
⁶³ Cu(n,2n)	0	0	0	0	0	0	0	0	0	0	0	0	0	8	8
³¹ P(n,p)	0	0	0	0	0	0	50	6	12	12	12	10	10	10	10
⁵⁶ Fe(n,p)	0	0	0	0	0	0	0	0	8	8	8	6	6	15	15
²⁷ Al(n,p)	0	0	0	0	0	0	0	50	10	8	8	8	8	20	20
¹¹⁵ In(n,n')	0	0	0	0	250	20	10	10	10	5	5	5	5	10	10
²³⁸ Pu(n,f)	10	10	10	10	12	10	10	10	10	10	10	10	10	10	10
⁵⁸ Fe(n,γ)	8	8	28	12	12	12	12	12	12	12	12	12	12	12	12
⁶³ Cu(n,α)	1000	1000	1000	1000	1000	1000	500	400	200	50	25	8	8	10	10
²³ Na(n,γ)	1	5	5	5	5	5	5	5	5	5	5	5	5	10	10
¹⁹⁷ Au(n,γ)	0.5	4	5	5	5	5	5	5	5	5	5	5	5	7	7
⁵⁹ Co(n,γ)	4	5	5	5	6	7	7	7	7	7	7	7	7	10	10
⁶³ Cu(n,γ)	5	5	5	5	5	5	5	5	7	7	7	7	7	7	7
⁴⁶ Ti(n,p)	0	0	0	0	0	0	1000	150	25	20	20	10	10	20	20
⁴⁷ Ti(n,p)	0	0	0	0	1000	500	100	50	15	15	15	15	15	15	15
⁴⁸ Ti(n,p)	0	0	0	0	0	0	0	0	1000	200	25	15	15	15	15

a. One standard deviation in percent.

b. SAND-II group numbers.

c. SAND-II group energy bounds. Note: 1⁻¹⁰ = 1x10⁻¹⁰, etc.

TABLE HEDL-12
 ^{235}U FISSION SPECTRUM AVERAGED
 CROSS SECTION EVALUATION OF A. FABRY (17)

Reaction (i)	$\bar{\sigma}(\chi_{25}, i)$ Spectral Averaged Cross Section (mb)
$^{197}\text{Au}(n, \gamma)^{198}\text{Au}$	88 ($\pm 5.1\%$)
$^{115}\text{In}(n, \gamma)^{116\text{m}}\text{In}$	146 ($\pm 3.4\%$)
$^{63}\text{Cu}(n, \gamma)^{64}\text{Cu}$	10.1 ($\pm 15.0\%$)
$^{235}\text{U}(n, f)\text{F.P.}$	1250 ^a
$^{239}\text{Pu}(n, f)\text{F.P.}$	1859 ($\pm 3.2\%$)
$^{237}\text{Np}(n, f)\text{F.P.}$	1370 ($\pm 5.5\%$)
$^{232}\text{Th}(n, f)\text{F.P.}$	83 ^d ($\pm 4.2\%$)
$^{238}\text{U}(n, f)\text{F.P.}$	328 ($\pm 3.0\%$)
$^{115}\text{In}(n, n')^{115\text{m}}\text{In}^c$	188 ($\pm 2.1\%$)
$^{31}\text{P}(n, p)^{31}\text{Si}$	36 ($\pm 5.6\%$)
$^{32}\text{S}(n, p)^{32}\text{P}$	69 ($\pm 2.9\%$)
$^{58}\text{Ni}(n, p)^{58}\text{Co}$	113 ($\pm 2.2\%$)
$^{58}\text{Ni}(n, p)^{58\text{m}}\text{Co}$	35.4 ($\pm 2.8\%$)
$^{54}\text{Fe}(n, p)^{54}\text{Mn}$	82.5 ($\pm 2.4\%$)
$^{64}\text{Zn}(n, p)^{64}\text{Cu}$	31 ($\pm 4.8\%$)
$^{27}\text{Al}(n, p)^{27}\text{Mg}$	4.0 ($\pm 10.0\%$)
$^{46}\text{Ti}(n, p)^{46}\text{Sc}$	12.3 ($\pm 4.1\%$)
$^{47}\text{Ti}(n, p)^{47}\text{Sc}$	20 ($\pm 10.0\%$)
$^{48}\text{Ti}(n, p)^{48}\text{Sc}$	0.315 ($\pm 6.3\%$)
$^{92}\text{Mo}(n, p)^{92\text{m}}\text{Nb}$	7.0 ($\pm 5.7\%$)
$^{56}\text{Fe}(n, p)^{56}\text{Mn}$	1.07 ($\pm 5.6\%$)
$^{24}\text{Mg}(n, p)^{24}\text{Na}$	1.53 ($\pm 2.0\%$)
$^{27}\text{Al}(n, \alpha)^{24}\text{Na}$	0.725 ($\pm 2.8\%$)
$^{63}\text{Cu}(n, \alpha)^{60}\text{Co}$	0.50 ($\pm 10.0\%$)
$^{59}\text{Co}(n, \alpha)^{56}\text{Mn}$	0.156 ($\pm 3.8\%$)
$^{63}\text{Cu}(n, 2n)^{62}\text{Cu}$	0.124 ($\pm 7.3\%$)
$^{93}\text{Nb}(n, 2n)^{92\text{m}}\text{Nb}$	0.47 ($\pm 6.4\%$)
$^{55}\text{Mn}(n, 2n)^{54}\text{Mn}$	0.253 ($\pm 4.0\%$)
$^{127}\text{I}(n, 2n)^{126}\text{I}$	1.09 ($\pm 4.6\%$)

- a. Reference reaction for assessment of the absolute cross section scale; if the true cross section is different from 1250 mb, all integral data would have to be normalized accordingly. The 1 σ errors of all other reactions are given relative to this value.
- b. All the experimental integral data below this dashed line might be affected in an inversely proportional way by any deviation of the integral ratio $\bar{\sigma}_f(\chi_{25}, ^{235}\text{U})/\bar{\sigma}_f(\chi_{25}, ^{238}\text{U})$ from the accepted value of 3.81 ± 0.15 ; the significance of such possible deviations for the data above the line is not clear.
- c. Gamma ray intensity of the 335 keV isometric transition fixed to 46%.
- d. Weighted mean value between the re-normalized result of Fabry et al.; i.e., 82 ± 3 mb, and the McElroy et al. retrieved Godiva measurement of 85.2 ± 7.7 mb (^{99}Mo counting).

to the SAND-II Monte Carlo code to derive a re-adjusted multiple foil representation of the ^{235}U fission spectrum, Figure 9, with assigned errors due to uncertainties in measured reaction rates and SAND-II evaluated cross sections.

The selected set of 16 reactions together with the ratios of measured to calculated reaction rates for the SAND-II 50th iteration solution are given in Table 13. The mean energy of this solution is 2.12 MeV as compared with 1.93 MeV for the Cranberg input form. The deviation of these ratios from unity is an indication of the difficulty of finding a solution spectrum that will exactly reproduce all of the measured values with the existing SAND-II evaluated energy dependent cross sections.* It should be emphasized that it is a difference of only about 15% to 20% between the measured and calculated (using the Cranberg form of the fission spectrum) ratios of the ^{235}U and ^{238}U fission rates that is primarily responsible for the harder form of the fission spectrum from activation data.

The multiple foil derived form of the fission spectrum of this study, Figure 9, is qualitatively the same as that given in references 19 and 21, but with a lower mean energy of 2.12 ± 0.07 (2σ) MeV as compared with previous values of 2.14 to 2.24 MeV.

Another difference from reference 21, but also observed in reference 19, is the appearance of a dip in the spectrum just below 1 MeV. The shaded bars in Figure 9, which designate the 90% energy response ranges for those detectors primarily responsible for this structure, show excellent foil coverage at the dip. However, although the multiple foil technique can detect the existence of such structure, uncertainties in reaction rates and reaction cross section are too large for precise shape definition. In a recent review paper, Grundl⁽²²⁾ brought attention to such structure in the photo-plate and time-of-flight measurements of Cranberg,⁽²⁰⁾ Rosen,⁽²³⁾ and Barnard.⁽²⁴⁾ Also, Zamyatin, et al.⁽²⁵⁾ have reported on the observation of structure in fission spectra below ~ 1 MeV. Using Grundl's⁽²²⁾ presentation of the differential experimental results, Figure 9, curves have been drawn through the data of Cranberg and Rosen to show this structure. It is apparent that the multiple

* The current SAND-II evaluated cross sections correspond most closely to those of Davey's⁽¹⁶⁾ for the fission reactions.

TABLE HEDL-13

RATIO OF MEASURED TO CALCULATED SPECTRAL AVERAGED CROSS SECTIONS FOR
THE 50th ITERATION SAND-II FISSION SPECTRUM SOLUTION
(2.12 MeV MEAN ENERGY)

Reaction	$\bar{\sigma}$ -Measured/ $\bar{\sigma}$ -Calculated ^a
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	0.9922
$^{235}\text{U}(n,f)\text{F.P.}$	1.0018
$^{239}\text{Pu}(n,f)\text{F.P.}$	1.0268
$^{237}\text{Np}(n,f)\text{F.P.}$	0.9935
$^{232}\text{Th}(n,f)\text{F.P.}$	1.0488
$^{238}\text{U}(n,f)\text{F.P.}$	1.0292
$^{115}\text{In}(n,n')^{115\text{m}}\text{In}$	0.9160
$^{31}\text{P}(n,p)^{31}\text{Si}$	0.9786
$^{32}\text{S}(n,p)^{32}\text{P}$	1.0200
$^{58}\text{Ni}(n,p)^{58}\text{Co}$	1.0060
$^{54}\text{Fe}(n,p)^{54}\text{Mn}$	0.9919
$^{27}\text{Al}(n,p)^{27}\text{Mg}$	1.0045
$^{56}\text{Fe}(n,p)^{56}\text{Mn}$	0.9840
$^{24}\text{Mg}(n,p)^{24}\text{Na}$	0.9837
$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	1.0238
$^{63}\text{Cu}(n,2n)^{62}\text{Cu}$	0.9992

Standard Deviation of All Measured Ratios = 2.96%

a. The measured values are those evaluated by Fabry, Table 12.

Figure HEDL-9 Fission Spectrum Comparisons

foil results support the previous spectrometer measurements of the possible existence of structure in the peak of the fission spectrum.

Such structure could contribute, but only in a secondary manner, to existing discrepancies between activation and spectrometer results because of possible structure in fission and non-fission reaction cross sections in this same region. In this same context, another contributing factor to inconsistencies for some detector reactions may be associated with subthreshold cross sections for exoergic reactions,^(12,13) as discussed in the previous Quarterly Report, reference 2.

The absolute uncertainty associated with the multiple foil results is shown by the shaded area in Figure 9. Uncertainties assigned to the activation measurements and the evaluated cross sections for the SAND-II Monte Carlo analysis are again those given in Table 12 and Table 11, respectively. Group averaged uncertainties from the same analysis are compared in Table 14 with values previously reported by Grundl⁽²⁶⁾ but based on eight of the listed reactions, Table 12. The present work, column 4 of Table 14, provides confirmation of Grundl's earlier findings and error assignments.

The effect of changes in SAND-II evaluated cross sections since the last study⁽²¹⁾ is shown in Table 15 by a comparison of previous with present calculated values of fission averaged cross sections using the Cranberg fission spectrum. The set of average measured values from Table 12 and the ratio to calculated values are also shown to illustrate the dilemma that exists in establishing agreement between measurement and calculation.

In reference 27, Fabry studied the fission spectrum problem with a view towards establishing better consistency between integral and differential data by effecting rather significant changes in evaluated energy dependent cross sections. For instance, for $^{238}\text{U}(n,f)$, he shifts all of Lamphere's⁽²⁸⁾ points about 100 keV towards lower energies. In addition, he imposes the constraint of a 3-parameter fit (in a least squares sense) for a more complicated, but theoretically based, analytical expression for the fission spectrum which considers scission neutrons. Using a selected set of eleven of the reaction rates given in Table 12, he then derives an activation foil form of the fission spectrum that is in much better agreement with the Watt form for energies below ~11 MeV.

TABLE HEDL-14
FIVE-GROUP REPRESENTATION OF
OBSERVATIONS OF THE ^{235}U FISSION-NEUTRON SPECTRUM

Energy Groups (MeV)	Maxwellian Description	Activation Detectors	
		Grundl ⁽²⁾ (8 Reactions)	Present Work (16 Reactions)
0 - 1.4	0.4621	0.373($\pm 22\%$) ^a	0.386($\pm 21\%$) ^a
1.4- 3.0	0.3386	0.360($\pm 21\%$)	0.382($\pm 13\%$)
3.0- 6.0	0.1737	0.241($\pm 28\%$)	0.209($\pm 9\%$)
6.0-11.0	0.0249	0.0257($\pm 11\%$)	0.0234($\pm 13\%$)
11.0- ∞	0.00070	0.00069($\pm 18\%$)	0.00074($\pm 16\%$)
Average Energy	1.935 MeV	2.20 ± 0.16 MeV	2.12 ± 0.07 MeV

^a. At the 95% confidence level.

TABLE HEDL-15
COMPARISON OF MEASURED ($\bar{\sigma}_m$) AND CALCULATED ($\bar{\sigma}_c$)
SPECTRAL AVERAGED CROSS SECTIONS FOR THE FISSION SPECTRUM

Reaction	$\bar{\sigma}_m(\text{mb})^a$	$\bar{\sigma}_c(\text{mb})^b$	$\bar{\sigma}_c(\text{mb})^c$	$(\bar{\sigma}_m/\bar{\sigma}_c)^d$
$^{235}\text{U}(n,f)\text{F.P.}$	1250	1233	1259	1.01
$^{237}\text{Np}(n,f)\text{F.P.}$	1370	1276	1269	1.07
$^{238}\text{U}(n,f)\text{F.P.}$	328	278	277	1.18
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	88	102	101	0.86
$^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$	10.1	11.1	11.5	0.91
$^{115}\text{In}(n,\gamma)^{115m}\text{In}$	188	180	178	1.04
$^{32}\text{S}(n,p)^{32}\text{P}$	69	58.2	57.3	1.19
$^{27}\text{Al}(n,p)^{27}\text{Mg}$	4.0	3.73	3.66	1.07
$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	0.725	0.696	0.683	1.04
$^{56}\text{Fe}(n,p)^{56}\text{Mn}$	1.07	1.10	1.08	0.97
$^{31}\text{P}(n,p)^{31}\text{Si}$	36	31.6	31.1	1.14
$^{63}\text{Cu}(n,2n)^{64}\text{Cu}$	0.124	0.118	0.116	1.05

^a. From Table 12.

^b. Calculated values using the Cranberg form of the fission spectrum and the current SAND-II evaluated $\sigma(E)$ library tape⁽⁹⁾ but with subsequent updating for $^{197}\text{Au}(n,\gamma)$.

^c. Taken from reference 21, same calculation as (b) above, but using previous SAND-II evaluated cross section library tape.

^d. Columns 2 and 3.

Qualitatively, this result is not inconsistent with the results of Grundl's and the present study. That is, it shows how changes in energy dependent cross sections and the acceptance of some distortion in the fission spectrum shape (resulting from the use of a three parameter analytical representation of the fission spectrum) may, at least partially, reconcile conflicting integral and differential data. Fabry's use of the $^{115}\text{In}(n,\gamma)$ reaction, omission of the $^{31}\text{P}(n,p)$ reaction, and use of different evaluated energy dependent cross sections for the other as well as the $^{238}\text{U}(n,f)$ reaction does not permit a direct quantitative comparison of his results with those of the present study.

In conclusion, and as stated elsewhere,^(11,19,21) inconsistencies between integral and differential quantities continue to cast doubt on the reliability of (1) the fission spectral form, (2) evaluated activation reaction cross section data, and (3) the multiple foil method. Previous studies⁽¹⁹⁾ indicate that measurements of reaction rates and lack of solution uniqueness are unlikely sources of significant error for the multiple foil method. This suggests that uncertainties in evaluated energy-dependent reaction cross sections are the major source of error for the multiple foil technique.

Relative to (1) and (2), above, the use of a larger number of foil reactions and updated evaluated cross sections⁽⁹⁾ reduced the mean energy of the SAND-II derived multiple foil form of the fission spectrum from 2.24⁽²¹⁾ to 2.12 (± 0.07) MeV. However, the relative uncertainties of one cross section to another for the larger set of reactions used in this study are not as firmly established as those used by Grundl in defining a fission spectrum with a mean energy of $2.2 \pm .16$ MeV^(26,29) by the activation technique. The values of ± 2.1 to 2.2 MeV based on integral measurements are to be compared with values of 1.93 to 2.00 MeV based on analysis of differential measurements by Cranberg,⁽²⁰⁾ Frye,⁽³⁰⁾ Watt,⁽³¹⁾ and others.

Expected Achievements

No further error analysis is planned for the ^{235}U fission spectrum until new values of reaction rates have been determined as part of the ILRR program.

A. Fabry's evaluated set of ^{235}U fission spectral averaged cross sections (with his documentation of the sources of experimental data) is expected to be submitted to the CSEWG Data Testing Subcommittee before the end of the next reporting period.

F. BACKGROUND INFORMATION ON THE CEN-SCK SECONDARY
INTERMEDIATE-ENERGY STANDARD NEUTRON SPECTRUM
FACILITY $\Sigma\Sigma$

A. Fabry^a

Objectives

- Provide measured integral reaction rate data for SAND-II Monte Carlo error analysis of the $\Sigma\Sigma$ central neutron flux spectrum for comparison with differential neutron spectrometry data obtained jointly by CEN-SCK (Mol), GFK (Karlsruhe) and RCN (Petten).
- Present background documentation for the CEN-SCK secondary standard neutron spectrum facility $\Sigma\Sigma$ for consideration of an operative link between the Inter-laboratory LMFBR Reaction Rate program and the joint NBS/CEN-SCK efforts to develop and/or characterize permanent intermediate-energy standard and reference neutron fields for fast reactor applications.

Summary

This first report summarizes the most relevant features of the secondary intermediate-energy standard neutron spectrum facility $\Sigma\Sigma$, presents preliminary CEN-SCK neutron spectrometry data, and outlines the integral reaction rate measurements performed thus far within the central exposure volume.

Accomplishments and Status

The secondary standard neutron spectrum facility $\Sigma\Sigma$ is a permanent, clean and reproducible, reference fast neutron source assembly of low power, characterized by a fast-breeder-like radiation field; it has been designed⁽³²⁾ and can⁽³³⁾ be depicted within the framework of one-dimensional neutron transport theory. Carbon, boron, and natural uranium are its main constitutive materials, and it is implemented within a conventional graphite thermal column.

The $\Sigma\Sigma$ facility was proposed by the CEN-SCK laboratories at Mol, Belgium, as a straightforward tool for the interlaboratory comparison and standardization of a number of reactor reaction rate, neutron spectrometry and dosimetry techniques; it is also directed toward the subsequent validation of some

a. Work performed at CEN-SCK essentially by M. DeCoster, G. DeLeeuw-Gierts, S. DeLeeuw, and A. Fabry (Reactor Study Department).

fundamental nuclear data; e.g., is aimed as a benchmark for specific cross sections. Finally, it is considered a valuable step in developing the methodology and obtaining technical experience for the design and future application of primary intermediate-energy standard neutron fields,^(34,35) a program which is now in the scope of a direct cooperative effort between CEN-SCK and the National Bureau of Standards.

The $\Sigma\Sigma$ facility has been used with success to intercompare a number of differential neutron spectrometers in a triplicate framework involving GFK (Karlsruhe, Germany), RCN (Petten, The Netherlands), and CEN-SCK; a closely similar facility, called NISUS,⁽³⁶⁾ has been developed at the Imperial College of Science and Technology (London) for similar purposes within the UKAEA; the Institute for Atomic Physics, IFA, Bucarest, Romania, is also operating such systems and working on its characterization in close relationship with CEN-SCK.⁽³⁷⁾

It is believed that the $\Sigma\Sigma$ facility is of relevance for consideration and planning of some ILRR program irradiations during FY-73 and beyond, especially in view of the interlaboratory emphasis placed on measuring its central neutron spectrum by means of state-of-the-art techniques and of the planned comparison efforts in relation to reaction rates and dosimetry.

I. Facility Performance and Major Features

A schematic in-pile view of the facility is displayed in Figure 10, and its computed central neutron spectrum is compared in Figure 11 with the best current approximation* developed by ANC for the CFRMF central core neutron spectrum.⁽³⁸⁾ The design and performance of the $\Sigma\Sigma$ facility have been extensively discussed in various progress reports⁽³³⁾ and in a forthcoming publication. They may be summarized as follows:

A. Facility Characterization

50 cm diameter spherical cavity hollowed out of the protruding end of the graphite horizontal thermal column of BR-1 reactor, CEN-SCK.

B. Source of Fast Neutrons and Access Capability

- 110 kg natural uranium (metal) spherical shell of 50 ± 0.1 mm thickness and outer diameter of 245 ± 0.2 mm.

* Collapsed here in the FD-2 group structure.

Figure HEDL-10 In-Pile View of the $\Sigma\Sigma$ Facility

Figure HEDL-11 Comparison of the CFRMF and $\Sigma\Sigma$ Central Neutron Spectra

- The power output of the source is approximately 5 watts at a reactor power of 1 MW.

- The unclad source is supported by an aluminum holder fixed to a movable graphite shielding plug; it is currently removed along a roller path up to an elementary disassembling cell; the gamma dose rate at a 30 cm distance 15 minutes after a 2-hour irradiation at a 1 MW reactor power is typically 30 Rad/hour.

- In the intersection plane of the two source hemispheres, three different matching plugs may be used according to the application:

1. A full plug for central reaction rate measurements by means of activation foils and track recorders.

2. A two-piece plug with five special exposure notches for reaction rate traverses within the source (any angular orientation with respect to the thermal column axis may be selected by rotation of the source during assembly).

3. A plug with a right axial hole of 15.5 mm diameter for differential neutron spectrometry.

Another plug with an axial access hole of 5 mm diameter will be developed for application of the NBS fission chamber⁽³⁹⁾ and others.⁽⁴⁰⁾

C. Boron Carbide Inner Screen

- Screen Purpose - Shaping of the low energy tail of the neutron spectrum.⁽³³⁾

- Screen Composition - Vibro-compacted natural boron carbide powder with an effective density of 1.499 ± 0.005 g/cm³; * homogeneous moisture content ≤ 500 ppm H₂O (computed effect of 500 ppm on the neutron spectrum: <1% in all energy groups).

* This is the final value as obtained from various independent measurements performed at CEN-SCK and at CBNM (Euratom, Geel) for up to 12 different hemispherical shells, some with access holes, some without; the error is a conservative estimate of the absolute uncertainty, but the relative dispersion for all 12 shells is less than ± 0.002 ; the quoted result never departs from the nominal expected value by more than +1.4%.

- Cladding - 1 mm aluminum 99.5%.
- Boron Carbide Thickness - 15.0 ± 0.1 mm.
- Access Hole - Shells without a hole and shells with two hole sizes, 17 mm and 27 mm diameter, respectively, are available; a shell with a hole diameter of 5 mm is considered for application of the NBS fission chamber and others.
- Inner Screen Diameter - 110 ± 0.1 mm.

D. Central Exposure Volume

The diameter of the central exposure sphere is 110 mm; however, flux and spectrum variations of the order of 10 to 15% exist due to the driving thermal neutron flux gradients and to the presence of the strong absorber screen. This restricts the useful exposure volume to a central sphere of approximately 4 cm diameter, which has been found quite satisfactory in practice. For activation foil applications, the exposures are generally done in the part of the central vertical plane intersected by that central sphere.

E. Total Neutron Flux and Fluence

The total central neutron flux is $\sim 6.4 \times 10^8$ n/cm²-sec at a reactor power of 1 MW; its ratio to the thermal neutron flux available in the empty cavity is 0.71 ± 0.05 .

The total neutron fluence is usually limited by the reactor operation policy to an upper value of 8×10^{12} n/cm², but under specific commitment, the pile can be and has been operated with $\Sigma\Sigma$ at a 3.5 MW power level so that fluences as high as $\sim 2 \times 10^{14}$ n/cm² might be achieved.

F. Central Gamma-to-Neutron Ratio

Measured as 6.2×10^{-7} Rad hour⁻¹/cm⁻² sec⁻¹. The major contribution to the central gamma dose rate originates from the source shell.

G. Monitoring

Redundancy is essential and is provided as follows:

1. Reactor instrumentation: Compensated ionization chamber checked against thermal flux activation foils irradiated in the core.

2. Special polar slot within the $\Sigma\Sigma$ source approximately at the peak of the fast flux, for use with threshold activation foils (usually inelastic scattering in ^{115}In).

3. Rotating graphite plug within the thermal column for thermal flux activation foils (usually Au).

4. A horizontal channel at the edge of and parallel to the axis of the thermal column, for ^{235}U fission chambers; an extracted adjacent thermal neutron beam is also available.

II. Neutron Spectrum Data

A. The coordinated effort undertaken so far to experimentally characterize the $\Sigma\Sigma$ central neutron spectrum is summarized in Table 16.

TABLE HEDL-16
 $\Sigma\Sigma$ NEUTRON SPECTRUM MEASUREMENTS - DECEMBER 1971

<u>Detection Process</u>	<u>Energy Range</u>	<u>Laboratory</u>
Proton Recoil	25 keV - 1.4 MeV	KFK ⁽⁴¹⁾
	Data Not Yet Available	RCN
	20 keV - 1.4 MeV	CEN-SCK
$^6\text{Li}(n,\alpha)$	400 keV - 10 MeV	RCN
	3 keV - 4 MeV	CEN-SCK ⁽⁴²⁾
$^3\text{He}(n,p)$	100 keV - 5.3 MeV	KFK ⁽⁴³⁾
	1.5 MeV - 10 MeV	RCN

The final analysis of these data is not yet complete, but it can already be seen that with an energy structure of $\Delta u = 0.5$ to 0.7 , the various measurements generally agree with each other to within $\pm 5\%$ between 25 keV and 4 MeV, over most of their range of overlap; with some systematic and reproducible deviations, not exceeding 10% for ^6Li spectrometry below 400 keV, which has been traced to inadequacies in the lithium cross section used (ENDF/B Version I).

Large or systematic deviations are observed, however, with the calculated spectrum below 40 keV and between 1.4 and 2.5 MeV. An illustration of the type of problems encountered is displayed on Figure 12 for earlier preliminary CEN-SCK measurements and calculations.

Figure HEDL-12 $\Sigma\Sigma$ Central Neutron Flux Spectra Used for Evaluation of Integral Reaction Rate Data

B. Calculations in the discrete-ordinates S_8 transport-corrected P_0 and P_1 approximations performed by means of the DTF-IV code using group cross sections derived from the KEDAK file⁽¹⁵⁾ fortuitously agree reasonably well with similar calculations based on the British FD-2 set.⁽⁴⁴⁾

A current investigation of the sensitivity of computed data to cross section inadequacies and numerical approximations indicates that the central neutron spectrum is most affected by uncertainties in the ^{238}U inelastic scattering data (especially the matrix elements describing the secondary neutron emission spectrum), by the depiction of fission neutron spectra, and by the selection of the weighting function within the source shell (particularly at energies below 2 keV); the contribution of graphite wall return neutrons and their attenuation by the source shell have been found to be the major cause of uncertainties in theoretical predictions below 50 keV.

Earlier detailed computations combined with specific measurements have shown that the carefully estimated uncertainties in all geometrical and physico-chemical data for this facility are unlikely to contribute for more than 1% to the error on theoretically derived neutronic data.

C. The low energy tail of the neutron spectrum below 10 keV is currently investigated by means of the resonance sandwich foil activation technique.

III. Integral Reaction Rate Data

A rather large and comprehensive program of integral measurements has been performed so far by CEN-SCK in order to assess reaction rate data for this system and to assist the theoretical analysis. This includes:

A. Systematic mapping of the thermal neutron flux incident on the source surface.

B. Spatial traverses within the source shell for various directions with respect to the axis of the thermal column and for the reactions $^{235}\text{U}(n,f)$, $^{238}\text{U}(n,\gamma)$, $^{115}\text{In}(n,n')^{115\text{m}}\text{In}$, $^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$, $^{164}\text{Dy}(n,\gamma)^{165\text{g}}\text{Dy}$, $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$, as well as for total fission rate; similar spatial traverses within the central exposure volume.

C. Measurement of the re-entrant hole perturbation as a function of space for two-hole sizes and the reactions $^{235}\text{U}(n,f)$, $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$, $^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$ and $^{23}\text{Na}(n,\gamma)^{24}\text{Na}$.

D. Double ratio comparisons by means of threshold detectors of the central neutron spectrum* and the ^{235}U thermal fission neutron spectrum.

E. Absolute measurements of the central reaction rates for a number of threshold and non-threshold reactions.**

In the present context, only data pertinent to items (D) and (E) are presented and shortly discussed hereafter. The usually small corrections for the re-entrant hole perturbation,⁽³³⁾ significant only in non-threshold fission chamber measurements, will be outlined in a subsequent contribution dealing with the use of the NBS fission chamber in $\Sigma\Sigma$.

1. High-Energy Central Spectral Indices

The relative spectral indices for reactions i and j with differential-energy cross sections $\sigma_i(E)$ and $\sigma_j(E)$ are defined as

$$R_{ij}(\Sigma\Sigma, \chi_{25}) = \frac{\bar{\sigma}_i(\Sigma\Sigma)/\bar{\sigma}_j(\Sigma\Sigma)}{\bar{\sigma}_i(\chi_{25})/\bar{\sigma}_j(\chi_{25})} \quad (1)$$

where χ_{25} means the ^{235}U thermal fission neutron spectrum and

$$\bar{\sigma}_i(\phi) = \int_0^\infty \phi(E) \sigma_i(E) dE / \int_0^\infty \phi(E) dE. \quad (2)$$

The source-detector assembly used for production of fission neutrons within the 50 cm diameter cavity to obtain the denominator values, equation (1), has been described and discussed elsewhere.⁽⁴⁵⁾ Obtaining the spectral indices R_{ij} only involves straightforward relative activation measurements.

The boron carbide shell for $\Sigma\Sigma$ was not used for most of these measurements. The results for $\Sigma\Sigma$ are displayed in Table 17 and are compared to present transport theory predictions. Also given are computed factors for relating these measurements in the absence of boron carbide to the actual $\Sigma\Sigma$ configuration: they were used to derive the absolute reaction rates for the

* In the absence of the boron carbide screen.

** Some absolute data have also been obtained with the boron carbide screen removed, but are not outlined here.

TABLE HEDL-17

SPECTRAL INDICES RATIOS^a IN THE CENTER OF THE $\Sigma\Sigma$ FACILITY

Reaction	Effective Threshold ^b (MeV)	Spectral Indices Ratios ^c		Computed ^f Conversion Ratio
		Measurements	Theory ^{d,e}	
$^{103}\text{Rh}(n,n')^{103m}\text{Rh}$	0.9	1.36 ($\pm 1\%$)	1.324 ⁽⁴⁸⁾	0.997
$^{115}\text{In}(n,n')^{115m}\text{In}$	1.1	1 ^g	1 ^{(45)g}	1 ^g
$^{238}\text{U}(n,f)$	1.6	1.045 ($\pm 1\%$)	0.906 ⁽⁴⁵⁾	0.999
$^{232}\text{Th}(n,f)$	1.9	0.903 ($\pm 2.5\%$)	0.869 ⁽¹⁵⁾	0.997
$^{58}\text{Ni}(n,p)^{58}\text{Co}$	2.8	0.817 ($\pm 5\%$)	0.842 ⁽⁴⁵⁾	0.984
$^{27}\text{Al}(n,p)^{27}\text{Mg}$	4.2	0.855 ($\pm 3\%$)	0.802 ⁽²⁹⁾	0.968
$^{56}\text{Fe}(n,p)^{56}\text{Mn}$	6.3	0.835 ($\pm 1.5\%$)	0.799 ^{(45)h}	0.949 ^h
$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	7.4	0.828 ($\pm 4\%$)	0.799 ^{(29)h}	0.949 ^h

a. Relative to the ^{235}U thermal fission neutron spectrum.

b. Computed in the fission spectrum according to the concept of reference 47.

c. Inner boron carbide screen removed.

d. S8 transport-corrected approximation, multigroup cross sections from the KEDAK file (reference 15) except the ^{238}U inelastic scattering matrix, taken from the Russian ABBN set.

e. Figures within brackets refer to the evaluation accepted for the differential energy cross sections of the reactions.

f. Multiplication factors allowing to derive the expected experimental indices ratios in the actual $\Sigma\Sigma$ configuration.

g. Reaction used as reference when forming the spectral indices ratios.

h. Only one theoretical group presently available for computation.

$^{232}\text{Th}(n,f)$ and $^{27}\text{Al}(n,p)^{27}\text{Mg}$ reactions as quoted in Table 18.

2. Absolute Central Reaction Rates

The measured absolute central reaction rates are summarized in Table 18 in terms of the thermal neutron flux available when the cavity is empty; in the lowest part of the table, a few absolute spectral indices, $\bar{\sigma}_i(\Sigma\Sigma)/\bar{\sigma}_j(\Sigma\Sigma)$, formed from the data in the upper part, are compared to present theoretical expectations. The discrepancies are sizeable; they are much decreased,⁽³³⁾ but not entirely removed, when the preliminary measured neutron spectrum (Figure 12), extrapolated by theory, is used to weight the differential energy cross sections. Presumably fruitful speculation on this will be possible when the final evaluated experimental neutron spectrum and new transport calculations with the most recent multigroup cross section sets will be available. The data gathered in Table 18 result from a very large number of irradiations at various neutron power levels, with various combinations of foils, various flux time profiles, counting conditions, etc.

The fission rates have been determined by the track recorder technique while all activation measurements were performed by means of a calibrated 3" x 3" NaI(Tl) gamma-ray spectrometer, except for capture in ^{238}U for which a 20 cc Ge(Li) diode has been used.

The samples for the capture and fission reactions were dilute alloy foils supplied by CBNM (Euratom, Geel) and nominally assessed to an accuracy of $\pm 1\%$. Extensive activation analysis checks of these foils have been performed by rotating most of them in a pure thermal neutron flux measured with Au: perfect agreement has been obtained within respective error margins in all but one case (4% Mn/Al alloy*). In any event, the foil assay does not enter the derivation of the final corresponding reaction rates because the samples have been exposed both in $\Sigma\Sigma$ and in the thermal neutron flux facility. These final data essentially rely on well known thermal cross sections and the precise measurement of a thermal neutron flux (except for fission in ^{238}U where the accurate mass spectrometry analysis provides a direct scaling factor in addition to the usual corrections). Furthermore, for each reaction,

* However, 1% Mn/Al and 4.5% Mn/Al agree well so that a simple clerical error is now suspected.

TABLE HEDL-18
ΣΣ CENTRAL REACTION RATES AND RATIOS

A. Central Reaction Rates Per Unit Available Thermal Neutron Flux^a

Reaction	Absolute Measurement	Deduced from Spectral Indices Ratios ^b	Recommended Value
$^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$	28.7 -27 ($\pm 3\%$)	-	28.7 -27 ($\pm 3\%$)
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	319 -27 ($\pm 2\%$)	-	319 -27 ($\pm 2\%$)
$^{238}\text{U}(n,\gamma)^{239}\text{U}$	140 -27 ($\pm 2.5\%$)	-	140 -27 ($\pm 2.5\%$)
$^{235}\text{U}(n,f)$	1096 -27 ($\pm 4\%$)	-	1096 -27 ($\pm 4\%$)
$^{239}\text{Pu}(n,f)$	1332 -27 ($\pm 3\%$)	-	1332 -27 ($\pm 3\%$)
$^{115}\text{In}(n,n')^{115\text{m}}\text{In}$	42.3 -27 ($\pm 2\%$) ^c	42.3 -27 ^{c,d}	42.3 -27 ($\pm 2\%$) ^c
$^{238}\text{U}(n,f)$	74.4 -27 ($\pm 3\%$)	77.0 -27 ($\pm 4\%$)	75.0 -27 ($\pm 3\%$)
$^{232}\text{Th}(n,f)$	-	16.8 -27 ($\pm 5\%$)	16.8 -27 ($\pm 5\%$)
$^{58}\text{Ni}(n,p)^{58}\text{Co}$	20.9 -27 ($\pm 2\%$)	20.4 -27 ($\pm 5.5\%$)	20.8 -27 ($\pm 2\%$)
$^{27}\text{Al}(n,p)^{27}\text{Mg}$	-	0.745 -27 ($\pm 10\%$)	0.745 -27 ($\pm 10\%$)
$^{56}\text{Fe}(n,p)^{56}\text{Mn}$	0.198 -27 ($\pm 3\%$)	0.191 -27 ($\pm 6\%$)	0.197 -27 ($\pm 3\%$)
$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	0.132 -27 ($\pm 3\%$)	0.129 -27 ($\pm 5\%$)	0.131 -27 ($\pm 3\%$)

B. Central Reaction Rate Ratios

Reaction Couple	Measurement	Theory ^e
$^{235}\text{U}(n,f) / ^{238}\text{U}(n,f)$	14.7 ($\pm 4\%$)	19.4
$^{239}\text{Pu}(n,f) / ^{235}\text{U}(n,f)$	1.21 ($\pm 5\%$)	1.09
$^{238}\text{U}(n,\gamma)^{239}\text{U} / ^{235}\text{U}(n,f)$	0.128 ($\pm 4.5\%$)	0.171
$^{238}\text{U}(n,f) / ^{115}\text{In}(n,n')^{115\text{m}}\text{In}$	1.76 ($\pm 3.5\%$)	1.45

- a. In BR1 50 cm diameter cavity, the available thermal neutron flux is $\sim 9 \times 10^8 \text{ n cm}^{-2} \text{ sec}^{-1}$ at a reactor power of 1 MW. Note: $28.7 - 27 = 28.7 \times 10^{-27}$.
- b. Converted spectral indices ratios from Table 17 multiplied by average fission spectrum cross section ratios from reference 17 and normalized to Indium(n,n') absolute reaction rate.
- c. The error band does not include the uncertainties in the internal conversion coefficient and level scheme: the gamma-ray intensity for the 335 keV isometric transition is fixed to 46%; $^{115}\text{In}(n,n')^{115\text{m}}\text{In}$ differential energy cross sections used for comparison must be normalized in the same way.
- d. Accepted for the normalization referred to under (b).
- e. S8 transport-corrected approximation; multigroup cross section data taken from the KEDAK file (reference 15) except the ^{238}U inelastic scattering matrix (Russian ABN) and the $^{115}\text{In}(n,n')^{115\text{m}}\text{In}$ cross section (reference 45).

two or more different alloy types have been used, with nominal compositions varying by at least a factor of 4: the resulting $\Sigma\Sigma$ central reaction rates agreed within the statistical spreads, which decisively confirms them as being effectively infinite dilution quantities.

For the threshold activation reactions, the absolute activity scale relates to the photo peak efficiency curve of the calibrated NaI(Tl) spectrometer [and in the case of the $^{56}\text{Fe}(n,p)^{56}\text{Mn}$, is substantiated further by thermal flux exposures of manganese foils]: this scale has been established for many years, backed by a number of interlaboratory cross-checks, and is one of the CEN-SCK sustained fission spectrum average cross section measurements.⁽⁴⁵⁾

Finally, it must be noted that when the relationship of the central reaction rate scale to the available thermal neutron flux was determined, only items (I-G-1) and (I-G-3) of the monitoring scheme indicated were effectively used; no later check has yet been done; the possible associated error ($\pm 2\%$) has not been included in Table 18.

Expected Achievements

By the end of June 1972, it is hoped that a first measurement of the ^{238}U and ^{235}U fission rates and ratio will have been performed in $\Sigma\Sigma$ by means of the NBS fission chamber and the results compared with existing solid state track recorder data.

It is expected that the SAND-II Monte Carlo error analysis⁽¹⁸⁾ of the $\Sigma\Sigma$ central neutron spectrum will be compared with proton recoil, $^6\text{Li}(n,\alpha)$, and $^3\text{He}(n,p)$ final neutron spectrometry results and with new, detailed transport theory computations based on ENDF/B Versions II and III, to be performed jointly at Oak Ridge⁽⁴⁶⁾ and at Mol.

REFERENCES FOR HEDL CHAPTER

1. W. N. McElroy, Editor, "LMFBR Reaction Rate and Dosimetry Quarterly Progress Report, June, July, August 1971," HEDL-TME 71-131, September 1971.
2. W. N. McElroy, Editor, "LMFBR Reaction Rate and Dosimetry Quarterly Progress Report, September, October, November 1971," HEDL-TME 71-179, December 1971.
3. S. B. Garfinkel, Private Communication, NBS (1972).
4. W/FTF Memo #72614, W. N. McElroy and L. S. Kellogg to Participants of ILRR Program, "Test Plan for the Non-Fission Foil Radiochemistry Measurements for the CFRMF Test and Irradiation Schedule for the NBS Fission Chamber, ANL Fission Track, AI Helium Mass Spectrometry Measurements," January 27, 1972.
5. J. W. Rogers, Private Communication, ANC (1972).
6. J. Grundl, Private Communications, NBS (1972).
7. W. Zimmer, Private Communication, ARHCO (1972).
8. N. Dudgey, Private Communication, ANL (1972).
9. R. L. Simons and W. N. McElroy, "Evaluated Reference Cross Section Library," BNWL-1312, Battelle-Northwest, 1970.
10. A. Paulsen and R. Wieders, " $^{58}\text{Ni}(n,p)^{58}\text{Co}$ and $^{54}\text{Fe}(n,p)^{54}\text{Mn}$ Cross Section Measurements for Use as Threshold Detectors," Central Bureau for Nuclear Measurements, Geel, Belgium (1971-1972).
11. W. N. McElroy and R. E. Dahl, Jr., "Neutron Dosimetry for Fast Reactor Applications," Irradiation Effects on Structural Alloys for Nuclear Reactor Applications, ASTM STP-484, 1970, pp. 375-399.
12. W. N. McElroy et al., "Neutron Energy Dependent Cross Section Evaluation by a Multiple Foil Activation Method," WHAN-SA-107, Paper presented at Third Conference on Neutron Cross Sections and Technology, Knoxville, Tennessee, March 15-17, 1971.
13. W. N. McElroy et al., "Low Energy Cross Section for $^{54}\text{Fe}(n,p)^{54}\text{Mn}$ and Other Exoergic Reactions," HEDL-SA-194 S, Trans. Am. Nucl. Soc. 14 Number 2, October 1971.
14. W. N. McElroy et al., "EBR-II Dosimetry Test Data Analysis," BNWL-1402, Battelle-Northwest (1970).
15. J. J. Schmidt, Neutron Cross Sections for Fast Reactor Materials, KFK-120 (EANDC-E-38-U) (1966). Available at AEC Depository Libraries.
16. W. G. Davey, "Selected Fission Cross Sections," Nucl. Sci. Eng. 32, 34-35 (1968).
17. A. Fabry, "Evaluation of Microscopic Integral Cross Sections Averaged on the ^{235}U Thermal Fission Neutron Spectrum," Unpublished document to be presented to the Euratom WGRD and CSEWG Committees.
18. C. A. Oster, W. N. McElroy and J. Marr, "A Monte Carlo Program for SAND-II Error Analysis," HEDL report in preparation.
19. W. N. McElroy, R. J. Armani and E. Tochilin, "The Spectral Distribution of Neutrons and Neutron Reaction Cross Sections in an Unreflected ^{235}U Critical Assembly and the Fission Neutron Spectrum," Nucl. Sci. Eng., May (1972).
20. L. Cranberg, G. Frye, N. Hereson and L. Rosen, Phys. Rev. 103, 662 (1956).
21. W. N. McElroy, Nucl. Sci. Eng. 36, 109 (1969).
22. J. A. Grundl, "Fission-Neutron Spectra: Macroscopic and Integral Results," in EANDC Symposium on Neutron Standards and Flux Normalization, CONF-701002, 417 (1971).
23. L. Rosen, in reference 22, above.
24. E. Barnard et al., Nucl. Phys. 71, 228 (1965).
25. Yu S. Zamyatin et al., CN-26/90, in Nuclear Data for Reactors, Vol. II, p. 183-193, IAEA, Vienna (1970).
26. J. A. Grundl, Nucl. Sci. Eng. 31, 191 (1968).
27. A. Fabry et al., "Some Nuclear Data and Considerations Related to Reactor Dosimetry with Threshold Detectors," 27.2.70, WGRD, Euratom-Ispra (1970).
28. R. W. Lamphere, Phys. Rev. 104, 1654 (1956).
29. J. A. Grundl, Nucl. Sci. Eng. 30, 39-53 (1967).
30. G. M. Frye, J. H. Gamel and L. Rosen, "Energy Spectrum of Neutrons From Thermal Neutron Fission of ^{235}U and From an Untamped Multiplying Assembly of ^{235}U ," LA-1670, Los Alamos Scientific Laboratory (1954).
31. B. E. Watt, Phys. Rev. 87, 1037 (1952).
32. A. Fabry, J. C. Schepers and P. Vandeplas, "Status of the Work About the Generation of Intermediate Standard Neutron Spectra at Mol," Report EACRP-A-97 (1969).
33. A. Fabry, M. DeCoster, G. DeLeeuw-Gierts and S. DeLeeuw, "The Intermediate-Energy Standard Neutron Spectrum Facility 22," Progress Report submitted at the 14th meeting of the EACRP and at the 30th meeting of the Euratom WGRD (1971).
A. Fabry, "The Role of Standard Neutron Fields in Relation to Fast Reactor Spectrometry and Reactor Design," IAEA Panel on "Fast Reactor Spectrum Measurements and Their Interpretation," Argonne National Laboratory, 10-13 November (1970).
34. A. Fabry and P. Vandeplas, "The Generation of Intermediate Standard Neutron Spectra and Their Applications in Fast Reactor Physics," Proceedings of the IAEA Karlsruhe Symposium, "Fast Reactor Physics," Vol. I, p. 389-411 (1968).
A. Fabry, "The Polythene-Boron-10 Intermediate-Energy Standard Neutron Spectra," Progress Report presented to the Euratom WGRD, March 9 (1971).
35. J. A. Grundl, "Intermediate-Energy Neutron Standard Field at the National Bureau of Standards," Proposal submitted to the EACRP (1970).
36. C. B. Besant, Private Communications, and "NISUS - A Neutron Intermediate Standard Uranium Source," (1971).
37. I. Girdle, Private Communications, Institute for Atomic Physics IFA (1971).
38. J. J. Scoville, Private Communications, Aerojet Nuclear Company (1971).
39. J. A. Grundl, "Absolute Fission Chamber Measurements in CFRMF," LMFBR Reaction Rate and Dosimetry Quarterly Progress Report, June, July, August 1971, HEDL-TME 71-131 (1971).
40. M. Guery and A. Jarrige, "Miniature Fission Monitors for the Measurements of the Fission Flux in Research Reactors," Revue de Physique Appliquée, Vol. 6, p. 121-127 (1971).
41. H. Blum, G. Fieg, and H. Werle, Private Communications to G. DeLeeuw-Gierts and S. DeLeeuw (1971).
42. G. DeLeeuw-Gierts and S. DeLeeuw, "Improvements in the Use of the $^6\text{Li}(n,\alpha)^3\text{He}$ Reaction for In-Core Neutron Spectrum Measurements," Report Big (1970).
43. R. J. S. Harry, Private Communications to G. DeLeeuw-Gierts and S. DeLeeuw (1971).
44. R. W. Smith, J. L. Rowlands and D. Wardleworth, "The FD2 Group Averaged Cross Section Set for Fast Neutron Calculations," Report AEEW-R491 (1966).
45. A. Fabry, M. DeCoster, G. Minsart, J. C. Schepers and P. Vandeplas, "Implications of Fundamental Integral Measurements on High-Energy Nuclear Data for Reactor Physics," Proceedings of the IAEA Conference, "Nuclear Data for Reactors," Vol. II, p. 535-569 (1970).
46. Personal contact between D. Jenkins (ORNL) and A. Fabry (CEN-SCK).
47. J. A. Grundl and A. A. Usner, "Spectral Comparisons with High-Energy Activation Detectors," Nucl. Sci. Eng. 8, 595 (1960).
48. P. Ribon, Unpublished evaluation document DRP-SMPNF-714-69, Saclay (1969).

LAWRENCE LIVERMORE LABORATORY

A. NUCLEAR CROSS SECTION CALCULATIONS*

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Objective

- We are accumulating and developing the computer programs necessary for calculating excitation functions of nuclear reactions induced by neutrons with energies up to several tens of MeVs.

Summary

During the past two Quarters we have continued the development and debugging of the statistical model codes COMNUC, CASCADE, and UHL. The coupled-channels optical model program FOURPLUS was put into operation and tested. Some preliminary results have been obtained for the neutron induced reactions on ^{197}Au and ^{239}Pu .

Accomplishments and Status

At the present time all of the known bugs are out of COMNUC, CASCADE, and FOURPLUS, although some additional changes are planned for COMNUC. The UHL code has been modified to accept transmission coefficients that are a function of J rather than l . We have attempted to include the ability to handle discrete states in the target nucleus and all daughter nuclei, but there are still errors in this version of the code.

To illustrate the usefulness of the COMNUC code, we include here recent preliminary results from the study of two reactions: $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ and $^{239}\text{Pu}(n,\text{fission})$. In Fig. 1 we compare the results of our Au capture calculation with an evaluation by HEDL, Westinghouse Hanford Company. We expect that a more complete calculation that we are planning to carry out in the energy region 0.1-2.0 MeV will result in somewhat lower calculated values, which will be more in line with the evaluation in this region.

Fig. 2 shows some other reactions that were calculated concurrently with the (n,capture) reaction.

Fig. 3 shows the results of our $^{239}\text{Pu}(n,\text{fission})$ calculation, which includes nine discrete fission channels and continuum fission in ^{240}Pu , as well as continuum fission in ^{239}Pu and ^{238}Pu . The fission barriers and the density of fissioning levels in the Pu nuclei appear to be reasonable, the (n,capture) and (n,2n) cross sections (shown in Fig. 4) that are calculated concurrently also agree well with experiment, and so we feel fairly confident that COMNUC can profitably be used in the evaluation of fission cross sections.

Expected Achievements

During the next Quarter we will continue to evaluate the COMNUC and CASCADE programs, and we hope to finish debugging the discrete levels version of UHL. Further in the future we plan to introduce precompound evaporation into the UHL code, and discrete fission channels for (n,n'f) and (n,2nf) reactions into the COMNUC code.

*Work performed under the auspices of the Atomic Energy Commission.

Figure LLL-1 Comparison of Calculated Excitation Function for the Reaction $^{197}\text{Au}(n, \gamma)^{198}\text{Au}$ with the Evaluation of HEDL

Figure LLL-2 Comparison of Calculated Nonelastic and Some Inelastic Cross Sections for ^{197}Au with Evaluations from BNL-325

Figure LLL-3 Comparison of Calculated ^{239}Pu (n, fission) Excitation Function With an Evaluation by R. Howerton. [The Calculated (n, capture) Excitation Function is also Shown]

Figure LLL-4 Calculated Excitation Functions for Several Reactions Induced by Fast Neutrons on ^{239}Pu

LOS ALAMOS SCIENTIFIC LABORATORY

A. NEUTRON FLUX SPECTRA ASSOCIATED WITH SEVERAL LASL CRITICAL ASSEMBLIES

G. E. Hansen

Objective

• Provide spectral information sufficient for estimation of various neutron reactions per unit fluence at central positions of the Big-10 (U-reflected U(10)) and Flattop (U-reflected ^{235}U , ^{233}U , or ^{239}Pu) critical assemblies.

Summary

Spectra are given in Table LASL-1

Accomplishments and Status

There is at present no direct experimental information on the central spectrum of Big-10. The spectrum listed in Table LASL-1 was obtained by a multi-group (1/4 lethargy widths) transport calculation based on ENDF-II cross sections.

The Flattop assemblies have provided numerous neutron bombardments over the past decade and many integral or mean activation cross sections for these spectra have been measured. Here we feel the listed central spectra accomplish the above objective.

Expected Achievements

These assemblies are potential neutron sources for ILRR measurements. Central fluences per bombardment are limited to the order of 10^{15} neutrons/cm². Access is via 1/2 inch diameter glory holes in Ft- ^{233}U and Ft- ^{239}Pu , up to a 1 inch diameter glory hole in Ft- ^{235}U , and up to a 3 inch diameter glory hole in Big-10.

Table LASL-1

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

<u>E(MeV)</u>	<u>Flux Fraction Above E(MeV)</u>			
	<u>Big-10</u>	<u>Ft-²³⁵U</u>	<u>Ft-²³³U</u>	<u>Ft-²³⁹Pu</u>
10	0.0002	0.0006	0.0007	0.0010
8	0.0009	0.0030	0.0037	0.0047
6	0.004	0.014	0.017	0.020
4	0.015	0.059	0.075	0.079
3	0.029	0.118	0.148	0.155
2	0.057	0.231	0.285	0.286
1.4	0.085	0.339	0.410	0.406
0.9	0.145	0.479	0.559	0.554
0.4	0.400	0.730	0.785	0.772
0.2	0.650	0.900	0.910	0.905
0.1	0.828	0.964	0.971	0.965
0.05	0.938	0.990	0.992	0.990
0.01	0.998	1.000	1.000	1.000

NATIONAL BUREAU OF STANDARDS

A. ABSOLUTE FISSION CHAMBER MEASUREMENTS IN CFRMF

J. A. Grundl

Objective

• A first-step measurement of absolute fission rates at the center of the CFRMF Facility will be performed for four fissionable isotopes: ^{235}U , ^{239}Pu , ^{238}U , and ^{237}Np . The measurement is to be carried out with the NBS double fission ionization chamber and fission foils that are part of the permanent set used in the NBS Fast-Neutron Standards Program. Specific efforts to identify and evaluate error sources will focus on chamber performance, foil assay, and flux perturbations.

Summary

Fission rate measurements at the CFRMF were performed during the first week of February 1972. The NBS Fission chamber and associated electronics was transported to Idaho without incident and performed reliably at the reactor. Reproducibility of fission chamber response and the CFRMF power level instrumentation was excellent and unambiguous. A dozen comparable fission rate determinations on successive days and involving chamber removal and disassembly gave fission rates which differed by less than 0.3%. Fission rates per nucleus are given but without final evaluation or error assignments. With the possible exception of ^{239}Pu , it is not anticipated that final values will depart from those given in this report by more than 3%. Fission ratios based on still incomplete foil weight determinations are as follows: $\bar{\sigma}_f(^{235}\text{U})/\bar{\sigma}_f(^{238}\text{U}) = 21.3$, $\bar{\sigma}_f(^{239}\text{Pu})/\bar{\sigma}_f(^{238}\text{U}) = 24.7$, and $\bar{\sigma}_f(^{237}\text{Np})/\bar{\sigma}_f(^{238}\text{U}) = 7.5$.

Accomplishments and Status

Fission rate measurements at the CFRMF Facility were performed during the first week of February. The NBS double fission chamber and the associated electronics--transported to Idaho as conventional airline luggage--was in place and operational the first day on site. Three and one-half days of measurements with more than 120 dual fission counts recorded provided the necessary data for specifying fission rates in ^{235}U , ^{239}Pu , ^{238}U , and ^{237}Np . This third report for the ILRR Program will summarize the important features of the measurement and present early fission rate results without final evaluation or error assignments.

1. Chamber Performance

General operational characteristics for the NBS double fission chamber were described in the first ILRR Quarterly Report. The only significant difference for the measurements performed at CFRMF was the use of two scalers instead of three to record the count rates from each side of the chamber. One scaler was biased near the alpha pulse height cut-off (V_α) and the second at $2V_\alpha$. The ratio of these two scaler counts, $S(V_\alpha)/S(2V_\alpha)$, is a measure of the correction required to obtain absolute fission rates from the counting data, the so-called extrapolation-to-zero. With P-10 gas (90% Argon--10% methane) flowing at 2 CFH, and with 30 feet of cable between the preamp and the fission chamber at the center of CFRMF, the average of scaler ratios observed for various thicknesses of foils were as follows:

	Foil thickness <u>($\mu\text{g}/\text{cm}^2$)</u>	<u>$S(V_\alpha)/S(2V_\alpha)$</u> (extrapolation-to-zero)
^{235}U	10	1.0015
^{235}U	100	1.0079
^{237}Np , ^{239}Pu	100	1.0035
^{237}Np , ^{238}U	200	1.0065

The smaller extrapolation-to-zero for ^{239}Pu and ^{237}Np compared to ^{235}U for the same foil thickness is not yet understood. Scaler ratios for the 120 dual counts taken during the course of the measurements were within ± 0.002 of

the above values. The ratio of scaler counts is also a sensitive monitor of noise. Minor ground loop problems and troublesome connections were apparent immediately in the observed increase of the scaler ratios. During each of the primary runs outputs were interchanged at the preamps at least once. No discernible difference was observed in the scaler readings corresponding to such interchanges.

2. Count Rate Losses

For well-resolved integral pulse counting at moderate rates count rate losses are proportional to the observed count rate, pulse pile-up is a negligible perturbation. The fraction of counts lost in this case is just the product of the count rate, C , and the effective dead time of the electronic network, τ . Output pulses for amplifiers were set at $\sim 1.5 \mu\text{sec}(\text{fwhm})$. Dead times were determined in two ways: (1) varying the pulse width and observing changes in count rate; and (2) comparing count rate ratios between two fission foils of different thicknesses at two power levels which differed by a factor of ten. Results were as follows:

- (1) pulse widths of 1.5 and 5 $\mu\text{sec}(\text{fwhm})$; $C = 4000 \text{ c/sec}$ (the maximum value for the primary measurements):

$$^{235}\text{U foil: } \tau = 1.4 \pm 0.2 \mu\text{sec}$$

$$^{239}\text{Pu foil: } \tau = 1.8 \pm 0.1 \mu\text{sec}$$

- (2) power levels of 0.6 kW ($C = 390$ and 4468 c/sec) and 6 kW ($C = 3733$ and 39020 c/sec); pulse width = 1.5 $\mu\text{sec}(\text{fwhm})$:

$$\tau = 2.7 \pm 1 \mu\text{sec.}$$

3. Flux Gradients

A cursory look at axial flux gradients near the center of CFRMF was undertaken as a complement to earlier measurements performed by ANC. The results were as follows:

position:	<u>center</u>	<u>+ 1.05"</u>	<u>+ 2.05"</u>	<u>+ 3.05"</u>	<u>+ 4.05"</u>
^{237}Np :	1.000	0.995	0.971	0.939	0.903
^{239}Pu :	1.000	0.993	0.974	0.946	0.907
$\frac{^{239}\text{Pu}}{\text{Np}}$:	1.000	0.999	1.003	1.007	1.004

Statistical errors of counting were no worse than ± 0.002 for the individual values. Thus, even without the assumption that the chamber is positioned at the maximum of an unperturbed cosine distribution, the axial flux gradient over the 0.5" diameter fission foils should be less than 0.4%.

4. Power Level Reproducibility and Linearity

The CFRMF reactor was taken up to power more than a dozen times during the three and one-half days of measurement and the fission chamber was removed from the reactor and disassembled for foil changes six times. Three foils were used repeatedly on successive days, paired with various of the other fission foils. Recorded fission rates for these foils at 0.6 kW were as follows (fiss./sec-nucleus):

<u>Isotope</u>	<u>Foil</u>	<u>2-1-72</u>	<u>2-2</u>	<u>2-3</u>	<u>2-4</u>	<u>Ctg.</u> <u>Statistics</u>	<u>Spread</u>
^{235}U	25S-1-1	1.284 $\times 10^{-14}$	1.286		1.284 1.281	<0.2%	1.0039
^{239}Pu	49-1-1&2		1.490	1.491 1.493	1.494	<0.2%	1.0029
^{238}U	28N-2-2			0.0603	0.0602 0.0604 0.0606	<0.3%	1.007

The observed reproducibility of the fission chamber, the associated electronics, and the CFRMF power level instrumentation is seen to be excellent and unambiguous. This was an important and welcome outcome of the weeks work at CFRMF.

The maximum power level for reliable operation of the fission chamber was chosen conservatively at 0.6 kW ($\phi \sim 8 \times 10^9$ n/sec-cm²). This corresponded to a maximum counting rate of around 4000/sec and requires dead time corrections of $\sim 1\%$. An appropriate power level for irradiating the activation detectors is 6 kW. Establishing these two power levels on a relative basis to a satisfactory and confident level of accuracy is an outstanding problem for the ILRR Program. In order to help with this problem and also to observe the behavior of the fission chamber at high fluxes, fission rates were obtained for a series of four power level changes. The

results were as follows:

Power level:	0.6 kW	2 kW	6 kW	0.6 kW
<u>relative:</u>	<u>1.000</u>	<u>3.33</u>	<u>10.0</u>	<u>1.00</u>
^{235}U :	1.000 (169.4c/s)	3.22 ± 0.024	9.60 ± 0.05 (1626c/s)	1.008 ± 0.006 (170.7c/s)
^{238}U :	1.000 (414.0c/s)	3.23 ± 0.012	9.63 ± 0.05 (3961c/s)	1.000 ± 0.004 (414.0c/s)

The power levels were taken in the order listed and the entire measurement lasted about 45 minutes.

The fission chamber results disagree with the CFRMF power level instrumentation by about 3% at 2 kW and by about 4% at 6 kW. The disagreement is the same for count rates which differ by a factor of $2\frac{1}{2}$ and for which count rate losses at the 6 kW level were 0.3% and 0.8%, respectively. In order to obtain agreement with the power level instrumentation a system dead time of around 10 μsec would be required. Observed pulse widths and the dead time measurements described above would not allow more than about 3 μsec . Depression of the pulse rate in the fission chamber due to gamma ray effects is possible but it is not consistent with the prompt return of the pulse rate to its initial level when the power level was returned to 0.6 kW (last column above). Another possibility is gas deterioration at high pulse rates. Even at moderate pulse rates significant deterioration of the pulse height will occur if the gas flow is interrupted for a few minutes (this is most pronounced for methane gas rather than the P-10 used at CFRMF). A quick check of this possibility was made by increasing the gas flow at 6 kW from 2 CFH to 4 CFH. No effect upon the pulse rate was observed to the extent of the $\pm 0.3\%$ counting statistics. The disagreement, therefore, cannot be explained by any known characteristics of the NBS fission chamber. It is not suggested by this statement or by the foregoing discussion that the NBS fission chamber response should be used in preference to the established CFRMF power level instrumentation.

5. Fission Rate Results at 0.6 kW

The fission chamber measurements at the center of CFRMF are essentially complete now. Nearly 500 individual scaler counts have been

summarized and additional foil weight measurements performed at NBS have been correlated with existing information. On this basis fission ratios and fission rates per nucleus have been calculated for all four fissionable isotopes. The results are presented in Table NBS-1. First listed are two runs of one-half and one hour duration which were carried out with gold and indium flux monitors placed on a 1.5" diameter disk attached to the chamber stem at a distance of 1.50" from the axis of the fission chamber (see HEDL chapter, Figure HEDL-2). Reaction rates given in the Table for the gold foils are those measured by the counting lab at HEDL. The remaining fission rate determinations are grouped according to individual foils rather than foil pairs as run; in part III of the Table fission ratios are listed without error assignments. The experimental values given in Table NBS-1 are subject to change: The correction of counting data to absolute fission rates is not fully evaluated and scattering perturbations must be included; and most important, foil weight determinations at NBS must be put in a first stage of completion and then documented. With the possible exception of ^{239}Pu , it is not anticipated that the final values will depart from those listed in Table NBS-1 by more than 3%.

Expected Achievements

By July 1, 1972 final values for absolute fission rates at the center of CFRMF will be specified and uncertainties assigned. Access to the Big Ten critical assembly at Los Alamos is not yet possible and the fission chamber measurements mentioned in previous ILRR reports have been postponed.

Table NBS-1

Fission Chamber Results at the Center of CFRMF

Isotope	Fission Foil	Fissions/sec Nucleus	* Precision of Count Rate Data (1 σ)
I. Runs monitored with gold and indium foils			
A. February 4, 1972 (Au(n, γ): 3.218×10^{-15} reactions/sec-nucleus)			
^{235}U	25S-1-1	1.284×10^{-14}	$\pm 0.1 \%$
^{238}U	28N-2-2	0.0602	$\pm 0.15\%$
B. February 2, 1972 (Au(n, γ): 3.173×10^{-15} reactions/sec-nucleus)			
^{239}Pu	49-1-1	1.490	$\sim 0.1 \%$
^{237}Np	37S-1-1	0.451	$\pm 0.2 \%$
II. Runs without monitor foils			
^{235}U	25S-1-1	1.283	$\sim \pm 0.1 \%$
	25S-01-1	1.276	$\pm 0.4 \%$
^{238}U	28N-2-2	0.0604	$\sim \pm 0.2 \%$
	28S-2-1	0.0599	$< \pm 0.2 \%$
^{239}Pu	49-1-1	1.492	$\pm 0.15\%$
	49-1-2	1.494	$\pm 0.15\%$
^{237}Np	37S-2-2	0.455	$\sim \pm 0.2 \%$
III. Fission Ratios			
$\bar{\sigma}_f(^{235}\text{U}) / \bar{\sigma}_f(^{238}\text{U}) = 21.3$			
$\bar{\sigma}_f(^{239}\text{Pu}) / \bar{\sigma}_f(^{238}\text{U}) = 24.7$			
$\bar{\sigma}_f(^{237}\text{Np}) / \bar{\sigma}_f(^{238}\text{U}) = 7.5$			

* Estimated on the basis of counting statistics and observed spread of individual counts.

B. THE NBS CALIFORNIUM-252 SPONTANEOUS FISSION NEUTRON SOURCES

J. A. Grundl

Objective

- To set down properties of the NBS ^{252}Cf spontaneous fission sources so far as they are known, and as they are relevant for activation detector calibration.

Summary

Two low mass and volume ^{252}Cf spontaneous fission sources will be at the National Bureau of Standards by April 1972. The source strength for the first to arrive has been measured at the NBS Manganous Bath Facility and a value of $2.154 \pm 0.035 \times 10^9$ n/sec is obtained. Isolated exposures of the source to the NBS double fission chamber have been carried out in the low-scatter target room of the NBS Van de Graaff. Scattering correction calculations for the 1.5 gram source capsule, the 7.0 gram fission chamber, and for other nearby structures at the irradiation facility are nearly finished. Fluences which could be furnished for detectors of interest to the ILRR Program will be a compromise involving accuracy requirements, detector efficiencies, and facility scheduling. A practical upper limit is probably 10^{13} . For the primary fission rate detectors, $^{239}\text{Pu}(n,f)$ and $^{235}\text{U}(n,f)$, present day uncertainties in the fission spectrum shape are unimportant, but for capture and threshold reactions these uncertainties are of concern. This issue is outlined, and it is suggested that it may be less expensive to face now rather than later the difficulties surrounding the establishment of fast-neutron energy distributions.

Accomplishments and Status

Two low mass and volume ^{252}Cf spontaneous fission neutron sources will be delivered to the NBS by April 1972. The source strength for the first to arrive has been measured at the NBS Manganous Bath Facility and a value of $2.154 \pm 0.035 \times 10^9$ n/sec is obtained. The second source, still to be fabricated, will be about twice this intensity. Three measurements to be performed with the sources are a part of the NBS Neutron Standards Program: (1) neutron source strength at the Manganous Sulfate Bath Facility; (2) neutron age-to-indium-resonance in ordinary water; (3) absolute fission-spectrum-averaged fission cross sections for ^{235}U , ^{239}Pu , ^{238}U , and ^{237}Np .

Description of Sources

Each californium source is a single cylindrical encapsulation of 0.51 mm thick stainless steel enclosing a disk-shaped deposit of californium. Total displacement of the capsule is 0.36 cc and the total mass is about 1.5 grams. More than 98% of the spontaneous fission neutrons are transmitted by the source encapsulation without collision, and the fraction absorbed or inelastically scattered is about 0.5%. Spontaneous fission in ^{254}Cf is the only contaminating neutron source; on arrival the ^{254}Cf in the first source was contributing $< 0.02\%$ of the total source strength. Specification of the mean position of the californium deposit relative to the front face of the 0.3" diam $\times$ 0.3" long cylindrical capsule is uncertain to ± 0.25 mm. This estimate is based on the known constraints of fabrication and is verified by means of an x-ray photograph.

Nuclear and physical properties of the sources are summarized in Table NBS-2.

Table NBS-2

Description of the NBS Californium-252 Spontaneous Fission Sources

Nuclear Properties

source strength (1 mg source):	2.15×10^9 n/sec
(2 mg source):	$\sim 4-5 \times 10^9$ n/sec
decay rate	2.3% per month
gamma emission rate	5 photons/neutron
flux @ 4 cm distance (2 mg source):	2×10^7 n/cm ² sec
fluence per day @ 4 cm	1.7×10^{12} n/cm ²
flux gradient @ 4 cm	1.3%/0.25 mm

Physical Description

cylindrical capsule	0.3" diam $\times$ 0.3" long
capsule mass (steel and aluminum)	1.5 gms
Californium deposit (disk-shaped)	≈ 2.5 mm diam $\times$ ≈ 1 mm thick
position of deposit relative to outside face of capsule	2.67 ± 0.25 mm

Facility for Irradiation

Isolated exposures of the NBS double fission chamber to the californium source have been carried out in the low-scatter target room of the NBS Van de Graaff. For these exposures an eight-foot long wand with the source attached rests upon a lightweight aluminum stand; the latter also supports the fission chamber. The distance to concrete foundations is twenty feet. Figure NBS-1 shows the essential features of the arrangement along with structural details of the source and of the fission chamber. Geometry calculations for all of the structures shown in the figure exist and single scattering corrections for thin detectors inside of the chamber may be estimated.

Activation detector foils may be placed in the fission chamber enclosure as was done at CFRMF. With the californium source, however, flux gradients will be considerable and the foil packages may have to be reduced significantly. The alternative of surrounding the source with an array of foils in a very lightweight fixture should be considered. Fission neutron fluences may be provided for the ILRR Program will be a matter of some serious compromise among conflicting priorities. Involved will be precision and accuracy of requested fluences, detector reactions and schemes of detection, facility scheduling at NBS and radiation exposure of personnel. It is suggested that a fluence range of 0.3 to 1.0×10^{13} is an appropriate region for the compromise.

Reaction Rates and Spectrum

Spontaneous fission in ^{252}Cf is a natural, well-defined, and isotropically emitting source of fast neutrons. The source is inexpensive, permanently available, and can be well-isolated for measurement. The neutron field intensity around the source may be established conveniently and without recourse to a nuclear cross section.

For the calibration of fission rate detectors involving fissile isotopes, the fission spectrum provides an independent calibration point in the high-energy range of fast reactor spectra. In this case an accurate knowledge of the fission spectrum shape is not necessary. For the calibration of reaction rate detectors with a threshold-like response, the fission spectrum is also relevant since it is the dominating component of fast-reactor spectra over the response range of these detectors. Here a difficulty may arise because of uncertainties in the relative shape of ^{235}U , ^{239}Pu , and ^{252}Cf fission spectra.

FIGURE NBS-1 NBS DOUBLE FISSION CHAMBER AND Cf-252 FISSION NEUTRON SOURCE

A large number of differential spectrum measurements for fission spectra exist in the literature, they cover a period of more than twenty years, and they employ just about every technique of neutron spectrometry ever attempted.¹⁻⁴ Predictably, there are discrepancies. A brief outline of the situation is necessary as will be apparent.

Measurements of fission spectrum shape are commonly fitted with the Maxwellian density function,

$$\chi(E, \bar{E}) = 3 \sqrt{\frac{3}{2\pi\bar{E}^2}} \bar{E}^{\frac{1}{2}} \exp\left(-1.5 \frac{E}{\bar{E}}\right), \quad (1)$$

so that an average energy parameter, $\bar{E}$, may be used to characterize individual spectrum measurements. For ^{252}Cf spontaneous fission, the observed average energy parameters vary from a low of $\bar{E} = 2.05$ MeV to a high of 2.35 MeV. The implied difference in neutron energy distributions for this range of parameters is shown in Table NBS-3. One way of reducing this parameter range is to presume that for the ^{235}U fission spectrum there exists a core of reliable measurements. Restricting consideration, then, to ^{252}Cf fission spectrum measurements which include ^{235}U , it is possible to specify an average energy parameter which is consistent with most of the measured spectra^{2,3} and also with theoretical prediction:⁴

$$\frac{\bar{E}(^{252}\text{Cf})}{\bar{E}(^{235}\text{U})} = 1.1 \pm 0.05; \quad \bar{E}(^{235}\text{U}) = 1.935 \text{ MeV}^{(4)}$$

$$\underline{\underline{\bar{E}(^{252}\text{Cf}) = 2.13 \pm 0.1 \text{ MeV.}}}$$

A discussion and a referencing of the above steps are not within the bounds of this report. It must be noted, however, that some well-documented time-of-flight measurements are excluded by this path of evaluation. Table NBS-3 shows integral flux spectra corresponding to the evaluated and to the extreme values of $\bar{E}$ in Eq. (1).

Table NBS-3

Integral Flux Spectra, $S(E)$, for ^{252}Cf Fission Neutrons

$$\left[S(E, \bar{E}) = \int_E^{\infty} \chi(E, \bar{E}) dE \right]$$

$E(\text{MeV})$	$\bar{E}(\text{MeV}) =$ 2.13±0.1 (evaluated)	$\bar{E}(\text{MeV}) =$ 2.05 (minimum)	$\bar{E}(\text{MeV}) =$ 2.35 (maximum)
0.25	0.95(±0.5%)	0.95	0.96
0.6	0.84(±1%)	0.83	0.86
1.4	0.58(±3%)	0.56	0.62
3	0.24(±8%)	0.22	0.28
6	0.038(±18%)	0.033	0.053

The uncertainty for the evaluated spectrum is seen to increase with energy. This is the result of applying directly to eq (1) the ± 0.1 MeV uncertainty assigned to the evaluated $\bar{E}$. It would be coincidental if this is actually consistent with the various experiments. More likely it is associated with the procedures used to impress eq (1) upon the experimental data. A comparison of point value tabulations, if they could be recovered, would be useful.

Table NBS-4 lists ^{252}Cf fission-spectrum-averaged cross sections for most of the reactions under study in the ILRR Program. Computations were carried out at the HEDL Laboratories with the Maxwellian of eq (1) set at $\bar{E} = 2.13$ MeV and with the HEDL cross section library. Also listed in the table are spectrum sensitivity factors corresponding to $\Delta\bar{E} = 0.1$ MeV.

Because of the inconclusiveness of the spectrum measurements, there is a sustained temptation to discard the fission spectrum as a fast-neutron calibration base. Such a step would not only ignore the attractive features of fission neutron sources mentioned at the beginning of this section, but would suppose that other specialized and less widely scrutinized neutron fields are more reliably established. In particular for the ILRR Program, it would assume incorrectly that high-energy spectrum components in some multiplying systems can be more accurately known than the fission spectrum itself. The difficulty of establishing fast-neutron energy distributions must be faced; it may be less expensive for engineering development programs like the ILRR to do this now rather than later.

Table NBS-4

Calculated Spectrum-Averaged Cross Sections for the
 ^{252}Cf Fission Spectrum (1)

<u>Reactions</u>	<u>Cross Section</u> <u>(millibarns)</u>	<u>Spectrum</u> <u>Sensitivity(2)</u>
<u>Primary fission</u>		
$^{235}\text{U}(\text{n}, \text{f})$	1231	< 0.1%
$^{238}\text{U}(\text{n}, \text{f})$	301	4 %
$^{237}\text{Np}(\text{n}, \text{f})$	1307	1.3%
$^{239}\text{Pu}(\text{n}, \text{f})$	1765	0.2%
<u>Primary non-fission</u>		
$^{238}\text{U}(\text{n}, \gamma)$	84.3	3 %
$^{197}\text{Au}(\text{n}, \gamma)$	93.2	4 %
$^{10}\text{B}(\text{n}, \alpha)$	445	1.8%
$^6\text{Li}(\text{n}, \alpha)$	364	$3\frac{1}{2}$ %
$^{115}\text{In}(\text{n}, \text{n}')$	190.8	3 %
$^{32}\text{S}(\text{n}, \text{p})$	68.5	8 %
$^{27}\text{Al}(\text{n}, \alpha)$	1.119	~25 %
$^{127}\text{I}(\text{n}, 2\text{n})$	1.714	
<u>Specialized for dosimetry</u>		
$^{63}\text{Cu}(\text{n}, \gamma)$	10.67	$2\frac{1}{2}$ %
$^{59}\text{Co}(\text{n}, \gamma)$	5.14	$4\frac{1}{2}$ %
$^{45}\text{Sc}(\text{n}, \gamma)$	6.07	
$^{58}\text{Fe}(\text{n}, \gamma)$	2.78	
$^{109}\text{Ag}(\text{n}, \gamma)$	11.35	
$^{181}\text{Ta}(\text{n}, \gamma)$	100.2	
$^{54}\text{Fe}(\text{n}, \text{p})$	87.9	$9\frac{1}{2}$ %
$^{58}\text{Ni}(\text{n}, \text{p})$	115.0	8 %
$^{46}\text{Ti}(\text{n}, \text{p})$	13.99	13 %
$^{47}\text{Ti}(\text{n}, \text{p})$	18.66	
$^{48}\text{Ti}(\text{n}, \text{p})$	0.427	
$^{55}\text{Mn}(\text{n}, 2\text{n})$	0.712	
$^{63}\text{Cu}(\text{n}, \alpha)$	0.730	
$^{238}\text{Pu}(\text{n}, \text{f})$	2513	0.8%
$^{232}\text{Th}(\text{n}, \text{f})$	75.8	5 %

Table NBS-4 (Cont'd)

<u>Reactions</u>	<u>Cross Section</u> (millibarns)	<u>Spectrum</u> <u>Sensitivity</u> (2)
<u>Additional for dosimetry</u>		
$^{115}\text{In}(n,\gamma)$	142.2	$2\frac{1}{2} \%$
$^{23}\text{Na}(n,\gamma)$	0.265	2 %
$^{31}\text{P}(n,p)$	36.1	$6\frac{1}{2} \%$
$^{56}\text{Fe}(n,p)$	1.645	21 %
$^{27}\text{Al}(n,p)$	4.93	
$^{63}\text{Cu}(n,2n)$	0.272	
$^{55}\text{Mn}(n,\gamma)$	3.49	
$^{24}\text{Mg}(n,p)$	2.45	

(1) Computed with HEDL cross section library and Eq. (1) set with $\bar{E} = 2.13 \text{ MeV}$.

(2) Approximate percent change in cross section for a 0.1 MeV change of $\bar{E}$ in Eq. (1).

The IAEA Specialist's Meeting on the Status of Prompt Fission Neutron Spectra, held in August 1971, made several recommendations which are appropriate for these issues:

High priority should be given to the determination of the Cf-252 fission-neutron spectrum to the best possible precision from a few keV to at least 10 MeV. The quality of the results should be such as to make it a "standard" fission-neutron spectrum.

It is recommended that data on fission neutron spectra should be transmitted by the measurers to their local Nuclear Data Centre in the form of tabulated point values as a function of energy.

Fundamental fission-spectrum-averaged, fission cross sections for ^{235}U and ^{238}U fission, and ^{235}U and ^{252}Cf fission spectra should be determined to an accuracy of $\pm 2\text{-}3\%$, preferably at more than one laboratory.

A goal of the ILRR Program--to achieve consistent results among participating laboratories for basic fission reaction rates--would be a step toward fulfilling these recommendations.

References for NBS Chapter

1. T. Wiedling, Microscopic Fission-Neutron-Spectra Measurements in "Neutron Standards and Flux Normalization," Proceedings of a Symposium, Argonne, Illinois, CONF701002 (1970).
2. J. A. Grundl, Fission Neutron Spectra: Macroscopic and Integral Results in "Neutron Standards and Flux Normalization," Proceedings of a Symposium, Argonne, Illinois, CONF701002 (1970).
3. IAEA Specialist's Meeting on the Status of Prompt Fission Neutron Spectra. (Aug. 1971). Proceedings to be published.
4. James Terrell, Phys. Rev., 113, 527 (1959).

OAK RIDGE NATIONAL LABORATORY

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

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

Objective

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

Summary

Batches of oxides and metals containing at least 200 g of each isotope ^{235}U , ^{238}U , ^{237}Np , and ^{239}Pu have been allocated by the AEC Division of Research for sole use in the various LMFBR associated dosimetry programs. Work is now in progress to form 20 samples of high purity metal from each of the isotopes noted. All oxide reductions to metal and subsequent rolling of 0.005-in. thick sheets of ^{235}U , ^{238}U , and ^{239}Pu are in progress or have been completed. Delivery of the ^{235}U and ^{238}U samples to HEDL is anticipated by March 31, 1972.

Accomplishments and Status

As agreed upon at the December 8, 1971, meeting with AEC Division of Research, Division of Reactor Development and Technology, HEDL, and ORNL personnel, five batches ^{235}U , ^{238}U , ^{237}Np , and ^{239}Pu of high assay have been received for exclusive use in LMFBR dosimetry programs. Table 1 lists these materials. One hundred grams of batches U-235-264C, U-ES-Z, and Np-21 (103 g) were transferred to Dow Chemical Company, Rocky Flats Division, for conversion to metal. The plutonium batch has already been reduced and is presently in inventory. All conversions to metal were completed and the metals returned to the ORNL Isotope Target Laboratory for processing into oxide-free metal foil.

ORNL TABLE 1
RESERVED ISOTOPES FOR LMFBR DOSIMETRY

Isotope	Form	Quantity, g	Assay, %	Batch No.
^{235}U	Oxide	206.42	99.887	U-235-264C
^{238}U	Oxide	300.00	99.999	U-ES-Z
^{237}Np	Oxide	103.00	99.99+	Np-21
	Oxide	200.00	99.9(?)	Np-5
^{239}Pu	Metal	209.648	99.11	453B

To date (March 1, 1972) the batches of ^{235}U and ^{238}U have been arc melted and etched six times each to remove oxide and calcium fluoride impurities from the reduced metals. After each arc melting, the metal buttons were scraped, filed, and chemically treated to remove the surface skin containing the undesirable impurities. The ^{235}U sample was initially hot rolled in stainless steel and finally cold rolled to the required 0.005-in. thickness. Storage of all metal samples in glass containers under an argon atmosphere is used as a routine procedure. The 0.500-in. diameter punch which will be used to form the circular metal samples has been received, and disk preparation will begin within the next few weeks.

Considerable effort has been expended in developing a suitable method for making a leak-tight seal on the 0.007-in. thick aluminum envelopes which will contain the radioactive dosimeter disks described above. Using 99.999% aluminum sheet (contained impurities being determined by HEDL using activation analysis), AC arc welding, plasma arc welding, ultrasonic welding, and cold pressure welding methods have been evaluated. Of these methods, arc welding has proven totally unsatisfactory. Some marginal success has been achieved with plasma arc welding, but the uneven contour of the edge (diameter and thickness) resulting from this process has been discouraging.

Both the ultrasonic welding and cold pressure welding procedures have resulted in apparently perfect welds. Unfortunately, the ultrasonic welding requires expensive equipment (~\$40,000) which is not obtainable because of limited budgets. However, the cold pressure welding procedure requires simply a die and fixture which has been designed and constructed on site. Using pressures of >100,000 psi leak tight pressure seals have been routinely accomplished while maintaining the close tolerances for the overall contained sample dimensions. A further advantage of easy manipulation of the welder in an inert atmosphere glove box is inherent in the process.

Encapsulation of disks of calcium metal by the cold pressure method has proven quite satisfactory. Resultant samples have been shelf tested in water at room temperature for 64 hours and then boiled for 20 min and rapidly cooled in cold water. Several samples were opened and no corrosion or leakage was noted. Other samples are still in storage under water and show no visible signs of deterioration after 240 hours. All samples passed leak testing in vacuum under ethylene glycol--a sensitive bubble test.

At present no plans have been made to produce archive samples of the oxides or metals used in this program, since the budgets are inadequate for this purpose. Similarly, 100 g of the ^{239}Pu metal (Batch 453B) is supposed to be converted to oxide for use in the EBR-II dosimeter program, but this will be postponed until monies become available.

Expected Achievements

Within March 1972 all samples of ^{235}U and ^{238}U metal disk dosimeters will be completed and encapsulated in aluminum. However, encapsulation will not begin until HEDL analysis of the aluminum material is completed (in progress). Samples of ^{239}Pu should be completed early in April or sooner and finally the ^{237}Np samples will be produced and available by late April or early May. Depending upon the extent of future funding, quality assurance and archive sample programs will be implemented.

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