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LMFBR
REACTION RATE
AND DOSIMETRY
QUARTERLY PROGRESS REPORT
JUNE, JULY, AUGUST 1971

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LMFBR
REACTION RATE
AND DOSIMETRY
QUARTERLY PROGRESS REPORT

JUNE, JULY, AUGUST 1971

Compiled by:
W.N. McElroy
September 1971

HANFORD ENGINEERING DEVELOPMENT LABORATORY
UNITED STATES ATOMIC ENERGY COMMISSION

Operated by

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ABSTRACT

This report was prepared at the Hanford Engineering Development Laboratory operated by WADCO Corporation, 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 LMFBF Reaction Rate Program in June, July and August 1971.

LMFBR Reaction Rate and Dosimetry Quarterly Progress Report
June, July, August 1971

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PREVIOUS REPORTS IN THIS SERIES

None

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NATIONAL BUREAU OF STANDARDS

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LMFBR Reaction Rate and Dosimetry Quarterly Progress Report

June, July, August 1971

FOREWORD

This report describes progress in the USAEC-sponsored Interlaboratory LMFBR Reaction Rate (ILRR) program. This program has been established by 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.

The accuracy goals of 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 accepted 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.

ACKNOWLEDGEMENTS

The following organizations are presently participating in the Inter-laboratory 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)
General Electric Company (GE)
Hanford Engineering Development Laboratory (HEDL)
Lawrence Livermore Laboratory (LLL)
Los Alamos Scientific Laboratory (LASL)
National Bureau of Standards (NBS)
Oak Ridge National Laboratory (ORNL)
Stanford University (SU)
University of California at Santa Barbara (UCSB)

Coordination activity and publication of a Quarterly Progress Report have been assigned to the Fast Reactor Materials Dosimetry Center (FRMDC) at HEDL.

Contributions from participating organizations are included in this report exactly as submitted with no editing or revisions by HEDL.

SUMMARY

AEROJET NUCLEAR COMPANY

Preliminary investigations have begun on several areas of concern to the ILRR program. Further flux gradient measurements employing both activation and fission chamber techniques are underway with preliminary results indicating the lack of any serious problem in the region of interest. Power level normalization procedures are being looked at by comparison of the normal reactor instrumentation (gamma-compensated ion chamber in the thermal driver region) output to the count rate observed in a ^{235}U fission chamber in the measurement position. So far these results indicate a one-to-one correspondence to within the accuracy of the fission chamber.

ARGONNE NATIONAL LABORATORY

A work plan for development, testing, and performance of fission rate measurements in CFRMF by the solid-state track-recorder method has been completed. A prototype track-recorder irradiation package suitable for the space limitations of CFRMF has been built. All gamma-ray detectors to be used for assay of irradiated foils have been recalibrated relative to IAEA standards.

ATLANTIC RICHFIELD HANFORD COMPANY

Previous interlaboratory tests have provided a calibration basis for assessing and improving the accuracy of gamma spectrometry measurement of reaction rates. Fission and non-fission foils are expected to be irradiated in the CFRMF facility before the end of December 1971 for the ARHCO measurements.

ATOMICS INTERNATIONAL

The initial contribution from Atomics International in the ILRR program will be to determine the integral (n, α) cross sections of ^6Li and ^{10}B by mass spectrometric measurement of the helium generated in small specimens. The first cooperative experimental irradiation will be conducted in the CFRMF facility at Idaho Falls.

The effort to date has been centered about the problem of encapsulating lithium and boron in a form compatible with methods subsequently used to extract the helium for measurement. After this has been resolved, materials will be procured and specimens will be irradiated for the maximum feasible duration, possibly at the same time and in the same assembly used by HEDL for dosimetry measurements.

BROOKHAVEN NATIONAL LABORATORY

No report this quarter.

GENERAL ELECTRIC COMPANY

No report this quarter.

HANFORD ENGINEERING DEVELOPMENT LABORATORY

Work is in progress at HEDL associated with the proper acquisition of reaction rates, flux-spectra, fluence, and burnup as related to the CFRMF program and LMFBR fuels and materials dosimetry. For the CFRMF and other ILRR tests, this work includes (1) the acquisition, quality assurance, and distribution of foil materials for radiochemistry measurements, (2) the preparation and distribution of a ^{140}Ba standard for radiochemistry measurements of fission rates, (3) the measurement of fission and non-fission reaction rates by radiochemistry, (4) the foil reaction cross sections that will be used for calculation of reaction rates and subsequent multiple foil definition of flux-spectra, and (5) a Monte Carlo technique being developed and used for the assignment of errors to the multiple foil results.

LAWRENCE LIVERMORE LABORATORY

During the past six months, this Laboratory has begun to accumulate and develop the necessary computer programs needed to calculate cross sections for neutron-induced reactions. The Laboratory can now handle the compound nucleus part of the cross sections, and expects to begin on the precompound calculations in the near future.

LOS ALAMOS SCIENTIFIC LABORATORY

It has been concluded that, in addition to reactor power level instrumentation, ^{235}U and ^{238}U fission foils should be used in the immediate vicinity of the CFRMF central test region to establish proper power normalization factors between reactor runs. Furthermore, before each irradiation, the reactor power level instrumentation should be checked against the response of a fission chamber placed in the central test region.

NATIONAL BUREAU OF STANDARDS

A double fission ionization chamber, convenient to disassemble and of minimum size and mass, has been constructed and the performance characteristics examined. For fission foils $200\text{ }\mu\text{g/cm}^2$ thick, the extrapolation-to-zero from just above the alpha pulse-height cut-off is less than 1%. A uranium weight ratio between normal and highly enriched uranium fission foils was measured by two independent methods, alpha counting and thermal-neutron-induced fission counting. Agreement between the two methods is better than 1%. Flux perturbations at the center of the CFRMF due to the presence of the fission chamber are predicted to be about 1% or less.

OAK RIDGE NATIONAL LABORATORY

No report this quarter.

STANFORD UNIVERSITY

No report this quarter.

UNIVERSITY OF CALIFORNIA AT SANTA BARBARA

Reported with HEDL this quarter.

AEROJET NUCLEAR COMPANY

AEROJET NUCLEAR COMPANY

A. CFRMF ILRR PROGRAM IRRADIATIONS SCHEDULE

J. J. Scoville

Objective

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

Summary

No experimental requests have been received from ILRR participants.

Accomplishments and Status

None

Expected Achievements

Given two to three weeks lead time, it is expected that any experimenter's request for time can be accommodated. However, requestors should give both a first and second choice, in the event that two or more requests are received for the same time period.

B. CFRMF FLUX-SPECTRAL, POWER LEVEL, GRADIENT DETERMINATIONS

J. J. Scoville, J. W. Rogers, and D. A. Pearson

Objective

In order that the over-all objectives of the ILRR program be accomplished, an accurate assessment of the test region environment of the CFRMF must be available. Important parameters are the shape of the neutron spectrum, flux gradient around the central test region, and the reproducibility of a given power level or multiples of it. These parameters need accurate identification in order that the results from different experimental procedures employed by the various laboratories involved in the ILRR program may be compared. The best possible information on these parameters will be obtained and disseminated.

Summary

Previously determined spectral information has been provided to each investigator and is included in the HEDL Chapter of this report. Preliminary measurements of flux gradients in the test region have been made, and indicate that any correction for this effect will be very small. However, more detailed investigations are planned. Power level stability during the course of an irradiation is well within acceptable limits due to basic design of the CFRMF static reactivity measurement control system. However, reproducibility of power level from day to day or month to month or obtaining accurate multiples of power level from experiment to experiment must still be investigated. Preliminary results are available which compare the output of the usual gamma compensated ion-chamber in the thermal driven region to the count rate of a ^{235}U fission chamber in the central test region.

Accomplishments and Status

Vertical flux gradient information heretofore has consisted of activation experiments using thick gold foils. Recent investigations using thin ^{235}U foils have been made and are compared to traverse measurements employing a ^{235}U fission chamber. These data are portrayed graphically in Figure 1. These data are still preliminary and will be compared to future work using threshold detectors both as foils and in fission chambers. However, the magnitude of the gradient in the small region of interest to ILRR measurements does not appear to be too distressing.

A very first attempt at comparing power level as measured by the ion chamber in the thermal driver core region with that of a fission chamber (^{235}U) positioned in the CFRMF test region is shown in Table I below.

Table ANC-I

Power Level Correlation

<u>Ion Chamber Signal</u>	<u>% Change from Previous Signal</u>	<u>Fission Chamber Count Rate</u>	<u>% Change from Previous Signal</u>
556	- - -	29918	- - -
440	20.9	23638	21.0
200	54.5	10890	53.9
90	55.0	4812	55.8
0 (sub-critical)		4	- - -

More measurements of this type will be done, particularly at higher power levels. These data must also be correlated with activation data obtained from monitor foils.

Expected Achievements

It is expected that gradient measurements with threshold foils and fission chambers should, together with the current information, adequately define vertical gradient effects. Further work is planned for radial gradient studies, however, it has been delayed until approval is granted for 10 kw operation of the facility. The higher power level will give greater activity to some of the threshold detectors to be used. An

Figure ANC-1. CFRMF Relative Reaction Rate (arbitrary unit)

effort is also underway to do some 1-dimensional transport theory studies of the gradient which can be compared to the measured effect.

Additional correlation of fluence measurements in the test region with power level as determined by the ion chamber needs to be done. It is expected that most experimentors will rely on the ion chamber level indication, but that monitor foils in the test region will also be used as a back-up verification of the total fluence. Also, some of the NBS fission chambers should be studied relative to the ion chamber current.

ARGONNE NATIONAL LABORATORY

ARGONNE NATIONAL LABORATORY

A. CFRMF REACTION RATES BY TRACK RECORDER METHOD

R. J. Popek and N. D. Dudey

Objective

The objective is to determine absolute fission rates of ^{235}U , ^{238}U , ^{237}Np , and ^{239}Pu in the CFRMF spectrum by means of solid-state track recorders. The goal is to achieve fission-rate measurements to an accuracy of $\pm 3\%$ (1 σ); however, an important objective is to define the true accuracies obtainable by this technique and to attempt to improve the technique to approach the accuracy goal.

Accomplishments and Status

The use of solid-state track recorders (SSTR) for measuring absolute fission rates has been demonstrated to be capable of 1-3% accuracy under ideal conditions.¹ These advances make SSTR's an attractive technique for in-reactor fission-rate measurements; however, the accuracies achievable in reactor environments have yet to be demonstrated. We have recently completed a series of SSTR measurements in critical assemblies, as part of the EBR-II mock-up experiments conducted in ZPR-3. A report on the first of these experiments² describes the use of SSTR's. Subsequently, results of SSTR fission-rate measurements have been compared with radiochemical fission-rate measurements and, in some cases, discrepancies as large as 12% were observed. These discrepancies can be attributed to a number of causes, e.g., errors associated with fast neutron fission yields, counting efficiencies, and the nuclear decay schemes used in the data analysis, but errors associated with SSTR's cannot be ruled out.

In order to meet the $\pm 3\%$ accuracy goal, it will be necessary to investigate the SSTR technique thoroughly. On the basis of the ZPR-3 results, we must examine all potential sources of error which may occur

in large, isotropic neutron fields, such as the variations in the optical efficiency of track recorders, as a function of both fission-fragment energy and angle incident upon the track recorder. A scattering chamber is being designed so that these optical-efficiency variables can be examined. A ^{252}Cf source has been obtained and fission fragments from this source will be collimated to produce a beam which can be degraded by absorbers and to which track recorders can be exposed at selected angles normal to the fragment beam.

A prototype irradiation package, which will hold the mica track recorders and the fissile material in intimate contact and meet the space requirements for the CFRMF tests, has been built. This package will be evaluated by sending a ^{235}U track-recorder set to J. Grundl for exposure in one-half of his fission chamber. This test will provide a direct comparison between the SSTR and a fission chamber.

Expected Achievements

The essential milestones of the program for track recorder measurements in CFRMF are outlined below.

1. Pre-CFRMF Irradiation Preparation
 - a. Prepare standard solutions of ^{235}U , ^{238}U , ^{237}Np , and ^{239}Pu
 - 1) Establish fissile-atom concentrations
 - 2) Electroplate nanogram amounts onto platinum
 - b. Design and build SSTR irradiation package
 - 1) Expose SSTR package in back-to-back configuration with Grundl's fission chamber
2. Irradiations and Results
 - a. Expose one set each of ^{235}U , ^{238}U , and ^{239}Pu in CFRMF
 - 1) Define amounts of fissile material, times, and accuracy relative to other ILRR measurements
 - b. Measure SSTR optical efficiencies vs angle and energy
 - c. Irradiate SSTR back-to-back with massive foil for absolute fission-yield and fission-rate determinations

- d. Analyze CFRMF fission-rate data (including error analysis)

Man-power limitations imposed by our work on the EBR-II flux mapping experiments will prohibit further work on this program until October.

B. CFRMF REACTION RATES BY RADIOCHEMISTRY

R. R. Heinrich and N. D. Dudey

Objective

The objective is to measure radiochemically the reaction rates of foils irradiated in CFRMF. The primary emphasis will be to determine the fission rates of ^{235}U , ^{238}U , ^{237}Np , and ^{239}Pu from massive foils irradiated simultaneously with track recorders. A secondary objective will be to provide reaction-rate measurements on dosimetry foils to complement the HEDL foil measurements.

Accomplishments and Status

A calibration of all Ge(Li) and NaI(Tl) detectors has been completed using IAEA absolute gamma-ray standards and an Amersham/Searle ^{140}Ba standard. The nuclide, energy, quoted accuracy, branching ratio, and half-life of each standard is presented in Table ANL-1. In the energy range 250-1350 keV, these standards agreed with previous calibrations to within $\pm 2\%$. The detector efficiencies have been incorporated into the computer-code library used for analysis of all Ge(Li) gamma-ray counting data. Our procedure for determining fission rates from irradiated foils will be identical to the procedures we used in recent critical assembly experiments.²

Expected Achievements

Man power limitations resulting from the EBR-II mapping study will prohibit us from further work on this program until October. If HEDL is able to supply the fissile foil material by October, we expect to begin foil irradiations in CFRMF in November.

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1. R. Gold, R. J. Armani, and J. H. Roberts, "Absolute Fission Rate Measurements with Solid State Track Recorders," Nucl. Sci. Eng. 34, 13-22 (1968).
2. N. D. Dudey, et al, "Activation Rate Measurements in the ZPR-3 Mock-up Critical Experiments," Part I, ANL-7781 (1971).

TABLE ANL-1
GAMMA-RAY STANDARDS FOR DETECTOR CALIBRATION

Nuclide	Energy (keV)	Quoted Accuracy (% at 2 σ)	γ /D (%)	Half-life
⁵⁷ Co	122.1	± 1.0	85.0	2.716 d
²⁰³ Hg	279.2	± 0.9	81.6	46.8 d
¹⁴⁰ La	328.8	± 1.7	n.c.	12.8 d
¹¹³ Sn	391.8	± 1.1	64.2	115.2 d
¹⁴⁰ La	487.0	± 1.7	n.c.	12.8 d
¹⁴⁰ Ba	537.3	± 1.7	n.c.	12.8 d
¹³⁷ Cs	661.6	± 1.3	85.1	29.90 y
¹⁴⁰ La	815.8	± 1.7	n.c.	12.8 d
⁵⁴ Mn	834.8	± 1.0	100.0	312.6 d
⁸⁸ Y	898.0	± 1.3	91.4	107.4 d
⁶⁰ Co	1173.0	± 0.6	99.87	5.275 y
²² Na	1274.6	± 0.9	99.95	2.602 y
⁶⁰ Co	1332.0	± 0.6	99.999	5.275 y
¹⁴⁰ La	1596.2	± 1.3	n.c.	12.8 d
⁸⁸ Y	1836.1	± 1.3	99.4	107.4 d

n.c. Not quoted by supplier

ATLANTIC RICHFIELD HANFORD COMPANY

ATLANTIC RICHFIELD HANFORD COMPANY

A. CFRMF REACTION RATES BY NON-DESTRUCTIVE ENERGY ANALYSIS

W. H. Zimmer

Objective

The non-destructive measurement of the reaction rates of fission and non-fission foils irradiated in CFRMF as a complement to measurements made by HEDL and other participants will be performed.

Summary

Previous interlaboratory tests have provided a calibration basis for assessing and improving the accuracy of gamma spectrometry measurement of reaction rates. Fission and non-fission foils are expected to be irradiated in the CFRMF facility before the end of December 1971 for the ARHCO measurements.

Accomplishments and Status

Previous ARHCO-HEDL work⁽¹⁻⁷⁾ involving interlaboratory gamma spectrometry determinations of fission and non-fission foil reaction rates for EBR-II and other dosimetry tests have provided a calibration basis for assessing and improving the accuracy of reaction rate measurements. The gamma spectrometry results obtained by ARHCO for previous interlaboratory tests and the procedures that will be used for the CFRMF and other ILRR tests are discussed in references 2, 8, and 9.

The EBR-II 31F dosimetry test documentation provides the most recent ARHCO radiochemistry results for comparison with other fission and non-fission foil radiochemistry results.⁽²⁾ The APFA-III⁽⁶⁾ and ECEL Core-16⁽⁷⁾ test results compare ARHCO radiochemistry results with those of other laboratories as well as with fission track recorders.

Expected Achievements

A fission and a non-fission foil set provided by HEDL are expected to

be irradiated in the CFRMF by late December 1971 for the ARHCO measurements.

References for ARHCO Chapter

1. H. H. Yoshikawa and W. H. Zimmer, "Fast Neutron Dosimetry: Intercalibration of Counting Facilities," USAEC Report HW-81871, General Electric Company (1964).
2. L. S. Kellogg and W. H. Zimmer, "EBR-II Dosimetry Test Reaction Rate Measurements," BNWL-1403, Battelle-Northwest (1970).
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ATOMICS INTERNATIONAL

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 LMFBR Reaction Rate (ILRR) Program is to determine integral (n, α) cross sections of ^6Li , ^{10}B , and possibly ^{27}Al and other isotopes, in a variety of well-established and permanent neutron fields, including several that cover the neutron energy range of interest for FTR and LMFBR's. The 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

The major goal of the ILRR program is to establish reliable and accepted values of several absolute integral reaction rates and ratios in various well-established and permanent neutron fields. These integral values, obtained by radiochemistry or by mass spectrometry, will then be used to adjust existing energy-dependent (differential) cross sections. The initial contribution from Atomics International will be to determine the integral (n, α) cross sections of ^6Li and ^{10}B by mass spectrometric measurement of the helium generated in small specimens. The first cooperative experimental irradiation will be conducted in the CFRMF facility at Idaho Falls.

The effort to date has been centered about the problem of encapsulating lithium and boron in a form compatible with methods subsequently used to extract the helium for measurement. After this has been resolved, materials will be procured and specimens will be irradiated for the maximum feasible duration, possibly at the same time and in the same assembly used by HEDL for dosimetry measurements.

Accomplishments and Status

The initial contribution to the ILRR program by Atomics International will be to study the integrated (n, α) cross sections of ^6Li and ^{10}B in the CFRMF neutron spectrum. The neutron flux in CFRMF, apart from being quite similar to that anticipated in a typical LMFBR core, is one of the more intense of those under consideration for irradiations in the program, and is therefore rather suitable for the first round of experiments.

Two "standard" (n, α) cross sections to be measured are for the $^6\text{Li}(n, \alpha)^3\text{H}$ and $^{10}\text{B}(n, \alpha)^7\text{Li}$ reactions. Gram quantities of separated ^6Li and ^{10}B will be procured with the highest enrichments available. To be sure that all the helium generated in these isotopes is retained for the mass spectrometric measurements, the milligram size specimens will be sealed under vacuum in thin capsules. The material to be used for the capsules has not been selected but it must be compatible with the contents, and must have a relatively low (n, α) cross section since present plans call for the entire capsules and their contents to be included in the mass spectrometric analysis. Identical sealed capsules without ^{10}B or ^6Li would be included for blank measurements.

Lithium's low melting point (179°C) makes this element unattractive to encapsulate in metallic form. Advantages and drawbacks of using various available enriched lithium compounds (LiF , LiH , LiD , Li_2CO_3) are now being considered. Considerations include elemental purity, stoichiometry, (n, α) cross sections of the other elements in the compound, available isotopic enrichments and mass spectrometer interferences. Boron and lithium compounds in crystalline form are preferable to powders because the crystals will also tend to contain the generated helium.

In the ILRR program, some of the experiments will be performed simultaneously while others will require separate reactor runs. The integral experiments of AI and HEDL both require the maximum feasible exposure (8 or more hours), and will be combined if flux depression (self-shielding) calculations at HEDL show that this can be accomplished without significant interference. To minimize the differences in the neutron spectrum experienced by the various experiments as a result of neutron scattering and absorption, it was agreed at the first planning session held at Idaho Falls on June 25, that each experimental assembly should be as similar as possible. The NBS aluminum fission chamber was the most bulky and least changeable, and was therefore chosen. Materials and plans needed to mock-up the fission chamber have been received from NBS, although they may not be needed if the Atomics International capsules are included in HEDL's assembly.

The possibility of using CFRMF at some later date for the determination of other (n, α) cross sections is under consideration. Of particular interest is the $^{27}\text{Al}(n, \alpha)^{24}\text{Na}$ reaction, because its high threshold energy will result in aluminum being a useful detector in an energy range (>6 Mev) where little is known about the ^6Li and ^{10}B (n, α) cross sections. Comparison of this integral cross section obtained by mass spectrometry with the same entity obtained by ^{24}Na radiochemistry at HEDL will also be of considerable interest.

The (n, α) cross section of aluminum in the CFRMF neutron spectrum is however, four orders of magnitude less than those of ^6Li and ^{10}B . To obtain an accuracy of 2-3% in a helium concentration measurement by mass spectrometer, however, a sample should contain at least $\sim 10^{11}$ atoms of ^4He . Preliminary calculations indicate that 5 mg samples of ^6Li and ^{10}B will produce this quantity after a CFRMF irradiation lasting 8 hours. For aluminum, on the other hand, 100g would be needed to generate this amount of helium. At present the mass spectrometer system can extract helium from samples weighing no more than 10 mg. A new high temperature furnace now being tested, however, should be able to accept samples up to 100 mg. Therefore, if the (n, α) cross section of aluminum is to be determined, 100 mg samples would have to be used, with a considerable loss of accuracy in the helium measurement. Even if the power of CFRMF is raised, as anticipated, to 10 kw (flux $\sim 10^{11}$ n/cm²-sec) the resulting number of ^4He atoms would be only $\sim 10^9$, which is barely

enough to give an absolute accuracy of 10%. In later experiments, where longer irradiations are made and after the mass spectrometer sensitivity has been increased by improvements planned for later this year, this accuracy should be improved, especially if several duplicate specimens are analyzed.

Expected Achievements

After the problems of encapsulation have been resolved, the short-term goal of obtaining (n, α) cross sections of ^{10}B and ^6Li integrated over the CFRMF neutron spectrum should present no difficulties. By the next reporting period the separated isotopes will have been procured and converted to a form suitable for encapsulation and irradiation. The material for encapsulation will have been selected and the capsules fabricated. Various analyses to characterize the materials will have been initiated.

After the irradiation, the small amounts of helium that are generated in milligram-size specimens will be measured precisely by high sensitivity gas mass spectrometer. These 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 the same integral cross sections obtained in this program by using radiochemical techniques, and both will then be reconciled with available energy-dependent (n, α) cross section data, providing the latter with an independent and absolute normalization. The sensitivity and absolute accuracy of the mass spectrometer system are expected to meet the requirements for the program, especially for irradiations in the higher-flux test reactors. The outcome of the ILRR program should go far in helping resolve some present inconsistencies in dosimetry.

**HANFORD ENGINEERING DEVELOPMENT
LABORATORY**

HANFORD ENGINEERING DEVELOPMENT LABORATORY

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

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

Objectives

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

Aluminum encapsulated metal fission foils will be obtained from the Isotope Target Center at ORNL. A formal request was submitted to the AEC to establish a pool of highly enriched fission foil material at ORNL for use by the ILRR program and for routine LMFBR dosimetry.

For non-fission foils, four suppliers are possibly qualified from the 13 investigated. Three separate Al alloy materials (Au, Ag and Ta) have been purchased. A preliminary activation analysis in NBS reactor RT-4 position together with the results of other analytical techniques shows the Au and Ag alloy content differ drastically from supplier certified values.

Accomplishments and Status

Twenty ^{235}U , ^{238}U , ^{237}Np , and ^{239}Pu aluminum encapsulated fission foils will be fabricated by ORNL for the CFRMF and other ILRR tests. It is anticipated that some of these foils can be developed as standard foils for

reuse in subsequent ILRR tests, such as in the NBS fission and Californium standard neutron fields.

These foils will be fabricated from a pool of highly enriched fissionable material that is to be established at ORNL for the ILRR program and routine LMFBR dosimetry. Traceable controlled archive samples will be maintained at ORNL. A formal request was made to the AEC to establish this pool of material at ORNL. A 99.9+% enrichment was requested for ^{235}U and ^{239}Pu and 99.999+% for ^{238}U and ^{237}Np . Table 1 provides a listing of the amounts of these as well as other materials requested for the fissionable material pool.

Table HEDL-1

ENRICHMENTS AND QUANTITIES OF FISSIONABLE
MATERIALS NEEDED TO MEET EXISTING PROGRAM REQUIREMENTS

<u>Material</u>	<u>Enrichment</u>	<u>Quantity (grams)</u>
^{235}U (Oxide)*	99.9+%	150
^{235}U (5 mil Metal)	99.9+%	50
^{239}Pu (Oxide)*	99.9+%	150
^{239}Pu (5 mil Metal)	99.9+%	50
^{238}U (Oxide)*	99.999+%	150
^{238}U (5 mil Metal)	99.999+%	50
^{237}Np (Oxide)*	99.999+%	150
^{237}Np (5 mil Metal)	99.999+%	50
^{238}Pu (Oxide)*	99.999+%	50
^{240}Pu (Oxide)*	99.999+%	10
^{241}Pu (Oxide)*	99.9+%	10
^{242}Pu (Oxide)*	99.999+%	5

* As U_3O_8 , NpO_2 , or PuO_2 .

Metal foil ~5 mil thick, 0.5" in diameter, weighing ~0.3 grams (with the exact weight of the metal specified to well within $\pm 0.5\%$ at the 95% confidence level), and encapsulated in 5 to 10 mil thick high purity aluminum with a finished outside diameter of 0.75" was specified for the ILRR program. The ^{239}Pu and ^{237}Np foils will be sealed by welding while the ^{235}U and ^{238}U foils will be mechanically sealed. Detailed specifications

and QA procedures are being developed with ORNL for these foils and the fissionable material pool. It will take approximately three months to acquire the 80 aluminum encapsulated foils for the ILRR program.

Two hundred (200) feet each of 0.092 wt %, 99.988% pure Ag/Al 20 mil wire and 0.096% Ta/Al 99.984% pure 20 mil wire and 500 feet of 0.093 wt % Au/Al 99.916% pure 20 mil wire were purchased. The values given above are the listed content and purity certified by the supplier.

An interlaboratory HEDL and NBS cooperative effort resulted in the selection and initial utilization of the NBS reactor's out-of-core RT-4 position for bare and cadmium covered thermal flux irradiations for QA work on these materials. The thermal flux level is $\sim 1.3 \times 10^{13} \text{ n/cm}^2 \cdot \text{sec}$ and the thermal to fast flux ratio is very high as a result of D₂O moderation. The RT-4 rabbit tube position is expected to become a well established and permanent NBS facility that can be used for high fluence thermal neutron irradiations, calibration, and QA studies.

Twelve (12) $\sim 1/4$ " segments of the Ag/Al and six (6) $\sim 1/4$ " segments of Ta/Al were taken throughout the material roll and were packaged and irradiated for one hour, 25 seconds in the NBS reactor RT-4 position. Reference standards of deposited pure AgNO₃ and pure Ta foil were also included.* The observed Ag content differed from the certification by $\sim 58\%$ (Table 2) while the Ta content differed by $\sim 10\%$. Homogeneity appears satisfactory for both materials as maximum ranges between results were $\sim 3.5\%$ in both instances, and counting precision due to the activity level was $\sim 1.3\%$, but further work is planned.

The new Au/Al material was not initially prepared for QA, and the ten samples were placed in five packages that were irradiated from ~ 30 minutes to ~ 8 hours in the NBS reactor RT-4 position. A 0.13% Au/Al 20 mil wire (previously characterized at another laboratory) was placed as an internal reference standard with each sample of alloy being analyzed. The observed

* Corrections for neutron self shielding in the standards have yet to be made but are expected to be small.

Table HEDL-2

OBSERVED ALLOY COMPOSITION WT %
ACTIVATION ANALYSIS

(Relative to Internal Standards)

<u>Ag/Al</u>	<u>Ta/Al</u>	<u>Au/Al</u>
0.145		0.173
0.147	0.0877	0.171
0.147	0.0857	0.176
0.145	0.0857	0.172
0.145	0.0865	0.173
0.146	0.0882	0.173
0.144	0.0867	0.173
0.146		0.176
0.145		0.173
0.146		0.172
0.145		
0.142		
$\bar{x}$ 0.145 + 0.004	$\bar{x}$ 0.0867 + 0.003	$\bar{x}$ 0.173 + 0.005
@ 99% Conf. Level	@ 99% Conf. Level	@ 99% Conf. Level
<u>Vendor Certified</u> <u>Composition</u>	<u>Vendor Certified</u> <u>Composition</u>	<u>Vendor Certified</u> <u>Composition</u>
<u>0.092 wt. %</u>	<u>0.096 wt. %</u>	<u>0.093 wt. %</u>

Au content of the new material appeared to differ from the certified value by ~86%.

The large deviation from certified values of both the Ag and Au prompted additional analysis. The new observed value of the Ag content was confirmed at another laboratory. Preliminary analysis of the new Au/Al material by other analytical techniques was done by extraction and subsequent spectrophotometric and X-ray analysis of the extracted Au and by atomic absorption analysis of the dissolved material.

The average value from each technique is summarized in Table 3. The first value listed for the Au/Al (0.093%) alloy is high, as are the corresponding relative values in Table 2, because they are based on the original value of 0.13% for the Au internal standard.

Table HEDL-3

SUMMARY OF ALLOY COMPOSITION WT% ANALYSES
BY VARIOUS TECHNIQUES

Material	Vendor Certification	Observed Wt. %
Au/Al	0.093	0.173 ^(a)
		0.137 ^(b)
		0.132 ^(c)
		0.138 ^(d)
		0.131 ^(e)
Ag/Al	0.092	0.145 ^(a,f)
		0.149 ^(b)
Ta/Al	0.096	0.087 ^(a)
		0.0851 ^(b)

- (a) Activation - compared with internal reference standards (0.13% Au/Al, AgNO₃, 0.5 mil Ta).
- (b) Activation - based on reference to NBS SMR-953 (0.116% 20 mil Co/Al wire) and thermal cross section values for ⁵⁹Co(n,γ)⁶⁰Co, ¹⁹⁷Au(n,γ)¹⁹⁸Au, ¹⁰⁹Ag(n,γ)^{110m}Ag and ¹⁸¹Ta(n,γ)¹⁸²Ta (37.2 ± 0.6, 98.8 ± 0.3, 4.1, and 20.7 barns, respectively).
- (c) Spectrophotometric analysis of the extracted Au.
- (d) X-ray analysis of the extracted Au.
- (e) Atomic Absorption analysis of the dissolved alloy.
- (f) Value confirmed (within ~3%) by another laboratory.

In addition to the internal standards of Au, Ag, and Ta already mentioned, pieces of NBS Standard Reference Material (SMR-953, 0.116% Co/Al, 20 mil wire) had been included in the irradiation packages. Relative to this standard and accepted values of thermal cross sections, values of 0.137%, 0.149%, and 0.0851% were obtained for the Au (0.093%), Ag (0.092%), and Ta (0.096%) alloys, respectively. For the 0.13% Au alloy used as an internal standard for gold, a value of 0.104% was obtained.

Relative to the NBS cobalt standard, the observed Au content differed by ~50% from that certified by the supplier.

The agreement among the results listed in Table 3 establishes increased confidence in the present accuracy of the various nuclear parameters and counting and calibration techniques being used for Au, Ag, and Ta.

While the above materials can now be used even though only partially characterized, these test data indicate the absolute necessity of a QA program on all materials used for Fast Reactor Dosimetry and the ILRR program. More extensive tests will be required to fully characterize homogeneity and establish final numbers for the Au, Ag, and Ta alloys.

Expected Achievements

By the end of the next reporting period, the acquisition and fabrication of fission and non-fission foils to be used for the CFRMF test should have been completed. Results of preliminary QA should also be available.

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

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

Objective

- 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. (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

^{140}Ba has been selected as a reference reaction rate standard for fission reactions. The HEDL Radiochemistry Standards Laboratory will provide a $^{140}\text{Ba}/^{140}\text{La}$ standard solution, calibration $\leq 2\%$ (95% confidence level), for the first phase study of the CFRMF program. Approximately two months' lead time will be required.

Accomplishments and Status

^{140}Ba has been selected as the reference reaction rate standard for correlation with total fission measurements using fission chamber and fission track techniques. It will thus serve to form part of the basis for readjusting evaluated fission cross sections. This selection was made because of the large amount of past work done on the $^{235}\text{U}(n,f)^{140}\text{Ba}$ reaction, the apparent minimal difference in ^{140}Ba fission yield (less than $\sim 2\%$) between thermal and different fast environments, $(1,2)^*$ and the relative ease of measurement using the ^{140}La daughter gamma spectrum.

* See page HEDL-26 for list of references

A $^{140}\text{Ba}/^{140}\text{La}$ standard solution will be supplied by the HEDL Radiochemistry Standards Laboratory for the CFRMF fission foil measurements within three months. The source material will be purchased, radiochemically purified, calibrated to $\sim 2\%$ (95% confidence level) and packaged in flame sealed glass ampoules. A sample will be sent to the NBS Radioactivity Section for a reference measurement in their " $4\pi\gamma$ " ionization chamber. This measurement could then be used to accurately compare the HEDL $^{140}\text{Ba}/^{140}\text{La}$ calibration with a NBS certified $^{140}\text{Ba}/^{140}\text{La}$ standard prepared later.

Six of ten vendors of ^{140}Ba listed in the 1971 Nuclear News Radioisotope Directory were contacted. Only one, The Radiochemical Centre - Great Britain (Amersham/Searle Corporation, the U.S. representative), appears to satisfy the program calibration requirements. They list an accuracy of $\leq 1.5\%$ (1σ) and have supplied this standard with certification of 3.4% (3σ). Unfortunately, this material is available only every six months (February and August).

The National Bureau of Standards Center for Radiation Research has expressed a definite interest in this aspect of the program but would be unable to schedule this standard in their routine radioactivity standards schedule until FY-73.

Expected Achievements

By the end of the next reporting period, a $^{140}\text{Ba}/^{140}\text{La}$ standard will be distributed to the appropriate participants of the CFRMF test.

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.]

Summary

The CFRMF spectrum provided by ANC was used with the SAND-II code to calculate reaction rates for proper sizing of fission and non-fission foils for the CFRMF test. Fission foil sizing, containment requirements, etc., were completed and foil acquisition from the Isotope Target Center at ORNL has been initiated. Nuclear data and information that will be needed or used for multiple foil results or data reduction are discussed.

Accomplishments and Status

Individual laboratory as well as interlaboratory cooperative tests have provided a large body of measured reaction rate data for fission and non-fission foils being used routinely by the FRMDC and the EBR-II project.⁽³⁻²⁵⁾ Comparison of radiochemistry with track recorder⁽²⁶⁾ and fission chamber⁽¹⁰⁾ results for these as well as other tests, such as those for the fission cavity⁽⁸⁻¹⁴⁾ and the C.E.N./S.C.K. secondary intermediate standard neutron facility $\Sigma\Sigma$,⁽¹⁵⁾ are helping to improve the understanding about the accuracy of current reaction rate measurements and calculations.

Reaction rate measurements in the CFRMF facility with its relatively high total flux level of between 10^{10} to 10^{11} n/cm²-sec will permit the attainment of more precise radiochemical reaction rates and ratios for both

fission and non-fission foil reactions in current use for fast reactor dosimetry. The rates and ratios can then be compared with ANL track and NBS fission chamber measurements. The CFRMF facility was selected for initial study because of the similarity of its spectrum (which is similar to the $\Sigma\Sigma$ spectrum) to a fast reactor core spectrum.

The CFRMF central core spectrum is shown in Figure 1.⁽²⁴⁾ A single spectrum, Figure 2, based on the results presented in Figure 1 was developed by ANC as the best current approximation to the true spectrum.⁽²⁷⁾ This spectrum was used as input to the SAND-II code to calculate reaction rates (saturated foil activities) for foil reactions of current interest to the participants of the ILRR program.

Results of these calculations for the reactions of primary concern (^{235}U , ^{238}U , ^{239}Pu , and ^{237}Np) as well as others of both short and long term interest are given in Table 4. The data in Table 4 were then used to provide estimates of foil activity 14 days and 2 days, respectively, after an 8-hour irradiation at a total flux level of 7×10^9 n/cm²-sec. (The value of 7×10^9 is considered a conservative lower boundary on the anticipated flux level of $\sim 10^{10}$ n/cm²-sec at 1 kW.) The results are given in Table 5.

If operation at 10 kW is possible, all the values in Table 5 can be increased by a factor of 10. ANC expects to obtain AEC approval to increase the operating power level of the reactor to ~ 10 kW within ~ 3 months. This would increase the total integral flux level from $\sim 10^{10}$ to $\sim 10^{11}$ n/cm²-sec and should significantly improve the overall accuracy of the radiochemistry measurements.

The data in Tables 4 and 5 are provided to help participants of the ILRR program in the selection and sizing of fission and non-fission foils for the CFRMF test. In applying the data of Table 5, corrections for self shielding and counting configuration should be investigated by the participant and applied where deemed necessary. Counting should be done in such a manner as to attempt to achieve a counting precision of $<0.5\%$ at 95% confidence level. Data listed for non-fission reactions are given on a per milligram foil material basis. Sample sizing will be made on those foils deemed feasible to optimize counting in relation to self absorption and self shielding.

Figure HEDL-1 CFRMF Neutron Spectrum Data

Figure HEDL-2 CFRMF Neutron Spectrum

TABLE HEDL-4
SAND-II CALCULATED SATURATED ACTIVITIES FOR THE CFRMF

Foil Reaction	Calculated Saturated Activity* (Dps or Fissions per Sec/Nucleus)	Nominal 5% Activity Limits (MeV)	
		Lower	Upper
<u>Primary Standards</u>			
$^{235}\text{U}(n,f)\text{F.P.}$	1.695 - 24*	3.60 - 4	2.40
$^{238}\text{U}(n,f)\text{F.P.}$	6.797 - 26	1.40	7.40
$^{238}\text{U}(n,\gamma)^{239}\text{U}$	2.428 - 25	8.40 - 5	1.00
$^{237}\text{Np}(n,f)\text{F.F.}$	4.980 - 25	4.25 - 1	5.40
$^{239}\text{Pu}(n,f)\text{F.P.}$	1.779 - 24	5.00 - 4	3.00
$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	6.408 - 25	1.60 - 4	4.75 - 1
$^{10}\text{B}(n,\alpha)^7\text{Li}$	1.914 - 24	2.10 - 4	6.30 - 1
$^6\text{Li}(n,\alpha)^3\text{H}$	1.034 - 24	4.75 - 4	8.00 - 1
$^{115}\text{In}(n,n')^{115\text{m}}\text{In}$	4.750 - 26	8.00 - 1	6.20
$^{32}\text{S}(n,p)^{32}\text{P}$	1.481 - 26	2.50	8.10
$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	2.484 - 28	6.50	12.2
$^{127}\text{I}(n,2n)^{126}\text{I}$	2.949 - 28	10.1	14.9
<u>Reactions for Dosimetry</u>			
$^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$	5.965 - 26	5.75 - 4	6.00 - 1
$^{59}\text{Co}(n,\gamma)^{60}\text{Co}$	1.019 - 25	1.20 - 4	2.20 - 1
$^{45}\text{Sc}(n,\gamma)^{46}\text{Sc}$	2.360 - 26	6.30 - 4	7.60 - 1
$^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$	9.283 - 27	3.20 - 4	9.20 - 1
$^{109}\text{Ag}(n,\gamma)^{110\text{m}}\text{Ag}$	2.998 - 26	3.00 - 4	7.60 - 1
$^{181}\text{Ta}(n,\gamma)^{182}\text{Ta}$	6.131 - 25	1.20 - 4	6.00 - 1
$^{54}\text{Fe}(n,p)^{54}\text{Mn}$	1.959 - 26	2.40	8.30
$^{58}\text{Ni}(n,p)^{58}\text{Co}$	2.535 - 26	2.20	8.00
$^{46}\text{Ti}(n,p)^{46}\text{Sc}$	3.170 - 27	3.50	9.70
$^{47}\text{Ti}(n,p)^{47}\text{Sc}$	4.029 - 27	2.10	7.60
$^{48}\text{Ti}(n,p)^{48}\text{Sc}$	9.032 - 29	6.70	13.2
$^{55}\text{Mn}(n,2n)^{54}\text{Mn}$	1.165 - 28	10.9	15.6
$^{63}\text{Cu}(n,\alpha)^{60}\text{Co}$	1.679 - 28	4.80	11.2
$^{238}\text{Pu}(n,f)\text{F.P.}$	1.499 - 24	5.00 - 2	4.30
$^{232}\text{Th}(n,f)\text{F.P.}$	1.699 - 26	1.50	7.70
<u>Additional Reactions for Dosimetry and Helium Production</u>			
$^{115}\text{In}(n,\gamma)^{116\text{m}}\text{In}$	3.135 - 25	3.40 - 4	1.30
$^{23}\text{Na}(n,\gamma)^{24}\text{Na}$	1.348 - 27	1.43 - 3	6.60 - 1
$^{31}\text{P}(n,p)^{31}\text{Si}$	7.755 - 27	2.20	7.60
$^{56}\text{Fe}(n,p)^{56}\text{Mn}$	3.868 - 28	5.60	11.4
$^{27}\text{Al}(n,p)^{27}\text{Mg}$	1.152 - 27	3.60	9.70
$^{63}\text{Cu}(n,2n)^{62}\text{Cu}$	4.104 - 29	11.9	16.6
$^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$	6.400 - 26	2.80 - 4	1.80 - 1
$^{24}\text{Mg}(n,p)^{24}\text{Na}$	5.571 - 28	6.60	11.8
$^{59}\text{Co}(n,\alpha)^{56}\text{Mn}$	5.398 - 29	5.90	12.2
$^{14}\text{N}(n,\alpha)^{11}\text{B}$	1.948 - 26	1.80	6.60
$^{28}\text{Si}(n,\alpha)$	1.477 - 27	6.10	10.3
$^{32}\text{S}(n,\alpha)$	1.294 - 26	1.90	7.00
$^{12}\text{C}(n,\alpha)$	2.374 - 28	7.10	12.3
$^{16}\text{O}(n,\alpha)$	1.968 - 27	4.00	8.80
$^{23}\text{Na}(n,\alpha)$	1.448 - 28	6.50	12.2
$\text{Cr}(n,\alpha)$	1.004 - 28	5.40	11.8
$\text{Fe}(n,\alpha)$	8.228 - 29	5.40	11.9
$\text{Ni}(n,\alpha)$	9.038 - 29	5.50	11.0

* $1.695 - 24 = 1.695 \times 10^{-24}$ per 1 n/cm²-sec; note: these values are also the spectral averaged cross sections for the CFRMF spectrum.

TABLE HEDL-5
ANTICIPATED FOIL ACTIVITIES

(After 8-hour irradiation in CFRMF at 1 kW and $\sim 7 \times 10^9$ n/cm²/sec)

Primary Standards	Reaction	(1) Foil Weight Used in Calc. (mg)	Disintegrations per Second		
			Nucleus @ TOR*	Foil @ TOR*	Foil @ 14 Days
Fission ⁽¹⁾ Foils	$^{235}\text{U}(n,f)^{140}\text{Ba}$	~ 307	1.3×10^{-17}	1×10^4	4.8×10^3
	$^{235}\text{U}(n,f)^{95}\text{Zr}$	~ 307	2.8×10^{-18}	2.2×10^3	1.9×10^3
	$^{238}\text{U}(n,f)^{140}\text{Ba}$	~ 307	5.1×10^{-19}	4×10^2	1.9×10^2
	$^{238}\text{U}(n,f)^{95}\text{Zr}$	~ 307	9.8×10^{-20}	7.6×10^1	6.6×10^1
	$^{237}\text{Np}(n,f)^{140}\text{Ba}$	~ 330	3.6×10^{-18}	3×10^3	1.4×10^3
	$^{239}\text{Pu}(n,f)^{140}\text{Ba}$	~ 320	1.1×10^{-17}	9.2×10^3	4.3×10^3
	$^{239}\text{Pu}(n,f)^{95}\text{Zr}$	~ 320	2.3×10^{-18}	1.9×10^3	1.6×10^3
Activation Foils				mg @ TOR*	mg @ 2 Days
	$^{238}\text{U}(n,\gamma)^{239}\text{U}$	~ 307	1.8×10^{-15}	4.3×10^3	2.4×10^3
	$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	1	5.4×10^{-19}	1.2×10^1	1.3
	$^{32}\text{S}(n,p)^{32}\text{P}$	1	1.7×10^{-18}	3×10^1	2.7×10^1
	$^{115}\text{In}(n,n')^{115\text{m}}\text{In}$	1	2.4×10^{-16}	1.2×10^3	7.3×10^{-1}
	$^{127}\text{I}(n,2n)^{126}\text{I}$	1	3.6×10^{-20}	1.3×10^{-1}	1.2×10^{-1}
	$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	1	3.7×10^{-16}	1.1×10^3	6.7×10^2
Dosimetry Foils	$^{45}\text{Sc}(n,\gamma)^{46}\text{Sc}$	1	4.5×10^{-19}	6.1	6.0
	$^{46}\text{Ti}(n,p)^{46}\text{Sc}$	1	6.1×10^{-20}	6.1×10^{-2}	6.0×10^{-2}
	$^{47}\text{Ti}(n,p)^{47}\text{Sc}$	1	1.8×10^{-18}	1.7	1.1
	$^{48}\text{Ti}(n,p)^{48}\text{Sc}$	1	7.5×10^{-20}	7×10^{-1}	3.3×10^{-1}
	$^{54}\text{Fe}(n,p)^{54}\text{Mn}$	1	1.0×10^{-19}	6.3×10^{-2}	6.3×10^{-2}
	$^{55}\text{Mn}(n,2n)^{54}\text{Mn}$	1	6.0×10^{-22}	6.6×10^{-3}	6.6×10^{-3}
	$^{58}\text{Ni}(n,p)^{58}\text{Co}$	1	5.8×10^{-19}	4.0	3.9
	$^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$	1	3.4×10^{-17}	1.2	1.2
	$^{59}\text{Co}(n,\gamma)^{60}\text{Co}$	1	8.6×10^{-20}	8.8×10^{-1}	8.8×10^{-1}
	$^{63}\text{Cu}(n,\alpha)^{60}\text{Co}$	1	1.4×10^{-21}	9.3×10^{-3}	9.3×10^{-3}
	$^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$	1	1.5×10^{-16}	9.6×10^2	7.24×10^1
	$^{109}\text{Ag}(n,\gamma)^{110\text{m}}\text{Ag}$	1	1.9×10^{-19}	5.2×10^{-1}	5.2×10^{-1}
	$^{181}\text{Ta}(n,\gamma)^{182}\text{Ta}$	1	8.6×10^{-18}	2.9×10^1	2.8×10^1
	$^{232}\text{Th}(n,f)^{140}\text{Ba}$	189	1.6×10^{-19}	4.2×10^{-1}	2×10^{-1} (a)

* TOR = Time of Removal

(a) @ 14 days

(1) For this calculation, it was assumed that 99.9+% metal, ~ 5 mil $\times 1/2"$ $\times 1/2"$ squares, will be used and the weight will be known to $\leq 0.5\%$. The foils ordered from ORNL will be ~ 5 mil by $1/2"$ diameter disks.

Based on a previous test, the fission fragment loss versus mil thickness is shown in Figure 3 for ^{235}U . Since one could anticipate as much as a 1-1/2% fragment loss for a 5 mil foil, it is necessary that analysis be made on the foil plus cover material.

It is estimated that the gamma self shielding correction for a 5 mil foil should be no more than $\sim 2\%$ for ^{140}La γ and $\sim 3\%$ for ^{95}Zr γ .

Reference (6) Provides more information on these as well as on other corrections.

Figure HEDL-3 Percent Loss of Fission Products From Different Thickness of ^{235}U Foils (0.4" x 0.4" Sq. Sheet).
Based on ZPR-III Test (FTR Type Spectrum)

Fission yields and fission product half lives in current use by HEDL for the FRMDC and that will be initially used to provide fission reaction rates for the radiochemistry results for comparison with track and fission chamber measurements are listed in Table HEDL-6. (2,4,29)

Table HEDL-6

FISSION REACTION HALF LIVES AND YIELDS

<u>Fission Reaction</u>	<u>Half-Life of Products</u>				<u>α Fission Yields, %</u>			
	<u>^{99}Mo</u>	<u>^{140}Ba</u>	<u>^{137}Cs</u>	<u>^{148}Nd</u>	<u>^{99}Mo</u>	<u>^{140}Ba</u>	<u>^{137}Cs</u>	<u>^{148}Nd</u>
$^{239}\text{Pu}(\text{n},\text{f})\text{F.P.}$	66.7	12.8	30.1	Stable	5.9	5.1	6.5	1.71
$^{235}\text{U}(\text{n},\text{f})\text{F.P.}$					6.1	6.1	6.2	1.75
$^{237}\text{Np}(\text{n},\text{f})\text{F.P.}$					-	5.8	6.4	-
$^{238}\text{U}(\text{n},\text{f})\text{F.P.}$					6.3	6.0	6.3	1.93

α . The uncertainty associated with these values of fission yield is estimated to be about 5 to 10% (1 standard error) depending on the isotope. Additional yield information for some of these reaction products will be available in the near future from program work now in progress at Argonne National Laboratory and Aerojet Nuclear Company.

Further discussion and comments on these fission yield values are encouraged. It is anticipated that the CFRMF test results as well as other work being accomplished at ANL, (1,5,6,28) HEDL, (3,4,19,20,29) and other laboratories will help to establish improved values of fission yields for the listed reaction products.

Expected Achievements

Selection, fabrication, and completion of preliminary quality assurance tests for fission and non-fission foils for the CFRMF radiochemistry measurements are expected before the end of November 1971. Distribution of foil sets to other participants will follow with initial irradiations tentatively scheduled before the end of December 1971.

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

Previous interlaboratory tests have provided reaction rate data which have been used to help develop a reasonably accurate and consistent set of evaluated energy-dependent cross sections for SAND-II data reduction.⁽²⁹⁻³²⁾ Work in progress to improve the accuracy and consistency of these cross sections is discussed.

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 $\sigma(E)$.⁽²⁹⁾

A reasonably consistent set of $\sigma(E)$ values has been developed and is in current use for the SAND-II code,⁽³⁰⁾ which will be used by HEDL to provide multiple foil flux spectral data for the ILRR program.⁽²⁹⁾ Better consistency and accuracy for these cross sections are required. Present accuracy is in the 20 to 40% range at the 95% confidence level.⁽²⁹⁻³²⁾ This accuracy can be improved if the sources of existing inconsistencies between calculated integral averaged cross section, $\bar{\sigma}_c$, using the current evaluated energy dependent cross sections $\sigma(E)$ and measured values, $\bar{\sigma}_m$, can be identified and effectively eliminated.^(3-25,29-37,44)

The last Cross Section Evaluation Working Group (CSEWG) meeting at BNL included a review of the cross section needs of the FRMDC and fuels and

materials programs. These needs have been discussed previously.⁽²⁹⁻³²⁾ As a result, arrangements were made with the National Neutron Cross Section Center (NNCSC) at BNL to compare the current SAND-II reference evaluated energy-dependent cross section library with existing ENDF/B data. NNCSC now has the SAND-II $\sigma(E)$'s and will prepare suitable plots for making the necessary comparisons.

As these comparisons are made, HEDL anticipates additional help and direction from NNCSC, as well as other participants of the ILRR program, in establishing a "best" set of evaluated energy-dependent reaction cross sections for improving 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 (σ_m values) are expected from the ILRR program to help in this difficult task, with first results from the CFRMF test.

Expected Achievements

By the end of the next reporting period, assistance will be obtained from appropriate participants of the ILRR program in establishing an improved evaluated energy-dependent cross section for $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ for subsequent studies of the CFRMF and other ILRR test results. $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ has been selected as one of the reactions that will be used as a reference standard for fast reactor environments.

E. MULTIGROUP COMPARISONS OF ^{197}Au CAPTURE CROSS SECTIONS

R. E. Schenter

Objective

- Obtain accurate microscopic capture cross sections of ^{197}Au .

Summary

$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ has been selected as a reaction that will be used as a reference standard for fast reactor environments. It is also used as a standard for measurements of integral capture reaction rates for the CFRMF.

Accomplishments and Status

Group averaged infinitely dilute capture cross sections of ^{197}Au have been calculated from three sources of microscopic data, SAND-II,⁽³⁰⁾ Fricke-70,⁽³⁸⁾ and ENDF/B-2. Results of these calculations are given in Figure 4, where it is seen that significant differences occur for energies below 10 keV.

Expected Achievements

These and other calculated microscopic capture cross sections of ^{197}Au will be used for comparison of calculated and measured integral reaction rates for the CFRMF and other ILRR tests.

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Figure HEDL-4 Group Averaged Capture Cross Sections of ^{197}Au

F. COMPARISON OF EXPERIMENTAL AND THEORETICAL CALCULATIONS OF INTEGRAL CAPTURE REACTION RATES FOR THE CFRMF FOR FISSION PRODUCT ISOTOPES

R. E. Schenter and F. A. Schmittroth

Objective

- Theoretical calculations of microscopic neutron capture cross sections have been continuing for fission product isotopes as part of a program to expand and improve ENDF/B files. There exist no experimental data of these cross sections in the "fast" energy region for most of the important fission product isotopes. Consequently, it is important to make use of any available integral measurements to compare with and help "normalize" the theoretical calculations.

Summary

A comparison of some previously measured and calculated CFRMF spectral averaged cross sections using theoretical $\sigma(E)$ data is presented.

Accomplishments and Status

The above described comparisons have been made using measurements from the CFRMF.⁽³⁹⁾ Table 7 shows these comparisons, where HEDL theoretical calculations and those of Cook⁽⁴⁰⁾ and Benzi⁽⁴¹⁾ integrated over the CFRMF spectrum are included.

Expected Achievements

Comparison of experimental and theoretical integral capture reaction rates for ILRR tests in the CFRMF and other facilities will be extended.

Table HEDL-7
INTEGRAL RESULTS

$\bar{\sigma}$ (barns)					
Z	Name	CFRMF $\bar{\sigma}$ Au = 0.45	HEDL	Cook	Benzi (Cook-E < 1 keV)
36	^{83}Kr		0.232	0.257	0.389
37	^{87}Rb	0.0153 $\pm$ 0.0011		0.0226	0.0237
40	^{95}Zn		0.0589	0.0485	
41	^{93}Nb	0.189 $\pm$ 0.019	0.145		
41	^{95}Nb		0.138	0.106	
42	^{95}Mo		0.186	0.188	0.176
42	^{96}Mo			0.0807	
42	^{97}Mo		0.178	0.156	0.196
42	^{98}Mo	0.062	0.062	0.107	0.0964
42	^{99}Mo		0.171	0.160	
42	^{100}Mo		0.0544	0.0762	0.0635
43	^{99}Tc	0.383 $\pm$ 0.063		0.301	
44	^{101}Ru		0.397	1.102	0.462
44	^{102}Ru	0.101 $\pm$ 0.007	0.229	0.137	0.192
44	^{103}Ru		0.266	0.296	
44	^{104}Ru	0.095 $\pm$ 0.005	0.0710	0.0789	0.0897
44	^{105}Ru		0.234	0.197	
44	^{106}Ru		0.0423	0.0408	
45	^{103}Rh	0.423	0.365	0.373	
45	^{105}Rh		0.293	9.263	
46	^{105}Pd		0.449	0.303	0.612
46	^{106}Pd		0.102	0.102	
46	^{107}Pd		0.378	0.310	
46	^{109}Pd		0.126	0.178	
47	^{107}Ag	0.468 $\pm$ 0.060		0.394	
47	^{109}Ag	0.563 $\pm$ 0.081			
48	^{113}Cd		0.300	0.186	
49	^{115}In	0.31 $\pm$ 0.03		0.348	0.366
53	^{127}I	0.308		0.341	0.386
53	^{129}I			0.187	
53	^{131}I		0.147	0.0991	
53	^{135}I		0.00166	0.00107	
54	^{131}Xe		0.286	0.276	0.276
54	^{132}Xe			0.0735	0.0651
54	^{133}Xe		0.0395	0.0682	
54	^{134}Xe			0.0147	0.0389
54	^{135}Xe		0.0588	0.00969	
54	^{136}Xe			0.00310	
55	^{135}Cs		0.0438	0.301	
55	^{137}Cs		0.00834	0.00957	
57	^{139}La	0.0207	0.326	0.314	0.0307
58	^{140}Ce	0.001			
58	^{141}Ce		0.102		
58	^{142}Ce	0.0207			

G. MULTIPLE FOIL TECHNIQUE ERROR ANALYSIS

W. N. McElroy, R. L. Simons, C. A. Oster* and G. R. Odette†

Objective

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

Summary

Procedures for the assignment of errors to SAND-II evaluated energy-dependent reaction cross sections are being developed and tested. Error assignments accomplished to date are discussed.

Accomplishments and Status

A SAND-II Monte Carlo Error Analysis Code has been developed⁽⁴²⁾ and is being used and tested for the assignment of errors to multiple foil flux-spectral results, re-evaluated energy-dependent cross section results, and empirically derived damage function results.⁽²⁹⁾ For the CFRMF and other ILRR tests, one of the more difficult aspects of this work is the proper assignment of errors for the current SAND-II energy-dependent evaluated cross sections.⁽²⁹⁾

Two methods of error assignment have been studied to date. First, assuming no separation in shape and magnitude, as will be discussed below, individual errors have been assigned to each of the 620 SAND-II energy groups based on a review of literature evaluations. However, a constraint was imposed that over certain fixed regions such as thermal, resonance, and fast, the total integrated error from the individual assignments must be consistent with anticipated experimental errors in the measured 2200 m/sec, resonance integral, and fission averaged cross sections, respectively.

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This procedure has been used primarily for analytical studies directed toward the general problem of understanding error propagation for the multiple foil technique. The Monte Carlo code is designed to permit the individual or simultaneous application of the two types of cross section errors and to combine these errors with those due to uncertainties in measured reaction rates (saturated activities). Thus the effect of a single type of error can be studied independently or combined with other errors.

A second approach to cross section error assignment is considered more realistic and practical in terms of present applied problems. The second approach assumes that over certain regions the relative uncertainty in the shape of a cross section is much smaller than in its magnitude, and a set of energy regions (based mainly on intervals where individual or consistent cross section measurements have been made) was selected within which the cross section shape was held constant but its magnitude was allowed to independently vary, again with certain constraints based on integral consistency requirements. The regions and tentative error assignments used in the second approach to derive a SAND-II error matrix are presented in Table 8.* Grundl's,^(9,43) Fabry's et al,⁽¹⁴⁾ and McElroy's et al^(29,31,32) works were primarily used as the guide in the selection of both the energy regions and subsequent assignment of these cross section errors.

Two types of significant errors which propagate in spectral unfolding are contained in the matrix of Table 8. The first is related to the uncertainties in the shape of each reaction cross section. The second is associated with the magnitude of all reaction cross sections in each of the energy intervals. Both of these may be considered as absolute errors in the sense that they are quantitative uncertainties associated with each particular cross section magnitude. However, if the analysis is concerned only with establishing spectral shapes, only the relative errors should be

* It is recognized that the group structure selected for the error matrix will have an effect on the propagation of errors. The group energy bounds have been set up as a variable, therefore, so that changes in group structure can be easily accommodated for the study of this effect.

Table HEDL-8

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	8	8	8	8	10	10	10	10	10	10	10	10	10
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.

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considered. That is, shape errors should be assigned relative to a reference cross section in an energy interval which is taken to be known exactly. Relative errors between reactions in each energy region should be assigned with respect to selected reference reaction cross sections and the larger overall absolute error on the barn scale ignored.

Thus the cross section error assignment in each energy region was guided both by recognizing the relative error reduction resulting from shape and magnitude cross section adjustment for all the reactions (accomplished by demanding integral consistency for a variety of spectra) and also from previous detailed investigations.^(9,14,29,31,32,43) For example, Grundl has effectively accomplished a separation of absolute and relative errors for a few of the fission and short half life reactions listed in Table 8 based on an extensive set of relative cross section measurements performed at the Los Alamos Van de Graaff.⁽⁴³⁾ It is anticipated that the CFRMF and subsequent ILRR tests will provide additional reaction rate data to help attain a similarly improved relative consistency for the longer half life reactions. Although it is very difficult at present to assign such relative errors precisely in terms of the two-dimensional input matrix to the Monte Carlo analysis, Table 8 is considered to contain conservative estimates reflecting both major sources of relative uncertainty.

Expected Achievements

By the end of the next reporting period, preliminary analytical results using the SAND-II Monte Carlo error code should be available for estimating the anticipated uncertainty of multiple foil flux spectral data for the CFRMF spectrum.

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LAWRENCE LIVERMORE LABORATORY

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A. NUCLEAR CROSS SECTION CALCULATIONS*

D. G. Gardner, J. L. Brownlee, and A. Delucchi

Objective

We are accumulating and developing computer programs for calculating excitation functions for essentially any neutron-induced reaction. These include statistical model programs for compound and precompound nucleus evaporation, a coupled-channels optical model program to compute transmission coefficients for nonspherical nuclei and cross sections for the direct excitation of inelastic levels, and various programs to compute isotope ratios following a gamma-ray cascade in the final nucleus. Our philosophy is that we wish to calculate concurrently all possible reactions that may occur, and to get them all right. This attitude is necessary because we are primarily interested in unstable target nuclei where confirming experimental data may never be available. If we cannot compute all competing reactions properly on stable nuclei where experimental data are available, then we will feel little confidence in our calculations for unstable nuclei.

Summary

During the past six months we have begun to accumulate and develop the necessary computer programs needed to calculate cross sections for neutron-induced reactions. We can now handle the compound nucleus part of the cross sections, and expect to begin on the precompound calculations in the near future.

Accomplishments and Status

In Radiochemistry Division of LLL we now have a number of codes for computing nuclear reaction cross sections. Most of these codes were obtained or developed during the past six months, and are not yet considered completely debugged. For several years we have used Moldauer's

ABACUS-NEARREX code to compute (n,γ) cross sections. This code has now been replaced by a completely revised version of Dunford's COMNUC code, which was developed primarily at LLL over the past six months. It possesses many advantages over the NEARREX code, including a better fission channel, charged particle competition, and multiple particle evaporation.

As supplementary programs we have improved versions of Dunford's CASCADE and FOURPLUS codes. The former code provides input to COMNUC for calculating reactions such as $(n,\gamma n')$, $(n,\gamma p)$, and (n,γ) fission). The FOURPLUS code is a coupled-channels program to be used in providing transmission coefficients for deformed nuclei, as well as direct inelastic excitation cross sections.

We have also obtained Uhl's code (called UHL here). This code permits up to four particles to be evaporated, and follows the gamma-ray cascade in the final nucleus. The code is operational, but we have not tested it thoroughly as yet.

Expected Achievements

We will continue the testing and debugging of our four new codes during the next Quarter. As time permits, a number of other codes will be developed and changes made in the existing codes. For example, UHL will now only accept transmission coefficients as a function of ℓ . We will modify the code so that a spin-orbit potential may be used. We will also modify UHL to follow more reaction sequences at one time. A new code, probably based on UHL, will be written to compute the precompound part of the cross section. Finally, we will complete a Monte Carlo gamma-ray cascade program, which was begun about one year ago. The latter will hopefully supply the best available estimates of isomer ratios.

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

LOS ALAMOS SCIENTIFIC LABORATORY

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A. POWER NORMALIZATION PROCEDURES

G. E. Hansen

Objective

To establish power normalization procedures for the CFRMF and other ILRR tests.

Summary

In addition to reactor power level instrumentation, ^{235}U and ^{238}U fission foils should be used in the immediate vicinity of the CFRMF central test region to establish proper power normalization factors between reactor runs.

Furthermore, before each irradiation, the reactor power level instrumentation should be checked against the response of a fission chamber placed in the central test region.

Accomplishments and Status

Discussions with participants of the ILRR program and others (experienced in the measurement of absolute and relative flux levels) have established that in addition to the CFRMF reactor power level instrumentation readout, there is a need for duplicate fission foil measurements for power normalization between reactor runs; i.e., one set of ^{235}U and ^{238}U foils should be counted by ANC and another by HEDL. The emphasis here is on relative rather than absolute measurements. These duplicate foils should be located within about one inch of the test region for the CFRMF irradiations.

A check on the reactor power level instrumentation is needed to further help in establishing the reactor run-to-run reproducibility. This can be accomplished by obtaining the response of a fission chamber placed in the central test region before each test irradiation. The fission chamber, the

electronics, and the procedures used for obtaining the fission count should be kept strictly the same during all of the CFRMF test irradiations.

Preliminary experimental measurements using these procedures should be obtained before the commencement of absolute measurements.

Expected Achievements

Further technical discussions will be held with participants of the ILRR program to work out the details of the fission foil placement, counting requirements, etc., for power normalization by the next reporting period.

NATIONAL BUREAU OF STANDARDS

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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 . Reaction rates in these isotopes provide a primary calibration base for neutron dosimetry and related problems of radiation damage. The measurement is to be carried out with a double fission ionization chamber designed to accommodate fission foils that are part of a 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

This first Interlaboratory Reaction Rate Program Report presents a summary look at each component of the proposed fission chamber measurements. A double fission ionization chamber, convenient to disassemble and of minimum size and mass, has been constructed and the performance characteristics examined. For fission foils $200\text{ }\mu\text{g}/\text{cm}^2$ thick, the extrapolation-to-zero from just above the alpha pulse-height cut-off is less than 1%. A uranium weight ratio between normal and highly enriched uranium fission foils was measured by two independent methods: alpha counting and thermal-neutron-induced fission counting. Agreement between the two methods is better than 1%. Flux perturbations at the center of the CFRMF due to the presence of the fission chamber are predicted to be about 1% or less.

Accomplishments and Status

This first report for the Interlaboratory Reaction Rate Program describes the results of a summary look at each component of the fission chamber measurement.

1. Fission Chamber Performance

A back-to-back fission ionization chamber designed to accommodate 0.75" diameter fission foils within an enclosure that is easily disassembled and of minimum size and mass has been constructed for the following applications:

Measurement of average fission cross sections for ^{252}Cf fission neutrons at NBS.

Measurement of average fission cross section ratios (i.e., spectral indexes) for basic fast critical assemblies at Los Alamos.

Fission rate measurements at the reference flux facility CFRMF.

Important design and operating characteristics for the chamber are as follows:

a. Chamber Enclosure

0.985" diameter x 0.60" high aluminum cylinder with 0.022" thick walls.

Chamber Stem

0.188" diameter x 13" long aluminum tube threaded into the chamber enclosure.

Chamber Manifold

1.18" diameter x 1.5" long cylinder with gas line connections and two subminiature RF connectors.

Fission Foils

0.5" diameter oxide deposits on 0.75" diameter platinum are mounted near the center of the enclosure and held at ground potential.

Ionization Collectors

Disk and cylinder shaped surfaces subtend 90% of the 2π solid angle presented by the fissionable deposit.

All chamber parts are assembled by smooth or light press fit for easy and repeated access; there are no soldered connections or seals. Final closure is secured by means of a spring ring and locking wire. An assembly sketch of the fission chamber and the chamber enclosure is reproduced in Figure 1.

b. Chamber Gas

Pure methane or P-10 argon-methane mixture at zero psig and 0.5 CFH flow.

Chamber Voltage

+200 volts.

Chamber output at the cut-off of natural alpha pulses is about 3 mv.

At the ILRR Program Meeting in June, it was agreed that the NBS fission chamber enclosure should be used for all irradiations at CFRMF in order to provide partial assurance that all detection techniques will be measuring the same flux. Accordingly, on August 10, 1971, design sketches of the fission chamber were sent to all ILRR Program participants, with fabricating materials included for the HEDL, ANL, and AI participants.

Performance of the double fission chamber was checked with two ^{235}U fission foils: a $200 \mu\text{g}/\text{cm}^2$ thick deposit on one side and $500 \mu\text{g}/\text{cm}^2$ on the other. Low-noise preamps followed by conventional solid-state electronics were employed to obtain pulse height distributions and to ascertain the proper bias settings for a triple-scaler arrangement. The latter technique is used for the routine acquisition of data and will be discussed in a later report. Pulse height distributions obtained from the chamber may be characterized simply in terms of the pulse height cut-off for alpha particles. With this alpha cut-off set at $V_\alpha \sim 0.5$ Volts, the single peak of the fission pulse distribution occurs at around 3 Volts, the valley between V_α and the bulk of the fission pulses extends to $\sim 3 V_\alpha$, and the peak-to-valley ratio is about 30 for the $200 \mu\text{g}/\text{cm}^2$ foil. The chamber voltage plateau is more than adequate as with any high resolution

FIGURE NBS-1. PEACE PIPE FISSION CHAMBER

ionization chamber: a 50 Volt reduction in chamber voltage produces less than a 0.1% change in the pulse count above a bias of $2 V_{\alpha}$. Similarly, for gain changes in general, the extrapolations-to-zero given below show that a 10% change in gain would produce, for the worst case listed, a 0.3% change in the integral pulse count above $2 V_{\alpha}$.

Extrapolation of the fission pulse height distribution to zero is the primary error source in the determination of an absolute fission chamber response. A convenient measure of this extrapolation-to-zero is the fraction of fission pulses which fall between the alpha cut-off, V_{α} , and twice V_{α} . In a preliminary check of chamber performance, the following results were obtained for this fraction.

Extrapolation-to-Zero for the NBS Double Fission Chamber

	<u>200 $\mu\text{g}/\text{cm}^2$ ^{235}U</u>	<u>500 $\mu\text{g}/\text{cm}^2$ ^{235}U</u>
Pure Methane:	0.9 ₃ %	1.8%
P-10 (90% Ar, 10% CH_4):	0.7 ₆ %	1.3%

For ^{239}Pu and ^{237}Np fission foils, the extrapolations will be notably larger because of greatly enhanced alpha disintegration rates. Of special concern for the CFRMF measurement is the placement of preamps. It was determined that 24 feet of cable between the chamber and preamp increased the lowest of the above extrapolation values from 0.76% to 0.83%.

2. Fission Foil Assay

A confident assignment of isotopic weights to fission foils will come only through redundancy of assay. An experimental check has been carried out in order to find out how easily or uneasily such a requirement may be realized. The weight ratio of a ^{235}U foil (99.7% ^{235}U , designation 25S-2-2) to that of a normal uranium foil (designation 28N-2-2) has been measured by two independent methods: 2π alpha counting, and thermal-neutron-induced fission counting.

Alpha counting of the two 200 $\mu\text{g}/\text{cm}^2$ thick foils was carried out with a conventional gas-flow proportional counter. With equivalent thickness,

size and backing materials, the counting efficiencies for the foils were assumed to be the same and, in this case, the uranium weight ratio is just the count rate ratio divided by the alpha disintegration rate ratio. A customary value for the disintegration rate in normal uranium, 1501 ± 3 dpm/mg, is given in Reference 1; for the enriched uranium, absolute determinations are reported in Reference 2. The result obtained was,

$$\text{uranium weight ratio, } \frac{^{25}\text{S}-2-2}{^{28}\text{N}-2-2} = 1.036 \pm 0.025 \text{ (alpha counting).}$$

Most of the assigned error is attributable to poor statistics for the alpha counting. Fission counting was carried out in a thermal-neutron beam extracted from the NBS Reactor Thermal Column. With a nominal value of 0.0072 for the isotopic concentrations of ^{235}U in normal uranium, the result was,

$$\text{uranium weight ratio, } \frac{^{25}\text{S}-2-2}{^{28}\text{N}-2-2} = 1.042 \pm 0.03 \text{ (fission counting).}$$

The agreement between the two methods is uncommonly good and it is possible that the errors were set too high for this, an exploratory experiment.

Fission foils for the CFRMF measurements will be selected from a permanent set which is part of a joint NBS-Los Alamos program to measure fission-spectrum-averaged cross sections for fissionable isotopes. The foils are 0.5" diameter oxide deposits vacuum evaporated (with double rotation) onto 0.75" diameter x 0.005" thick polished platinum backings. Three deposit thicknesses are generally available: 100, 200, and 500 $\mu\text{g}/\text{cm}^2$. An abridged listing of the isotopic content of stock materials used is given below in atom percent.

Isotopic Content of Fissionable Deposits (Abridged Listing)

	<u>^{235}U</u>	<u>^{238}U</u>		<u>^{239}Pu</u>	<u>^{240}Pu</u>	<u>^{237}Np</u>
Enriched Uranium:	99.7%	0.13%	^{239}Pu :	94.0%	6.5%	
Normal Uranium:	0.7%	99.3%	^{237}Np :			100%
Depleted Uranium:	0.1%	99.9%				

3. Flux Perturbations

Elastic scattering effects are generally small in an isotropic flux where in-scatter and out-scatter compensate well. Capture and inelastic scattering can be important and for this reason, aluminum was chosen as the fabricating material for the fission chamber. The largest fission chamber piece, the chamber enclosure as shown in Figure 1, will capture not more than 0.1% of the incident flux and it is less than a 1% source for inelastic scattered neutrons. The platinum foil backings will capture less than 0.2% of the incident flux. The hydrogen in a pure methane filling of the chamber will scatter about 0.5% of the incident flux.

Expected Achievements

By the end of the next quarter, December 1971, it is expected that the fission chamber measurements at CFRMF and at Los Alamos will have been performed.

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