LA-5284-PR

PROGRESS REPORT

C.3

Quarterly Report

Advanced Plutonium Fuels Program

January 1 to March 31, 1973

los alamos
scientific laboratory
of the University of California
LOS ALAMOS, NEW MEXICO 87544

UNITED STATES
ATOMIC ENERGY COMMISSION
CONTRACT W-7405-ENG. 36

This report presents the status of the LASL Advanced Plutonium Fuels program. The four most recent reports in this series, all unclassified, are:

LA-4993-PR LA-5106-PR
LA-5067-PR LA-5193-PR

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Printed in the United States of America. Available from
National Technical Information Service
U. S. Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22151
Price: Printed Copy $3.00; Microfiche $0.95

LA-5284-PR
PROGRESS REPORT
UC-79b
ISSUED: May 1973

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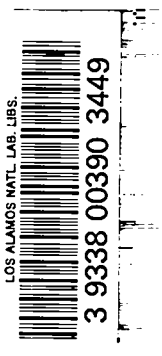

Compiled by

R. D. Baker

This work supported by the U.S. Atomic Energy Commission's
Division of Reactor Development and Technology

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ABSTRACT

This is the 26th quarterly report on the Advanced Plutonium Fuels Program at the Los Alamos Scientific Laboratory.

Most of the investigations discussed here are of the continuing type. Results and conclusions described may therefore be changed or augmented as the work continues. Published reference to results cited in this report should not be made without obtaining explicit permission to do so from the person in charge of the work.

PROJECT 401

EXAMINATION OF FAST REACTOR FUELS

Person in Charge: R. D. Baker
Principal Investigators: J. W. Schulte
K. A. Johnson
G. R. Waterbury

I. INTRODUCTION

This project is directed toward the examination and comparison of the effects of neutron irradiation on LMFBR Program fuel materials. Unirradiated and irradiated materials will be examined as requested by the Fuels and Materials Branch of DRDT. Capabilities are established and are being expanded for providing conventional preirradiation and postirradiation examinations. Nondestructive tests will be conducted in a hot cell facility specifically modified for examining irradiated prototype fuel pins at a rate commensurate with schedules established by DRDT.

Characterization of unirradiated and irradiated fuels by analytical chemistry methods will continue, and additional methods will be modified and mechanized for hot cell application. Macro- and micro-examinations will be made on fuel and cladding using the shielded electron microprobe, emission spectrograph, radiochemistry, gamma scanner, mass spectrometers, and other analytical facilities. New capabilities will be developed in: gamma scanning, analyses to assess spatial distributions of fuel and fission products, mass spectrometric measurements of burnup and fission gas constituents, chemical analyses, and measurement of carbon in irradiated fuels.

Microstructural analyses of unirradiated and irradiated materials will continue using optical and electron microscopy, and autoradiographic and x-ray techniques. Special emphasis will be placed on numerical representation of microstructures and its relationship to fabrication and irradiation parameters. New etching and mounting techniques will be developed for high burnup materials.

II. EQUIPMENT DEVELOPMENT

A. In-Cell Equipment

(R. W. Basinger, G. R. Brewer, E. L. Ekberg, K. W. R. Johnson, M. E. Lazarus, P. A. Mason, C. D. Montgomery, J. R. Trujillo, L. A. Waldschmidt)

1. Mechanical Profilometer

The new profilometer for measuring the diameters of breached fuel pins following irradiation has been completed and installed. Preliminary operation indicates that it has many improvements over the first model.

2. Electro-Optical Profilometer

The new Electro-Optical Unit (Optron) has exhibited an unusually high and unacceptable noise level. Should the manufacturer be unable to rectify the difficulty in a reasonable length of time, the order will be canceled.

3. Macro-Photography Unit

The three-view photography equipment does not work on bowed fuel elements since it is impossible to line up the montage of the three views without separating each view. The equipment has now been modified to take single views of the elements.

4. Fuel Element Leak Detection and Location

A device has been designed and fabricated to detect and locate cladding leaks by pressurizing the fuel element. The pressure, up to 200 psi, is applied through a seal assembly which is placed over the fission gas

puncture hole. Leakage is determed by system pressure drop and/or helium leak detection. Location is determined by submerging the fuel element in a liquid or by the use of a "sniffer" on the helium leak detector. This is similar to a device developed at ANL-East.

5. Measurement of Fuel Pin Length

In response to a request from GE to develop a device for precise measurement of fuel pin length, a device was fabricated and put into service. The complete fixtures for making measurements on pins up to 60 in. in length (to ± 0.004 in.) are shown in Fig. 401-1.

6. Storage of Irradiated Fuel Pins

To satisfy the need for interim storage of fuel pins at various stages of the examination and to provide long-term storage capabilities, the floor holes at the DP-West Facility had been modified to accept pins up to 61 in. in length.

The insert, centering fixture, plug, and lifting device are shown in Fig. 401-2. There are 22 storage holes with a criticality limitation of 25 pins per hole. Inserts have been provided for both the 40-in.- and the 61-in.-long fuel pins.

7. Supplementary Equipment for Metallography

A new vacuum potting fixture was designed, fabricated and installed in the DTA Cell along with a vacuum pump assembly. This permits potting metallography samples immediately after being processed through the Sodium Distillation Furnace.

A replacement adapter for the autoradiography and waste compactor press was fabricated and installed.

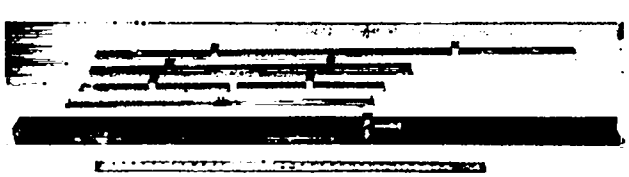

Fig. 401-1. Device for Measuring Fuel Pin Length

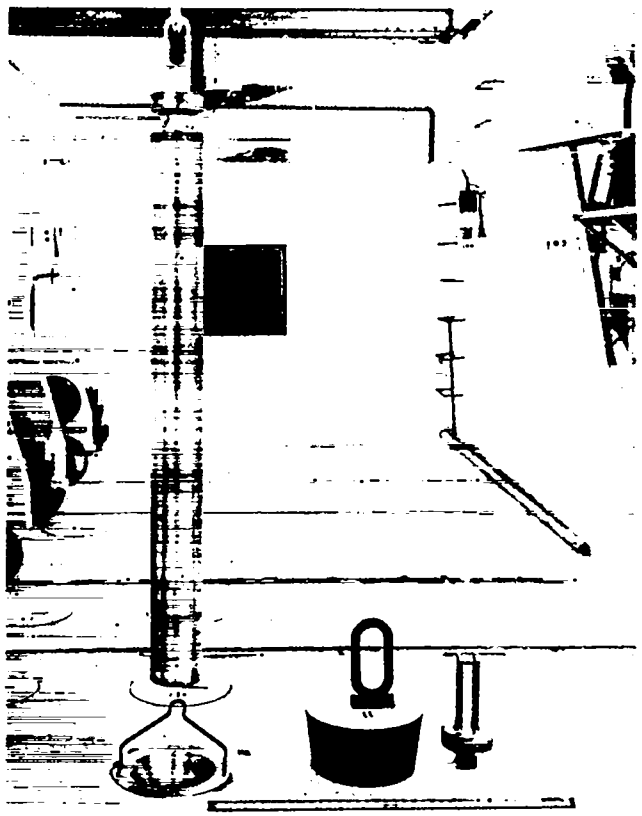

Fig. 401-2. Storage Holes Located in DP-West Facility

8. Sodium Distillation System

The high vacuum system of the DTA Cell was completely disassembled for cleaning and replacement of defective parts. Modifications were made to the sodium still and cold trap assembly to permit the maintenance of a vacuum tight seal during the distillation operation (to remove Na from specimens prior to metallographic examination).

B. Inert Atmosphere Systems
(P. A. Mason)

Replacement oxygen and moisture gettering agents were received and installed in the recirculating purifier for the Disassembly Cell. The purifier system is presently maintaining an atmosphere containing typical concentrations of 20 ppm O_2 and < 1 ppm H_2O.

A replacement boost pump for the Disassembly Cell atmosphere sampling line has been received. The pump has been temporarily installed at tbe operator station pending removal of fuel pins from the cell to permit permanent installation under the alpha box.

The installation of the Butyl Acetate Removal System for the Metallography Cells has been completed and the system is now operational. An average of 0.9 liters of butyl acetate is being collected in a 24 hour period.

C. Manipulator Maintenance and Development
(G. R. Brewer, P. A. Mason, E. L. Mills)

Overhaul of the AMF manipulators at Wing 9 and the CRL Model 8 manipulators at DP-West has continued.

The re-installation of AMF manipulators in the Metallography Cells has eliminated the frequent breakdowns experienced with the CRL Model "L" manipulators. Only one AMF manipulator required removal from the cells for repair during the report period. As a result of discussions with Central Research Laboratories regarding the difficulties experienced with the Model "L" manipulators, tests were performed at the factory. "Slack tape eliminators" were designed by the factory and recommended as a solution to the problems. One set of the eliminators was purchased and will be installed to determine the effectiveness of the modification.

The AMF manipulators are being converted to a CRL slave hand assembly, type SRL, to reduce wear prob lems and provide a more secure gripping action.

The spare CRL Mini-Manipulator has been modified to permit straightening the slave arm during installation in the Metallograph Compartment.

The experimental polyurethane boots obtained from Central Research Laboratories proved satisfactory after installation in the Disassembly and Metallography Cells. An order for 24 of the boots has been received.

Dural pads for the metallography cell manipulators were designed, fabricated and installed. Butyl acetate solvent used in the metallography sample preparation causes rapid deterioration of the standard neoprene pads.

D. Fuel Pin Handling System for Betatron Radiography
(J. M. Ledbetter, P. A. Mason, C. D. Montgomery T. Romanik, J. R. Trujillo)

Several modifications were incorporated in the design of the fuel pin handling cart. The electrical wiring was revised and a new control console fabricated. Testing of the system to insure high reliability is continuing with availability for operations expected by the second week of April. The Radiography Cask and trunnion stand are shown in Fig. 401-3. The cart containing the equipment for elevation of the pin is shown in Fig. 401-4.

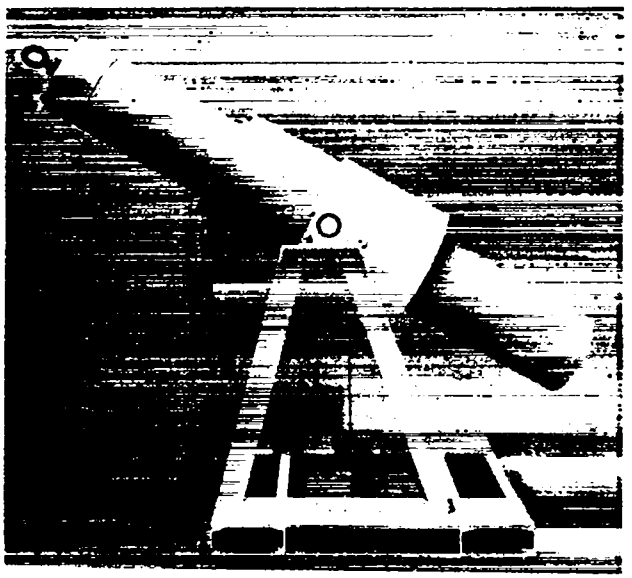

Fig. 401-3. New Radiography Cask Shown in Tilting Fixture

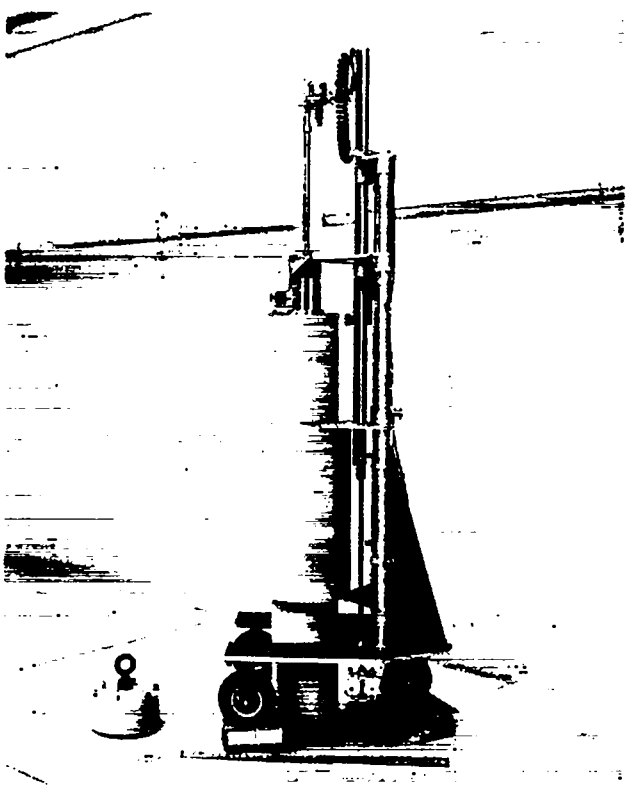

Fig. 401-4. New Radiography Cask with Elevating Mechanism Used at Betatron Building

E. Status of Shipping Cask

Following a meeting at EBR-II, it was determined that LASL could handle the following casks: T-2, Hanford 14-Ton, Murphy, and possibly the TREAT units, which may present some problems in unloading.

In order to maintain adequate shipping capacity, and in view of the TAN Facility being phased out, it appeared advisable to modify the two LASL vertical casks (capacity for 19 pins) to be compatible with the HFEF-S and HFEF-N loading ports.

Preliminary design was started in March 1973 to alter the cask arrangement to provide a separable base-skid concept thus reducing the effective diameter of the cask to < 33 in. which is a limiting dimension of the loading ports. Cost estimates for the modification are ~ $2000 per cask.

When approval of the proposed modifications are obtained from DOT, the fabrication would be started. It is hoped that an early completion date can be obtained since the phase-out of TAN Facility may be imminent and also since the loading costs at TAN were ~ $1800 per shipment in the Third Quarter of FY 1973.

III. ANALYTICAL CHEMISTRY

1. Gamma Scanning

(J. R. Phillips, T. K. Marshall, G. H. Mottaz, J. R. Netuschil, J. N. Quintana

a. Separation of ^{95}Zr(756 keV) and ^{95}Nb(765 keV) gamma-ray peaks: The two adjoining gamma-ray peaks of ^{95}Zr (756 keV) and ^{95}Nb (765 keV) are often summed to obtain the relative burnup as a function of axial position on an irradiated fuel pin. These two gamma photons are separated by only 9 keV, thereby precluding the use of single channel analyzers for determining their individual isotopic distributions. The net areas can be obtained by fitting each of the two peaks with a summation function consisting of a Gaussian function and three exponential functions.[1] A typical fitted spectrum of the ^{95}Zr(756 keV) and ^{95}Nb (765 keV) gamma peak is shown in Fig. 401-5.

The capability of separating adjoining gamma peaks, in particular the ^{95}Zr 756 keV and ^{95}Nb 765 keV peaks, is essential for the investigation of their individual migration properties. This capability was used in

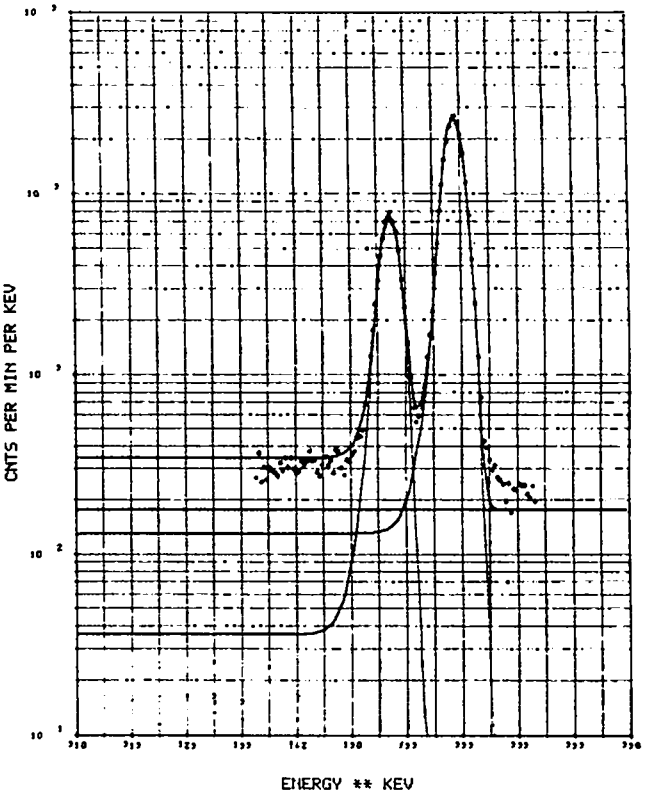

Fig. 401-5. The fitted spectra for the ^{95}Zr 756 keV and ^{95}Nb 765 keV gamma-ray peaks.

measuring the ^{95}Zr and ^{95}Nb axial isotopic distributions individually for a failed mixed-oxide fuel pin. The ^{95}Zr isotopic distribution was determined by the analysis of its 724 and 756 keV gamma-ray peaks (Fig. 401-6). Each data point is represented by error bars equivalent to one standard deviation. There were four prominent spikes in the ^{95}Zr activity at 48.78, 51.25, 52.62, and 56.26 in. The ^{95}Nb axial isotopic distribution (Fig. 401-7) over the same fuel region was significantly different, with six regions of high activity: 48.78, 51.25, 52.62, 56.26, 58.24, and 58.96 in. Four of these regions,48.78, 51.25, 52.62, and 56.26 in., corresponded to spikes in the ^{95}Zr isotopic distribution. However, two of the ^{95}Nb spikes, at 58.24 and 58.96 in., did not have correspondingly high ^{95}Zr concentrations. Also, the relative magnitudes of the ^{95}Nb spikes were significantly different from the ^{95}Zr spikes.

The results indicate that the migration properties of ^{95}Zr and ^{95}Nb are significantly different, at least in this failed fuel pin, with the ^{95}Nb isotope undergoing

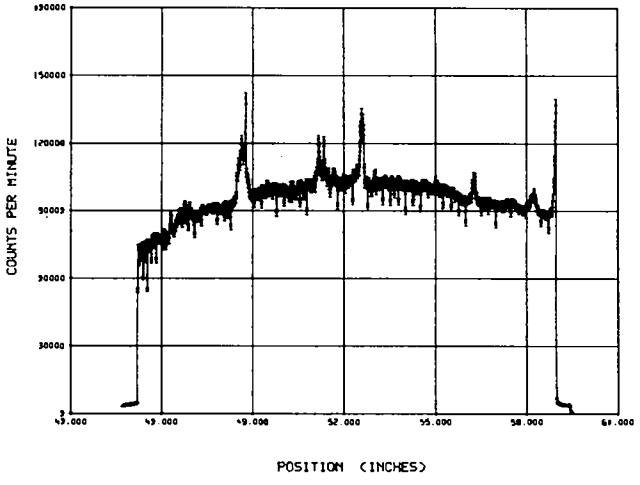

Fig. 401-6. The ^{95}Nb axial isotopic distribution of a failed mixed-oxide fuel pin.

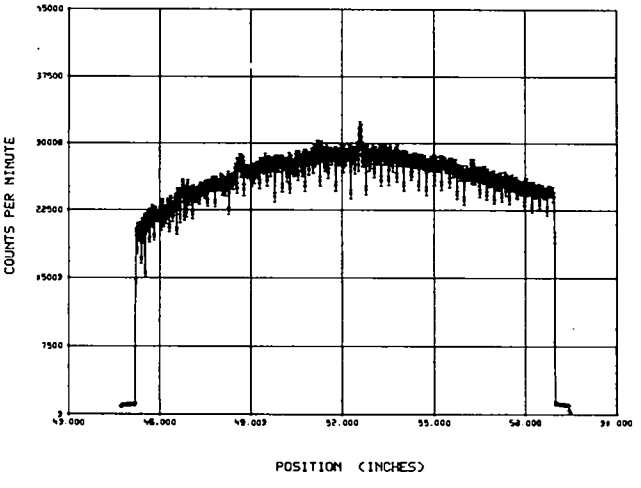

Fig. 401-7. The ^{95}Zr axial isotopic distribution of a failed mixed-oxide fuel pin.

considerable axial relocation. The precision spectral unfolding capability, essential for the detailed analysis of adjacent gamma-ray peaks, will be used in examinations of other fuel pins to determine if differences in ^{95}Zr and ^{95}Nb are common.

b. Data Processing Computer Code: The basic data processing computer code, SURVEY, has been modified to improve the data analysis. An optional data routine has been incorporated for the automatic computation of standard deviations for gross gamma and isotopic distributions.

2. Sealed-Tube Dissolution of Irradiated Fuels[2]
(J. W. Dahlby, R. R. Geoffrion)

The sealed-tube dissolution equipment is being modified to improve sample throughput and to provide for safe operation. With this equipment, difficult soluble materials are dissolved in HCl at 300°C and 4000 psi. A new improved cooling system has been designed to eliminate possible flooding of the hot cell. Also, an attachment is being designed to allow the pressure furnace to be disconnected from the pressure source tank when the sample is at the high temperature and pressure necessary for dissolution, thereby eliminating the possibility of contamination spread due to pressure surges.

3. Determination of Fission Gases Retained in Irradiated Fuels
(J. W. Dahlby, R. R. Geoffrion)

A system is being assembled to dissolve irradiated fuel remotely and collect the released fission gases. The collected fission gases will be analyzed using a mass spectrometer or gas chromatograph. The system for dissolving the irradiated fuel and collecting the fission gases has been built and will be checked by determining the recovery of a calibrated gas standard. The system will be installed in the hot cell, tested under remote conditions, and then used to determine the retained fission gases of irradiated fuel materials.

4. Determination of Water in Mineral Oil
(G. E. Meadows, R. G. Bryan)

A heated mineral oil bath is used for melting the sodium bonding between the pin cladding and capsule before the extraction of the fuel pin. Water in the oil could adversely affect this routine process, even to the extent of being a safety hazard. Therefore, the mineral oil must be periodically analyzed for water contamination and either dried or discarded if found to be wet.

The amount of water is determined by mixing the oil sample with a pretitrated methanol solution of Karl Fisher Reagent (KFR) and then titrating with standard KRF using an automatic titrator. The precision of this method was determined by repeatedly analyzing known amounts of water between 0.25 and 9.48 mg added to the titration

vessel. The relative standard deviation was 1, 8, and 16%, in titrating 9.48, 0.49, and 0.25 mg of water, respectively. The poorer precision of the method for small quantities of water is a result of a variation of ± 0.02 ml in dispensing of the KFR. The method had a positive bias of 9% in measuring 0.25 to 0.5 mg of water.

The solubility of water in mineral oil at 74°F was estimated by equilibrating mineral oil with excess water, centrifuging to remove entrained water, and analyzing the mineral oil for water using KFR. The oil contained 32 ± 5 μg of water per milliliter of oil. Mineral oil samples received to date have contained lesser amounts of water than this value.

IV. REQUESTS FOR DRDT

A. Examination of Irradiated Materials

(R. N. Abernathey, K. A. Johnson, M. E. Lazarus, R. A. Morris, J. R. Phillips, J. W. Schulte, G. R. Waterbury, W. F. Zelezny)

During this Third Quarter of FY 1973 forty-six irradiated capsules were received. The distribution is as follows: GE - 17; HEDL - 20; BMI - 5; LASL - 2; and WARD - 2.

General Electric Company: Examinations performed on irradiated fuel assemblies received on February 14, 1972, October 22, 1972, and March 21, 1973 are shown as follows:

Visual examination was performed on GE capsules F8L, F8N, F8O, F8P, F8Q, FOB, and FOD.

Sectioning was done on pins F4A and E2R.

Three samples of fuel from pins numbered GE-FOA-F, GE-FOA-J, and GE-FOC-J and two samples of cladding from pins numbered GE-FOA and GE-FOC were analyzed for oxygen content. A spectrochemical analysis for impurities in the fuel was done on section F of GE-FOC.

Sections B, D, E, and F of GE-E2R and sections E of both GE-FOA and GE-FOC were examined using the shielded microprobe.

The atom percent burnup was determined on each of the following pins: GE-FOA, GE-FOC, GE-F12P, GE-F12Q, and GE-E2R, using an isotope dilution mass spectrometric method.

Microstructural examinations consisting of macrophotography, alpha and beta-gamma autoradiography and optical microscopy (including mosaics) were carried out in an Ar atmosphere on specimens listed in Table 401-I.

TABLE 401-I

MICROSTRUCTURAL ANALYSIS OF GE MATERIALS

GE Pin No.	No. Samples Fuel and Clad	EMX (Sample Prep.)
FOC	2	1
E2R	7	2
F4A	1	

Ten structural capsules (L-16 Series) were received from GE on February 16, 1973 and are being stored until time is available to work these into the priority schedule.

Hanford Engineering Development Laboratory: During this report period (3rd Quarter FY 1973) shipments were received as follows: 9 capsules on February 1 and 11 capsules on March 21. Examinations performed on HEDL materials during this period are listed in this section. (Tables 401-II and 401-III).

The fuel pins listed in Table 401-IV were nondestructively examined using the gamma scanner.

Atom percent burnup was determined by dissolving and analyzing a cross-section sample of fuel and clad on HEDL P-17A-33.

The pin gas was analyzed mass spectrometrically in HEDL-P-20-21, while both the cover gas and pin gas were analyzed in the following pins: HEDL-P-19-1R, P-19-14, P-20-5, P-20-12, P-20-15, P-20-28, P-20-36, and PNL-2-13.

Microstructural examinations consisting of macrophotography, alpha and beta-gamma autoradiography, and optical microscopy (including mosaics) were carried out in an Ar atmosphere on the specimens as listed in Table 401-V.

Los Alamos Scientific Laboratory: This section contains carbide and nitride fuel pins, the technical evaluation of which is being carried out by LASL personnel under the Advanced Pu Fuels Program.

TABLE 401-II

POSTIRRADIATION EXAMINATIONS
OF HEDL CAPSULES

	Examination	Capsule Identity, HEDL
1.	Visual Inspection	PNL-2-13, P-19-1R, P-19-14, P-20-1, P-20-3, P-20-5, P-20-9, P-20-11, P-20-12, P-20-15, P-20-18, P-20-20, P-19-23, P-20-21, P-20-24, P-20-26, P-20-28, P-20-32, P-20-34R, P-20-36
2.	Preliminary Measurements	PNL-2-13, P-19-1R, P-19-14, P-20-5, P-20-12, P-20-15, P-20-21, P-20-28, P-20-36
3.	Profilometry, Optical	PNL-2-13
4.	Photography, Max. Bow	PNL-2-13
5.	Radiography	Same as Item 2
6.	Gamma Scan	PNL-2-13
7.	Eddy Current	Same as Item 2
8.	Cover Gas Sample	Same as Item 2
9.	Na Removal	Same as Item 2
10.	Clad Removal	Same as Item 2

TABLE 401-III

POSTIRRADIATION EXAMINATIONS
OF HEDL PINS

	Examination	Pin Identity, HEDL
1.	Visual Examination	P-19-1R, P-19-14, P-20-5, P-20-12, P-20-15, P-20-21, P-20-28, P-20-36
2.	Profilometry, Optical	Same as Item 1
3.	Profilometry, Mechanical	PNL-2-13
4.	Radiography	Same as Item 1
5.	Photography, Incremental	PNL-2-13
6.	Gamma Scanning	Same as Item 1
7.	Eddy Current	PNL-2-13
8.	Wire Wrap Removal	Same as Item 1
9.	Fission Gas Sample	P-20-5, P-20-12, P-20-15, P-20-21, P-19-1R, P-19-14, P-20-28, P-20-36, PNL-2-13
10.	Sectioning	P-20-5, P-20-12, P-20-15, P-20-21, P-19-1R, P-19-14, P-20-28, P-20-36

TABLE 401-IV

GAMMA SCANNING OF HEDL PINS

Fuel Pin Number	No. of Gamma Scans		No. of Isotopic Distributions Calculated
	Gross	Multispectral	
P-20-5	8	-	-
P-20-12	4	1	9
P-20-15	4	-	-
P-20-21	4	-	-
P-20-28	4	2	12
P-20-36	4	1	10
P-19-1R	4	-	-
P-19-14	4	-	-
PNL-2-13	4	2	10

TABLE 401-V

MICROSTRUCTURAL ANALYSES
OF HEDL MATERIALS

HEDL Pin No.	No. Samples Fuel and Clad
P-20-12	3
P-20-5	3
P-20-15	3
P-20-21	3

BMI Experiments --- Examinations performed on nine irradiated capsules received on October 11, 1972 and February 16, 1973, are shown in Table 401-VI.

A summary of the gamma scanning which was applied to the nondestructive examination of five fuel pins is outlined in Table 401-VII.

Cover gas and pin gas samples were analyzed in each of the three fuel pins, BMI-3-2, BMI-3-4, and BMI-3-5.

Microstructural examinations consisting of macrophotography, alpha and beta-gamma autoradiography, and optical microscopy (including mosaics) were carried out in an Ar atmosphere on six specimens from BMI-3-5.

Gulf United Experiments --- Postirradiation examinations made on irradiated fuel pins received from the Gulf United program are shown in Table 401-VIII.

TABLE 401-VI

POSTIRRADIATION EXAMINATIONS OF CAPSULES AND PINS FROM BMI

Examination	Capsule Identity, BMI	Pin Identity, BMI
1. Visual Inspection	1-1, 1-2, 2-2, 2-6, 2-7	3-2, 3-4, 3-5
2. Prelim. Measurements	Same as Item 1	
3. Profilometry - Optical	Same as Item 1	
4. Photography, Full Length	Same as Item 1	3-2, 3-4, 3-5
5. Photography, Incremental	Same as Item 1	3-2, 3-4, 3-5
6. Radiography	Same as Item 1	
7. Gamma Scanning	Same as Item 1	
8. Eddy Current	Same as Item 1	3-2
9. Cover Gas Sampling	2-2, 2-6, 3-2, 3-4, 3-5	
10. Na Removal	3-2, 3-4, 3-5	
11. Clad Removal	3-2, 3-4, 3-5	
12. Profilometry, Mechanical		3-2, 3-4
13. Micrometer Measurements		3-5
14. Sectioning		3-2, 3-4, 3-5

TABLE 401-VII

GAMMA SCANNING OF BMI FUEL PINS

Fuel Pin Number	No. of Gamma Scans Gross	No. of Gamma Scans Multispectral	No. of Isotopic Distributions Calculated
BMI-1-1	4	1	10
BMI-1-2	4	-	-
BMI-2-2	4	-	-
BMI-2-6	4	-	-
BMI-2-7	4	-	-

Gamma scanning, including gross and complete spectral scans, was applied to the examination of the fuel pins listed in Table 401-IX.

The following four samples were examined using the shielded electron microprobe: UNC-189-F, -198-K, -200-G, and -206-E.

Cover gas samples in UNC-97 and UNC-113 and pin gas samples in UNC-93, UNC-94, and UNC-105 were analyzed mass spectrometrically.

Atom percent burnup was determined on each of the following ten fuel pins: UNC-187, -189, -191, -192, -195, -197, -198, -200, -206, and -208.

Clad samples from the following ten pins were cleaned and prepared for density measurement: UNC-187, -189, -191, -192, -195, -197, -198, -200, -206, and -208.

TABLE 401-VIII

POSTIRRADIATION EXAMINATIONS OF GU CAPSULES AND PINS

Examination	Capsule Identity, UNC	Pin Identity, UNC
1. Photography, Full Length and Maximum	114, 185, 186, 202, 203, 204, 205	
2. Gas Sampling	93, 94, 97, 105, 110, 113	93, 94, 105
3. Na Removal	93, 94, 97, 105, 110, 113	
4. Clad Removal	93, 94, 97, 105, 110, 113	
5. Gamma Scan	202, 203, 204, 205, 106	
6. Eddy Current	186, 202, 203, 204, 205	191, 192, 195, 197, 198, 200, 208
7. Visual Inspection		93, 94, 97, 105, 110, 113
8. Preliminary Measurements		93, 94, 97, 105, 110, 113
9. Profilometry, Optical		93, 94, 97, 106, 110, 113
10. Photography, Full Length and Maximum Bow		93, 94, 97, 105, 110, 113
11. Photography, Incremental		93, 94, 97, 105, 110, 113
12. Sectioning		93, 94, 106, 191, 192, 195, 197, 198, 200, 208
13. Na Distillation		192, 195, 197
14. Density Measuremeots		187, 189, 191, 192, 194, 195, 197, 198, 200, 206, 208

TABLE 401-IX

GAMMA SCANNING OF GU PINS

Fuel Pin Number	No. of Gamma Scans Gross	No. of Gamma Scans Multispectral	No. of Isotopic Distributions Calculated
UNC-106	-	1	8
UNC-202	4	-	-
UNC-203	4	-	-
UNC-204	4	-	-
UNC-205	4	-	-

Microstructural examinations consisting of macrophotography, alpha and beta-gamma autoradiography, and optical microscopy (including mosaics) were carried out in an Ar atmosphere on the specimens listed in Table 401-X.

LASL Experiments --- Examinations performed on irradiated fuel pins from the LASL-identified pins are listed in Table 401-XI.

Four gross gamma scans were obtained on each of two fuel pins, LASL-K-38B and LASL-K-43.

TABLE 401-X

MICROSTRUCTURAL ANALYSES OF GU MATERIALS

Pin Identity	No. Samples Fuel and Clad	Clad	EMX Prep.
UNC-194	3		
UNC-187	3	1	
UNC-189	3	1	2
UNC-206	2	2	1
UNC-192	1	2	
UNC-195	2	2	
UNC-197	2	2	
UNC-200	2	1	1
UNC-208	3	1	
UNC-191	4	-	
UNC-198	3	-	1
UNC-93	3	-	
UNC-94	4	-	
UNC-105	3	-	
UNC-192	1	-	

TABLE 401-XI

POSTIRRADIATION EXAMINATIONS OF CAPSULES FROM LASL

	Examination	Capsule Identity, LASL	Pin Identity, LASL
1.	Visual Inspection	K-36B, K-43	K-36B, K-46
2.	Profilometry, Optical	K-36B, K-43	
3.	Radiography	K-36B, K-43	
4.	Gamma Scanning	K-36B, K-43	
5.	Eddy Current	K-36B, K-43	
6.	Photography, Full Length	K-36B, K-43	
7.	Photography, Maximum Bow	K-36B, K-43	K-36B, K-40
8.	Photography, Incremental	K-36B, K-43	K-36B, K-46
9.	Gas Sampling	K-36B, K-38B, K-43, K-46, K-49, K-50, K-51	K-36B, K-38B, K-43, K-46, K-49, K-50, K-51
10.	Na Removal	K-36B, K-46	
11.	Clad Removal	K-46	
12.	Profilometry, Mechanical		K-36B
13.	Sectioning		K-36B, K-49

Section K of LASL-K-49 was examined using the shielded electron microprobe.

The water and chlorine contents were determined in a sample of mineral oil used as the heat transfer medium in melting the sodium bonds in irradiated fuel pins.

Mass spectrometry was applied to the analysis of cover gas and pin gas in each of the following fuel pins: LASL-K-36B, -K-38B, -K-43, -K-46, -K-49, -K-50, and -K-51.

Two clad samples from LASL-K-49 were cleaned and prepared for density measurement.

Microstructural examinations consisting of macrophotography, alpha and beta-gamma autoradiography, optical microscopy (including mosaics) were carried out in an Ar atmosphere on 6 fuel and clad specimens from LASL K-49 and 5 fuel and clad samples from LASL K-36B. One sample was also prepared for EMX from LASL pin K-49.

ORNL Experiments --- Pulsed eddy current scanning was made of ORNL 43-N1.

WARD Experiments --- Examinations performed on irradiated fuel pins received on February 1, 1973 are listed in Table 401-XII.

TABLE 401-XII

POSTIRRADIATION EXAMINATIONS OF WARD CAPSULES AND PINS

	Examination	Capsule Identity, WARD	Pin Identity, WARD
1.	Visual Inspection	W4F, W8F	W4F, W8F
2.	Preliminary Measurements	W4F, W8F	
3.	Profilometry, Optical	W4F, W8F	W4F, W8F
4.	Photography, Full Length	W4F, W8F	
5.	Photography, Incremental	W4F, W8F	
6.	Photography, Maximum Bow	W4F, W8F	
7.	Radiography	W4F, W8F	
8.	Gamma Scanning	W4F, W8F	
9.	Eddy Current	W4F, W8F	
10.	Cover Gas Sampling	W4F, W8F	
11.	Na Removal	W4F, W8F	
12.	Clad Removal	W4F, W8F	
13.	Temperature Measurements		W4F, W8F

V. QUALITY ASSURANCE

Microstructural Analysis: The Microstructural Analysis procedure for diagnostic examinations has been revised and updated. Traveler documentation has been expanded and is in use. A training program for operators has been completed and certification records prepared. Calibration procedures have been written. Instruments have been calibrated, labeled, and required documents have been supplied to the Quality Assurance Manager.

An audit conducted by the Quality Assurance Manager has been completed and an audit report prepared.

Hot Cell Examinations: The Quality Assurance Plan for Hot Cell Examinations has been revised. Procedures for Hot Cell Diagnostic Examinations have been revised and updated. New procedures have been added. The training program has been extended to include the use of designated trainees under the supervision of certified operators.

Calibrated instruments are in use, labels in place and records have been supplied to the Quality Assurance Manager.

An audit was conducted by the Quality Assurance Manager in both the Wing 9 and DP West areas and an audit report was prepared.

Chemical Analysis: The changes made in the Analytical Quality Assurance Plans have been approved and are being used. The traveler system has been revised and new forms prepared. The revised Analytical Chemistry Quality Assurance Procedures have been accepted by all sponsors of diagnostic examinations and are incorporated into the quality assurance system. Certifications for analysts have been revised and updated.

The recall system for scheduled recalibration of instruments is being used and documentation is being supplied to the Quality Assurance Manager.

An audit was conducted by the Quality Assurance Manager in all of the Analytical Chemistry areas and an audit was prepared.

VI. REFERENCES

1. W. M. Sanders and D. M. Holm, "An Analytical Method for Unfolding Gamma-Ray Spectra," Los Alamos Scientific Laboratory report LA-4030 (March 1969).
2. "Quarterly Status Report on the Advanced Plutonium Fuels Program, January 1 - March 31, 1971," Los Alamos Scientific Laboratory report LA-4693-MS, p. 6, (1971).

PROJECT 463

HIGH PERFORMANCE LMFBR FUEL MATERIALS

Person in Charge: R. D. Baker
Principal Investigator: J. L. Green

I. INTRODUCTION

The primary objective of this program is the overall evaluation of the most promising of the candidate fuel systems for advanced LMFBR application. Emphasis currently is placed on the study of the relative merits of stainless steel clad nitride and carbide fuels under conditions that appropriately exploit the potential of these materials to operate to high burnup at high power densities. The major portion of the program is the evaluation of the irradiation performance of these fuel element systems. A continuing series of irradiation experiments is being carried out under steady state conditions in fast reactor environments to assess the effects of damage and burnup on stainless steel clad, carbide and nitride fuel elements. These experiments are designed to investigate fuel swelling, interactions between the fuel and clad and thermal bonding medium, fission gas release, and the migration of fuel material and fission products as a function of burnup and irradiation conditions. In addition, experiments are being designed to allow the study of the effects of rapid, overpower, reactor transients on carbide and nitride fuel assemblies. Contiguous efforts are necessary in the development of fuel material preparation and fabrication procedures as well as the techniques required for the characterization of fuel materials both before and after irradiation.

A second objective in the program is the determination of thermophysical, mechanical and chemical properties and characteristics of plutonium-containing ceramics that are required for their evaluation and use as fuel materials. A broad range of capabilities in this area has been developed, including the study of (1) phase relationships using differential thermal analysis, (2) thermal transport, (3) thermal stability and compatibility, (4) hot hardness and its temperature dependence, (5) structure and phase relationships using high temperature x-ray and neutron diffraction, (6) thermal expansion, and (7) compressive creep rates as a function of temperature and stress. Several of these techniques are available for use with irradiated fuels.

II. IRRADIATION TESTING

The objective of the irradiation testing program is the overall evaluation of the most promising of the candidate fuel systems for advanced LMFBR application. The irradiation experiments are carried out under conditions that take advantage of the potential of these materials to operate to high burnup at high power densities.

A. Synthesis and Fabrication

(K. W. R. Johnson, J. G. Reavis, H. Moore, R. Walker, C. Baker)

1. Process Development. Development work on a process for the preparation of high density, single phase $U_{0.8}Pu_{0.2}C$ by reduction of UO_2-PuO_2 mixtures by heating in vacuo with graphite has previously been reported.[1,2] Three additional production scale (250 g) batches of carbide have been produced by this method for the purpose of further investigation of the effects of processing variables. The products of the reduction step

have been split into smaller batches for the refinement of the additional processing steps of comminution, H_2 treatment, pressing, and sintering. The parameters which have been the basis for evaluation are chemical composition, density, and microstructure.

Results of these studies are presented in Table 463-I. Since very few batches of carbide were prepared, the degree of uncertainty in concentrations and concentration changes which can potentially occur in the preparation of additional batches is significant. Certain trends, however, seem to be present. For instance, the average oxygen concentration in the reduction products (before H_2 treatment) as shown by batches 12, 13, and 14 was 160 ppm. It appears that oxygen concentrations of less than 200 ppm can be achieved routinely by the carbothermic reduction process on the 250 g scale. All subsequent processes, including comminution, H_2 treatment, pressing and sintering, can be expected to add less than 200 ppm oxygen to the carbide. Batches other than 12, 13 and 14 listed in Table 463-I are smaller than production size 30 g to 150 g) and the oxygen contamination of such small batches could be relatively more serious than for larger batches.

Plutonium losses, possibly because of selective volatilization, appear to be larger than usual in these experiments. The processing implications of this Pu loss are not clear, since the path of the loss was not determined in these experiments. It appears that about half the loss is in the reduction step and half is in all subsequent steps.

TABLE 463-I

COMPOSITION OF (U, Pu)C PRODUCED USING THE CARBOTHERMIC REDUCTION PROCESS

Batch No.	Oxygen, ppm	Pu / U + Pu Oxide + C	Pu / U + Pu Product	H_2 Treatment Time, h	C/M
12	160	19.7	19.3	0	--
12-I	385	19.7	19.0	65	0.97
12-III	290	19.7	19.0	66	0.98
12-IV	365	19.7	--	4	0.98
13	90	20.5	20.3	0	--
13-I	--	20.5	19.5	3.5	1.00
13-II	325	20.5	19.6	50	0.95
13-III	365	20.5	19.4	30	0.97
14	100	20.5	20.0	0	--
14-I	350	20.5	19.7	30	0.98

Another factor considered in Table 463-I is the change of carbon/metal atomic ratios with the time of H_2 treatment. As the process is usually run, the H_2 treatment time is 0.6 h per gram of carbide. This was not the procedure used for the preparations listed in Table 463-I. In these runs, H_2 treatment was terminated after the times listed and pellets prepared from this material were analyzed to determine the C/M atomic ratio. The pellets were examined by use of metallographic techniques to determine the number of phases present. Examination of the data of Table 463-I does not reveal a close correlation between the H_2 treatment times and the C/M ratios. At the extreme, the shortest H_2 treatment time (3.5 h) gave the product with the highest C/M ratio (1.00); little further correlation exists. The sample with the shortest H_2 treatment time was also the only product which contained traces of M_2C_3 and C-rich platelets, indicating a high C content. All the other H_2-treated products were single phase monocarbide.

Another type of measurement which promises to be useful in process evaluation and control is analysis of the exhaust gas from the H_2 treatment furnace by gas chromatography. Methane was periodically determined in this gas stream during preparation of the last four carbide batches. It was found that of the order of 90% of the CH_4 formed during 30 to 50 h runs was liberated during the first 10 h of treatment. About 95% of the CH_4 was evolved during the first 20 h, but a detectable amount of CH_4 was still being formed even after 50 h. It appears from the CH_4 evolution rate and from data in Table 463-I that single phase monocarbide can be produced in much shorter than normal H_2 treatment periods.

Densities achieved in this series were randomly distributed over the range 87 to 94% of the theoretical density, but there was no apparent correlation between densities and H_2 treatment times. At present, the production of fuel pellets consistently having the desired densities (95 ± 2% of theoretical) requires the use of the arc melting process. Further study of the effect of processing parameters on the sintered density of

carbothermically produced material will be necessary.

A major effort has been directed toward developing and applying QA procedures for use in the fuel preparation task in the program. A number of formalized process control procedures have been prepared. These include detailed processing instructions, data forms, transfer and storage documentation, sampling procedures, etc. Considerable progress has been made in the area of instrument control and calibration. A large number of instruments have been designated as data gathering devices, and, therefore, must be calibrated prior to use. The calibration system involves several different areas:

a. identifying data gathering devices,

b. preparing calibration procedures for instruments to be calibrated by project personnel,

c. training and certifying personnel,

d. procuring certified standards and devices to be calibrated and certified by the manufacturer,

e. obtaining calibrations and certifications from standards laboratories when required,

f. carrying out routine calibrations and tagging, and

g. maintaining local records of calibration activities.

All calibrated instruments and standards are included in the central files for calibrated instruments and are part of the QA recall system.

B. EBR-II Irradiation Testing

(J. O. Barner, K. W. R. Johnson, J. F. Kerrisk, T. W. Latimer, L. L. Marriott, H. E. Strohm)

The purpose of the EBR-II testing program is the evaluation of the steady-state irradiation behavior of high performance fuel element systems for application in advanced LMFBR reactors. Several series of carbide- and nitride-fueled experiments have been initiated in the past several years. The main objectives of the irradiations are: (1) the development of fuel element designs for use with each fuel type; (2) the determination of the irradiation behavior of the fuel materials; (3) a comparison of sodium and helium bonding; (4) a comparison of different cladding alloys; and (5) the evaluation of the overall irradiation performance of the fuel element systems. The majority of the experiments under test or that have been completed have been encapsulated. Most of the experiments that are currently available for irradiation or that are being designed are singly clad.

Fourteen series of experiments have been originated. The description and status of these series are summarized in Tables 463-II to 463-IX. In order to better define the status of those experiments which are undergoing postirradiation examination, the following steps are referenced in the tables:

1. Capsule Examination
   - 1.1 Visual examination
   - 1.2 Preliminary Measurements (radiation measurements, etc.)
   - 1.3 Profilometry
   - 1.4 Photography
   - 1.5 Radiography
   - 1.6 Eddy Current Test
   - 1.7 Gamma Scan
   - 1.8 Cover Gas Analysis
   - 1.9 Deencapsulation
2. Element Examination
   - 2.1 Visual Examination
   - 2.2 Profilometry
   - 2.3 Photography
   - 2.4 Eddy Current Test
   - 2.5 Fission Gas Analysis
   - 2.6 Sectioning
   - 2.7 Autoradiography
   - 2.8 Metallography
   - 2.9 Burnup
   - 2.10 Clad Density
   - 2.11 Special Tests
   - 2.12 Data Reduction
   - 2.13 Report Preparation

Table 463-II describes the K1, K2, and K3 series tests. In these experiments single-phase, high-purity, uranium-plutonium monocarbide pellets are sodium bonded to Type 316 stainless steel cladding. In general, the operating linear power ratings of the capsules are relatively high (approximately 30 Kw/ft). Three tests at very high power (> 45 Kw/ft) were included to determine the effect of high thermal stresses and high fuel temperatures on fuel element behavior. Indications of element cladding

TABLE 463-II

SERIES K1, K2, AND K3 ENCAPSULATED CARBIDE EXPERIMENT

Expmt. No.	Fuel Type[a]	Fuel Density, % Theo.	Bond and Diametral Gap, in.	Clad[h] Type	Clad O.D. x I.D.,[g] in.	Max. Linear Power, Kw/ft	Maximum Centerline Temp., °C	Goal Burnup, at.%	Current Burnup, at.%	Status
				Series K1						
K-36B	$(U_{0.8}Pu_{0.2})C$	90	Na-0.015	SA-316SS	0.300 x 0.280	30	1165	6	6.8	Exam, 2.8[e]
K-37B	$(U_{0.8}Pu_{0.2})C$	90	Na-0.015	SA-316SS	0.300 x 0.280	30	1165	6	3.2	Exam, 1.7[b,e]
K-38B	$(U_{0.8}Pu_{0.2})C$	90	Na-0.015	SA-316SS	0.300 x 0.280	30	1165	6	6.4	Exam, 1.7[c,e]
K-39B	$(U_{0.8}Pu_{0.2})C$	90	Na-0.015	SA-316SS	0.300 x 0.280	30	1165	10[i]	6.4	EBR-II, Unassigned
K-42B	$(U_{0.8}Pu_{0.2})C$	90	Na-0.015	SA-316SS	0.300 x 0.280	30	1165	6	5.0	Completed[d]
				Series K2						
K-49	$(U_{0.8}Pu_{0.2})C$	95	Na-0.020	SA-316SS	0.300 x 0.280	45-50	1400	5	4.0	Exam, 2.8[e]
K-50	$(U_{0.8}Pu_{0.2})C$	95	Na-0.020	SA-316SS	0.300 x 0.280	45-50	1400	6.5	4.0	Exam, 1.9[e]
K-51	$(U_{0.8}Pu_{0.2})C$	95	Na-0.020	SA-316SS	0.300 x 0.280	45-50	1400	8	3.9	Exam, 1.9[e]
				Series K3						
K-43	$(U_{0.8}Pu_{0.2})C$	94	Na-0.020	SA-316SS	0.300 x 0.280	30	1150	8	6.1	Exam, 1.8[e]
K-44	$(U_{0.8}Pu_{0.2})C$	94	Na-0.020	SA-316SS	0.300 x 0.280	30	1150	8	6.1	EBR-II, X182
K-45	$(U_{0.8}Pu_{0.2})C$	94	Na-0.020	SA-316SS	0.300 x 0.280	30	1150	5	3.0	Exam, 1.7[f]
K-46	$(U_{0.8}Pu_{0.2})C$	94	Na-0.020	SA-316SS	0.300 x 0.280	30	1150	5	2.9	Exam, 2.4[e]

[a] Series 1 and 3 experiments are 93% enriched in ^{235}U. Series 2 experiments are 97% enriched in ^{233}U. All fuel is single phase.

[b] K-37B was damaged during reconstitution of X152 to the extent that it cannot be irradiated further.

[c] K-38B was damaged during reconstitution of X152. Additional irradiation was completed.

[d] Reported in LA-4669-MS

[e] Element cladding failure indicated.

[f] Nondestructive examination completed. Capsule stored for possible irradiation of failed element.

[g] Dimensions are nominal.

[h] SA = solution annealed

[i] Original goal burnup was 6 at.%. New AIP request for further irradiation in preparation.

failure were found at EBR-II in several experiments from these series; five in subassembly X119B, one from X142, and two from X152, using γ-scanning for ^{133}Xe. Examinations of these experiments in the LASL hot cells confirmed these failures. Complete postirradiation examination of the failed experiments is currently under way.

Table 463-III describes the Series U1300 experiments. These experiments contain two-phase, uranium-plutonium carbide fuel pellets which are helium bonded to either Type 316 stainless steel or Incoloy 800 cladding. Two methods for the accommodation of fuel swelling were investigated in this series, i.e., the introduction of internal porosity by the use of either low-density solid fuel pellets or high-density cored pellets. These experiments reached their goal burnup of 10 at.% in subassembly X142 after operation at moderate linear power ratings (approximately 20 Kw/ft). Indications of element cladding failure for three experiments were found at EBR-II using γ-scanning for ^{133}Xe. These element failures have been confirmed by γ-scanning for ^{137}Cs at LASL. All of the capsules in the series are currently undergoing nondestructive or destructive examination in the LASL hot cells.

The Series U1950 experiments are described in Table 463-IV. In these experiments, either two-phase or single-phase carbide fuel is helium bonded to Type 304 or 316 stainless steel or to Incoloy 800 cladding. Fuel densities range from 75 to 99% of theoretical. These experiments are currently at about three-fourths of their goal burnup after operation at low linear power (10 to 15 Kw/ft).

TABLE 463-III

SERIES 1300 ENCAPSULATED CARBIDE EXPERIMENTS

Expmt. No.	Fuel Type[a]	Fuel Density, % Theo.	Bond and Diametral Gap, in.	Clad[d] Type	Clad O.D. x I.D., in.	Max. Linear Power, Kw/ft	Maximum Centerline Temp., °C	Goal Burnup, at.%	Current Burnup, at.%	Status
U93	MC+5% M_2C_3	84	He-0.004	SA-316SS	0.302 x 0.248	18.0	1750	11	11.1	Exam, 2.8
U94	MC+5% M_2C_3	83	He-0.007	SA-316SS	0.305 x 0.274	21.9	1680	11	10.7	Exam, 2.8
U97	MC+5% M_2C_3	84	He-0.004	SA-INC-800	0.299 x 0.245	18.0	1750	11	11.0	Exam, 1.8
U98	MC+5% M_2C_3	84	He-0.007	SA-INC-800	0.299 x 0.269	21.9	1680	11	10.6	Exam, 1.8[c]
U105	MC+5% M_2C_3	76	He-0.008	SA-INC-800	0.300 x 0.246	15.1	1900	11	11.5	Exam, 2.8
U106	MC+5% M_2C_3	75	He-0.009	SA-INC-800	0.304 x 0.274	19.8	1825	11	10.9	Exam, 1.8[c]
U110	MC+10% M_2C_3	96[b]	He-0.014	SA-INC-800	0.304 x 0.274	21.9	1960	10	10.1	Exam, 1.8
U113	MC+10% M_2C_3	96[b]	He-0.010	SA-INC-800	0.300 x 0.246	16.9	1880	11	11.4	Exam, 1.8
U114	MC+10% M_2C_3	96[b]	He-0.007	SA-INC-800	0.304 x 0.274	22.1	1575	10	10.4	Exam, 1.8[c]

[a] $M = (U_{0.85}Pu_{0.15})$

[b] Cored pellet with nominal 0.080 in. diameter axial hole.

[c] Element cladding failure indicated.

[d] SA = Solution annealed

During interim examination at EBR-II after run 58, ^{137}Cs was detected by γ-scanning in the sodium reservoir of capsule U136. Release of fission gas from a breached helium-bonded element would be expected. However, no ^{133}Xe was detected in the capsule plenum. The lack of fission gas in the capsule and the presence of ^{137}Cs in the capsule sodium present a contradictory picture and the failure of the element in capsule 136 can only be considered tentative and of a low degree. None of the other capsules indicated fuel element failure during the examinations at EBR-II. All 19 capsules were reconstituted into subassembly X055B which is currently being irradiated.

The Series U1930 and U1960 experiments are described in Table 463-V. Experimental parameters include fuel type, fuel density, bond type, and cladding type. The operating linear power ratings for the experiment are relatively high (30-35 Kw/ft). Nondestructive examination of the eleven experiments listed in part A of Table 463-V was completed several months ago. The results of these examinations showed that fuel elements U194 and U200 had failed. Destructive examination of this group of experiments has been completed. Data reduction and interpretation are currently under way.

The experiments listed in part B of Table 463-V are currently in subassembly X182 which is in the EBR-II storage basket. The subassembly is to be inserted for run 63 in May, 1973. No element cladding failures have been indicated in this group of capsules.

The experiments listed in part C of Table 463-V were used as replacement capsules in order to allow the irradiation to be continued to the desired burnup in lead experiments from other series. Only a cursory postirradiation examination is planned for these elements. Nondestructive examination of the experiment is complete. The experiments listed in part D of Table 463-V are awaiting insertion into the reactor. Capsule U261 will be returned to LASL for rework of an apparent sodium bond defect in the capsule-element annulus.

Table 463-VI describes the Series WF experiments. These sodium-bonded, carbide capsules were designed to evaluate the effects of (1) various amounts of sesquicarbide in the fuel, (2) linear power rating, and (3) cladding cold work on element performance. The amount of sesquicarbide reported to be in the fuel varies from 0 to 24 vol%. Results from γ-scanning for ^{133}Xe at EBR-II indicate that the element cladding for all of these experiments

TABLE 463-IV

SERIES U1950 ENCAPSULATED CARBIDE EXPERIMENTS

Expmt. No.	Fuel Type[a]	Fuel Density % Theo.	Bond and Diametral Gap, in.[e]	Clad Type[f]	Clad O.D. x I.D., in.	Max. Linear Power, Kw/ft	Maximum Centerline Temp., °C	Goal Burnup, at.%	Current Burnup, at.%	Status
U129	MC+5% M_2C_3	84	He-0.022	SA-316SS	0.303 x 0.260	12.8	1755	11	8.5	EBR-II, X055B
U130	MC+5% M_2C_3	75	He-0.022	SA-316SS	0.303 x 0.260	13.1	1500	11	8.5	EBR-II, X055B
U131	MC+5% M_2C_3	84	He-0.022	SA-316SS	0.303 x 0.260	13.1	1495	11	8.4	EBR-II, X055B
U132	MC+5% M_2C_3	84	He-0.022	SA-316SS	0.303 x 0.260	12.8	1495	11	8.3	EBR-II, X055B
U133	MC+5% M_2C_3	84	He-0.022	SA-316SS	0.303 x 0.260	12.8	1495	11	8.1	EBR-II, X055B
U134	MC+5% M_2C_3	84	He-0.022	SA-316SS	0.303 x 0.260	12.8	1495	11	8.1	EBR-II, X055B
U135	MC+5% M_2C_3	84	He-0.022	SA-INC-800	0.302 x 0.260	12.8	1475	11	6.4	EBR-II, X055B
U136[d]	MC+5% M_2C_3	84	He-0.022	SA-INC-800	0.302 x 0.260	13.3	1475	11	8.0	EBR-II, X055B
U137	MC+10% M_2C_3	99	He-0.022	SA-316SS	0.303 x 0.260	13.4	1440	10	7.1	EBR-II, X055B
U138A[b]	MC+10% M_2C_3	99	He-0.022	SA-316SS	0.293 x 0.260	14.8	1440	8	3.5	EBR-II, X055B
U139	MC+10% M_2C_3	99	He-0.022	SA-INC-800	0.304 x 0.260	14.8	1440	10	7.2	EBR-II, X055B
U140	MC	93	He-0.022	SA-INC-800	0.302 x 0.260	13.9	1460	10	7.7	EBR-II, X055B
U141	MC	93	He-0.022	SA-316SS	0.303 x 0.260	14.3	1460	10	7.6	EBR-II, X055B
U142	MC	93	He-0.022	SA-316SS	0.304 x 0.260	14.5	1460	11	7.7	EBR-II, X055B
U143	MC+10% M_2C_3	99[c]	He-0.022	SA-INC-800	0.302 x 0.260	12.8	1395	11	7.9	EBR-II, X055B
U144	MC+10% M_2C_3	99[c]	He-0.022	SA-316SS	0.304 x 0.260	13.1	1395	11	8.0	EBR-II, X055B
U145	MC	93	Na-0.030	SA-304SS	0.305 x 0.270	13.4	820	10	7.5	EBR-II, X055B
U146A[b]	MC+10% M_2C_3	99	Na-0.030	SA-304SS	0.300 x 0.270	13.7	810	8	3.5	EBR-II, X055B
U147	MC+10% M_2C_3	99	Na-0.030	SA-INC-800	0.304 x 0.270	14.2	810	10	7.6	EBR-II, X055B

[a] $M = (U_{0.85}Pu_{0.15})$

[b] Capsules 138 and 146 were removed at 45,000 MWD/MT for Treat testing. Duplicates replaced the originals.

[c] Pellets cored with nominal 0.080-in. diam axial hole.

[d] Possible element cladding failure indicated.

[e] All cladding I.D. and gap measurements are nominal.

[f] SA = Solution annealed.

is intact. Two of the eight capsules are currently being destructively examined at LASL. Three experiments are to continue irradiation in subassembly X182 starting in May, 1973. It is planned that the remaining three capsules will be reinserted into the reactor. Further irradiation is pending the preparation and approval of a request for approval-in-principle from the AEC to extend the burnup limit beyond 6 at.%.

Table 463-VII describes the Series B-1, B-2 and B-3 experiments. These capsules are fueled with single-phase, uranium-plutonium mononitride. All the elements in Series B-1 and B-2 are sodium-bonded and clad with either Type 304 or 316 welded stainless steel tubing. Operating linear power ratings for the experiments are relatively high (25-35 Kw/ft). Capsules B-1-4 and B-2-5 have been examined using γ-scanning techniques for the detection of ^{137}Cs, and both elements are apparently intact. Further irradiation of these two capsules is planned. The remaining experiments from this series were recently removed from subassembly X152. During the interim examination, capsules B-1-1, -1-2, -2-2, -2-6, and -2-7 were found to have failed as indicated by γ-scanning for ^{133}Xe at EBR-II. The elements in capsules B-2-1 and B-2-3 were found to be intact. Capsule B-2-3 will continue irradiation in subassembly X182. Further irradiation of capsule B-2-1 is planned, but reinsertion is pending the preparation and approval of a request to the AEC to extend the burnup limit beyond 6 at.%. The failed experiments in this group have been nondestructively examined and destructive examination has been scheduled.

Series B-3 is similar to the B-1 and B-2 series except that three helium bonded experiments are included and the average operating linear power ratings are slightly

TABLE 463-V
SERIES U1930 AND U1960 ENCAPSULATED CARBIDE EXPERIMENTS

Expmt. No.	Fuel Type[a]	Fuel Density, % Theo.	Bond and Diametral[f] Gap, in.	Clad[d] Type	Clad O.D. x I.D.,[f] in.	Max. Linear Power, Kw/ft	Maximum Centerline Temp., °C	Goal Burnup, at.%	Current Burnup, at.%	Status
					A					
U187	MC+5% M_2C_3	84	He-0.007	SA-316SS	0.304x0.264	30.0	1935	5	4.5	Exam, 2.12
U189	MC+5% M_2C_3	84	He-0.007	SA-INC-800	0.302x0.262	30.0	1935	5	4.5	Exam, 2.12
U191	MC	92	Na-0.030	SA-304SS	0.304x0.276	31.7	1148	5	4.7	Exam, 2.12
U192	MC	92	Na-0.030	SA-304SS	0.305x0.277	31.7	1148	5	4.7	Exam, 2.12
U194	MC+10% M_2C_3	98	Na-0.030	SA-304SS	0.305x0.277	33.1	1132	5	5.0	Exam, 2.12[c]
U195	MC+10%M_2C_3	98	Na-0.030	SA-304SS	0.305x0.276	33.1	1132	5	5.0	Exam, 2.12
U197	MC+10% M_2C_3	98	Na-0.030	SA-INC-800	0.305x0.277	33.4	1132	5	5.0	Exam, 2.12
U198	MC+10% M_2C_3	98	Na-0.030	SA-INC-800	0.305x0.277	33.4	1132	5	5.0	Exam, 2.12
U200	MC+5% M_2C_3	85	He-0.008	SA-304SS	0.288x0.260	30.8	2042	5	4.6	Exam, 2.12[c]
U206	MC+5% M_2C_3	90	He-0.008	SA-316SS	0.288x0.252	31.5	2084	5	4.7	Exam, 2.12
U208	MC+10% M_2C_3	97[b]	He-0.009	SA-316SS	0.293x0.257	31.9	1912	5	4.8	Exam, 2.12
					B					
U188	MC+5% M_2C_3	84	He-0.007	SA-316SS	0.304x0.264	30.0	1935	11	8.7	EBR-II, X182
U190	MC+5% M_2C_3	84	He-0.007	SA-INC-800	0.302x0.262	30.0	1935	11	8.8	EBR-II, X182
U193	MC	92	Na-0.030	SA-304SS	0.305x0.277	31.7	1148	11	8.7	EBR-II, X182
U196	MC+10% M_2C_3	98	Na-0.030	SA-304SS	0.305x0.277	32.6	1132	11	8.6	EBR-II, X182
U199	MC+10% M_2C_3	98	Na-0.030	SA-INC-800	0.305x0.277	33.5	1132	11	5.0	EBR-II, X182
U201	MC+5% M_2C_3	85	He-0.008	SA-304SS	0.288x0.260	30.0	2042	11	4.5	EBR-II, X182
U207	MC+5% M_2C_3	90	He-0.008	SA-316SS	0.293x0.257	31.7	2088	11	4.7	EBR-II, X182
U209	MC+10% M_2C_3	97[b]	He-0.009	SA-316SS	0.293x0.257	30.9	1909	11	4.6	EBR-II, X182
					C					
U185	MC+10% M_2C_3	96	He-0.011	SA-316SS	0.304x0.264	30.0	2195	3	3.0	Exam, 1.7
U186	MC+10% M_2C_3	96	He-0.011	SA-316SS	0.304x0.264	30.0	2195	3	3.0	Exam, 1.7
U202	MC+5% M_2C_3	85	He-0	SA-316SS	0.269x0.251	31.7	1270	3	2.8	Exam, 1.7
U203	MC+5% M_2C_3	85	He-0	SA-316SS	0.288x0.252	31.4	1260	3	2.8	Exam, 1.7
U204	MC+10% M_2C_3	97[b]	He-0	SA-316SS	0.266x0.248	32.2	1131	3	2.9	Exam, 1.7
U205	MC+10% M_2C_3	97[b]	0	SA-316SS	0.284x0.248	31.9	1124	3	2.9	Exam, 1.7
					D					
U260	MC+10% M_2C_3	98	He-0.015	20CW-316SS	0.298x0.264	34.1	2590	12	---	EBR-II, unassigned
U261[e]	MC+10%M_2C_3	98	He-0.015	SA-316SS	0.290x0.260	34.1	2590	12	---	
U262	MC+10%M_2C_3	97	He-0.015	SA-INC-800	0.290x0.260	34.1	2590	12	---	

[a] $M = (U_{0.85}Pu_{0.15})$

[b] Cored pellets with nominal 0.080-in. diam axial hole.

[c] Element cladding for 194 and 200 has failed

[d] SA = Solution annealed; 20CW = 20% cold-worked.

[e] Eddy current test at EBR-II indicates capsule bond discontinuity.

[f] All cladding I.D. and gap measurements are nominal.

higher. Gamma-scans made at EBR-II for ^{133}Xe indicated that the four sodium-bonded elements have failed, while the three helium-bonded elements have not failed. Nondestructive examination of the failed elements is currently under way. The three unfailed helium-bonded experiments are to continue irradiation in subassembly X182 starting in May, 1973.

The Series U5100 singly-clad experiments are described in Table 463-VIII. In this group, either single-phase or two-phase carbide fuel is sodium bonded to Type 304 or 316 stainless steel or to Incoloy 800. In seven of the elements, a shroud is incorporated primarily to test the retention of fuel fragments by close fitting tubes. A secondary objective of the shroud is to study the effectiveness of the shroud alloy as a carbon getter. These elements will be irradiated in subassembly X156 starting with run 63 in May, 1973. The first interim examination will be made at a burnup of 2.5 at.%.

TABLE 463-VI

SERIES WF ENCAPSULATED CARBIDE EXPERIMENTS

Expmt. No.	Fuel Type[a]	Fuel Density, % Theo.	Bond and Diametral Gap, in.	Clad[b] Type	Clad O.D. x I.D., in.	Max. Linear Power, Kw/ft	Maximum Centerline Temp., °C	Goal Burnup, at.%	Current Burnup, at.%	Status
W3F	MC+6-15% M_2C_3	91	Na-0.025	SA-316SS	0.300x0.276	27	1075	10[c]	6.0	EBR-II, Unassigned
W4F	MC+7-17% M_2C_3	93	Na-0.027	SA-316SS	0.300x0.276	28	1100	6	6.1	Exam, 1.7
W5F	MC+7-10% M_2C_3	89	Na-0.010	SA-316SS	0.250x0.230	20	975	6	2.9	EBR-II, X182
W6F	MC+9-10% M_2C_3	89	Na-0.011	SA-316SS	0.251x0.231	20	975	6	2.9	EBR-II, X182
W7F	MC+5% M_2C_3	89	Na-0.027	20CW-316SS	0.300x0.276	27	1075	10[c]	6.0	EBR-II, Unassigned
W8F	MC+3-6% M_2C_3	93	Na-0.025	20CW-316SS	0.300x0.276	28	1100	6	6.6	Exam, 2.1
W10F	MC+0-3% M_2C_3	88	Na-0.012	SA-316SS	0.251x0.231	20	975	10[c]	5.9	EBR-II, Unassigned
W12F	MC+19-24% M_2C_3	95	Na-0.013	SA-316SS	0.251x0.231	21	1000	6	2.8	EBR-II, X182

[a] $M = U_{0.8}Pu_{0.2}$

[b] SA = Solution annealed, 20 CW = 20% cold worked

[c] Original goal burnup was 6 at.%. New AIP request for further irradiation in preparation.

The C-5 and ON-1 series of singly-clad experiments are described in Table 463-IX. Single-phase nitride fuel is sodium bonded to 20% cold-worked Type 316 stainless steel cladding in all of the fuel elements in this group. Profilometry measurements of the C-5 series elements have been made using the same equipment that will be used for the postirradiation examination. Shipment of selected elements to EBR-II is pending LASL review of the experiments from a quality assurance standpoint.

The O-N1 series of singly-clad experiments is similar to the C-5 series. The elements are fueled with $(U_{0.8}Pu_{0.2})N$ which is sodium bonded to 20% cold-worked Type 316 stainless steel cladding. Three elements have been rejected because of large fuel chips in the sodium annulus. The diameters of the elements have been measured on the same profilometer that will be used after irradiation. The status of shipment of these experiments to EBR-II is the same as for the C-5 series. Four of the fuel pins from Series O-N1 will be irradiated with fifteen pins from Series C-5.

Possible design parameters for a new group of experiments, Series K-4, have been described in previous quarterly reports. The final design of this subassembly has been deferred pending the analysis of the irradiation experiments currently being examined.

In addition to the experiments described above, two nitride fueled thermal irradiation experiments from ORNL (43N1 and 43N2) will be examined. Results and status will be reported in future reports.

Engineering Test Plans have been prepared and approved in order to satisfy the Quality Assurance requirements for the irradiation and examination for the experiments in the K1, K2, K3, U1300, U1950, U1930, U1960, U5100, B-1, B-2, B-3, C-5, and O-N1 series of tests.

A major effort has been expended in the overall evaluation of all of the high performance irradiation experiments under LASL direction. All available data on each experiment has been cataloged and evaluated with respect to both technical characterization and quality control completeness. An overall evaluation for each

TABLE 463-VII

SERIES B-1, B-2 AND B-3 ENCAPSULATED NITRIDE EXPERIMENTS

Expmt. No.	Fuel Type	Fuel Density, % Theo.	Bond and Diametral Gap, in.	Clad[e] Type	Clad O.D. x I.D., in.	Max. Linear Power, Kw/ft	Maximum Centerline Temp., °C	Goal Burnup, at.%	Current Burnup, at.%	Status
				Series B-1						
B-1-1	$(U_{0.8}Pu_{0.2})N$	80	Na-0.019	SA-304SS	0.290x0.250	27.9	1125	5	6.5	Exam, 1.7[b]
B-1-2	$(U_{0.8}Pu_{0.2})N$	81	Na-0.018	SA-304SS	0.290x0.250	27.1	1125	9	6.3	Exam, 1.7[b]
B-1-4	$(U_{0.8}Pu_{0.2})N$	85	Na-0,012	SA-304SS	0.290x0.250	28.6	1150	10[f]	3.0	Exam, 1.7[c]
				Series B-2						
B-2-1	$(U_{0.8}Pu_{0.2})N$	82	Na-0.021	SA-316SS	0.316x0.275	32.7	1230	10[f]	6.3	EBR-II, unassigned
B-2-2	$(U_{0.8}Pu_{0.2})N$	82	Na-0.020	SA-316SS	0.316x0.275	32.5	1230	9	6.2	Exam, 1.7[b]
B-2-3	$(U_{0.8}Pu_{0.2})N$	81	Na-0.020	SA-316SS	0.315x0.275	32.4	1230	12	6.3	EBR-II, X182
B-2-5	$(U_{0.8}Pu_{0.2})N$	76	Na-0.028	SA-316SS	0.315x0.284	32.4	1230	12	3.0	Exam, 1.7[c]
B-2-6	$(U_{0.8}Pu_{0.2})N$	82	Na-0.021	SA-316SS	0.316x0.295	36.6	1230	6	6.2	Exam, 1.7[b]
B-2-7	$(U_{0.8}Pu_{0.2})N$	82	Na-0.020	SA-316SS	0.316x0.295	36.7	1230	12	6.1	Exam, 1.7[b]
				Series B-3						
B-3-2	$(U_{0.8}Pu_{0.2})N$	88	Na-0.009	SA-304SS	0.315x0.284	37.7	1250	9	3.1	Exam, 2.7[b]
B-3-3	$(U_{0.8}Pu_{0.2})N$	91	Na-0.010	SA-304SS	0.315x0.284	38.9	1280	12	3.1	Exam, 1.7[b,d]
B-3-4	$(U_{0.8}Pu_{0.2})N$	94	Na-0.013	SA-304SS	0.316x0.284	38.9	1280	12	3.1	Exam, 2.6[b]
B-3-5	$(U_{0.8}Pu_{0.2})N$	90	Na-0.010	SA-304SS	0.316x0.295	41.5	1310	6	3.1	Exam, 2.7[b]
B-3-6	$(U_{0.8}Pu_{0.2})N$	95[a]	He-0.005	SA-304SS	0.315x0.275	34.2	1925	6	3.0	EBR-II, X182
B-3-7	$(U_{0.8}Pu_{0.2})N$	89	He-0.005	SA-304SS	0.315x0.275	34.2	1925	6	3.0	EBR-II, X182
B-3-8	$(U_{0.8}Pu_{0.2})N$	90[a]	He-0.005	SA-304SS	0.315x0.275	32.4	1875	6	2.9	EBR-II, X182

[a] Pellets are annular with a 0.070-in. diam axial hole.

[b] Element cladding failure indicated.

[c] Available for further irradiation.

[d] Nondestructive examination completed. Capsule stored for possible irradiation of failed element.

[e] Cladding is welded tubing. SA = Solution annealed.

[f] Original goal burnups were 3 to 5 at.%. New AIP request for further irradiation in preparation.

series (except the O-N1 series) from a quality control standpoint will be completed during the first part of next quarter. The transfer of information from ORNL, the original experimenter, is still incomplete for the O-N1 series; consequently that evaluation will necessarily take somewhat longer than for the other series. A summary report of the evaluations and LASL recommendations will be issued for RDT review.

C. TREAT Irradiation Testing

(J. F. Kerrisk, D. G. Clifton, R. E. Alcouffe)

In order to assess the behavior of (U, Pu)C and (U, Pu)N fueled elements under fast reactor accident conditions, transient irradiations will be conducted in the TREAT facility. Investigations will be carried out on both irradiated and unirradiated fuel pins to determine (1) the threshold power levels at which damage or failure occurs, (2) the effect of bond and cladding defects, and (3) the failure propagation mechanism in multipin assemblies.

1. Series UL Tests. A cooperative effort has been carried out with Gulf United Nuclear Fuels Corporation in the area of TREAT testing. A series of four tests, designated LASL Series UL, will determine the effect of irradiation on the behavior of helium and sodium bonded advanced fuel elements (fabricated by GUNFC) under possible LMFBR accident conditions. Table 463-X describes the fuel elements and test objectives. LASL has assumed complete responsibility for these tests in fiscal year 1973.

a. Test LASL-UL-1. Test LASL-UL-1 was performed November 16, 1972. The TREAT capsule from this test has been returned to LASL and the postirradiation examination has been started. At present, only radiographs of the TREAT capsule have been reviewed. No indication of fuel element failure has been seen.

b. Test LASL-UL-2. Test LASL-UL-2 (TREAT transient number 1495) was performed on February 20,

TABLE 463-VIII

SERIES U5100 SINGLY CLAD CARBIDE EXPERIMENTS

Expmt. No.	Fuel Type[a]	Fuel Density, % Theo.	Bond and Diametral[b] Gap, in.	Clad[c] Type	Clad O.D. x I.D., in.	Max. Linear Power, Kw/ft	Maximum Centerline Temp., °C	Goal Burnup, at.%	Current Burnup, at.%	Status
U241	MC	92	Na-0.018	SA-304SS	0.310x0.281	35.8	1175	6	0	EBR-II, X156
U242	MC	92	Na-0.017	SA-304SS	0.310x0.281	35.8	1175	9	0	EBR-II, X156
U243	MC	92	Na-0.031	SA-304SS	0.310x0.281	33.8	1150	6	0	EBR-II, X156
U244	MC	92	Na-0.017	SA-304SS	0.310x0.281	35.8	1175	9	0	EBR-II, X156
U245	MC	91	Na-0.032	SA-304SS	0.310x0.281	33.8	1150	12	0	EBR-II, X156
U246	MC	92	Na-0.017	SA-316SS	0.310x0.281	36.4	1190	6	0	EBR-II, X158
U247	MC	92	Na-0.032	SA-316SS	0.310x0.281	33.8	1150	6	0	EBR-II, X156
U248	MC	91	Na-0.032	SA-316SS	0.310x0.281	36.4	1140	12	0	EBR-II, X156
U249	MC	92	Na-0.017	SA-INC-800	0.309x0.281	36.4	1210	6	0	EBR-II, X156
U250	MC	91	Na-0.032	SA-INC-800	0.309x0.281	36.4	1145	6	0	EBR-II, X156
U251	MC	92	Na-0.031	SA-304SS	0.310x0.281	36.4	1145	12	0	EBR-II, X156
U252	MC	92	Na-0.024	SA-304SS	0.310x0.281	36.4	1140	12	0	EBR-II, X156
U253	MC	92	Na-0.024	SA-304SS	0.310x0.281	33.8	1145	12	0	EBR-II, X156
U254	MC	92	Na-0.024	SA-304SS	0.310x0.281	33.8	1140	12	0	EBR-II, X156
U256	MC+10% M_2C_3	95	Na-0.024	SA-304SS	0.309x0.281	34.0	1140	12	0	EBR-II, X156
U257	MC+10% M_2C_3	95	Na-0.024	SA-INC-800	0.309x0.281	33.5	1135	12	0	EBR-II, X156
U258	MC+10% M_2C_3	95	Na-0.024	SA-304SS	0.310x0.281	33.5	1145	6	0	EBR-II, X156
U259	MC+10% M_2C_3	95	Na-0.024	SA-INC-800	0.309x0.281	34.6	1150	12	0	EBR-II, X156

[a] MC = $U_{0.85}Pu_{0.15}$.

[b] Elements U-252, -253, -254, -256, -257, -258, and -259 have shrouds ~ 0.0035 in. thick made from V, Fe, 304SS, 304SS, V, Ta, 304SS, and 304SS, respectively. The shrouds are alotted.

[c] SA = Solution Annealed

1973. A preliminary review of the capsule temperatures indicated that the test was performed as requested. The TREAT capsule from this test has been returned to LASL.

c. Tests LASL-UL-3 and LASL-UL-4. The assembly of the inner capsules for tests 3 and 4 of the UL Series was done by GUNFC up to the point of insertion of the fuel elements into the inner capsules. Since the fuel elements were preirradiated in EBR-II, the completion of the assembly of the inner capsule and the assembly of the inner capsules into the TREAT capsules requires hot cell facilities. LASL, in conjunction with RDT, has arranged for the completion of the assembly by the Radiometallurgy Group of Hanford Engineering Development Laboratory (HEDL). A purchase order covering the assembly work has been written to HEDL. Additional welding samples required by HEDL for weld development have been fabricated and shipped. A quality assurance plan covering the assembly and inspection of the capsules, the tests, and the postirradiation examination has been prepared and submitted to TREAT for approval.

2. Series 1 Tests. A group of eight tests using LASL fabricated fuel elements has been designated LASL Series 1 tests. The tests are designed to determine if any significant safety related behavioral problems exist for sodium bonded, stainless steel clad (U, Pu)C and (U, Pu)N fuels by defining failure thresholds and the types of failure experienced by these fuels. Table 463-XI summarizes the test parameters and objectives. Approval-in-principle has been received from the AEC for this series of tests.

An Engineering Test Plan, an Inspection and Test Plan, and detail fabrication and assembly drawings have been completed for the initial tests of this series. Due to a reduction in funding, work on this series of tests has been postponed.

3. Fuel-Coolant Interaction Tests. At the request of RDT, an investigation into the feasibility of performing fuel-coolant interaction tests with high performance fuels was started. The initial objective of these tests would be to study the manner in which molten

TABLE 463-IX

SERIES C-5 AND O-N1 SINGLY CLAD NITRIDE EXPERIMENTS

Expmt. No.	Fuel Type	Fuel Density, % Theo.	Bond and Diametral Gap, in.	Clad[a] Type	Clad O.D. x I.D., in.	Max. Linear Power, Kw/ft	Maximum Centerline Temp., °C	Goal Burnup, at.%	Current Burnup, at.%	Status
					Series C-5					
C-5-1	$(U_{0.8}Pu_{0.2})N$	93	Na-0.020	20CW-316SS	0.310x0.280	--	--	--	-	Reject[b,c]
C-5-2	$(U_{0.8}Pu_{0.2})N$	93	Na-0.020	20CW-316SS	0.310x0.280	--	--	--	-	Reject[b,c]
C-5-3	$(U_{0.8}Pu_{0.2})N$	94	Na-0.020	20CW-316SS	0.310x0.280	--	--	--	-	Reject[b,c]
C-5-4	$(U_{0.8}Pu_{0.2})N$	95	Na-0.021	20CW-316SS	0.310x0.280	33.2	719	12	0	At LASL[d]
C-5-5	$(U_{0.8}Pu_{0.2})N$	95	Na-0.020	20CW-316SS	0.310x0.280	-------Spare------		12	0	At LASL[d]
C-5-6	$(U_{0.8}Pu_{0.2})N$	93	Na-0.021	20CW-316SS	0.310x0.280	33.3	720	12	0	At LASL[d]
C-5-7	$(U_{0.8}Pu_{0.2})N$	94	Na-0.020	20CW-316SS	0.310x0.280	33.7	724	12	0	At LASL[d]
C-5-8	$(U_{0.8}Pu_{0.2})N$	94	Na-0.030	20CW-316SS	0.310x0.280	32.7	623	12	0	At LASL[d]
C-5-9	$(U_{0.8}Pu_{0.2})N$	94	Na-0.020	20CW-316SS	0.310x0.280	33.5	651	12	0	At LASL[d]
C-5-10	$(U_{0.8}Pu_{0.2})N$	94	Na-0.020	20CW-316SS	0.310x0.280	32.6	625	12	0	At LASL[d]
C-5-11	$(U_{0.8}Pu_{0.2})N$	94	Na-0.020	20CW-316SS	0.310x0.280	33.4	629	12	0	At LASL[d]
C-5-12	$(U_{0.8}Pu_{0.2})N$	94	Na-0.030	20CW-316SS	0.310x0.280	32.5	644	12	0	At LASL[d]
C-5-13	$(U_{0.8}Pu_{0.2})N$	96	Na-0.030	20CW-316SS	0.310x0.280	32.1	623	12	0	At LASL[d]
C-5-14	$(U_{0.8}Pu_{0.2})N$	96	Na-0.030	20CW-316SS	0.310x0.280	32.0	711	12	0	At LASL[d]
C-5-15	$(U_{0.8}Pu_{0.2})N$	95	Na-0.030	20CW-316SS	0.310x0.280	32.1	642	12	0	At LASL[d]
C-5-16	$(U_{0.8}Pu_{0.2})N$	96	Na-0.030	20CW-316SS	0.310x0.280	-------Spare------		12	0	At LASL[d]
C-5-17	$(U_{0.8}Pu_{0.2})N$	96	Na-0.030	20CW-316SS	0.310x0.280	--	--	--	-	Reject[c]
C-5-18	$(U_{0.8}Pu_{0.2})N$	94	Na-0.021	20CW-316SS	0.310x0.280	32.6	645	12	0	At LASL[d]
C-5-19	$(U_{0.8}Pu_{0.2})N$	94	Na-0.021	20CW-316SS	0.310x0.280	33.0	649	12	0	At LASL[d]
C-5-20	$(U_{0.8}Pu_{0.2})N$	95	Na-0.021	20CW-316SS	0.310x0.280	32.4	624	12	0	At LASL[d]
					Series O-N1					
O-N1-1	$(U_{0.8}Pu_{0.2})N$	90	Na-0.020	20CW-316SS	0.310x0.280	32.9	649	12	0	At LASL[d]
O-N1-2	$(U_{0.8}Pu_{0.2})N$	90	Na-0.020	20CW-316SS	0.310x0.280	--	--	--	-	Reject[b]
O-N1-3	$(U_{0.8}Pu_{0.2})N$	90	Na-0.020	20CW-316SS	0.310x0.280	32.6	717	12	0	At LASL[d]
O-N1-4	$(U_{0.8}Pu_{0.2})N$	90	Na-0.020	20CW-316SS	0.310x0.280	32.8	717	12	0	At LASL[d]
O-N1-5	$(U_{0.8}Pu_{0.2})N$	90	Na-0.020	20CW-316SS	0.310x0.280	--	--	--	-	Reject[b]
O-N1-6	$(U_{0.8}Pu_{0.2})N$	89	Na-0.020	20CW-316SS	0.310x0.280	--	--	--	-	Reject[b]
O-N1-8	$(U_{0.8}Pu_{0.2})N$	90	Na-0.020	20CW-316SS	0.310x0.280	33.1	718	12	0	At LASL[d]

[a] 20CW = 20% cold worked [b] Chips in bond [c] Air in plenum [d] QA evaluation in progress

carbide or nitride fuel interacts with liquid sodium, and to measure the pressures generated and the conversion of thermal energy to mechanical energy due to the interaction. The fuel-coolant interaction tests performed on oxide fuels were reviewed. Due to a reduction in funding, fuel-coolant interaction work on high performance fuels will not be pursued at the present time.

III. ANALYTICAL CHEMISTRY

1. X-Ray Fluorescence Spectrometric Determination of Ta in Stainless Steel

(G. B. Nelson, J. M. Hansel, E. A. Hakkila)

Chemical characterization of stainless steels for FFTF Program applications requires reliable measurements of several specified impurities including Ta. Modifications of an existing solvent extraction method for separation of Ta at low concentrations from stainless steel[3] permits final measurement by an x-ray fluorescence method. This separation involves extraction of Ta from a 6M H_2SO_4-1M HF mixture, followed by a second extraction from 6M H_2SO_4-0.4M HF. Previously, relative standard deviations were reported to be 4.6% for determining 500 or 1000 μg (1000 or 2000 ppm) of Ta in 500 mg samples, with a limit of detection of approximately

TABLE 463-X

LASL SERIES UL TESTS

	TEST			
	LASL-UL-1	LASL-UL-2	LASL-UL-3	LASL-UL-4
Fuel Element[a]	263 (138 A)	264 (146 A)	265 (138)	266 (146)
Fuel Material[b]	90 vol% $(U_{0.85}Pu_{0.15})C$ + 10 vol% $(U_{0.85}Pu_{0.15})_2C_3$			
Fuel Pellet O.D., in.	0.246	0.240	0.246	0.240
Bond Material	He	Na	He	Na
Bond Thickness (Radial), in.	0.005	0.015	0.005	0.015
Clad Material	316SS	304SS	316SS	304SS
Clad Thickness, in.	0.022	0.015	0.022	0.015
Smear Density, % Theoretical	90	77	90	77
Fuel Column Length, in.	13.75 ± 0.125			
Burnup, MWD/MTM[c]	0	0	45,000	45,000
Test Objective	Fuel Melting	Fuel Melting	Same Transient as 263	Same Transient as 263

[a] Fuel element numbers reassigned by Gulf United. Old numbers shown in parentheses.

[b] Uranium enriched to 60% in ^{235}U.

[c] Irradiated in EBR-II at 10 to 15 Kw/ft in subassembly X055.

TABLE 463-XI

LASL SERIES 1 EXPERIMENTS

Test	Fuel Material[a]	Burnup	TREAT Transient[b]	Test Objective
1A-1	$(U_{0.8}Pu_{0.2})C$	0	Fast	Na bond ejection-incipient fuel melting
1A-2	$(U_{0.8}Pu_{0.2})C$	0	Slow	Same as 1A-1
1B-1	$(U_{0.8}Pu_{0.2})C$	0	Fast	50% fuel melting
1B-2	$(U_{0.8}Pu_{0.2})N$	0	Fast	Same as 1B-1[d]
1B-3	$(U_{0.8}Pu_{0.2})C$	0	Slow	Same as 1B-1
1B-4	$(U_{0.8}Pu_{0.2})N$	0	Slow	Same as 1B-1[d]
1C-1	$(U_{0.8}Pu_{0.2})C$	8%	c	Same as 1B-1
1C-2	$(U_{0.8}Pu_{0.2})N$	8%	c	Same as 1B-1[d]

[a] The fuel will be pellets, contained in 0.310 in. o.d. by 0.012 in. wall 316 stainless steel cladding at 80% smear density. The uranium is enriched to 93% in ^{235}U.

[b] Fast transients will deposit energy in time periods of the order of 1 sec while slow transients will be on the order of 10 sec.

[c] The type of transient to be used will be determined by the results of the unirradiated element tests.

[d] Since (U, Pu)N does not melt, but decomposes to metal and nitrogen, the test objectives are described in terms of the energy required to produce a given melting in (U, Pu)C.

3.3 μg (7 ppm). During this report period the precisions were measured for Ta concentrations between 10 and 100 μg in 500 mg samples (Table 463-XII).

TABLE 463-XII

PRECISION OF X-RAY SPECTROMETRIC DETERMINATION OF Ta IN STAINLESS STEEL (500 mg Samples)

Ta Added, μg	Number of Determinations	Standard Deviation, μg	Relative Standard Deviation, %
10	10	3.2	32
25	9	3.7	15
100	8	11.9	12

Triplicate analyses of two National Bureau of Standards SRM samples indicated that the measured precisions were accurate. There was a small negative bias in the method which was eliminated by chemical calibration. Measured Ta contents were 330 ± 21 ppm in one sample reported to contain 360 ppm, and 2230 ± 160 ppm in a sample certified at 2300 ppm Ta.

The effects of various elements that may be present in stainless steel are being tested to determine possible interferences. Up to 0.5 wt% of Mo was shown to have no effect, and tests of other elements are under way.

2. Coulometric Titrations of Pu and U
(N. Fawcett)

In the coulometric titration of U in the presence of Pu, a pre-electrolysis at +0.085V vs. the saturated calomel electrode (SCE) is sufficient to oxidize Hg^{o} to Hg^{+} if even trace quantities of Cl^{-} ion are present. At the potential of the final electrolysis, which is -0.325V vs. the SCE, Hg^{+} is reduced to Hg^{o}. It is imperative to avoid contamination of the sample solution with Cl^{-}, or electrolysis of Hg^{+} to Hg^{o} will contribute to the current during the final electrolysis of U(IV) to U(VI). Furthermore, if there is more than a trace of Cl^{-} present, Hg_2Cl_2 precipitates on the mercury surface forming a film which decreases the rate at which soluble ions are electrolyzed.

It appears that the most likely source of chloride contamination is the SCE which is almost universally used as a reference half cell. Even though the SCE is isolated from the titrant by an intervening 1N H_2SO_4 salt bridge, Cl^{-} leaking from the fiber junction in the SCE may eventually diffuse through the salt bridge and into the working electrode compartment.

To avoid Cl^{-} contamination, the SCE was replaced with a Pb(Hg)/$PbSO_4$ reference half cell. The lead sulfate electrode (LSE) is commercially available and is as stable and long-lived as the SCE. The supporting electrolyte for the LSE can consist of 1N H_2SO_4, making the chemical potential of sulfate ion the same in the reference electrode, the Vycor tube salt bridge, and the working electrode compartment. Use of the LSE in titrating known quantities of U has proved satisfactory.

Coulometric determination of Pu(VI) by reduction at Pt to Pu(III) followed by subsequent oxidation to Pu(IV) does not proceed satisfactorily in aqueous HCl at room temperature. Reduction of Pu(VI) on Pt at +0.310V vs. the SCE required more than 1 h in 1N HCl at 25°C. More negative potentials, up to the point of H_2 evolution, were also ineffectual. As the HCl electrolyte effectively reduces interference caused by small amounts of Fe impurity, conditions were investigated to produce satisfactory titrations of Pu in HCl.

Warming the sample solution to approximately 50°C was found to produce a much more rapid reduction. Thus, 5.759 mg of Pu(VI) in 1N HCl at +0.310V was reduced to Pu(III) in less than 30 min as indicated by recession of the current to a background level of 30 μa. Subsequent oxidation to Pu(IV) at +0.86V and at 25°C resulted in a titer of 5.758 mg Pu after subtraction of a measured blank correction. For solution samples not adversely affected by elevated temperature, warming the solution during the prereduction step may make it possible to titrate hydrochloric acid solutions of Pu(VI) without preliminary chemical reduction of the Pu(VI). Further work is in progress to verify this observation.

IV. QUALITY ASSURANCE

Fuel Preparation: Procedures for fuel preparation are being revised. A traveler system is being prepared to document all processing and inspection data during the preparation and characterization of the material.

The calibration of instruments has continued and records are being made for these instruments. Quality

Assurance Procedures for instrument calibration have been prepared, approved, and are being used.

Fuel Pin Fabrication: Procedures for inspection and overchecking of items have been prepared and the inspection and overchecking are in process. A traveler system to provide material and component control is being prepared.

Archive and material storage facilities meeting quality assurance requirements are being prepared. Items are being stored, identified and provisions are being set up for inventory control.

Dummy fuel elements were fabricated, inspected and shipped using quality assurance procedures, reviews, and documentation.

EBR-II Irradiation Testing: Engineering Test Plans have been prepared for each series of irradiation experiments. The postirradiation examination requests are based on the requirements of these Engineering Test Plans.

Files and data packages are being prepared for the retention of quality assurance and technical information relating to the EBR-II tests and the postirradiation examinations.

An audit by the Quality Assurance Manager was conducted of the records and procedures for EBR-II Testing and an audit report was written.

TREAT Irradiation Testing: An Engineering Test Plan has been prepared for the Series 1 TREAT tests. Procedures for TREAT fabrication, and material inspection and overchecking have been prepared.

Welding samples for work at HEDL were fabricated, inspected and shipped using quality assurance procedures, reviews, and documentation.

An audit of the TREAT Irradiation Testing was conducted by the Quality Assurance Manager and an audit report was written.

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PROJECT 472

ANALYTICAL STANDARDS FOR FAST BREEDER REACTOR OXIDE FUEL

Person in Charge: R. D. Baker
Principal Investigator: G. R. Waterbury

I. INTRODUCTION

Necessary to the development of the high quality fuels and control rods required by the LMFBR program are highly reliable analytical methods for the chemical characterization of the source materials and the pellet products, and for the measurement of burnup and fission products on irradiated fuels. Objectives concerned with ensuring the production of these high quality reactor materials are: (1) the continued preparation of carefully characterized calibration materials for the various analytical methods, (2) the preparation of quality control samples used for surveillance of analytical chemistry laboratory operations during periods of production, (3) continued assistance and guidance of quality assurance programs for chemical specification sampling and analysis, (4) the development of more reliable methods of chemical analysis with emphasis on O/M measurement, (5) the preparation of continuously updated compilations of analytical methods for these materials, and (6) the analysis, as a referee laboratory, of those samples in dispute between a vendor and a purchaser. For the immediate future these objectives are centered on the FFTF. Later they will be extended to the LMFBR demonstration and large production plants.

Objectives concerned with irradiated LMFBR fuel examination are: (1) the development of burnup methods based on conventional mass spectrometry measurements and later by spark source mass spectrometry; (2) the development of faster burnup methods using chemical analysis techniques; and (3) start development of analytical methods for the determination of gases on pre and postirradiated fuels and control rod absorber materials to provide data on gas retention properties and cladding stability.

II. ANALYTICAL CHEMISTRY PROGRAM FOR BORON CARBIDE

A program equivalent to that established for the production of FFTF mixed oxide fuel is in progress for the production of FFTF boron carbide pellets.

A. Status of Analytical Methods and Qualification of Analytical Laboratories

(J. E. Rein, W. H. Ashley, G. R. Waterbury)

Nitrogen, probably as BN, has been found as a significant impurity in various manufactured lots of B_4C. For this reason, HEDL plans to add to the specifications a maximum acceptable content for nitride N impurity in the B_4C pellets to be manufactured for the FFTF.

As a high priority task, HEDL and LASL will cooperatively recommend an analytical method for the determination of nitride nitrogen in B_4C that will be made available to the industrial companies. To expedite the issuance of this method, HEDL and LASL exchanged descriptions of their present state-of-the-art methods. The desirable features of each were selected and HEDL started writing a recommended method that included these features. (A discussion of the LASL method is presented in Section II. D below.) To evaluate the reliability and status of this

recommended method when used in different laboratories, the industrial companies, HEDL, and LASL will analyze a test lot of B_4C pellets, using prepared blends to calibrate the methods.

B. Preparation of Calibration Materials and Quality Control Samples
(J. V. Pena, H. J. Kavanaugh, J. E. Rein)

On a high priority basis, LASL is preparing a series of calibration blends for nitride N in B_4C. This series consists of five powder blends using starting materials of well-characterized BN and low-nitrogen-content B_4C. Sufficient amounts are being prepared for the immediate laboratory evaluation and for future use by vendor and HEDL laboratories during production of B_4C pellets for the FFTF.

Three lots of B_4C pellets, supplied by HEDL, are being characterized for nitride nitrogen content. If proven satisfactory relative to nitrogen levels and homogeneity, these lots will be packaged and used as the quality control samples for the FFTF Quality Assurance Program.

C. Status of RDT Standards
(J. E. Rein, W. H. Ashley, O. R. Simi, G. R. Waterbury)

RDT F11-2, "Analytical Chemistry Methods for Control Rod Absorber Material," reviewed by RDT, is being revised by HEDL to incorporate recommended revisions. A draft of the revised document is being reviewed at LASL by all contributors.

D. Studies and Improvements of Analytical Methods
(R. E. Perrin, T. K. Marshall, A. Zerwekh, G. E. Meadows, A. L. Henicksman, C. H. Ward, W. H. Ashley, and R. G. Bryan)

1. Determination of Nitrogen. The evaluation of the separation of nitride nitrogen from B_4C by caustic fusion and then titration of the evolved NH_3 with acid has shown the need for several modifications to improve the reliability of the measurement: 1. Substitution of a silver crucible liner for the original nickel liner to eliminate formation of unwanted nitrogen compounds that cause low results. 2. Substitution of lithium hydroxide monohydrate ($LiOH \cdot H_2O$) for NaOH as the flux. 3. Addition of a water bubbler to the gas pretreatment apparatus to supply the moisture needed to give complete conversion of evolved N to NH_3. 4. Specification of a schedule for heating the sample slowly to avoid violent early reactions that may spatter sample and flux into the cooler parts of the tube. Following these modifications, five portions of a BN sample were analyzed. The recovery of N was 99.3%, assuming theoretical stoichiometry of the BN, and the relative standard deviation was 0.08%.

An independent method for the N determination was developed in which the BN and B_4C were completely dissolved in 80% H_2SO_4 in a sealed, fused-silica tube at 370°C. Little or no residual gas pressure remained in the cooled tubes. An aliquot of this solution was made strongly basic, and the N was separated by distillation as NH_3 which was titrated with standard acid. Seven portions of the previously analyzed BN were analyzed by this method with an average nitrogen recovery of 99.3% and a relative standard deviation of 0.14%.

The basic fusion and the sealed-tube acid dissolution methods also were compared by repeatedly analyzing the same lot of B_4C. The average of seven determinations of the N by the fusion method was 0.71% N with a relative standard deviation of 1%; the average of six determinations by the sealed tube dissolution method also was 0.71% with a relative standard deviation of 0.8%.

Additional work on the LiOH fusion and the sealed tube dissolution is needed to determine the effect of particle size variations. In particular, the possibility of sealed tube dissolution of unground boron carbide is intriguing because of the application to the assay of boron.

The measurement of total N (nitride N plus adsorbed N) also was investigated. The usual Dumas technique was found to be satisfactory for this purpose. In the Dumas analysis the sample is mixed with CuO, burned in an atmosphere of pure CO_2 in a tube containing Cu and CuO, and the N is measured volumetrically after the rest of the gases are adsorbed in strong NaOH. The reaction with B_4C is slow, however, making the Dumas method unattractive for routine analyses.

Tests showed that the total N in B_4C can also be determined using a LECO-Nitrox-6 analyzer that is modified to attain 2300°C. In this analyzer, the B_4C is heated inductively in a carbon crucible to a temperature of 2300°C; oxygen in the sample is converted to CO_2 and absorbed on Ascarite; and the nitrogen is released for measurement by

gas chromatography. Analyses of samples of B_4C containing added BN showed that 94% of the N was recovered. The relative standard deviation in measuring 700 to 900 ppm of N was about 6%. Results obtained on the same lot of B_4C were 785 ± 50 ppm by the Dumas method and 760 ± 40 ppm by the LECO analyzer. Further work will be required to determine the effect of sample size and to determine the optimum sample weight.

2. Soluble Carbon. Soluble carbon is measured as CO_2 which is generated by wet oxidation of the sample in hot chromic acid in a glass reaction flask. The CO_2 is swept from the flask by a stream of N introduced through an inlet tube which extends down from the top of the flask into the acid solution. Results for soluble C were found to be low unless the N inlet tube extended near enough to the bottom of the reaction vessel to give effective stirring. Otherwise, the B_4C powder settled in a thick layer and did not react completely with the reagent. To avoid further difficulty, the reaction vessel was described explicitly in the RDT standard being prepared. The recommended vessel had a funnel-shaped bottom with flat sloping walls, and the gas inlet tube had a 2 or 3 mm i.d. and reached within 1 to 2 mm of the bottom tip.

3. Fluoride and Chloride. Fluoride is separated from B_4C by pyrohydrolysis, collected in water or aqueous solution in a receiver, and measured using a fluoride-selective electrode. As some boric acid (H_3BO_3) is formed during the pyrohydrolysis and steam distills into the receiver, the effect of H_3BO_3 on the measurement of F^- was investigated. This was done by measuring with the fluoride-selective electrode known quantities of F^- under the following six conditions: (1) added to water, (2) pyrohydrolyzed from U_3O_8 accelerator into water, (3) pyrohydrolyzed from U_3O_8 plus B_4C into water, (4) added to saturated H_3BO_3 solution, (5) pyrohydrolyzed from U_3O_8 accelerator into saturated H_3BO_3 solution, and (6) pyrohydrolyzed from U_3O_8 plus B_4C into saturated H_3BO_3 solution. It was found that the measurements in saturated H_3BO_3 solutions were only about 25% as sensitive as measurements of F^- in water but the electrode response was faster. The measurements of F^- pyrohydrolyzed from U_3O_8 differed by 10 to 15% from the results for unpyrohydrolyzed F^- in either water or saturated H_3BO_3 solution. Addition of B_4C to the U_3O_8 reduced the measured amount of F^- in either water or saturated H_3BO_3 solution by about 10%.

Because of the indicated differences in recoveries of F^-, all future samples of B_4C will be analyzed under carefully controlled conditions. The F^- will be pyrohydrolyzed into saturated H_3BO_3 solution and the electrode will be calibrated by measuring known amounts of F^- pyrohydrolyzed from U_3O_8 into saturated H_3BO_3 solution. A correction for the 10% low bias will be valid because of the uniformity of analysis conditions.

III. ANALYTICAL CHEMISTRY PROGRAM FOR FBR MIXED OXIDE FUEL

A. Qualification of Analytical Laboratories

(R. K. Zeigler, J. E. Rein, G. R. Waterbury)

The analytical laboratories of the two vendors (Kerr-McGee and NUMEC) and of the PuO_2 supplier (ARHCO) have been qualified for FFTF fuel production analysis. Analysis of quality control samples in accordance with the HEDL Quality Assurance Program has been initiated at these facilities as well as at the HEDL Analytical Laboratory.

B. Calibration Materials and Quality Control Samples

(J. V. Pena, H. J. Kavanaugh, L. A. Maestas, J. E. Rein)

Quantities of calibration materials and quality control samples adequate for two quarters of fuel production have been packaged and are awaiting shipment. A change in the shipment policy is pending. Previously, all such materials were sent to HEDL where they were repackaged and distributed to the various participating laboratories. A request is being submitted to the AEC for approval to ship the materials to the individual laboratories directly from LASL. This will provide cost savings, both in transportation charges and HEDL manpower effort.

Additional powder blends of calibration materials and quality control samples have been prepared for future use. The supply of the various matrix materials that are used for these materials has been depleted. HEDL has initiated the preparation of new matrices of $(U, Pu)O_2$ and PuO_2 required for the next series of blends. A large

batch of highly pure UO_2 is being prepared at LASL for use as the matrix material for future blends.

C. Development of Analytical Methods

1. Determination of Burnup.
(S. F. Marsh, J. E. Rein)

a. Mass Spectrometric Method. The chemical separation procedure developed in this laboratory to provide separated fractions of U, Pu, and Nd for mass spectrometric measurements has been in routine use for two years with no inadequacies.[1] However, investigators in other laboratories have reported difficulties with various operations in the procedure. We, therefore, investigated possible causes for these reported difficulties.

The procedure involves two sequential anion exchange column treatments. Hexavalent U and Pu are retained on the first column as chloride complexes while the rare earths and other fission products are eluted. Plutonium then is eluted with a HI-strong HCl mixture followed by the elution of U with dilute HCl. The rare earth effluent is converted to nitrate salts and loaded onto the second column in a relatively strong HNO_3-methanol solution. Neodymium is chromatographically separated as the nitrate complex with a dilute HNO_3-methanol elutriant.

No difficulties were observed for the first column under the conditions prescribed. The recovery of Pu was consistently greater than 90% with insignificant contamination of uranium. The recovery of U was greater than 85%. A tailing of Pu was observed, verifying that a wash with the hydriodic acid-hydrochloric acid eluant solution was necessary to minimize contamination of the uranium fraction.

Observations of the performance of the second column separation were that at least 99.9% of Ce elutes in the first 5 ml and that the elution of Nd centers between the 15th and 22nd elution ml. (The procedure prescribed collection of the 15th through 24th ml.) These second column experiments were done with the same batch of AG-1-2x (200-400 mesh) resin that had been used for the original development study of the procedure. Other batches of this same resin gave widely different flow rates. Microscopic studies showed significant discrepancies between the actual size and size distribution of the resin and the values provided by the manufacturer. It is well known that the elution characteristics of Nd and other rare earths are very sensitive to elutriant flow rate in chromatographic separations. It therefore is necessary to characterize each lot of resin for flow rate (and elution characteristics) using Nd tracer before use for samples. Fines (or heavy particles) are removed as necessary to obtain a 10 ± 2 min/ml flow rate at the prescribed operating conditions of the second column.

b. Development of Spectrophotometric Method Using Low-Cost Apparatus.

Ion exchange resin separation of U, Pu, and the rare earth group followed by their measurements using optical spectrophotometry form the basis for an alternate burnup method. The total rare earth fission products represent the number of fissions; U and Pu represent the residual fuel. Total rare earths as the fission product monitor have the advantage of high and relatively constant fission yield for the various fissioning U and Pu nuclides in FBR fuel. A desirable feature of the ion exchange separation for the rare earth fraction is freedom from the transplutonium actinides, mainly Am and Cm, which are present in significant levels in FBR mixed U-Pu fuel. Americium and Cm are trivalent ions in solution chemistry and are, therefore, difficult to separate from the rare earth group.

An excellent separation, with batch separation factors exceeding 100, of rare earths from Am has been achieved with a cation exchange resin in an ethanol-HCl medium. It seemed reasonable to expect better separation factors when this system was extended to column operation; however, poorer separations have been observed which are attributed to slow kinetics. Separations are improved by using macro-porous resins with large surface areas, but equilibration is still too slow. The batch separation is adequate but less convenient to use. If satisfactory kinetics cannot be established soon with columns, the batch technique will be used.

A spectrophotometer ordered for this project was received at the end of this quarter. This instrument will be used for continued spectrophotometric studies of colored rare earth complexes, such as the arsenazo III complexes described previously.

c. Evaluation of Technique for Distributing Isotope Dilution Spikes.

Isotope dilution mass spectrometry is based on the addition of an accurately known internal standard (or spike) to the sample, treatment to ensure chemical identity between sample and spike, chemical separation, and mass spectrometric measurement of the sample relative to the added spike. Thus the accuracy of the overall measurement can be no better than the accuracy to which the spike is known. Spikes have been distributed into plastic vials on a weight basis from a plastic squeeze bottle which contains enough spike solution for about 100 one-gram aliquots. It is possible that the concentration of the spike solution may increase slightly (due to solvent evaporation) between the first and last delivery, and thereby introduce an uncertainty in the delivered amount of spike.

This possibility was evaluated by delivering 100 one-gram aliquots of a 0.5 M Zn solution from a plastic squeeze bottle at a rate equal to that used for the delivery of spikes. The first three and last three aliquots were retained for measurement by weight titration with 0.25M EDTA. The average values of the first and last set of three aliquots agreed to 0.02%, well within the experimental uncertainty of the measurement. The spike distributing technique was thus shown to be satisfactory.

2. Determination of O/M
(G. C. Swanson)

Thermogravimetric methods for determination of oxygen-to-heavy metal atom (O/M) ratios are sensitive and precise. The oxides are weighed, subjected to conditions that produce stoichiometry (O/M = 2.000), reweighed, and the initial O/M calculated. The accuracy of such methods depends upon how closely the O/M of the product oxide approaches 2.000. Highly pure U_3O_8 and PuO_2 prepared by careful ignition of the respective metals were used at LASL to determine the conditions of stoichiometry for UO_2, PuO_2 and mechanical mixtures of the two. A theoretical model[2] is being applied to determine conditions producing stoichiometry for UO_2, PuO_2 and solid solution $(U,Pu)O_2$.

A Mettler thermobalance equipped with an Al_2O_3 furnace tube and gas entry tubes was used to study high temperature reduction of UO_2 in highly purified H_2. Spurious weight increases caused by interaction of the Pt sample pan with Al_2O_3 reduction products vaporized or spalled from the furnace tube were prevented by use of a Mo liner and Mo gas entry tubes. Preliminary data generated with UO_2 have shown that pure H_2 reduction gas produces O/M ratios as low as 1.80. A safer system using an Ar-6% H_2 gas will replace the H_2 gas if sufficiently reducing conditions can still be achieved.

Preliminary results on UO_2 at temperatures from 600°C to 1400°C indicate that equilibrium of the gas phase with the solid material is rapidly achieved at temperatures above 1200°C. At lower temperatures, equilibrium is achieved less rapidly due to slow diffusion of O_2 interstitials or vacancies from the UO_2 particle interiors to the surface where reaction occurs.

A study of applicability of the theoretical model to "standard" U_3O_8 at high temperatures is awaiting completion of fabrication of a solid state O_2 concentration cell to be situated in the thermobalance. This cell will allow in situ measurement of equilibrium O_2 potentials of fuel materials simultaneously with the measurement of their O/M ratio.

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CM:257(25)