LA-5390-PR

PROGRESS REPORT

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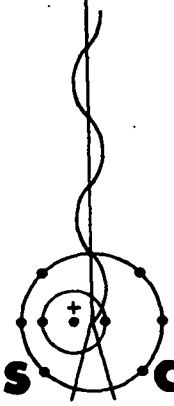
Quarterly Report

Advanced Plutonium Fuels Program

April 1 through June 30, 1973

and Seventh Annual Report, FY 1973



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

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

LA-5067-PR	LA-5193-PR
LA-5106-PR	LA-5284-PR

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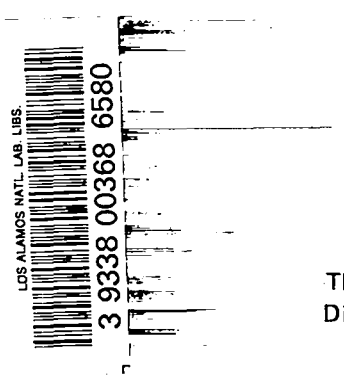

Compiled by

R. D. Baker

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

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ABSTRACT

This is the seventh annual report on the Advanced Plutonium Fuels Program conducted at the Los Alamos Scientific Laboratory. Results of the current quarter's work have been, in most cases, incorporated into the summary of the year's work, and are therefore specifically identified.

Most of the investigations discussed 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 the report should not be made without obtaining explicit permission to do so from the persons 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 DRRD. 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 DRRD.

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, J. H. Bender, E. L. Ekberg, F. J. Fitzgibbon, K. W. R. Johnson, M. E. Lazarus, J. M. Ledbetter, P. A. Mason, C. D. Montgomery, F. H. Newbury, T. J. Romero, P. R. Sena, J. R. Trujillo, L. A. Waldschmidt, W. T. Wood)

1. Mechanical Profilometer

A new mechanical profilometer for measuring the diameters of breached fuel pins following irradiation has been completed and installed in the disassembly cell. Due to the urgent need for replacing the old unit, only a few modifications were made to the previous design. These changes allow easier maintenance and operation of the unit. A new design is under consideration which will automatically correct for errors introduced when bowed pins are measured.

2. Electro-Optical Profilometer

A new Electro-Optical sensor unit (Optron) has been installed at the DP West Facility and will be used in lieu of the Physitech unit whenever profilometry is required on fuel elements that cannot be properly centered. This difficulty occurs when the fuel element is bowed by more than 1/8 in. per 10 in. of length or when the centering roller cannot be used. The latter occurred with the GE structural fuel pins which are multisectional and which have undercuts in diameter between each section.

3. Fission Gas Sampling System

A new fission gas drill has been designed and is now under construction. The new drill was designed to improve capsule and pin void volume measurements by minimizing the gas volume of the first expansion chamber. This improvement resulted in limiting the maximum fuel element diameter from 1-1/8 in. to 7/16 in. Sampling gas from large diameter experimental capsules will be performed with special fixtures.

4. Butyl Acetate Removal System

The refrigeration cold trap unit for removing condensable impurities (mostly butyl acetate) from the atmosphere of the metallography cell has been built, installed, and operated. The equipment has removed as much as 1 liter of liquid in a 24-h period.

5. Fuel Pin Leak Detection and Location

A device has been designed and fabricated to detect and locate cladding leaks by pressurizing the fuel pin. The pressure, up to 200 psi (this may be increased to 400 psi), is applied through a seal assembly which is placed over the fission gas puncture hole. Leakage is determined by a pressure drop in the system and/or helium leak detection. Location is determined by submerging the fuel element in a liquid or by the use of a "sniffer" attached to the leak detector.

6. Macro-Photography Unit

The "three-view" photography equipment was not suitable for use on bowed fuel capsules or pins, since it is impossible to line up the montage of the three views without separating each view. The equipment has been modified to take single views of the elements.

7. Fuel Pin Weighing and Density Apparatus

The apparatus has been designed and an order issued to provide an 8-in.-diameter hole 30 in. deep in a cell at the Wing 9 Facility to contain the lower end of the liquid container. Fuel pins up to 61 in. in length can be accommodated. The unit is expected to be operational about mid-August 1973.

8. Sodium Distillation System

An improved sodium distillation unit has been designed, and a more reliable vacuum pumping system has been ordered. Installation is expected to be completed in late September 1973. The present system is operational and capable of producing satisfactory results until the new system is installed.

9. Miscellaneous Equipment

A special jig was fabricated for sectioning 0.230-in.-diam pins.

Special caps were fabricated for placement over the ends of fuel-clad sections to minimize loss of fragments and pellets from "low burnup pins."

Four additional 4-hole specimen holders were fabricated for use on the modified Unipol grinding-polishing units.

A device for measuring the length of irradiated pins was designed, fabricated, and placed in 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.

B. Inert Atmosphere Systems
(R. W. Basinger, P. A. Mason)

1. The molecular sieve was replaced in the recirculating purifier for the metallography cells.

2. The Ar atmosphere in the Disassembly Cell has been maintained at a higher negative pressure differential during a portion of the report period; this resulted in an increase in the O_2 concentration. This differential was necessary to prevent venting contaminated dust through the manipulator penetration ports. The cell is scheduled for return to a high purity atmosphere (< 50 ppm O_2, < 10 ppm H_2O) prior to the re-introduction of sodium bonded fuel pins or failed pins into the cell.

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

1. Frequent "breakdowns" were experienced with the installation of CRL Model "L" master-slave

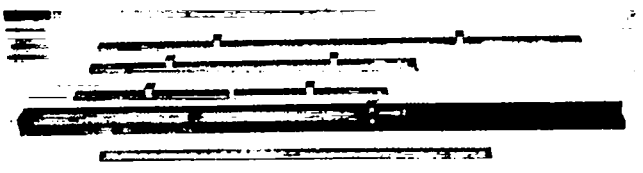

Fig. 401-1. Device for Measuring Fuel Pin Length.

manipulators in the metallography cells. In-cell equipment was modified and heavier tapes were installed in the manipulators without attaining sufficient durability for the more strenuous work requirements. The Model "L" was designed for loads up to 10 lbs, compared to a 40-lb rating for the AMF Standard Duty manipulators which were originally installed.

To prevent delays in the metallography work schedule, the AMF manipulators were re-installed.

Special "slack tape eliminators" were developed by CRL after consultation, and one set was purchased for installation and evaluation.

2. A modified version of a CRL polyurethane manipulator boot design, which is compatible with the CRL Model "L" and the AMF Standard Duty Manipulators was developed. The boots have been found satisfactory, especially in resisting deterioration from exposure to the solvent vapors (primarily butyl acetate) present in the Metallography Cells.

3. A new seal system is being developed for the AMF manipulators installed in the inert atmosphere cells. The seal should provide a more positive method of preventing contaminated dust, which may have penetrated the booting material, from entering the Operating Area.

D. Fuel Pin Handling System for Betatron Radiography

Several modifications were incorporated in the design of the fuel pin handling cart. The electrical wiring was revised and a new control console fabricated. The Radiography Cask and trunnion stand are shown in Fig. 401-2. The cart containing the equipment for elevation of the pin is shown in Fig. 401-3. The cask and cart were used routinely in the fourth quarter of FY 1973.

E. Status of Shipping Casks

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. The latter, however, 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.

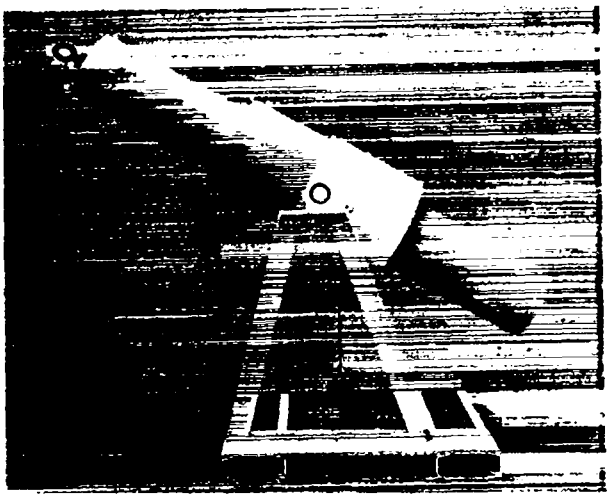

Fig. 401-2. New Radiography Cask Shown in Tilting Fixture.

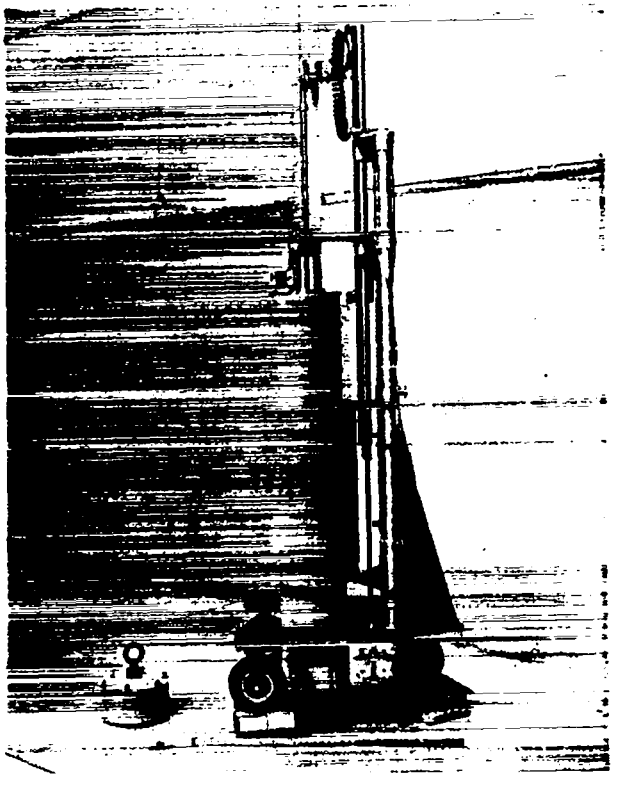

Fig. 401-3. New Radiography Cask with Elevating Mechanism Used at Betatron Building.

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 is obtained from DOT, the fabrication will be started.

Since the TAN Facility is being phased out on July 1, 1973, it is hoped to have the revised casks in operation during the first quarter of FY 1974.

One of the two LASL vertical casks, designated as DOT SP-5885, was made available to the University of Missouri for transporting an irradiated reactor fuel element to Idaho for examination by Aerojet Nuclear personnel. These casks have been made available for use in meeting other AEC-related shipping requirements.

The inserts used in the two vertical casks have been modified to receive the 1/2-in.-diam tubes used for handling in the Argon Cell at EBR-II.

F. Hot Cell Facility at DP-West

The DP-West Hot Cell Facility was completed in the first quarter of FY 1973. It is now considered operational. A view of the Operating area is shown in Fig. 401-4.

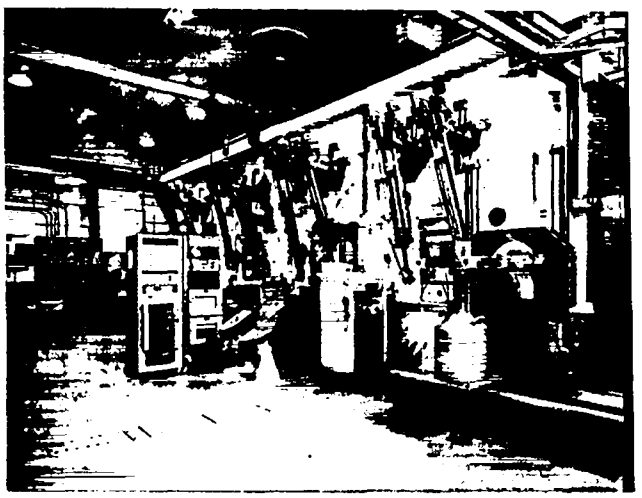

Fig. 401-4. View of Operating Area at the DP-West Facility.

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 were modified to accept pins up to 61. in.in length.

The insert, centering fixture, plug, and lifting device are shown in Fig. 401-5. 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

III. ANALYTICAL CHEMISTRY

1. Gamma Scanning
(J. R. Phillips, T. K. Marshall, J. N. Quintana, J. R. Netuschil, G. H. Mottaz)

A precision gamma scanning system representing the state-of-the-art was installed and operated at the DP-West hot cells. The system was precisely calibrated and tested, including: the calibration of two scanning mechanisms using certified length standards, the radiography of the precision collimators for the presence of voids, and the determination of the surface response function and efficiency of the Ge(Li) detectors as a function of gamma-ray energy. The length measurement

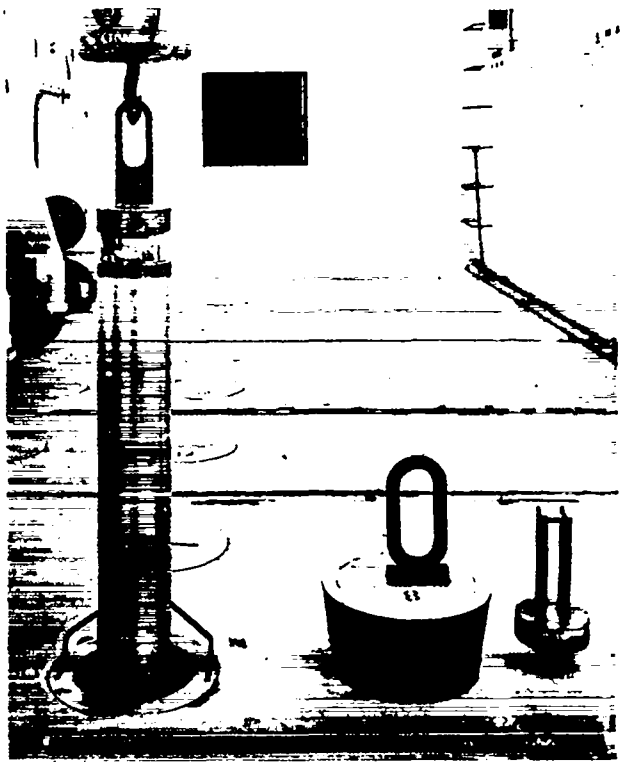

Fig. 401-5. Storage Holes Located in DP-West Facility

calibrations showed that lengths of 11 in. were measured within ± 0.003 in. of the certified value. Significant voids were not found in the collimators. The new system was applied to the determination of the axial, diametral, and two-dimensional radial isotopic distributions of irradiated $(U,Pu)O_2$ and (U,Pu)C fuel pins.

The principal data processing computer code, SURVEY, was expanded to include logarithmic and linear plotting routines and statistical routines for the determination of standard deviation. The efficiency of the code was increased by a factor of two by improving the logic patterns and reducing the magnetic tape manipulation requirements.

The nondestructive determination of the two-dimensional radial isotopic distributions of specific fission products was routinely applied to the examination of some irradiated $(U,Pu)O_2$ fuel pins.[1] This relatively new technique complements axial and diametral gamma scanning to provide more complete characterizations of fission product distributions within a fuel pin.

The migration properties of the volatile fission products ^{134}Cs and ^{137}Cs were investigated in the axial UO_2 blankets. The two isotopes had significantly different distributions which were related to the O/M ratio of the $(U,Pu)O_2$ fuel, the half-lives of the precursor isotopes, the oxygen content of the UO_2 pellets, and to the temperature gradient.

The relative axial burnup profiles over the enriched fuel columns are often determined by summing the two adjoining gamma-ray peaks of ^{95}Zr (756 keV) and ^{95}Nb (765 keV). The net areas of the two gamma peaks were unfolded using a spectral fitting routine to determine the specific isotopic distributions for a failed $(U,Pu)O_2$ fuel pin.[2] The two isotopes had different axial profiles (Figs. 401-6 and 401-7). The ^{95}Zr appeared to have remained with the fuel, while the ^{95}Nb had undergone significant axial relocation, concentrating at six axial positions. The radial isotopic distributions are being determined to characterize further the difference between ^{95}Zr and ^{95}Nb. The new gamma scanning system will be used in continuing nondestructive investigations of the migration properties of fission products.

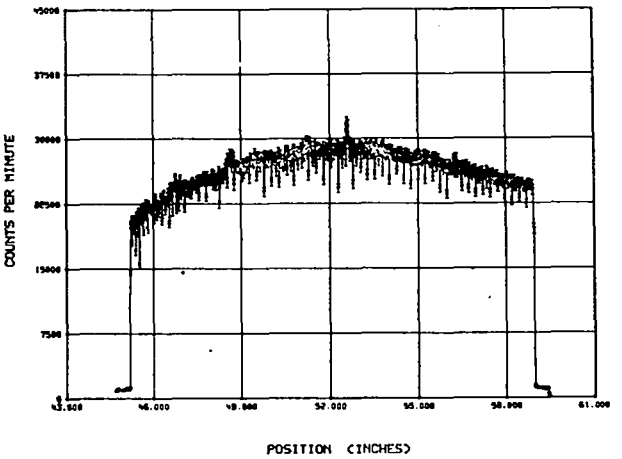

Fig. 401-6. The ^{95}Zr axial isotopic distribution of a failed mixed-oxide fuel pin.

2. Determination of U and Pu in Irradiated Fuels (J. W. Dahlby, R. R. Geoffrion)

Controlled-potential coulometry provided the capability of measuring U and Pu without chemical separation in solutions of irradiated $(U,Pu)O_2$ fuels. The U was measured by integrating the current while reducing U(VI) to to U(IV), and a blank was obtained by again reducing the same sample after a 6-min waiting period. The Pu was determined by integrating the current used to electrolytically oxidize Pu(III) to Pu(IV). The Pu(IV) was then reduced to Pu(III) which was again oxidized to Pu(IV). This oxidation-reduction cycle was repeated until successive

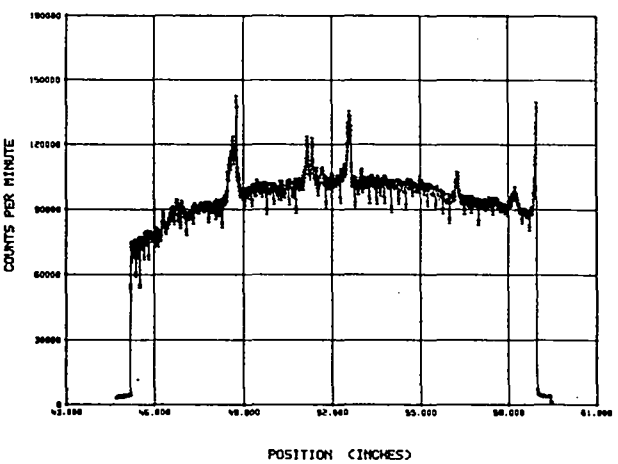

Fig. 401-7. The ^{95}Nb axial isotopic distribution of a failed mixed-oxide fuel pin.

integrated oxidizing currents agreed within 2 mV or 2 μg of Pu. Small samples containing approximately 1 mg of fuel having up to 6 at.% burnup were analyzed for U and Pu with a precision of 0.5% relative standard deviation.

The coulometric analysis of fuels having burnups greater than 6 at.% was adversely affected by the high β-γ radioactivity and the presence of interfering fission products. Because of this, separation of the U and Pu from the highly radioactive fission products was investigated. Precipitation separations, being quick and easy to perform under remote conditions, were investigated first. In the most promising precipitation separation method, the U and Pu were precipitated with NH_4OH, filtered, then the U was selectively dissolved in a basic solution of NH_2OH. Greater than 95% of the β-γ active fission products were removed from the U using this separation scheme. Recoveries of U were 99.6%. Most of the 0.4% loss was caused by a small amount of U (< 1 mg of the original 200 mg of U) not being dissolved by the NH_2OH when Pu was present.

Several techniques were tried to improve the dissolution and recovery of U without reducing the efficiency of the separation from fission products. Slight improvement in the U recovery to the 99.7-99.8% range was attained.

3. Sealed Tube Dissolution of Irradiated Fuels
(J. W. Dahlby and R. R. Geoffrion)

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

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

A system was assembled to dissolve irradiated fuel remotely and to collect the released fission gases for analysis using a mass spectrometer. The system was tested by determining the recovery of a calibrated gas standard. A trap containing silica gel cooled in liquid nitrogen was found to be superior to the conventional molecular sieve trap because the collected gas could be quantitatively desorbed quickly permitting rapid analysis of the gas composition. The system will be installed in a hot cell, tested under remote conditions, and then used to determine the retained fission gases of irradiated fuel materials.

5. Determination of Tritium in Irradiated Fuels and Cladding Materials
(C. S. MacDougall, L. G. Atencio)

As quantities of tritium retained by irradiated fuels and cladding are important reactor operating parameters, measurement techniques were developed and remotized to obtain these data. Tritium is released by burning the sample in moist oxygen at approximately 1000°C. The tritiated water is recovered in a spiral trap cooled to -80°C, and an aliquot is pipetted into a vial filled with a liquid scintillator (Aquasol). The low-energy (0.018 MeV) beta particles are counted in a sensitive scintillation counter. This method applies to the measurement of quantities of tritium ranging from 0.001 to 10 μg. The precision of the method is ± 10 relative percent for 0.1 μg of tritium.

Two irradiated (U,Pu)C fuels were analyzed for tritium. The amount of tritium found, 0.008 ± 0.002 and 0.004 ± 0.002 ppm, was less than the theoretical quantities produced (~ 0.1 ppm). These results indicate that (U,Pu)C fuels do not retain tritium to any great extent. Further analyses are scheduled.

6. Determination of Nitrogen in $(U,Pu)O_2$
(T. K. Marshall, R.G. Bryan, C. H. Ward)

Nitrogen in $(U,Pu)O_2$ is determined in a LECO Nitrox-6 nitrogen analyzer by heating the sample inductively in a carbon crucible containing a platinum-tin bath to a temperature of 2000°C to 2500°C, removing O_2 by trapping as CO_2, and measuring the N_2 following chromatographic separation using a thermistor detector. Using this high temperature and the platinum-tin bath, blank determinations were obtained equivalent to 20 ± 2 μg of nitrogen and repeated measurements on standards at the

60 μg of C level showed that the relative standard deviation was ± 6%.

Plans are being completed to adapt the Nitrox-6 analyzer for hot cell operation and to test the method for analyzing N_2 in irradiated oxide samples.

7. Determination of Water and Chloride in Mineral Oil
(G. E. Meadows, C. H. Ward)

A heated mineral oil bath is used for melting the sodium bonding between the pin cladding and capsule before extracting the fuel pin. As water in the oil could adversely affect this routine process and chlorine could contaminate the sample, the mineral oil must be analyzed periodically for these contaminants. A method for measuring O_2 in LiH was modified for this application. In this method, the oil sample was mixed with a pre-titrated methanol solution of Karl Fischer Reagent (KFR) and the water was titrated with standard KFR using an automatic titrator. The relative standard deviation for determining 9.48, 0.49, and 0.25 mg of water was 1, 8, and 16%, respectively. The poorer precision for small quantities of water was a result of a variation of ± 0.02 ml in the dispensing of the KFR.

Chloride in microgram quantities was measured spectrophotometrically following separation from the mineral oil by extraction. The mineral oil was mixed with benzene prior to extraction of the chloride into an aqueous solution of ferric ammonium sulfate and mercuric thiocyanate. The chloride complexed the mercury, freeing an equivalent quantity of thiocyanate which formed the colored ferric thiocyanate complex. The relative standard deviation was 5% for a single determination of 6 to 50 μg/ml of chloride and 10% for 4 μg/ml.

IV. MICROSTRUCTURAL ANALYSIS

(J. A. Bender, D. D. Jeffries, K. A. Johnson, J. L. Lehmann, H. D. Lewis, K. L. Walters)

A. General Developments

The autoprinter and print processor are now in routine usage and are of significant assistance in handling the photographic work.

Development work on the ion gun etcher continued this year on a low priority basis. A prototype hot-cell model has been designed, fabricated, and operates satisfactorily. Operating parameters are still being investigated.

Techniques were developed and applied for the scanning electron microscope (SEM) examination of beta-gamma active materials (which are alpha active also) up to the level of 25 r/h at contact.

The SEM has been relocated, resulting in a significant increase in resolution due to a major reduction in the strength of ambient electrical fields.

Two polishers were extensively modified and installed in the metallographic cells to give increased capability in processing individual samples.

A new chemical etchant was developed for irradiated fuels which is also an excellent cladding etch, see item 7 under Publications, Section VII.

A study was made of grinding papers with respect to grinding and damage rates versus the grit size, particle density, and size control. This study has increased the remote sample grinding efficiency with minimum damage. A topical report is in preparation.

B. Image Analysis Developments

The Spektor analysis, descriptive code IMAGE has been completed. In addition, a streamline version of this code has been incorporated in an emerging master areal analysis code TRYSAL.

Though in its early stages, TRYSAL currently can perform (1) Spektor analysis with descriptive statistics, (2) classical Saltykov coefficient generation and analysis, and (3) areal analysis based on the general modification of the "Saltykov" type technique. Algorithms for error analysis of this latter technique have also been in part developed.

BASIC SALTYKOV, A PDP 11/20 Basic language algorithm for simple calculation of classic Saltykov coefficients, has also been developed. This code provides the first compact mini-computer algorithm implementing these techniques as well as the first detailed, computational scheme for Saltykov coefficients.

V. REQUESTS FROM DRRD

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 FY 1973 one hundred eleven irradiated capsules were received as follows: Four oxide capsules and 27 advanced fuel capsules were received in the second quarter; 27 oxide fuel capsules, 9 advanced fuel capsules, and 10 structural capsules in the third quarter; and 34 oxide fuel capsules in the fourth quarter. The distribution was: BMI-9; GE-35; GU-16; HEDL-39; LASL-8; ORNL-2; and WARD-2.

Argonne National Laboratory (West): Two samples from two separate pins were examined with the SEM.

General Electric Company: Examinations performed on forty irradiated fuel capsules received on February 14, 1972, October 11, 1972, March 21, 1973, and April 7, 1973, are listed in Table 401-I.

Hanford Engineering Development Laboratory: During this Fiscal Year, shipments were received as follows: 9 capsules on February 1, 1973; 11 capsules on March 21, 1973; and 19 on April 7, 1973. Examinations performed on HEDL materials are listed in Table 401-II.

TABLE 401-I

POSTIRRADIATION EXAMINATIONS OF CAPSULES AND PINS FROM GE

	Examination	No. of Capsules	No. of Pins
1.	Visual Examination	35	13
2.	Preliminary Measurements	25	3
3.	Profilometry, Optical	10	13
4.	Radiography	17	1
5.	Photography, Full Length	35	13
6.	Photograpay, Maximum Bow	22	—
7.	Photography, Incremental	35	3
8.	Photography, Full Length w/wire	—	2
9.	Photography, Incremental w/wire	—	2
10.	Gamma Scanning,[a] Gross	7	—
	Spectral	5	—
11.	Wire Wrap Removal	—	12
12.	Length Measurement	—	5
13.	Gas Sampling and Analysis	2	2
14.	Na Removal	3	—
15.	Clad Removal	13	—
16.	Profilometry (capsule cladding)	2	—
17.	Temperature Measurement (capsule cladding)	2	—
18.	Sectioning	2	9
19.	Photography (sectioned faces)	—	5
20.	Density Measurements	2	6
21.	Burnup	—	10 samples
22.	Microprobe	—	8 samples
23.	Oxygen in Fuel	—	3 samples
	Oxygen in Clad	—	2 samples
24.	Microstructural Analysis[b]	—	—
	a. Optical Microscopy	—	10 pins (28 samples)
	b. Prep'n. for EMX	—	6 samples
	c. Imago Analysis	—	6 samples

[a] 28 Gross gamma scans, 7 complete spectral scans, and 72 distributions of isotopes were calculated.

[b] The optical microscopy includes macrophotography, alpha autoradiography, beta-gamma autoradiography, and as-polished and etched photomicroscopy, (including mosaics) in inert (Ar) atmosphere. Specimens from other experimentor's fuel pins were examined in like manner.

TABLE 401-II

POSTIRRADIATION EXAMINATIONS OF HEDL CAPSULES AND PINS

	Examination	No. of Capsules	No. of Pins
1.	Visual Inspection	39	38
2.	Preliminary Measurements	39	—
3.	Profilometry	1	45
4.	Photography (Maximum Bow)	31	—
5.	Photography (Incremental)	—	3
6.	Radiography	18	37
7.	Gamma Scan[a]	—	—
8.	Eddy Current	9	1
9.	Gas Sample	39	39
9a.	Gas Analysis	38	35
10.	Na Removal	39	—
11.	Clad Removal	39	—
12.	Wire Wrap Removal	—	38
13.	Photography (Maximum Bow) w/o wire	—	30
14.	Na Melting and Pressurizing Tests	—	1
15.	Sectioning	—	20
16.	Burnup	—	15 samples
17.	Microprobe	—	8 samples
18.	Microstructural Analysis	—	—
	a. Optical Microscopy	—	27 pins (95 samples)
	b. Prep'n. for EMX	—	13 samples

[a] 177 Gross, 35 complete spectral scans on 42 and 13 pins, respectively; 136 isotopic distribution calculations on 13 pins; radial distribution of 48 isotopes (using TWODIM computer code) on 6 pins and corresponding density plots and isometric projections.

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 Fuel Program.

1. BMI Experiments -- Examinations performed on nine BMI irradiated capsules received on October 11, 1973, are shown in Table 401-III.

2. Gulf United Experiments -- Examinations performed on twenty-nine GU irradiated fuel capsules received on December 28, 1970, September 10, 1971 and November 6, 1972, are shown in Table 401-IV.

3. LASL Experiments -- Examinations performed on nine LASL irradiated fuel pins received on February 7, 1969, October 11, 1972, and February 16, 1973, are listed in Table 401-V.

Chemical analysis for water and halogens was made on mineral oil used in decladding operations.

On November 1, 1972, two cladding sections from the fuel area of a pin were shipped to ANL-East for examination with an ion probe.

TABLE 401-III

POSTIRRADIATION EXAMINATION OF CAPSULES AND PINS FROM BMI

	Examination	No. of Capsules	No. of Pins
1.	Visual Inspection	9	3
2.	Preliminary Measurements	9	—
3.	Profilometry, Optical	9	—
4.	Photography, Full Length	9	3
5.	Photography, Incremental	9	3
6.	Radiography	9	—
7.	Gamma Scanning[a]	—	—
8.	Eddy Current	9	1
9.	Cover Gas Sample and Analysis	6	—
10.	Fission Gas Sample	—	6
11.	Na Removal	3	—
12.	Clad Removal	3	—
13.	Profilometry.	—	2
14.	Micrometer measurements	—	1
15.	Sectioning	—	3
16.	Density	—	3
17.	Burnup	—	3 samples
18.	Microprobe	—	3 samples
19.	Microstructural Analysis	—	—
	a. Optical Microscopy	—	3 pins (12 samples)
	b. Prep'n. for EMX	—	3 samples

[a] 36 Gross scans on 9 pins; 4 multispectral and 34 isotopic distribution calculations on 3 pins

TABLE 401-IV

POSTIRRADIATION EXAMINATIONS OF GU CAPSULES AND PINS

	Examination	No. of Capsules	No. of Pins
1.	Visual Examination	16	6
2.	Preliminary Measurements	16	6
3.	Profilometry(Optical)	16	8
4.	Radiography	16	—
5.	Photography (Full Length)	16	6
6.	Photography (Maximum Bow)	7	6
7.	Photography (Incremental)	16	7
8.	Gamma Scan[a]	—	—
9.	Gas Sample	11	3
9a	Gas Analysis	11	3
10.	Eddy Current	16	8
11.	Sectioning	—	16
12.	Na Distillation	—	4
13.	Na Melting and Pressurizing	—	1
14.	Density Measurements	—	14
15.	Microprobe	—	6 samples
16.	Burnup	—	10 samples
17.	Tritium Analysis	—	9 samples
18.	Spectrography	—	1 sample
19.	Microstructural Aanlysis	—	—
	a. Optical Microscopy	—	17 pins (52 samples)
	b. Prep'n. for EMX	—	6 samples

[a] 64 Gross gamma scans on 16 pins; 8 multispectral on 8 pins; and 41 isotopic distributions calculated for 8 pins

TABLE 401-V

POSTIRRADIATION EXAMINATIONS OF CAPSULES AND PINS FROM LASL

	Examination	No. of Capsules	No. of Pins
1.	Visual Inspection	2	8
2.	Preliminary Measurements	2	6
3.	Profilometry (Optical)	2	6
4.	Photography (Full Length)	2	6
5.	Photography (Maximum Bow)	2	2
6.	Photography (Incremental)	2	6
7.	Radiography	2	6
8.	Gamma Scan[a]	—	—
9.	Eddy Current	2	6
10.	Gas Sample	7	7
10a	Gas Analysis	7	7
11.	Na Removal	2	—
12.	Clad Removal	1	—
13.	Profilometry (Mechanical)	—	1
14.	Sectioning	—	2
15.	Density	1	2
16.	Microprobe	—	3 samples
17.	Microstructural Analysis	—	—
	a. Optical Microscopy	—	3 pins (12 samples)
	b. Prep'n. for EMX	—	4 samples
	c. Image Analysis	—	3 samples

[a] Gross gamma scans on 8 pins; 6 multispectral scans on 4 pins; 56 isotopic distributions calculated from the 4 multispectral scans

4. NUMEC Experiments -- Density measurements were made on cladding specimens from four pins. A one-inch length of fuel from a pin was removed for chemical requirements.

5. ORNL Experiments -- Examinations performed on two ORNL irradiated fuel pins received on October 11, 1972,are listed in Table 401-VI.

TABLE 401-VI

POSTIRRADIATION EXAMINATIONS OF ORNL CAPSULES

	Examination	No. of Capsules
1.	Visual Inspection	2
2.	Preliminary Measurements	2
3.	Photography (Full Length)	2
4.	Photography (Incremental)	1
5.	Gamma Scanning	2
6.	Radiography	2
7.	Gamma Scan[a]	2
8.	Eddy Current	1
9.	Fission Gas Analysis	4

[a] Eight gross and 5 complete spectral gamma scans, along with the calculation of the distribution of 20 isotopes, were applied in the examination of one fuel pin, while 5 gross gamma scans were done on a second pin.

6. WARD Experiments -- The examinations associated with these experiments are shown in Table 401-VII.

TABLE 401-VII

POSTIRRADIATION EXAMINATION OF WARD CAPSULES AND PINS

	Examination	No. of Capsules	No. of Pins
1.	Visual Inspection	2	2
2.	Preliminary Measurements	2	—
3.	Profilometry (Optical)	2	2
4.	Photography (Full Length)	2	—
5.	Photography (Maximum Bow)	2	—
6.	Photography (Incremental)	2	—
7.	Radiography	2	—
8.	Gamma Scanning[a]	—	—
9.	Eddy Current	2	—
10.	Cover Gas Sample and Analysis	2	—
11.	Na Removal	2	—
12.	Clad Removal	2	—
13.	Temperature Measurements	—	2

[a] Four gross gamma scans were obtained in the nondestructive analysis of 2 fuel pins. In addition, 1 multispectral scan was obtained, from which 8 isotopic distributions were calculated.

VI. QUALITY ASSURANCE
(L. E. Lanham)

General: An audit conducted by RRD included an evaluation of Project 401 operations. The following actions have been taken on the findings of this audit team.

A letter has been submitted to RRD advising them of the additional program cost of implementing independent QA surveillance and project QA Representative functions.

A letter has been submitted to each of the external experimenters advising them of the findings in regard to a more extensive delineation of quality assurance requirements and consideration of assigning a resident experimenter-engineer at LASL. A response in writing has been received from one of the experimenters.

Procedures have been initiated to provide an independent overcheck of data and a signature by a responsible person as to the acceptability and completeness of data packages.

Actions in process on other findings include revisions to the Quality Assurance Manual, preparation of additional sections, and preparation of supporting documents such as check-lists and forms for corrective action requests from outside organizations.

Microstructural Analysis: The RRD finding expressed concern in regard to metallographic data for a specific fuel element examination. The samples prepared were the first of this type. Actions have been initiated to check procedures with prototypical samples before their application to critical samples.

Some adjustments have been made in instrument calibration frequency to accommodate changes in optics. Calibration procedures require recalibration of these instruments when such a change is made. Surveillance by QA has determined that this and other recalibrations are being documented by updated tags, and information recorded in the appropriate LASL notebook.

Hot Cell Examinations: The Quality Assurance Plan for Hot Cell operations is being reviewed to determine if changes may be required to meet the new requirements of the latest amendment to RDT Standard F2-2T.

Quality Assurance procedures are being reviewed to determine the requirements of combining the documents for Quality Assurance Diagnostic Examinations and Standard Operating Procedures into a single document.

The training program for operators has continued. Documents showing the current status of operator certification have been supplied to the Quality Assurance Manager.

Chemical Analysis: All procedures are being reviewed and will be revised as required. The Quality Assurance Plans have been rewritten to meet the requirements of the latest amendment to RDT Standard F2-2T and are being edited.

A significant number of instruments have gone through a recalibration cycle. The QA recall system is effective as a means of checking and assuring that instruments are being recalibrated. All calibration information is being recorded in the appropriate LASL notebook.

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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. G. Moore, C. Baker, R. W. Walker)

1. Fuel Production. With the adoption of a Quality Assurance Program it became necessary to prepare formal fuel specifications prior to the production of any fuel for EBR-II irradiation testing or for Treat experiments. The status of design specifications is presented in subsequent sections. At the time the QA program was put into effect, four batches of high purity, single phase, high density $U_{0.80}Pu_{0.20}C_{0.98}$ were in process, two in the form of annealed arc melted ingots and two in the form of hydrogen treated powder. The annealed ingots showed signs of surface oxidation after 6 months storage

in the inert glovebox atmosphere. One batch of powder stored in a high vacuum furnace deteriorated due to repeated removal from the vacuum system when the furnace was needed for other purposes. The second batch of powder was sampled for oxygen after 3 months storage in a high vacuum container and showed $\leq$ 75 ppm oxygen.

A total of 8 batches of $U_{0.8}Pu_{0.2}N$ was received from Battelle Columbus Laboratories. Each pellet was examined and classified according to its physical appearance. Additional physical characterization was provided by radiographic examination of each pellet. A sampling plan based on statistical methods was developed and samples were submitted for U, Pu, C, N, O, spectrochemical examination, microstructural examination, isotopic analysis, and immersion density determination. Analyses are not yet complete.

2. Process Development

a. Fuel Densification: Previous carbide preparation procedures specified that pellets be sintered for 8 h at 1800°C followed by annealing for 2 h at 1400°C in a flowing Ar atmosphere. These conditions usually produced material which was 95 ± 2% theoretical density, but a few batches of pellets had slightly lower densities. Shown in Table 463-I are the results of resintering these batches at 1850°C for 8 h and at 1400°C for 2 h. It can be seen that an increase of 1.1 to 2.4% TD occurred, and with but one exception, all batches were within a tolerance of 95 ± 2% TD. As anticipated, the resintered pellets showed an increased grain size.

TABLE 463-I

CHANGE IN DENSITY OF $U_{0.8}Pu_{0.2}C$ PELLETS PRODUCED BY RESINTERING

Lot No.	Percent of Theoretical Density	
	Sintered	Resintered
2-10-1	91.9	93.1
2-11-1	92.9	94.0
2-12-1	92.4	93.9
2-16-1	91.5	92.6
2-17-1	91.8	93.5
2-37	92.9	95.3
2-38	92.5	94.0

No trace of sesquicarbide formation or melting was observed in microstructural examination of the specimens. Chemical analysis verified the carbon content and the high purity of the product pellets.

b. Carbide Production by Carbothermic Reduction of UO_2-PuO_2: Progress has been made in development of a process for production of $U_{0.8}Pu_{0.2}C$ by carbothermic reduction of mixtures of UO_2-PuO_2, a process which is expected to have significant economic advantages over the arc melting process. Since the process has been described previously,[1] only a brief description is presented below. The principal processing steps are:

1. Ball mill and blend UO_2, PuO_2 and excess graphite,
2. Press low density powder compacts,
3. Heat in a high vacuum system at 1400-1700°C until CO evolution ceases,
4. Crush and comminute,
5. Heat in flowing H_2 at 800°C to remove higher carbides,
6. Press into pellets,
7. Sinter in Ar.

The unique features of the process developed at LASL include:

1. The intentional use of excess carbon in the initial UO_2-PuO_2-C mixture to ensure the presence of sesquicarbide in the reduction product. This is done to maintain a large carbon activity during the final stages of the reduction in order to maximize the overpressure of CO.
2. The use of high vacuum during the final, high-temperature stage of the reduction step.
3. The use of the LASL hydrogen reduction procedure to remove all higher carbides from the product.

Other experimenters have produced (U,Pu)C using established carbothermic reduction techniques, but in general the carbon content of the product was difficult to control and oxygen contents were normally large. This new procedure was devised to eliminate these problems.

In continuing development work, it has been found that the dynamic system pressure during the final stages

of the carbothermic reduction is a useful indication of the completeness of the reaction. The correlation between the final dynamic pressure and the oxygen concentration in the reduction product is shown in Table 463-II. It can be seen that as the pressure in the system dropped below 5×10^{-5} torr at a furnace temperature of 1600°C, the concentration of oxygen in the product dropped to an acceptable level.

TABLE 463-II

CORRELATION OF OXYGEN CONCENTRATION IN THE REDUCTION PRODUCT WITH FINAL PRESSURE AT 1600°C

Batch	Final Pressure, torr	Oxygen Conc. in Product, ppm
CR-10	3×10^{-5}	< 25
CR-8	3×10^{-5}	30
CR-14	3×10^{-5}	60
CR-13	3×10^{-5}	80
CR-12	3×10^{-5}	160
CR-11	1×10^{-4}	290
CR-7	3×10^{-4}	2000

Temperature gradients within the heated zone of the reduction furnace were found to contribute to an inhomogeneous distribution of oxygen in the reduction product. Seven shallow, cylindrical, graphite trays were used as containers for the UO_2-PuO_2-C compacts during reduction. These trays were stacked and supported in the center of the vertical, cylindrical hot zone of the furnace. As a precaution against possible temperature gradients, the top tray was left empty. Even this was not sufficient, as shown by the data in Table 463-III. Had the entire batch been mixed homogeneously for subsequent processing, the average oxygen concentration would have increased to 77 ppm.

If the carbide from the second tray only had been used in the fabrication of fuel pellets, the oxygen content of pellets derived from this tray would have been unacceptably high. To overcome this potential difficulty, the inside diameter of the trays was increased slightly so that a 250 g batch could be accommodated in the lower five trays.

TABLE 463-III

CORRELATION OF OXYGEN CONCENTRATION IN THE REDUCTION PRODUCT WITH POSITION IN THE FURNACE

Tray No.	Oxygen Conc., ppm
1 (top-empty)	--
2	360
3-6 (combined)	17
7 (bottom)	35

Although processing steps 4 through 7 were basically similar for both arc melted and carbothermic carbide, some differences were observed. These differences appeared in the rate of excess carbon removal by hydrogen treatment and in the attainable densification. Arc melted carbide powders have been routinely subjected to hydrogen treatment at 800°C for periods of 0.6 $h\text{-}g^{-1}$. Reduction of this period to 0.3 $h\text{-}g^{-1}$ led to the presence of sesquicarbide in the product. Table 463-IV shows the results of microstructural examinations of seven batches of

TABLE 463-IV

VARIATION OF CARBON CONTENT OF $U_{0.8}Pu_{0.2}C$ WITH TIME OF HYDROGEN TREATMENT

Batch No.	Batch Size, g	Hydrogen Treatment h	Time $h\text{-}g^{-1}$	Microstructure phases
12-I	100	66	0.66	1
12-III	56	66	1.18	1
12-IV	32	4.5	0.14	1
13-I	117	3.5	0.03	2
13-II	110	50	0.45	1
13-IV	60	30	0.50	1
14-I	134	30	0.22	1

sintered pellets prepared by the carbothermic process. The batches were hydrogen treated for varying periods at 800°C. These data suggest that the period of hydrogen treatment for powders produced by the carbothermic reduction process may be significantly shorter than that used in the arc melting process. The optimum time for hydrogen treatment of carbon rich powders has not yet been established.

Several factors may have contributed to the shorter periods of hydrogen treatment needed for powders from the carbothermic reduction process. These powders had a considerably smaller average particle size than powders from arc melted material. As such, a larger surface area and a shorter diffusion path were presented to both the H_2 reductant and CH_4 product gases. In addition, these powders may have been inherently more reactive with H_2 gas.

Preliminary results indicate that product pellets from carbothermic material were less dense than those from arc melted material when subjected to identical hydrogen treatment, pressing, and sintering conditions. Additional work will be directed toward optimization of conditions for the production of the same high percentage of sound, dense pellets as with the arc melting process. The average oxygen concentration in the pellets represented in Table 463-IV was 347 ± 26 ppm. Although this amount of oxygen may be acceptable in fuel materials, improvement should be possible in routine production of larger batches.

c. Nitride Development: Equipment checks and scoping experiments were begun in the development of a process for the production of nitride fuel for reactor experiments. Powdered UN was prepared by reacting U + N_2 at 1400°C followed by heating at 1450°C in high vacuum. The process for producing UN end pellets will be developed before initiating work on solid solution $U_{0.8}Pu_{0.2}N$.

3. Equipment Development

a. Carbide Facility: A new hydrogen treatment apparatus was designed, fabricated and tested. Insofar as possible, the system incorporated high vacuum and UHV design principles and components. After assembly, the entire system was leak-free when tested with a He mass spectrometer leak detector. The original reduction furnace design was based on a 3-in.-diam x 18-in.-long reaction chamber, but heat losses to the box from that unit were larger than the inert atmosphere recirculator could accommodate. A smaller furnace was subsequently designed, fabricated and installed. Heat losses from this system do not present any problems.

A titanium chip furnace reduces the impurities in the Ar to a few ppm and a palladium diffuser reduces the impurities in the H_2 to a few ppb. Included in the system are back flow preventors, safety interlocks, and monitors for H_2, O_2, and H_2O. The hydrogen detector samples the atmosphere both inside the glovebox and above the exterior gas manifold. These monitors contain alarms which alert personnel in the event of a leak or a malfunction. Prior to operating the system a comprehensive standard operating procedure and safety analysis was written and then reviewed and approved by LASL safety personnel. The manual has been recommended as a guide for others using gaseous H_2 systems.

Solid solution $U_{0.8}Pu_{0.2}C_{1+x}$ was subjected to hydrogen treatment in the new system and the effluent gas was monitored on a gas chromatograph. Preliminary results suggest that it may be possible to reduce the time previously required to adjust the stoichiometry of this carbon-rich material.

b. Nitride Facility: The remaining components of the nitride fuel production system were installed and equipment check-out was begun. The facility consists of a high temperature, tungsten-mesh resistance furnace in one recirculating inert atmosphere glovebox and a hydriding-nitriding furnace, mill, press, and balance in another recirculating inert atmosphere glovebox.

Atmospheres of both Ar and vacuum at temperatures up to 2000°C have been used in conjunction with the sintering furnace. A recently installed Nb-1% Zr chip furnace will be used to purify the N_2 supplied to the furnace. The hydriding-nitriding furnace incorporates all of the features of the hydrogen treatment apparatus plus the capability of introducing purified N_2. Preliminary tests with flowing N_2 indicated that the system was ready for operation. Tests with pure H_2 have been delayed pending the completion and approval of a standard operating procedure and safety analysis. A new centrifugal mill is being tested which should substantially reduce the time previously required for comminution.

c. Miscellaneous Equipment: Inert atmosphere recirculation units on three gloveboxes were modified by replacing the recirculating pumps and furnaces with new, improved components. In addition to an increased

circulation rate the glovebox atmosphere is superior to that previously obtained.

An electronic analytical balance was obtained and evaluated for glovebox use. The balance head within the glovebox is connected to a digital readout exterior to the glovebox. Replacement of the glovebox head due to normal wear and environmental deterioration represents a considerable saving over replacement of an entire analytical balance. In addition, individual weighings can be made in a fraction of the time required in the operation of a mechanical analytical balance. This unit should greatly facilitate the time consuming process of weighing powder charges and product pellets.

4. Associated Activities

A significant portion of the Synthesis and Fabrication task was dedicated to the development and implementation of the RRD QA Program. Formalized documents were produced and approved which provided detailed processing procedures and parametric instructions, sampling plans and procedures, transfer and storage requirements for components, etc. All meters, gauges, measuring devices and instruments associated with any process or component variable were grouped collectively as data gathering devices. In accordance with the recommendations of the Director of Surveys and Audits for the Measurements Standards Department of the Quality Assurance Department of Sandia Laboratories, Albuquerque, New Mexico, a plan was developed for the calibration of all data gathering devices. These devices were brought into calibration by employing one of the following schemes:

a. Devices calibrated against standards traceable to the U. S. Bureau of Standards were purchased.

b. Devices were calibrated at standards laboratories.

c. Calibration systems were designed and fabricated and procedures were developed and approved for the calibration of devices either in situ or in support areas. Approximately 200 data gathering devices were calibrated and placed under the Master Inventory and Recall system.

Associated with the QA Program was the training of operating personnel in the application of all pertinent procedures and in the maintenance and control of QA documentation.

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.

1. Experiment Description and Status

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

a. Capsule Examination

a.1 Visual examination

a.2 Preliminary Measurements (radiation measurements, etc.)

a.3 Profilometry

a.4 Photography

a.5 Radiography

a.6 Eddy Current Test

a.7 Gamma Scan

a.8 Cover Gas Analysis

a.9 Deencapsulation

TABLE 463-V

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[e,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.

b. Element Examination

- b.1 Visual Examination
- b.2 Profilometry
- b.3 Photography
- b.4 Eddy Current Test
- b.5 Fission Gas Analysis
- b.6 Sectioning
- b.7 Autoradiography
- b.8 Metallography
- b.9 Burnup
- b.10 Clad Density
- b.11 Special Tests
- b.12 Data Reduction
- b.13 Report Preparation

All listed burnups are those calculated from EBR-II operation data.

Table 463-V 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 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.

TABLE 463-VI

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

Examinations of these experiments in the LASL hot cells confirmed the failures. Complete postirradiation examination of the failed experiments is currently under way. One unfailed experiment, K-44, is undergoing irradiation in subassembly X182, while a second unfailed experiment, K-39B, is at EBR-II awaiting further irradiation pending preparation of a request for approval in principle to extend the goal burnup.

Table 463-VI 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-VII. 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% theoretical. These experiments are currently at about three-fourths of their goal burnup after operation at low linear power (10 to 15 Kw/ft). 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.

TABLE 463-VII

SERIES U1950 ENCAPSULATED CARBIDE EXPERIMENTS

Expmt. No.	Fuel Type[a]	Fuel Density % Theo.	Bond and[e] Diametral Gap, in.	Clad[f] Type	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	8.4	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.

The Series U1930 and U1960 experiments are described in Table 463-VIII. 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-VIII 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-VIII are currently undergoing irradiation to their goal burnup in subassembly X182. No element cladding failures have been indicated in this group of capsules.

The experiments listed in part C of Table 463-VIII 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-VIII 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-IX 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 is intact. Two of the eight capsules are currently being destructively examined at LASL. Three experiments are continuing irradiation in subassembly X182. 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-VIII

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.

Table 463-X 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 is continuing irradiation in subassembly X182. Further irradiation of capsule B-2-1 is planned, but reinsertion

TABLE 463-IX

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.

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 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 continuing irradiation in subassembly X182.

The Series U5100 singly-clad experiments are described in Table 463-XI. 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 started irradiation in subassembly X156 in May 1973. The first interim examination will be made at a burnup of 2.5 at.%.

The C-5 and ON-1 series of singly-clad experiments are described in Table 463-XII. 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

TABLE 463-X

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-316SS	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-316SS	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-316SS	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-316SS	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-316SS	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-316SS	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-316SS	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.

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.

2. Postirradiation Examination Results

As indicated in the previous section, most of the elements undergoing postirradiation examination are in the intermediate stages of their examination. As a compromise between reporting piecemeal results on all elements as they are obtained and waiting for complete results on a related series of experiments before reporting, this section will report significant trends in examination results as they become apparent. These trends should be considered as preliminary, when reported in progress reports, since additional examination results may alter initial ideas. Final examination results will be reported in topical reports.

a. Severe Failure Mechanism: During the postirradiation examination of failed elements, the overall condition of each element has been described by assigning a "degree of failure" to the element. Three classifications have been used: (1) a slight failure being an element known to have failed from evidence such as γ-scanning but with no readily observable cladding defects, (2) a moderate failure being an element with several individual cladding cracks, and (3) a severe failure being an element with many interconnected cladding cracks and

TABLE 463-XI

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, X156
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 slotted.

[c] SA = Solution Annealed

in some cases melted cladding. It should be pointed out that all failures discussed are failures of the element cladding. In no case has capsule failure been observed. An initial goal of the postirradiation examination process was to establish a cause or mechanism for the severe failures. A mechanism by which an element that has previously undergone cladding failure and would be considered as a slight or moderate failure can progress to a severe failure has been developed. The mechanism postulates the release of fission gas from a cladding defect into the capsule annulus. The entrapment or lodging of fission gas bubbles in the capsule annulus effectively gas blankets the element cladding in isolated areas, causing severe overheating of the cladding and possible melting. The continued irradiation of encapsulated elements after initial cladding failure leads to a progression of the element condition from slight through moderate to severe failure as local overheating of the cladding leads to more cladding cracks or melting which then provide additional areas where fission gas may be released from the element into the capsule annulus. The remainder of this section describes the proposed mechanism in more detail and outlines the postirradiation examination observations which have contributed to defining this mechanism.

There are three specific areas where postirradiation observations have contributed evidence to support this mechanism of element-capsule interaction which leads to the severe failures observed. They are (1) the behavior of the element and capsule sodium levels in sodium-bonded elements as determined during interim and final examinations, (2) the quantity and isotopic distribution of fission gas in the capsule plenum of failed sodium-bonded elements, and (3) the condition of the cladding and capsule of severely failed elements. The observations on sodium level changes, along with an estimation of the degree of failure of failed element currently under examination and for some elements previously examined by Gulf United Nuclear Fuels Corporation are shown in Table 463-XIII. The degree

TABLE 463-XII

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	1156	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	1158	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	1184	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	1127	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	1133	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	1105	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	1121	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	1121	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	1113	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	1142	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	1113	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	1115	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	1123	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	1107	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	1140	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	1164	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	1164	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	1117	12	0	At LASL[d]

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

of failure for experiments that have not been deencapsulated must be considered as only an estimation and may change when the visual examination of the element can be completed. The table lists the results of the observations of the sodium level changes in the capsule plenum for sodium- and helium-bonded elements and in the element for sodium-bonded elements. Where possible, level changes are tabulated, but in some cases initial or final levels are not known well enough to calculate actual changes. Where available, results from neutron radiographs taken during interim examinations are noted.

In the seven "moderately" or "slightly" failed sodium-bonded experiments for which sufficient preirradiation data are available for either the element or the capsule, the element sodium level appeared to rise in only one case, and all of the capsule sodium levels either rose or remained at the original level. For these experiments, the data indicate a sodium transfer from the element to the capsule. In seven of the nine "severely" failed sodium-bonded pins for which preirradiation data are available, the sodium level in the fuel element has risen during the irradiation. In five of the seven, the sodium level in the

TABLE 463-XIII

SODIUM LEVEL CHANGES IN FAILED EXPERIMENTS

Capsule No.	Capsule sodium level with respect to initial level, in.[e]	Element sodium level with respect to initial level, in.[e]	Degree of Failure[a]
A. Sodium Bonded Experiment			
K-38B	up	down 3.0 (down 2)	severe[b,c]
K-43	insufficient data	down 0.3	severe[b]
K-49	down	up 2.5 (up 1.3)	severe
K-50	down	up 2.9 (up 0.8)	severe[b]
K-51	down	up 1.2 (down 0.7)	severe[b]
B-2-6	up 0.2	up 0.4	severe[b]
B-2-7	up 0.7	up 1.6[d]	severe[b]
B-3-4	down (down)	up 2.5 (up 2)	severe
B-3-5	down 5.5 (up 0.15)	up 2.0	severe
B-2-2	up 1.0	up 4[d]	moderate[b]
B-3-2	up 0.5	down	moderate
K-36B	up	down 2.3 (down 1.0)	slight
K-45	insufficient data	down 0.3	slight[b]
K-46	insufficient data	down 0.5 (no change)	slight
B-3-3	up 0.8	insufficient data	slight[a]
U-104	up 3.6	down	slight
B-1-1	insufficient data	insufficient data	moderate[b]
B-1-2	insufficient data	insufficient data	severe[b]
B. Helium Bonded Experiment			
U-96	up 0.4	——	moderate[b]
U-106	no change	——	moderate[b]
U-114	down 1.5	——	moderate[b]
U-200	up 0.4	——	slight[b]
U-101	no change	——	severe
U-107	down 0.3 (up 0.5)	——	severe
U-108	down 0.6 (up 0.3)	——	severe
U-109	no change	——	slight
U-111	up 0.2	——	slight
U-112	up 0.2	——	moderate

a. Slight - no readily observable cladding defects
Moderate - several individual cladding cracks
Severe - many interconnected cracks or melted cladding.

b. Failure classification based on visual examination. Classifications of the other elements are based on radiographic data.

c. K-38B was damaged during reconstitution at EBR-II.

d. Inner fuel hold-down spring interfered with positive definition of sodium meniscus.

e. Observations in parentheses are for data obtained from neutron radiographs at interim examinations.

capsule has clearly lowered. This indicates that sodium has been transferred from the capsule to the element, which is opposite to what would normally be expected. The helium-bonded experiments show a similar trend. For three of the seven "moderately" or "severely" failed elements, the sodium level in the capsule has lowered during the irradiation. For the three experiments classified as slight failures, the sodium levels in the capsules have either risen or remained constant.

In the eight experiments in which the final sodium level in the capsule was lower than the initial level, in four cases (K-51, B-3-5, U-107, and U-108) data from interim examinations showed that the capsule sodium level had originally risen. The transfer of sodium from the pin to the capsule, therefore, occurred during the later stages of the irradiation.

The behavior of the sodium levels can be explained by a discussion of the disposition of fission gas in the capsule-element system. Figure 463-1 shows a schematic representation of a sodium-bonded element and capsule at three different stages of its lifetime. Figure 463-1 (a) represents the system at the time of fabrication. The overpressure in the element plenum and capsule plenum is the same at this time. As irradiation proceeds, some fission gas is released from the fuel into the element plenum. Both the element plenum and the capsule plenum operate at essentially the same temperature during irradiation. Therefore the fission gas in the element plenum results in a higher pressure in the element with respect

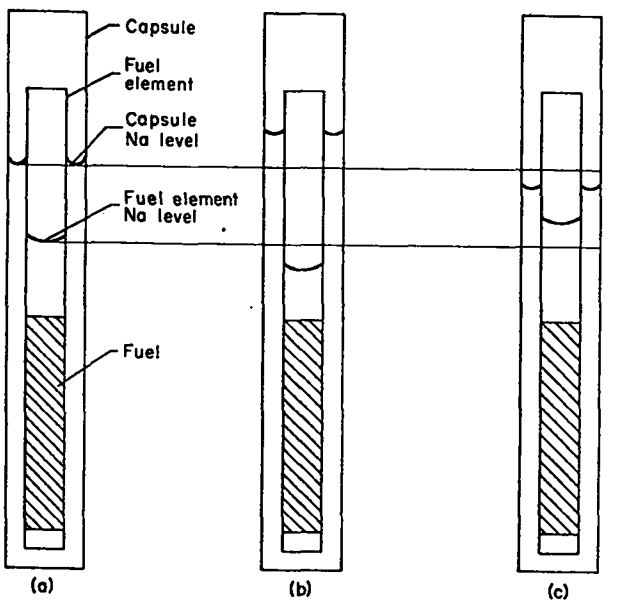

Fig. 463-1. Schematic of capsule and element sodium levels, (a) as-fabricated, (b) after initial failure, and (c) after failure propagation caused by encapsulation

to the capsule as irradiation proceeds. When a breach occurs in the element cladding, the pressures can equalize by the flow of material from the element into the capsule. In the case of a sodium-bonded element with a cladding breach in the fuel region, sodium from the element will most likely be displaced into the capsule. This is shown in Fig. 463-1 (b) by a drop in the element sodium level and an increase in the capsule sodium level. As noted previously this condition has been observed in slight and some moderate failures, and in interim examinations of some severely failed elements. It is also possible that fission gas may be trapped in the element bond in the vicinity of a cladding breach, and thus when initial failure occurs a mixture of fission gas and element sodium can be displaced into the capsule. In a helium bonded element a mixture of fission gas and element cover gas would be released into the capsule on initial failure. In that case, the capsule sodium level would probably not change.

As irradiation of a slight failure proceeds, the fission gas which is normally released from the fuel can now move into the capsule as well as into the element plenum. If all the fission gas released from the fuel moved into the element plenum, the element sodium level would continue to drop in order to maintain a pressure equal to that in the capsule plenum. But contrary to this scheme, the element sodium level of severely failed elements increases and the capsule level drops (See Figure 463-1(c)). This indicates that a large fraction of the fission gas normally released from the fuel moves to the capsule plenum, thus displacing sodium out of the capsule and back into the element. This is probably due to fission gas being able to move through the cladding breach and up the capsule annulus easier than up through the element bond and fragmented fuel. Thus the observed sodium level changes in slight through severe failures provide evidence that significant quantities of fission gas are released into the capsule annulus during irradiation after initial failure.

An examination of the quantity of fission gas and its isotopic distribution in the capsule plenum can provide some information about the material displaced from the element to the capsule during initial failure of a sodium-bonded element. Sodium-bonded elements classified as slight failures have very small quantities of fission gas in the capsule plenum, much less than 1% of the total capsule plenum gas. The isotopic distribution of Xe and Kr in the capsule plenum of slight failures is very different from the isotopic distribution expected (and found in the element plenum). In general the Xe isotopes with short-lived precursors are depleted from their expected equilibrium content while those with long-lived precursors are enhanced. A qualitative explanation of these observations is that on initial failure of a sodium-bonded element in the fuel region, only sodium is displaced from the element to the capsule. The fission gas observed in the capsule plenum comes from the decay of precursors which are dissolved in the element sodium. Until such time as the initial cladding breach is enlarged to the point where fission gas can move to the capsule more easily than up the fuel stack, little or no fission gas will be released into the capsule. In severely failed elements large quantities of fission gas are found in the capsule plenum. The isotopic distribution of Xe and Kr is essentially the same in the element and the capsule of these severe failures, indicating that no selection mechanism is occurring.

The cladding of severe failures was described as having many interconnected cracks and possible melting. Figure 463-2 shows three views of a section of B-3-4, which is typical of a severe failure. The center of the views corresponds to a position slightly above the midplane of the reactor. Generally, the cladding in the lower part of the 0^{o} view and the top part of the 120^{o} view appeared shiny with no defects. The remaining surface is severely cracked with one area in which the cladding has melted. Typically, the severely failed elements have about one-half of the circumference at any axial position in the failed region in a condition which closely resembles the as-fabricated appearance. The remaining one-half of the circumference has large axial and circumferential cracks and some melted cladding areas. This general crack pattern often has a varying amount of a helical nature (the elements had no spirally wrapped spacer wire). Usually, no defects occur in the cladding on the lower 25% of that part of the pin in the core region.

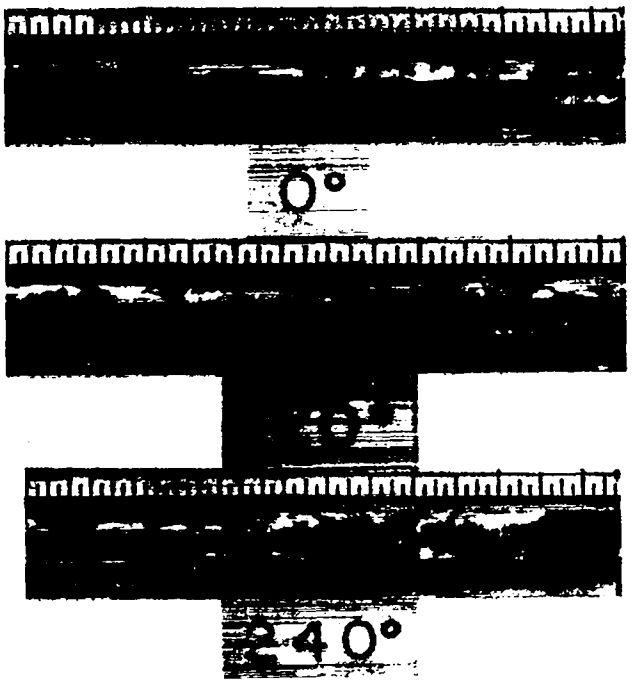

Fig. 463-2. Appearance of a portion of fuel element cladding from B-3-4. Note cracks and melt area on one side of element and essentially undamaged cladding on the other side. No fuel was lost through the melted area.

The capsule which contained B-3-4 was sectioned longitudinally and examined for signs of melting opposite the cladding melt shown in the 0^{o} view of Fig. 463-2. No indication of melting was found. Another severely failed element (K-49) was sectioned for metallography while still encapsulated. An examination of the capsule wall adjacent to areas where the cladding had melted gave no indication of interaction with the cladding or of overheating.

The fact that clad melting has occurred in severe failures with no capsule deterioration in adjacent areas indicates that some resistance to heat transfer has been interposed between the element cladding and the capsule. This relates directly to the previous discussion of the release of fission gas into the capsule annulus, since the thermal conductivity of fission gas is more than an order of magnitude less than that of liquid sodium. Thus fission gas bubbles in the capsule annulus can cause local overheating of the cladding. Local overheating of the fuel adjacent to the gas bubble can also cause increased fuel swelling which results in large forces on cladding that is weakened due to the high temperatures caused by the fission gas bubble. Thus, the presence of fission gas bubbles in the capsule annulus can cause more cladding cracks which then allow additional paths for fission gas to move into the capsule. Thus, the mechanism may be autocatalytic. The continued release of fission gas to the capsule may lower the capsule sodium level below the top of the fuel stack in a very severely failed element. In the postirradiation examination of three experiments (K-49, B-3-4, and B-3-5) no definite capsule sodium level could be detected by eddy-current testing. These capsule sodium levels were probably at or below the top of the fuel stack.

Essentially all the discussion and evidence presented for the element-capsule interaction mechanism has been for sodium-bonded elements. None of the helium-bonded elements now under examination have been classed as severe failures at this time. Some helium bonded elements examined earlier by Gulf United Nuclear Fuels Corporation would be classed as severe failures. Clad melting was observed in at least one case, U-107. Thus helium-bonded encapsulated elements can be subject to this same mechanism.

In summary, the severe failures observed in the current series of examinations appear to be caused by an element-capsule interaction that allows fission gas to be released into the capsule annulus and maintained long enough to cause local overheating and in some cases melting of the cladding. If the element were not encapsulated, any fission gas released from the element would be swept away by the flow of reactor coolant. Therefore, heat transfer from the cladding would have been maintained (except for very short periods as the bubbles passed by) and no significant overheating would occur. Unfortunately little can be learned about the initial failure mechanism from severely failed elements. Elements classed as slight or moderate failures are expected to provide more information in this area.

Further analyses of this element-capsule interaction mechanism are currently in progress. These analyses are mainly heat transfer calculations simulating gas bubbles in the capsule bond.

b. Circumferential Element Scratches: In the majority of the fuel elements that have been deencapsulated to date, both failed and unfailed, circumferential scratches have been observed on the outside surface of the cladding. These scratches may or may not be related to initial cladding failure and are apparently due to the element being encapsulated. Three of these scratches are shown in Fig. 463-3. The element illustrated in Fig. 463-3 is K-43 and the scratches are in the upper half of the fueled region of the element. The single scratch is one of the worst that has been observed. The majority of the scratches that have been observed have been in the plenum regions of the fuel elements, but a significant number have been seen in the fueled regions. As many as fifty circumferential scratches have been observed on a single element. The severity or depth of the scratches appears to be inversely correlatable with the total number of scratches. For at least those experiments fabricated at LASL (the K Series), the scratches were not present when the fuel element was loaded into the capsule. Since none of the elements from any of the fabricators has a mechanism to prevent rotation of the element with respect to the capsule, it appears that the scratches are formed during irradiation by the rotation of the element inside the capsule, possibly due to vibration. All the scratches observed to date are purely circumferential and have no helical nature, which implies that no significant axial movement of the element inside the capsule occurred. No extraneous material has been found inside the capsules which could cause these scratches, but the effect will be investigated further during future deencapsulations.

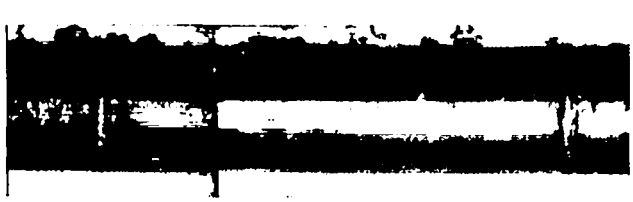

Fig. 463-3. Circumferential scratches on the outer surface of fuel element K-43.

3. Miscellaneous Items

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, WF, ORNL-43, 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 series (except the O-N1 series) from a quality control standpoint has been completed. The transfer of information from ORNL, the original experimenter, has been received recently 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 RRD 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-XIV describes the fuel elements and test objectives. LASL assumed complete responsibility for these tests in fiscal year 1973.

TABLE 463-XIV

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.

a. Tests LASL-UL-1 and LASL-UL-2: The capsules for these tests were received from GUNFC in July 1972. A series of operations and tests was required at LASL due to the quality assurance requirements of TREAT and to leaks which had been found in both TREAT capsule head, potted seals and to the quality assurance requirements of TREAT. Two new TREAT capsule heads were prepared. The heads and capsule bodies were hydrostatically tested to 15,000 psi and helium leak tested. The inner capsules and TREAT capsules were assembled, subjected to a helium leak test, an electrical continuity test, a visual examination, and a trial heat-up to pretest temperatures using the capsule heaters. During this same time period a quality assurance plan covering these tests was written and accepted by the TREAT staff. LASL received permission to ship the capsules to TREAT in September and both capsules were shipped in early October.

The capsule for test LASL-UL-1 was loaded into the reactor and a calibration transient (TREAT transient number 1476) was run on November 15. The object of the calibration transient was to experimentally determine a figure of merit for these tests by the heat balance method. The figure of merit is defined as the ratio of the power density in the fuel to the reactor power. This quantity is calculated from the total reactor energy deposited in the fuel and inner capsule as determined by the temperature rise of the inner capsule. The TREAT capsule-inner capsule system is designed so that the inner capsule loses little heat over a time period required to equilibrate temperatures radially in the inner capsule over the fuel column length.

The inner capsule thermocouples indicated spurious signals during this calibration transient. These thermocouples have ungrounded junctions and these signals were

traced to the fact that these thermocouples had not been externally grounded prior to the transient.[2] The thermocouples were grounded and a second calibration transient (TREAT transient number 1477) was run. This calibration transient generated 82 MW-sec of reactor energy using a computer controlled transient. Figure 463-4 shows a plot of reactor power as a function of time for this transient. The temperature rise of the inner capsule, adjusting for heat losses, was 152°F. The energy deposited in the inner capsule was calculated from the heat capacity of the inner capsule components as 467.5 cal/cm of length. If all this energy were generated in the fuel, the figure of merit would be 7.75×10^{-5} W/(cc fuel) (W reactor power). This estimate would ignore γ heating in the nickel heat sink and stainless steel capsule components which must be considered to accurately estimate the figure of merit due to fission heating in the fuel. Using an estimate of γ heating as 2.51×10^{-6} W/(cc) (W reactor power),[3] the figure of merit due to fission heating in the fuel was calculated as 7.34×10^{-5} W/(cc fuel) (W reactor power). This is 20% below the calculated value of 9.17×10^{-5} reported previously.[4]

Test UL-1 (TREAT transient number 1478) was run shortly after the calibration transient. The total reactor energy requested was increased over that originally requested to account for the lower figure of merit determined during the calibration transient. The reactor energy totaled 159 MW-sec. Figure 463-5 shows a plot of reactor power as a function of time. The temperature rise of the inner capsule, accounting for heat losses, was 284°F. A calculation of the figure of merit from this transient (fission heating only) results in 7.38×10^{-5} W/(cc fuel) (W reactor power), which is excellent agreement with the result obtained from the calibration transient. The response of the fuel element and inner capsule to the reactor power shown in Fig. 463-5 was calculated using CINDA heat transfer code. Figure 463-6 shows the results for three selected temperatures, the fuel center (curve 1), the clad inside surface (curve 2), and the inner thermocouple location (curve 3). The data points are measured thermocouple temperatures for the inner thermocouple located at the axial center of the fuel column (TC-4). The break in the fuel center temperature at 4.0 and 4.8 sec corresponds to the start of fuel melting and the end of fuel melting (complete resolidification) at the fuel center. The maximum clad temperature of 1435°F occurs at 4.3 sec.

Figure 463-7 shows an expanded plot of the calculated and measured thermocouple temperatures. Since the

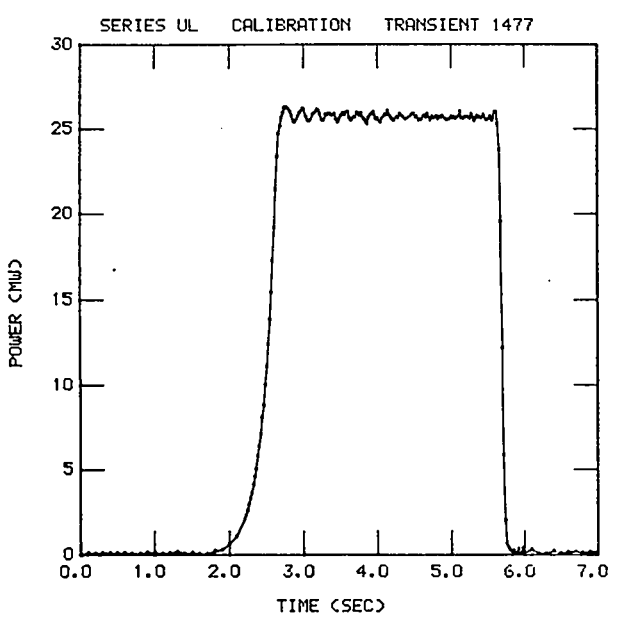

Fig. 463-4. TREAT reactor power as a function of time for calibration transient for Test UL-1.

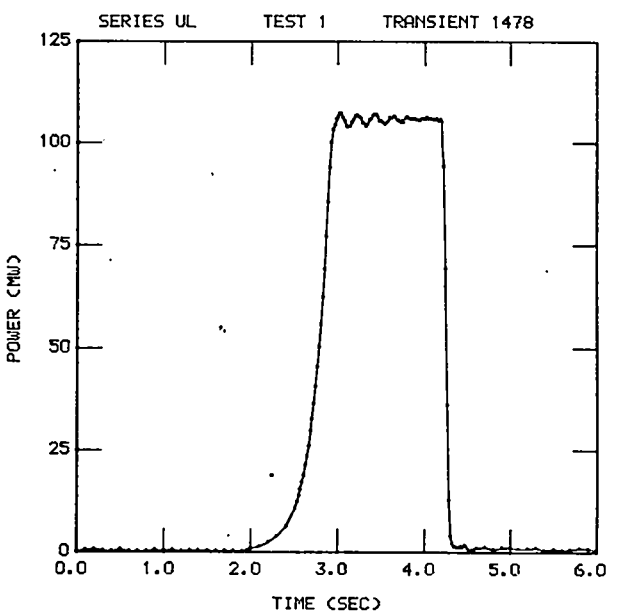

Fig. 463-5. TREAT reactor power as a function of time for test transient for Test UL-1.

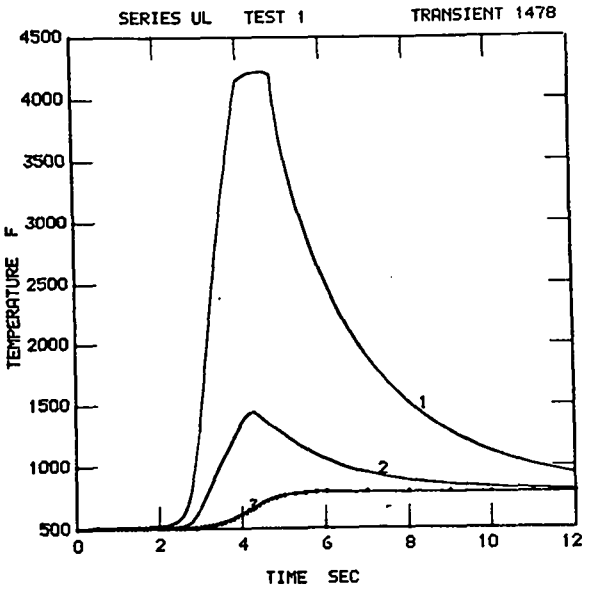

Fig. 463-6. Temperature as a function of time for Test UL-1.
Legend: Curve 1 = calculated fuel center, Curve 2 = calculated clad inside surface, Curve 3 = calculated thermocouple location, * = measured inner thermocouple temperature.

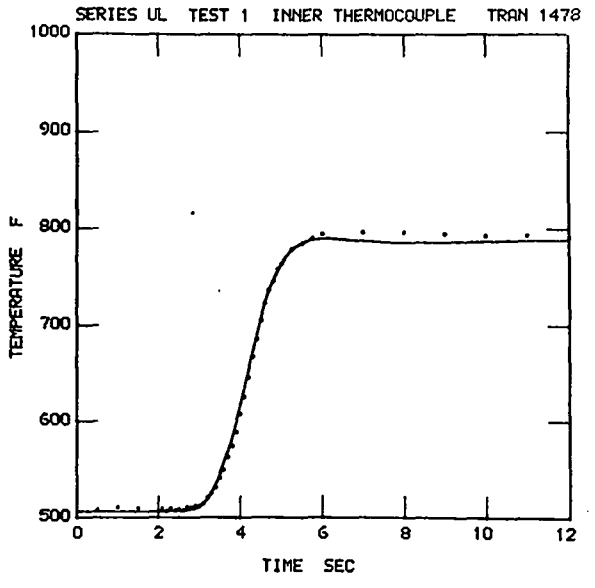

Fig. 463-7. Inner thermocouple temperature as a function of time for test UL-1
Legend: Curve = calculated temperature, * = measured temperature.

thermocouples in the inner capsule have ungrounded junctions, the actual location of the thermocouple junction relative to the end of the thermocouple sheath and the response time of the thermocouple junction must be considered. Thermocouple structure (junction location, junction size, sheath thickness, insulation thickness, and wire thickness) was determined from radiographs of the actual thermocouples used. The calculated thermocouple junction temperatures shown in Fig. 463-6 and 463-7 were obtained from numerical heat transfer calculations on a model of the thermocouple using dimensions and properties of the sheath, insulation, and wires. Sheath surface temperatures were assigned as the temperature of the surrounding medium (NaK in the inner annulus or nickel in the heat sink) at that radial location. The time constant of the model thermocouple was calculated as approximately 300 μsec which is in good agreement with measurements on similar thermocouples.[5]

The capsule from test UL-1 was returned to LASL in January 1973. The TREAT capsule was radiographed at TREAT (neutron radiograph) and at LASL (x-radiograph). The inner capsule was removed from the TREAT capsule and also x-radiographed. A comparison of the pre-test and post-test radiographs shows that considerable pellet cracking occurred during the test. No indication of fuel element failure has been seen.

The TREAT capsule for test UL-2 was loaded into the reactor on November 17 but the test was not run owing to unfavorable weather conditions through November 27. At that time the capsule was unloaded from the reactor and placed in storage because of higher priority tests at TREAT. The test was rescheduled for early 1973, and was performed on February 20, 1973 (TREAT transient number 1495). The reactor transient requested for test UL-2 was the same as for test UL-1. A preliminary review of the capsule temperatures indicated that the test was performed as requested. The capsule was returned to LASL in March. The TREAT capsule neutron radiographs and x-radiographs, and the inner capsule x-radiograph were compared with similar pre-test radiographs. Little or no change was observed. Although calculations indicated that the fuel element Na bond would boil during the test, there

is no indication at this time that bond boiling occurred.

The inner capsules from tests UL-1 and UL-2 are now in storage. Further examination will require use of the LASL hot cells, which are currently unavailable due to higher priority work. A hold has been placed on any destructive examination of the inner capsules until after tests UL-3 and UL-4. At that time a decision will be made whether to continue examination or request further tests of the elements 263 and 265 under more severe conditions.

b. 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 pre-irradiated in EBR-II, the completion of the assembly of the inner capsules and the assembly of the inner capsules into the TREAT capsules requires hot cell facilities. LASL, in conjunction with RRD, has arranged for the completion of the assembly by the Radiometallurgy Group of Hanford Engineering Development Laboratory.

A preliminary discussion between LASL and HEDL staff indicated that only minor changes were required in the normal HEDL assembly procedures used for these operations. The fuel elements and partially assembled inner capsules were shipped to HEDL during August and September 1972. A purchase order was written to HEDL to cover the assembly costs, and welding samples for weld development work on the inner capsule welds were supplied. The TREAT capsules (TR-1 and TR-2 which had been used for tests UL-1 and UL-2) were inspected at LASL and shipped to HEDL in April 1973. A QA plan covering the assembly work at HEDL and the post-test examination was written and approved by the TREAT staff. The assembly work at HEDL was completed with no significant problems. The two capsules are currently awaiting a QA review of the HEDL work prior to shipment to TREAT.

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-XV summarizes the test parameters and objectives. Approval-in-principle has been received from the AEC for this series of tests.

Design work on the inner capsules for these tests was carried out and completed during this fiscal year. This design incorporated a pressure transducer to monitor fuel element plenum pressures during the transient. Design of the modifications of the TREAT capsules to incorporate a thermal neutron filter was also completed. Materials required for all the modification work were purchased. 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 RRD, 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 carbide or nitride fuel interacts with liquid sodium, and to measure the pressure 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.

4. Postirradiation Disassembly Facilities. LASL began preparation of postirradiation disassembly facilities for TREAT tests. These facilities would allow the disassembly of TREAT capsules and inner capsules containing fuel elements with no other irradiation except the TREAT irradiation through the point of sectioning of the fuel element in preparation for metallography. Engineering drawings for the modification of two existing evacuable gloveboxes and for transfer devices between the boxes were prepared. Modification of one of the boxes has been completed. Further work on the disassembly facility has been postponed following a reduction in funding.

TABLE 463-XV

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.

III. ANALYTICAL CHEMISTRY

1. X-Ray Fluorescence Spectrometric Analyses of Stainless Steel for As and Ta

(J. M. Hansel, G. B. Nelson, V. D. Jolin, R. E. Smith, E. A. Hakkila)

Chemical evaluation of stainless steels for FFTF Program applications requires reliable measurements of several specified trace impurities including Ta and As. As these two impurities are difficult to measure with the necessary sensitivity by existing emission spectrochemical methods, x-ray fluorescence techniques are being applied. For the determination of 20 to 1000 ppm of As, solutions containing 500 mg of sample plus Ga internal standards are evaporated to dryness, ignited to 300°C for 2 h, and the solid residue is mixed thoroughly before irradiation with x-rays from a tungsten-target tube. The intensities of the Kα x-ray for As and the Kβ x-ray for Ga are measured and compared to intensities from standards to obtain the As content. The relative standard deviation ranges between 30% in measuring 20 ppm of As to 3% at the 600 or 1000 ppm As level (Table 463-XVI). The limit of detection, defined as three times the standard deviation of measuring a blank, is approximately 10 μg, or 20 ppm for a 500 mg sample.

TABLE 463-XVI

PRECISION OF X-RAY FLUORESCENCE DETERMINATION OF As IN STAINLESS STEEL (500 mg SAMPLE)

As Added, μg	No. of Determinations	Standard Deviation, μg	Relative Standard Deviation, %
0	7	3.0	-
10	8	3.4	29
50	8	1.7	3.4
100	7	3.0	3.0
300	8	8.0	2.7

A study of the effects of several elements which might interfere with measurement of As or Ga x-ray intensities showed that up to 0.3% of Ta, 0.1% of Cu or Bi, 400 ppm of Pd, 100 ppm of Hf, 40 ppm of W, or 20 ppm of Tl did not cause serious interference with the procedure. Although Pb interferes, a correction can be made based upon 10 μg of Pb being equivalent to 5 μg of As in this measurement.

An existing solvent extraction separation of Ta[6] was modified to permit final measurement by x-ray fluorescence spectrometry. The Ta in a solution of 0.5-g of stainless steel in 6$\underline{M}$ H_2SO_4-1$\underline{M}$ HF was extracted into hexone for a preliminary separation. A second extraction of the Ta into hexone from 6$\underline{M}$ H_2SO_4-0.4$\underline{M}$ HF completed the separation. The hexone extract plus 50 μg of Hf were evaporated on a microscope cover glass, excited using a tungsten-target x-ray tube, and the intensities of the $L\alpha_1$ x-rays for Ta and Hf were measured and compared to intensities for known samples. The relative standard deviation varied between 32 and 4.6%, and as little as 10 μg of Ta could be detected (Table 463-XVII). Among the elements that might interfere, up to 0.5% of Mo and 0.1% of W, Nb, As, and Co were shown to have no effect.

Quality Assurance Documents describing these two x-ray fluorescence measurements have been written.

TABLE 463-XVII

PRECISION OF DOUBLE EXTRACTION-X-RAY FLUORESCENCE SPECTROMETRIC DETERMINATION OF Ta IN STAINLESS STEEL (500 mg SAMPLE)

As Added, μg	No. of Determinations	Standard Deviation, μg	Relative Standard Deviation, %
0	9	1.1	
10	10	3.2	32
25	9	3.7	15
100	8	11.9	12
250	10	11.3	4.5
500	8	23	4.6
1000	9	46	4.6

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

In the coulometric titration of U in the presence of Pu, trace quantities of Cl^- ion catalyze oxidation of Hg^o to Hg^+ during pre-electrolysis at +0.085V vs the saturated calomel electrode (SCE). At the potential of the final electrolysis, which is -0.325V vs the SCE, Hg^+ is reduced to Hg^o. It is imperative, therefore, 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). As the most likely source of Cl^- contamination is the SCE, it was replaced with a Pb (Hg)/$PbSO_4$ reference half cell (LSE). Use of the LSE in titrating known quantities of U was satisfactory.

Coulometric determination of Pu(VI) by reduction at a Pt electrode did not proceed satisfactorily in aqueous HCl at room temperature. 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 1$\underline{N}$ HCl at + 0.310V was reduced to Pu(III) in less than 30 min. 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).

3. Analytical Chemistry Quality Assurance Program
(J. W. Dahlby)

Changes were made in the Analytical Chemistry Quality Assurance Plans to increase the assurance that all reported results are completely documented and traceable to certified chemical and physical standards. The Analytical Chemistry Quality Assurance Procedures were revised and new procedures written to comply with the additional requirements in the Quality Assurance Plans. These revised and new procedures were incorporated into our quality assurance system.

All measuring and testing equipment has been labeled and the equipment on a fixed calibration schedule has been calibrated and added to the Quality Assurance Manager's recall system to ensure recalibration as scheduled.

IV. QUALITY ASSURANCE
(L. E. Lanham)

General: An audit conducted by an AEC Audit Team included an evaluation of Project 463 status and plans. The following actions have been taken on the findings of this audit team.

Efforts are being made to develop a list of qualified laboratories to perform tests required by specifications. In continuing the overcheck of cladding tubing, it is being considered that an outside laboratory will be used for ultrasonic inspection. Surveillance will be provided by certified nondestructive testing personnel from LASL.

A letter has been submitted to RRD advising them of the additional program cost of implementing independent QA surveillance and project QA Representative functions.

Actions in process on other findings include revisions to the Quality Assurance Manual, preparation of additional sections, and preparation of supporting documents such as check-lists and forms for corrective action requests from outside organizations.

Fuel Preparation: The work on instrument calibration procedures has continued.

Procedures and traveler documents which will be used in fuel preparation have been prepared and approved.

Fuel Pin Fabrication: Archive and material storage facilities meeting Quality Assurance requirements have been completed and are being used.

A Quality Assurance Plan for the characterization of BMI nitride fuel has been written and approved. Samples for characterization have been prepared by Fuel Preparation and sent to Chemical Analysis. Work instructions or traveler documents for both sample preparation and chemical analysis have been reviewed by the Quality Assurance Manager.

TREAT Irradiation Testing: An addendum has been prepared and approved for the Quality Assurance Plan which will be used for the UL-3 and UL-4 TREAT Tests. This addendum shows the new HEDL numbers for their procedures and lists the three procedures that have been added.

Additional welding samples for work being done at HEDL were fabricated, inspected, and shipped using Quality Assurance procedures, reviews, and documentation.

V. REFERENCES

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2. J. F. Boland, Nuclear Reactor Instrumentation, Gordon and Breach Science Publishers (N.Y.) 1970, p. 200.

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4. R. D. Baker, "Quarterly Progress Report on the Advanced Plutonium Fuels Program, July 1 to September 30, 1972", Los Alamos Scientific Laboratory Report LA-5106-PR (1972).

5. P. G. Bently and R. Rowley, "Comparison of the Frequency Responses and Time Constants of Mineral Insulated Thermocouples in Sodium and Water", TRG Report 1259 (R) (1967).

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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)

After two round robin programs and a consultation visit to three potential producers of boron carbide pellets, analytical measurements for nine FFTF specifications were deemed satisfactory. These nine measurements are (1) total boron, (2) isotopic boron, (3) total carbon, (4) HCl soluble boron, (5) HNO_3 soluble boron, (6) soluble carbon, (7) chloride, (8) fluoride, and (9) metallic impurities. When HEDL added nitride nitrogen as a tenth specification, HEDL and LASL cooperatively prepared a method writeup for nitride nitrogen which was distributed to the three commercial laboratories. A round robin program is now in progress to evaluate this method in use at the three laboratories and at HEDL and LASL.

B. Preparation of Calibration Materials and Quality Control Samples

(J. V. Pena, H. J. Kavanaugh, J. E. Rein)

The preparation of quality assurance materials in amounts deemed adequate for the production of about 150,000 boron carbide pellets has been completed except for nitride nitrogen quality control materials. The completed materials were sent to HEDL with a detailed report that lists the concentration values for all materials and recommended control limits for quality control samples.

Although the planned quality control samples for nitride nitrogen analysis were pellets, a large between-pellet heterogeneity within batches precluded their use for this purpose. Powder blends prepared from batches of pellets have been prepared. Analysis of these blends is in progress to verify homogeneity.

C. Status of RDT Standards

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

Cooperatively with HEDL, RDT Standards F2-8 "Qualification and Control of Analytical Chemistry Laboratories for Control Rod Absorber Material Analysis" and F11-2 "Analytical Chemistry Methods for Control Rod Absorber Material" were prepared. Revisions will be necessary to incorporate information for nitride nitrogen analysis.

D. Studies and Improvements of Analytical Methods

(R. D. Gardner, A. L. Henicksman, W. H. Ashley, R. E. Perrin, A. Zerwekh, G. E. Meadows, C. H. Ward, R. G. Bryant)

1. Determination of Soluble B. Soluble B is expected to exist in B_4C as B_2O_3 and elemental B. These minor components are determined after selective dissolution of B_2O_3 in 0.1M HCl from one sample, and both B_2O_3 and B in 1.6M HNO_3 from a second sample. As the boron carbide materials cover a composition range from about $B_{12}C_3$ to $B_{13}C_2$, the dissolution of B from high boron content carbides (near $B_{13}C_2$) was investigated by making two successive refluxings with 1.6M HNO_3, each for 4 h according to the method. Essentially all of the soluble B was dissolved in the first refluxing, indicating that the method was valid for materials having a wide range of boron-carbon compositions.

The double refluxing experiment described above was repeated using 10% H_2O_2 as the solvent and 2 h as the leaching period. The soluble B was again essentially all dissolved in the first refluxing. Either method seemed to be equally satisfactory for dissolving the soluble B.

2. Determination of Total B. Summations of total boron, and the measured impurities have been consistently less than 100%. In an attempt to determine the cause for the low results, we analyzed the hydrous oxides, which are discarded in the boron procedure, and found only negligible amounts of boron. The probable cause for low results was found to be the iron contamination, which ranged from 0.2 to 0.6%, that was introduced during grinding in a diamond (steel) mortar. Although boron results could be corrected for the interference, if each sample portion were analyzed for Fe, such a correction is not applicable for the carbon determination. The use of a different mortar, possibly a boron carbide mortar, is recommended for grinding the samples.

3. Determination of Soluble C. 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. To avoid further difficulty, the reaction vessel was described explicitly in the RDT standard being prepared.

4. Determination of F^- and Cl^-. The determination of F^- and Cl^- in fuel materials was modified in order to analyze B_4C. In this determination, the halides are separated from B_4C by pyrohydrolysis, and collected in water or aqueous solution in a receiver for measurement using a fluoride-selective electrode or by a spectrophotometric method for chloride. 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. Based on the data obtained, the determination was modified to include pyrohydrolysis at a temperature no lower than 950°C, and

distillation of the halides into saturated H_3BO_3. The saturated H_3BO_3 did not affect the measurement of Cl^-, and it improved reproducibility in measuring F^-.

Although the saturated H_3BO_3 reduces the sensitivity of the F determination, it is still adequate. The recovery of F^- is greater than 90% and the recovery of Cl^- is greater than 85%.

5. Determination of N. A study of the measurement of N in B_4C produced several improvements. In the measurement involving caustic fusion to liberate the N as NH_3 which is titrated, modifications included:

(1) Substitution of a Ag crucible liner for the original Ni liner to eliminate formation of unwanted nitrogen compounds that cause low results.

(2) Substitution of $LiOH \cdot H_2O$ for NaOH as the flux.

(3) Addition of a water bubbler to the gas pretreatment apparatus to supply needed moisture. Following these modifications, five portions of a BN sample were analyzed with a recovery of N of 99.3%, and a relative standard deviation of 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, and the N was separated by distillation as NH_3 which was titrated with standard acid. Seven portions of BN were analyzed by this method and an average nitrogen recovery of 99.3% and a relative standard deviation of 0.14%.

Tests showed that the total N in B_4C can also be determined using a LECO-Nitrox-6 analyzer that is modified to attain a temperature of 2300°C. The B_4C is heated inductively to 2300°C to release the nitrogen which is separated by gas chromatography and measured in a thermal conductivity analyzer. 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%.

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 the PuO_2 supplier (ARHCO) have been qualified for FFTF fuel production analysis. The use of LASL-supplied calibration materials and quality control samples in accordance with the HEDL quality assurance program is under way at these facilities and at the HEDL analytical laboratory. Aid is being provided to the HEDL Quality Assurance organization in identifying difficulties and interpreting technical data. To date, only minor problems have occurred which have been resolved.

B. Calibration Materials and Quality Control Samples

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

Changes were made in late FY 1972 by HEDL in the quality assurance program covering the production of FFTF fuel that doubled the previous estimated quantities of these materials. In accordance with the new schedule, a one-quarter shipment of materials has been made to the four facilities in the program. An additional quantity of materials has been packaged for another quarter shipment. The quantities of materials prepared have depleted the supply of mixed oxide and plutonium dioxide matrix materials that are used for the preparation of powder blends. The preparation of further blends will be resumed immediately after receipt of satisfactory material from HEDL.

Included in the materials supplied was a series of mixed oxide pellets with substoichiometric O/M ratios to be used as quality control samples for the O/M ratio measurement. These pellets were individually sealed in glass ampoules at a low air pressure such that O/M upward drift will be insignificant.

C. Status of RDT Standards

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

Cooperatively with HEDL, RDT Standards F2-6 "Qualification and Control of Analytical Chemistry Laboratories for Mixed Oxide Fuel Analysis" and F 11-1 "Analytical Chemistry Methods for Mixed Oxide Fuel" were prepared.

D. Development of Analytical Methods

1. Determination of Burnup

a. Study of Chemical Separation Procedure for Burnup Determination by Mass Spectrometry

(S. F. Marsh, M. R. Ortiz, J. E. Rein, R. M. Abernathey)

Chemical separation to obtain fractions of Pu, U, and Nd for mass spectrometric analysis requires two sequential ion exchange columns. The first column contains Pu and U from 12M HCl while most fission products pass through. The second column, which chromatographically separates Nd from other fission products, including the other rare earths, uses a mixed solvent of methanol-HNO_3. To ensure the correct elutriant mixture by minimizing evaporation of methanol, the elutriant was metered out in small increments during the 4.5-h elution period. The apparatus was modified to provide an automated continuous flow of the elutriant from an inverted plastic bottle thereby maintaining a constant liquid head in the column reservoir, and, hence, a constant flow rate. Evaporation of methanol from the elutriant is prevented by operating the columns in an enclosure having a methanol-saturated atmosphere. The 4.5-h of automatic operation frees the analyst for other work.

Some reported difficulties with various portions of the ion exchange separation prompted recent investigations that revealed significant differences between batches of ion-exchange resin of identical type, cross-linkage, mesh-size designation, and manufacturer. Microscopic examination showed significant discrepancies in mesh-size range and distribution within the range. These mesh-size differences seriously affect flow rate (and elution characteristics). However, the between-batch differences in the resin performance seem much larger than resin particle size alone would explain. It is, therefore, necessary that each new batch of resin be characterized with suitable radionuclide tracers to verify that the separations occur as anticipated. Recent experience has shown that substantial modifications to the procedure can be required. As an example, no presently available batch of the AG1-X2 (200-400 mesh) resin originally used provides a usable Nd fraction when the established procedure is followed. It has been necessary to change both the resin, from AG1-X2 to AGMP-1 (macroporous), and the elutriant, from 90% methanol-0.0078M HNO_3 to 80% methanol-0.094M HNO_3, in order to obtain a well-separated neodymium fraction in a reasonable time. Based on this experience it is recommended that a large supply of any satisfactory resin be obtained and reserved for burnup separations.

b. Modification of Chemical Separation Procedure for Low Burnup Samples

(M. R. Ortiz, R. M. Abernathey, J. E. Rein)

The separation described above was designed for fuel samples having undergone at least 0.1% burnup. For analyzing samples having lower burnup, two modifications were required: (1) Two sample aliquots were used, one containing a necessary amount of fission product Nd for mass spectrometric analysis with one spike, and the other being a suitable weight dilution of the sample for the measurement of U and Pu using another spike. (2) A "pellicular" cation-exchange resin column was used between the two usual columns to obtain a hundred-fold reduction in the Am content of the large aliquot taken for Nd measurement. This aliquot contained more Am than could be removed by the usual two column separation, and the carried-through Am constituted a health hazard and caused beam instability in the mass spectrometric determination of Nd. With these modifications, burnups less than 0.01 at.% have been successfully analyzed.

c. Development of Burnup Method Using Conventional Low-Cost Apparatus

(S. F. Marsh, M. R. Ortiz, J. E. Rein)

Mass spectrometry is the most reliable technique for determining nuclear fuel burnup using the triple spike technique. Laboratories with limited mass spectrometer availability need a chemical burnup method which requires only conventional, low-cost equipment. Such a method might measure a selected fission product or group of fission products as the fission monitor. A proposed scheme is a chemical separation of the selected fission monitor, U, and Pu from a dissolved fuel sample followed by chemical measurements of each of the three components by a conventional technique such as spectrophotometry.

The fission products that are most useful as fission monitors for FBR fuels and that can be determined

by conventional chemical techniques are total rare earths and elemental Zr. Of these, total rare earths are preferable because their yield from fast fission is higher and more constant for different fissile nuclides, and no evidence has been reported to indicate that they migrate in FBR oxide fuels.

A promising separation scheme being investigated is retention of U and Pu on an anion-exchange column from 12M HCl while the rare earths and most fission products pass through. The rare earths then are retained on a cation-exchange column from anethanol-HCl mixture while transplutonium actinides and extraneous fission products pass through. The Pu and U are sequentially eluted from the first column with a 0.1M HI-12M HCl mixture and 0.1M HCl, respectively. The total rare earths are eluted from the second column with 5M HCl.

Of greater difficulty is separation of the rare earths from the trivalent actinides. This separation is needed to reduce the radiological hazard and to avoid the interference in the measurement from the chemically similar trivalent actinides.

Preliminary experiments of the cation-exchange behavior of Eu (typical of rare earths) and Am (typical of trivalent actinides) in ethanol-HCl mixtures indicated that large separation factors were attainable. Unfortunately, the equilibrium rates in these mixtures were much slower than for aqueous media. Macroporous cation-exchange resin was investigated as a means to obtain more rapid equilibrium between the ions in solution and the resin in ethanol-HCl media. Improved kinetics were obtained, but equilibrium times were still slower than desired for column operation. Column elution peaks were broad with considerable tailing, and incomplete recoveries were experienced.

A third type of ion exchange resin, "Pellicular," was investigated. These resin beads, consisting of spherical silica beads of 40-μ diam, coated with a 2-μ layer of resin, should greatly reduce the long equilibrium times attributed to diffusion of ions to the interior of conventional ion-exchange resin beads.

Separations on 2-cm-long by 0.6-cm-diam columns of pellicular cation-exchange resin have provided > 99% recovery of Eu with less than 1% of the initial Am. The separation of other fission products also looks promising, based on gamma spectrometry of mixed fission product samples.

As the neodymium separation aspect of the problem nears satisfactory completion, the adequacy of the U and Pu separation will be verified. Attention then will be given to the development of chemical methods for the determinations of the separated fractions of total rare earths, U, and Pu. Preliminary experiments indicate that all three can be determined by spectrophotometry using Arsenazo III as the chromogenic agent.

2. The Determination of O/M Ratio in Solid Solution $(U,Pu)O_2$
(G. C. Swanson and G. R. Waterbury)

The most used methods of determining O/M on unirradiated fuels are thermogravimetric: the fuel is weighed, subjected to conditions to produce stoichiometry, reweighed, and the initial O/M is calculated from the change in weight. The method used at LASL is calibrated with UO_2 and PuO_2 prepared by the careful oxidation of highly pure U and Pu metals. Mechanical mixtures of the reference UO_2 and PuO_2 are used for the calibration, but such mechanical mixtures probably do not display the same chemical properties as a true solid-solution, sintered material. The goal of the present effort is to provide method calibration applicable to solid solution $(U,Pu)O_2$, both from a theoretical study of the redox behavior of the solid solution oxide and by preparation of appropriate solid solution "standard" materials.

Equipment was assembled and tested for studying the thermogravimetric measurement of O/M. This included a Mettler thermogravimetric analyzer suitable for analysis of α-radioactive materials, a digital data-logging system to provide a punched tape data output from the thermobalance for computer processing, and a gas manifold in which the oxygen potential is controlled by coulometric generation of O_2 gas in a high-temperature, solid-state, electrochemical cell. This manifold allows control of a dynamically varying oxygen potential which is required for the theoretical study of solid solution $(U,Pu)O_2$ stoichiometry.

Preliminary data on the O/M of UO_2 vs gaseous oxygen potential agree well with the data of Markin and Bones,[1] but the calibration of the gaseous oxygen potentials generated is not sufficient for the more extensive studies planned. Several prototype oxygen concentration electrochemical cells have been fabricated for measurement of the oxygen potentials of gases in equilibrium with the fuel samples. These cells employ a solid electrolyte to measure the gaseous oxygen potential on the cell exterior relative to a reference oxygen potential developed by a mixture of a metal and its oxide on the cell interior. Such measurements will not only confirm the data for the theoretical interpretation of the solid solution $(U,Pu)O_2$ stoichiometry, but may also be applicable to determination of oxygen potentials of irradiated fuel samples. None of the prototypes has displayed a theoretical response. Methods of calibration for the non-theoretical response are under development; electrodes having different configurations are being considered. Satisfactory methods are not now available for determination of O/M on irradiated fuels, and none of the several proposed methods to determine oxygen potentials of irradiated fuels is in general usage. Extension of the present studies to determination of O/M and oxygen potentials of irradiated fuel samples is planned.

IV. QUALITY ASSURANCE

A finding of the AEC Audit Team was that the Project 472 preparation of analytical chemistry standards for the vendor procurement program should be under a QA plan meeting the requirements of RDT Standard F2-2T.

A plan is being prepared and QA procedures are being written. Calibration materials and quality control samples are being shipped using procedures described in the CMB-RDT Quality Assurance Manual.

V. REFERENCES

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