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Quarterly Report

# Advanced Plutonium Fuels Program

July 1 through September 30, 1973

LOS ALAMOS NATIONAL LABORATORY  
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**Los Alamos**  
**scientific laboratory**  
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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-5106-PR  
LA-5193-PR

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Quarterly Report

# Advanced Plutonium Fuels Program

July 1 through September 30, 1973

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 28th quarterly report on the Advanced Plutonium Fuels Program at the Los Alamos Scientific Laboratory.

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

## PROJECT 401

### EXAMINATION OF FAST REACTOR FUELS

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

#### I. INTRODUCTION

This project is directed toward the examination and comparison of the effects of neutron irradiation on LMFBR Program fuel materials. Unirradiated and irradiated materials will be examined as requested by the Fuels and Materials Branch of 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, F. J. Fitzgibbon, M. E. Lazarus, P. A. Mason, F. H. Newbury, W. T. Wood)

##### 1. Electro-Optical Profilometer

A new thermocouple assembly is being installed on the profilometer. This will allow a temperature profile to be obtained from any fuel element within the capacity of the profilometer. The earlier thermocouple could only be used on smooth surfaces where there were no undercuts or protrusions on the surface.

Many of the fuel elements we are now receiving are badly bowed. In a few cases the bow was great enough to disengage the profilometer centering assembly during the profiling operation. Several design modifications are now being made to the profilometer positioning stand which will allow the ends of the fuel element to "float."

##### 2. Fission Gas Sampling System

In order to improve the accuracy of the void volume measurements obtained from fuel pins, a new drilling system was designed for pins up to 0.375 in. in diameter. The improved device utilizes a smaller chamber to house the drill. The preceding unit had the flexibility of puncturing capsules or pins from 0.125 to 1.25 in. in diameter, and thus the drill housing had to be large. The new device has been used successfully on several fuel pins.

##### 3. Pulsed Eddy Current Scanner

An indexing mechanism has been incorporated into

the Pulsed Eddy Current Scanner to permit the rotational orientation of fuel pins.

#### 4. System for Obtaining Weight and Density of Fuel Pin

A system has been partially installed to measure in air or in a suitable fluid the weights of irradiated fuel pins up to 61 in. in length in a vertical position. Using the weights in air and in a fluid, the density of irradiated fuel pins will then be determined. Dry weights have been taken of a dummy fuel pin, and a procedure is presently being written. A method for taking weights of submerged pins will be developed when the plumbing for filling and draining the system has been completed.

The system consists of a 1000-g analytical balance, with a sensitivity of 0.1 mg, mounted on a cell roof plug directly above a small hole through the plug. A piano wire is attached to the balance and hangs into the operating cell. The fuel pins are weighed by attaching them to a holding device on the end of the piano wire and suspending them into a 6-in.-diameter tube installed in the cell. The tube will be filled with a suitable liquid for obtaining fuel pin weights during immersion.

#### 5. Miscellaneous Equipment

Modifications were designed for the swinging arm mechanism of the 7-in. Transfer Systems to prevent disengagement of the drive gears. The necessary new hardware has been fabricated for the five units in operation. The modifications have been installed on the Disassembly Cell unit. The remaining units will be completed when personnel access is permitted.

A collapsible holder for photographing full length breached pins in the disassembly cell was designed and fabricated.

The Veeco MS-90 Helium-Leak Detectors used with the Fission Gas Sampling Systems have been modified to insure all radioactive gases vented by the detector vacuum pumps are ducted to the cell venting system.

#### B. Inert Atmosphere Systems

(R. W. Basinger, P. A. Mason)

A new assembly for sealing the interior of the through-tube of the AMF Manipulators has been developed with considerably improved sealing characteristics ( $> 10$  times  $\Delta P$  for given boot purge rate) over the previous

design. The seal assemblies have been installed in the manipulators of the Disassembly Cell and in two additional spare manipulators. A total of ten manipulators is scheduled to be modified for use in all of the inert atmosphere cells.

A special assembly has been designed for sealing the exterior of the through-tube of the AMF Manipulators to the alpha box penetration ports. Fabrication and testing are presently in progress.

#### C. Processing of Data from the Electro-Optical Profilometer

(M. E. Lazarus, T. R. Wilson)

Two computer programs are now available for processing profilometry data. The first provides trace outputs on 35-mm film and on Calcomp graph paper, and a magnetic tape output of corrected data. It also provides a printout of data averaged over any specified length of fuel element together with the standard deviation of the numbers averaged. The second program was developed specifically for HEDL. It utilizes the magnetic tape generated by the first computer program to perform the following operations:

- a) Averages  $0^\circ$ ,  $45^\circ$ ,  $90^\circ$ ,  $135^\circ$ , and  $180^\circ$  outputs.
- b) Subtracts the  $180^\circ$  run from the  $0^\circ$  run (this gives an indication of instrument drift and of errors introduced in the data by element bow in the  $0^\circ - 180^\circ$  plane).
- c) Prints the maximum at each 0.1-in. length average.
- d) Prints the minimum at each 0.1-in. length average.
- e) Subtracts the minimum from the maximum.
- f) Subtracts a preirradiated constant, provided by Experimenter, from the average.
- g) Obtains percent change of (f).
- h) Subtracts a postirradiated plenum average from the average.
- i) Obtains a percent change of (h).

The above data processing was requested by HEDL together with Calcomp plots of  $0^\circ$ ,  $45^\circ$ ,  $90^\circ$ ,  $135^\circ$ , and  $180^\circ$ . The Calcomp plots were obtained by modifying the first computer code.

Further changes will be made, as time allows, to permit accurately referencing the fuel element length positions to either the bottom of the fuel element or the fuel element location slot in the bottom end plug. The bottom of the fuel element is a better reference point than the top since the bottom is fixed with respect to the reactor core and the top position will vary with fuel element expansion.

#### D. Shipping Casks

(F. J. Fitzgibbon, J. W. Schulte)

The T-2 casks have been successfully unloaded several times at LASL using special fixtures to aid in removing the inserts.

One of the two LASL vertical casks having a capacity for handling 19 irradiated EBR-II pins (40 in. long) is being modified. The minor changes, as discussed previously, will permit handling at the HFEF north and south facilities. The first unit will be completed by November 1, 1973; it will be scheduled to return an irradiation experiment from EBR-II to LASL.

The small (2000 lb) "Analytical Sample" cask was used during this report period for shipping an irradiated fuel pellet to HEDL for burnup analysis.

### III. ANALYTICAL CHEMISTRY

#### A. Gamma Scanning

(J. R. Phillips, T. K. Marshall,  
J. R. Netuschil, J. N. Quintana)

The axial burnup profiles of six low burnup (< 1%) (U,Pu)O<sub>2</sub> fuel pins were determined by measuring non-destructively the relative axial isotopic distributions of <sup>95</sup>Zr (724 and 756 keV) and <sup>95</sup>Nb (765 keV). The <sup>95</sup>Zr and <sup>95</sup>Nb isotopes were selected because of their relative gamma-ray activities, uncomplicated gamma-ray spectra, and tendency to remain with the fuel material. The two isotopes did not migrate axially as has been noted before under certain circumstances.<sup>1</sup> The fuel pins were recently discharged from the EBR-II reactor; therefore the relatively short half-lives of the two isotopes (<sup>95</sup>Zr, 65.5d; <sup>95</sup>Nb, 35.1d) did not adversely affect the calculated burnup profiles.

A nonlinear least squares fitting routine<sup>2</sup> was used to calculate the smoothed <sup>95</sup>Zr and <sup>95</sup>Nb axial isotopic distributions. The calculated axial burnup profile of a

(U,Pu)O<sub>2</sub> fuel region (Fig. 401-1) is superimposed on the <sup>95</sup>Nb axial distribution. Several levels of numerical filters were applied to determine the effect of the interpellet gaps, indicated by the periodic dips in the activity, upon the resulting smoothed functions. The inclusion of the interpellet gap values did not alter substantially the computed burnup profiles of the six fuel pins.

The axial burnup profile shown in Fig. 401-1 illustrates the effect of the axial reflector in EBR-II upon the axial neutron flux profile for this particular fuel pin. Five burnup samples have been excised at specified axial locations. These samples will be analyzed mass spectrometrically for comparison with the gamma scanning results.

The principal data processing computer codes for the presentation of gross gamma and isotopic distributions are being converted to the International System (SI) of Units.

#### B. Determination of Fission Gases Retained in Irradiated Fuels

(R. M. Abernathy, J. W. Dahlby,  
R. R. Geoffrion)

Measurement of the quantity of gaseous fission products in the fuel pin plenum relative to that trapped or retained in the (U,Pu)O<sub>2</sub> fuel matrix is necessary in studies of the gas-release mechanisms. The measurement of the gaseous fission products in the fuel pin is routinely done at LASL in postirradiation examinations of fuel pins. Specialized equipment was assembled to collect and measure the retained fission gases in irradiated (U,Pu)O<sub>2</sub>

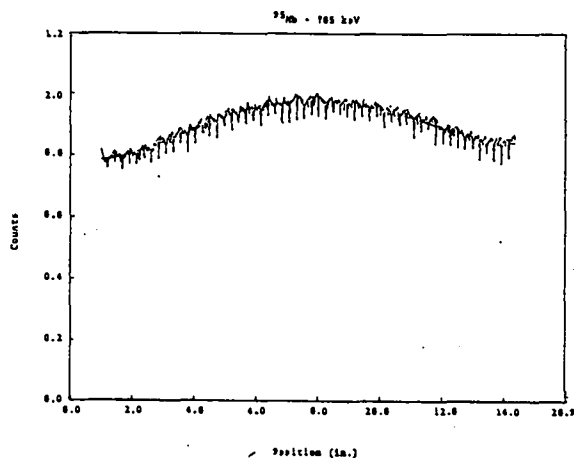


Fig. 401-1. The calculated axial burnup profile of a (U,Pu)O<sub>2</sub> fuel region superimposed on the <sup>95</sup>Nb axial distribution.



fuel materials. The total quantities and isotopic compositions were determined by isotope dilution mass spectrometry.

In this determination, the (U,Pu)O<sub>2</sub> fuel sample is dissolved remotely in a HNO<sub>3</sub>-HF acid mixture to release the retained fission product gases from the fuel. Known amounts of Kr and Xe enriched in nonfission product isotopes are added to the gases which are carried by He out of the hot cell through a purification system that removes acid fumes, H<sub>2</sub>O, N<sub>2</sub>, O<sub>2</sub>, and CO<sub>2</sub>. A silica gel trap maintained at 77° K is used to collect the fission product gases. This trap has advantages as compared to the conventional molecular sieve trap of quantitative recovery and fast desorption. The desorbed gases are purified using a gas chromatograph with a charcoal column, and the Kr and Xe fractions are analyzed mass spectrometrically for total quantities and isotopic distributions.

The gas collection system was tested by injecting known quantities of natural Kr and Xe, then determining the recovery factor. Repeated determinations showed the average recovery was 95%. In the isotope dilution technique, losses up to 10% are considered insignificant because the spike and sample which are collected and measured concurrently have essentially the same recovery factor.

Five 2-g cross-sectional samples of (U,Pu)O<sub>2</sub> fuel with < 1% burnup were excised at specified axial locations along the fuel column and were analyzed using this method. An interesting finding was the different isotopic distributions for the retained fission gases relative to the gaseous fission products in the pin plenum. The <sup>83</sup>Kr, <sup>131</sup>Xe, and <sup>132</sup>Xe isotopes were higher in abundance in the retained gases. The precursors of the Kr and Xe isotopes in these chains have the longest half-lives and tend to preferentially remain in the fuel matrix.

#### IV. MICROSTRUCTURAL ANALYSIS

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

A photo darkroom trailer has been acquired and will be installed. It will provide an autoprocessor for negatives and films. This unit when installed will help us shift more photographic work to roll and cut film

away from the limitations of instant type film-prints in many applications.

The results of the grinding experiments and developments have been applied to the hot cell metallographic grinding with a significant increase in throughput rate at this step.

Ion gun etching development has continued this quarter with the solution to the small etched spot problem being achieved through a combination of a minor gun design change and somewhat different operating conditions.

#### V. REQUESTS FROM DRRD

##### A. Examination of Irradiated Materials

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

During the First Quarter of FY 1974 seventeen irradiated capsules were received. The distribution is as follows: GE-5; HEDL-10; and LASL-2.

General Electric Company: Examinations performed on twenty-six irradiated fuel capsules received on March 21, 1973, April 7, 1973, and September 1, 1973, are listed in Table 401-I.

Hanford Engineering Development Laboratory: Examinations performed on eighteen irradiated fuel capsules received on February 1, 1973, April 7, 1973, and August 31, 1973, are listed in Table 401-II.

Dried fuel solutions from four pins and one 6.4-mm cross-section sample from another fuel pin were sent to HEDL as part of a burnup correlation study.

Seventeen irradiated fuel pins were shipped to HEDL following nondestructive (and some destructive) testing at LASL.

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 Fuel Program.

1. BMI Experiments -- Examinations on one BMI irradiated fuel capsule received on February 16, 1973, are shown in Table 401-III,

2. LASL Experiments -- Examinations performed on three LASL irradiated fuel capsules received on October 11, 1973, and September 6, 1973, are listed in Table 401-IV.

TABLE 401-I

POSTIRRADIATION EXAMINATIONS OF CAPSULES  
AND PINS FROM GE

<u>Examination</u>	No. of Capsules	No. of Pins
1. Visual Inspection	4	5
2. Preliminary Measurements	14	3
3. Profilometry, Optical	22	4
4. Radiography	13	1
5. Gamma Scan <sup>a</sup>	24	--
6. Gas Sampling and Analysis	8	2
7. Na Removal	4	--
8. Clad Removal	4	--
9. Profilometry, (Capsule Cladding)	4	--
10. Photography, Full Length	5	5
11. Photography, Maximum Bow	5	3
12. Photography, Incremental	5	5
13. Wire Wrap Removal	3	5
14. Photography, Incremental w/o Wire	--	1
15. Eddy Current	--	3
16. Length Measurement	--	3
17. Sectioning	--	2
18. Photography (Sectioned Faces)	--	2

<sup>a</sup> Ninety-six gross gamma scans, 24 complete spectral scans, and 188 distributions of isotopes were calculated.

3. WARD Experiments -- Examinations performed on two WARD irradiated fuel capsules received on February 16, 1973, are shown in Table 401-V.

4. Gulf United Experiments -- Examinations performed are shown in Table 401-VI.

## VI. QUALITY ASSURANCE

(L. E. Lanham)

General: The CMB-RRD Quality Assurance Manual has been revised to incorporate changes required to comply with the findings of the AEC audit team. Audit checklists and supporting documents for corrective action procedures have been prepared.

The quality assurance organization has reviewed and commented on changes in plans and procedures

TABLE 401-II

POSTIRRADIATION EXAMINATIONS OF CAPSULES  
AND PINS FROM HEDL

<u>Examination</u>	No. of Capsules	No. of Pins
1. Visual Examination	1	10
2. Preliminary Measurements	--	10
3. Profilometry, Optical	--	2
4. Radiography	8 <sup>a</sup>	10
5. Gamma Scan <sup>b</sup>	10	--
6. Photography, Full Length	9	10
7. Photography, Maximum Bow	9	10
8. Photography, Incremental	9	10
9. Gas Sampling and Analysis	1	--
10. Na Removal	1	--
11. Clad Removal	1	--
12. Wire Wrap Removal	--	2
13. Eddy Current	--	2
14. Sectioning	--	6
15. Na Melting and Pressurizing Tests	--	1
16. Electron Microprobe	--	3 (8 Samples)
17. Burnup	--	4 (4 samples)
18. Microstructural Analysis Optical Microscopy <sup>c</sup>	--	11 (22 Longit. Samples) (16 Trans. Samples)
Preparation for EMX		4 (9 samples)

<sup>a</sup> GETR Capsule Assembly is included.

<sup>b</sup> Gross gamma scans, 10 multispectral scans, and 68 distributions of isotopes were calculated. Also, axial burnup profiles over the enriched fuel columns of six capsules were determined from gamma scan data.

<sup>c</sup> 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 experimenter's fuel pins were examined in like manner.

TABLE 401-III

POSTIRRADIATION EXAMINATIONS OF BMI  
CAPSULES AND PINS

<u>Examination</u>	No. of Capsules	No. of Pins
1. Profilometry, Optical	1	--
2. Photography, Full Length	--	1
3. Photography, Incremental	--	1
4. Gas Sampling and Analysis	1	1
5. Na Removal	1	--
6. Clad Removal	1	--
7. Profilometry, Mechanical	--	1
8. Eddy Current	--	1
9. Sectioning	--	1
10. Electron Microprobe	--	1 (1 Sample)
11. Microstructural Analysis		
Optical Microscopy	--	2 (6 Samples)
Preparation for EMX	--	1 (1 Sample)

TABLE 401-IV

POSTIRRADIATION EXAMINATIONS OF LASL  
PINS AND CAPSULES

<u>Examination</u>	No. of Capsules	No. of Pins
1. Gas Sampling and Analysis	1	1
2. Photography, Full Length	--	1
3. Photography, Incremental	--	1
4. Radiography	2 <sup>a</sup>	--
5. Na Removal	1	--
6. Clad Removal	1	--
7. Na Melting and Pressurizing Tests	--	1
8. Eddy Current	--	1
9. Profilometry, Mechanical	--	1
10. Electron Microprobe	--	1
11. Microstructural Analysis		
Optical Microscopy	--	1 (5 Samples)
Preparation for EMX	--	1 (1 Sample)

<sup>a</sup>TREAT Capsule Assemblies.

TABLE 401-V

## POSTIRRADIATION OF WARD PINS AND CAPSULES

<u>Examination</u>	No. of Pins
1. Photography, Full Length	1
2. Photography, Incremental	1
3. Photography, Full Length w/o Wire Wrap	1
4. Photography, Maximum Bow w/o Wire	1
5. Photography, Incremental, w/o Wire	1
6. Gas Sampling and Analysis	1
7. Sectioning	1

TABLE 401-VI

POSTIRRADIATION EXAMINATIONS  
OF GU EXPERIMENTS

<u>Examination</u>	No. of Pins
1. Electron Microprobe	2 (2 Samples)
2. Burnup	4 (4 Samples)
3. Microstructural Analysis	
Optical Microscopy	1 (1 Sample)
Preparation for EMX	1 (1 Sample)

required as a result of the findings of the AEC audit team and the additional requirements of the latest issue of RDT Standard F2-2. Procurement packages have been reviewed and surveillance has been performed to determine compliance to shipping requirements and procedures.

**Microstructural Analysis:** An audit of Microstructural Analysis was conducted by the Quality Assurance Manager, and an audit report has been prepared.

A position description for a Quality Assurance Representative has been made a part of the Quality Assurance Plan and member of the Microstructural Analysis Section has been designated as the Quality Assurance Representative. The requirements of the latest issue of RDT Standard F2-2 have been included in the Quality Assurance Plan.

**Chemical Analysis:** An audit was conducted by the Quality Assurance Manager of all Chemical Analysis operations and a report has been prepared.

All procedures are being reviewed and will be revised as required. The numbering system for procedures has been changed and a common Chemical Analysis designation will be used. This will eliminate the need for duplication when a procedure is used for more than one project. The Chemical Analysis Quality Assurance Plan has been revised to meet the requirements of the current issue of RDT Standard F2-2.

Hot Cell Examination: An audit was conducted by the Quality Assurance Manager of the Hot Cell operations and a report has been prepared. Corrective actions are being taken on the findings of this audit.

The Hot Cell Quality Assurance Plan has been revised to include the requirements of the current issue of RDT Standard F2-2. A position description of a Quality Assurance Representative has been included to meet one of the findings of the AEC audit team. The traveler is being reviewed and will be revised to include the new numbers for the Chemical Analysis procedures, and a more visible method of documenting the independent over-check of inspection data.

## VII. REFERENCES

1. R. D. Baker, "Quarterly Report on Advanced Plutonium Fuels Program, April 1 to June 30, 1973, and Seventh Annual Report, FY 1973," Los Alamos Scientific Laboratory report LA-5390-PR, p. 5, (1973).
2. R. H. Moore and R. K. Zeigler, "Solution of General Least Squares Problem with Special Reference to High-Speed Computers," Los Alamos Scientific Laboratory report LA-2367 (1959).

## VIII. PUBLICATIONS

1. C. S. MacDougall, T. K. Marshall, G. M. Matlack, and G. R. Waterbury, "Determination of Oxygen, Hydrogen, and Tritium in Irradiated Reactor Fuels and Cladding Materials," to be presented at the 17th Conference on Analytical Chemistry in Nuclear Technology, Gatlinburg, TN, October 23-25, 1973.
2. T. K. Marshall, J. R. Phillips, B. K. Barnes, and G. R. Waterbury, "New Techniques in Two-Dimensional Gamma Scanning," to be presented at the 17th Conference on Analytical Chemistry in Nuclear Technology, Gatlinburg, TN, October 23-25, 1973.
3. J. W. Dahlby, T. K. Marshall, G. R. Waterbury, G. C. Swanson, "Measurement of Oxygen-to-Metal Atom Ratios in Uranium and Plutonium Oxides," Los Alamos Scientific Laboratory report LA-5329, (Aug. 1973).

## PROJECT 463

### HIGH PERFORMANCE LMFBR FUEL MATERIALS

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

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#### 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 considered which would 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) vapor pressure using mass spectrometry, (5) heat content using drop calorimetry, (6) elastic properties using sonic modulus measurements, (7) hot hardness and its temperature dependence, (8) structure and phase relationships using high temperature x-ray and neutron diffraction, (9) thermal expansion, and (10) compressive creep rate 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. Fuel Synthesis and Fabrication

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

##### 1. Carbide Fuel Production

A special 250-g batch of  $U_{0.8}Pu_{0.2}C$  was prepared as a critical test of the carbide synthesis procedures and equipment relative to the requirements of the RRD Quality Assurance Program. The deficiencies discovered were principally related to the procedures used to document the preparation. The problem areas were eliminated with no alteration to the technical procedures. Fuel specifications have not been finalized for the LASL

Series K-4 irradiation experiments, but the product from this preparation will be used if possible.

## 2. Carbide Grain Growth Studies

Grain size may be one of the factors influencing the behavior of reactor fuels during irradiation. If grain size is to be investigated as an experimental parameter, a method must be developed to produce various controlled grain sizes in  $U_{0.8}Pu_{0.2}C$ .

A series of experiments has been started to determine the effectiveness of additional sintering to increase grain size. The starting material for this series was a batch of  $U_{0.8}Pu_{0.2}C$  pellets which were originally sintered 8 h at  $1800^{\circ}C$  and annealed 4 h at  $1400^{\circ}C$ . Small batches of these pellets were thermally cycled. The densities of the pellets were then measured and the microstructures were examined. Average grain sizes of the original and the thermally cycled pellets were estimated by use of the comparison procedure described in ASTM Procedure E-112. The thermal cycles used and the grain size and density of the initial and product pellets are listed in Table 463-I.

TABLE 463-I

### EVALUATION OF THE EFFECT OF ADDITIONAL SINTERING OF $U_{0.8}Pu_{0.2}C$

Expt. No.	Additional Thermal Cycle	Estimated Grain Size, $\mu m$	Density % T.D.
0	None	36	93.1
1	0.1 h at $2100^{\circ}C$	37	93.1
1a	0.1 h at $2100^{\circ}C$ + 16 h at $1400^{\circ}C$	41	93.0
2	0.5 h at $2000^{\circ}C$	38	93.2
2a	0.5 h at $2000^{\circ}C$ + 16 h at $1400^{\circ}C$	34	92.4
3	2 h at $1900^{\circ}C$	32	92.6
3a	2 h at $1900^{\circ}C$ + 16 h at $1400^{\circ}C$	38	92.5
4	Cycled $1400-1800^{\circ}C$ , 9 cycles	37	93.3
5	24 h at $1600^{\circ}C$	-	93.6
6	24 h at $1600^{\circ}C$	-	93.3

The results shown in Table 463-I indicate that little or no change of either grain size or density was produced by the thermal cycling in this series of experiments. The precision of this method of grain size estimation is approximately  $\pm 5 \mu m$ , and none of the grain size changes observed are greater than this. Similarly, it does not appear that significant changes in density have occurred. Grain sizes produced in experiments 5 and 6 will be measured in the near future.

## 3. Nitride Fuel Development

The primary aim of experiments in this effort is the development of a method of production of high purity, high density, single phase  $U_{0.8}Pu_{0.2}N$  fuel pellets on a 250-g scale. Also of importance is the development of a method of production of UN insulator pellets of the same high quality. Equipment to synthesize, grind, press, and sample nitrides has been set up in an inert atmosphere glovebox, and the sintering furnace has been put into operation in another inert atmosphere glovebox. Exploratory experiments performed in this equipment were made to produce UN pellets.

Two batches of UN were prepared on the 100-g scale by reaction of chunks of U metal with  $N_2$  in the temperature range  $800-1065^{\circ}C$ . This reaction was found to be slow. Consequently, two additional batches of UN were prepared by first breaking down the chunks of uranium to a fine powder by forming the hydride in the temperature range  $25-300^{\circ}C$ , and decomposing it by heating above  $400^{\circ}C$  in vacuum. This reaction is rapid (after a variable and somewhat unpredictable induction period), as is the reaction between  $N_2$  and the metal powder produced. The optimum temperature range of nitride formation appears to be  $400-600^{\circ}C$ . This two-step process requires much less time for completion than did the direct reaction between  $N_2$  and chunks of metal. Weight changes indicated that the product of this process was  $U_2N_3$ .

The  $U_2N_3$  was pressed into pellets and heated in vacuum to the temperature range  $1350-1500^{\circ}C$  to convert it to UN. Continued heating of some of these pellets through a  $2100^{\circ}C$  sintering cycle produced pellets having densities in the range 77-85% of theoretical, and having widely variable diameters. For the remaining material,

the process included grinding and pressing. The UN produced in the vacuum decomposition was ground 3-5 min in an oscillatory mill, and the resulting powder was pressed into pellets which were sintered 16-18 h at 2100°C under ~ 78 KPa N<sub>2</sub>. The pellets were cooled to ~1500°C under N<sub>2</sub>, then in vacuum to room temperature.

Although chemical characterization of the pellets produced is incomplete, the available information is listed in Table 463-II. The high oxygen concentrations found in these pellets is attributed to reaction during transfer of green and sintered pellets in air. The glovebox atmosphere in which the sintering furnace is located will be changed to Ar in the near future, so exposure to air will be avoided, and lower oxygen concentrations can be expected.

Metallographic examination showed no detectable second phase was present, and the porosity observed is in qualitative agreement with measured immersion densities.

#### 4. Equipment Development

The original gas handling system for the hydride-nitride furnace was cumbersome and could not readily be modified to meet new safety standards. A new manifold was designed, fabricated, and installed and proved to be superior to the original installation. On completion and approval of a standard operating procedure, this system was used in the previously described nitride development work. After a brief period of operation, the drive motor on the new centrifugal mill used to grind nitrides

failed to function properly. Special silver-doped graphite brushes designed for use in an inert atmosphere were ordered and installed to correct the problem. Preliminary observations suggest that considerable time may be saved by using the centrifugal mill rather than the older grinding equipment. Data are not yet available on potential contamination of the powder by the mill components.

The nitride sintering furnace was tested at temperature according to recommended procedures using a purified nitrogen atmosphere in the chamber. Furnace and glovebox windows were calibrated for temperature corrections using an optical pyrometer comparator apparatus. The inert recirculating atmosphere glovebox containing this furnace was sealed and, as soon as minor piping alterations are complete, will be tested and placed in service.

All measuring and test equipment was brought into the Q.A. recall system by completing calibrations and calibration procedures. Preventative maintenance activities and routine replacement of components due to normal attrition were continued. The fuel synthesis and fabrication operations were audited by the LASL Quality Assurance Manager. All suggestions and recommended corrective actions stemming from the audits were accomplished.

#### B. Fuel Element Fabrication

(K. W. R. Johnson, D. G. Clifton, H. E. Strohm, L. L. Marriott)

##### 1. Programmable Welder

Future fuel element welds will be made using a programmable welder for a power supply. A programmable welder has been installed and all associated data gathering devices have been calibrated using standards traceable to the U. S. Bureau of Standards. A new gas manifold to control the introduction of spectroscopic grade Ar to the welding head was fabricated and leak tested. Gas lines supplying the manifold were modified to permit evacuation and flushing. As a precursory operation to developing a welding procedure for the new system, two capsules containing archival fuel pellets were hand welded closed using a manual power supply. Data were gathered during the operation to provide a point of departure for the development of the programmed welding procedure.

TABLE 463-II

#### RESULTS OF PARTIAL CHARACTERIZATION OF UN PELLETS

Batch	Oxygen Concentration in Product, ppm	No. of Phases	Density % of T.D. <sup>a</sup>
3	550	1	92
4	650	1	94

<sup>a</sup>Based on T.D. = 14.32 Mg·m<sup>-3</sup>.

## 2. Grinding Facility

In the event that it becomes necessary to provide fuel pellets with extremely small diametral tolerances, a facility for grinding pellets is being installed. A centerless grinder has been fabricated and tested and will be installed in a vacuum-inertable recirculating glovebox. The glovebox has been located at the fuel element fabrication facility. During this quarter the pumping system, recirculation unit, gas supply manifold, and exhaust filter were installed and leak tested. With the replacement of window gaskets and installation of atmosphere monitoring instrumentation the unit will be ready for testing.

## 3. Xenon Tagging Device

A xenon tagging device was fabricated, leak tested, and is currently being calibrated. The device provides a means of adding a controlled amount of xenon having a known isotopic composition to the atmosphere in the plenum of fuel elements being fabricated and a mechanism for insertion of the top end plug into the fuel element. Essential to the operation of this device is a knowledge of the pressure in the element at various stages during tagging. Volumetric limitations preclude the use of precise Bourdon type gauges, so a pressure transducer was incorporated into the system. The first pressure transducer employed utilized vacuum as a reference point. In the range of pressure where measurements were critical, the sensitivity of this transducer had decreased noticeably. Replacement of this transducer by one with an atmospheric-pressure reference point appears to have improved sensitivity in the pressure range of interest.

## 4. Associated Activities

Spring wire to be used in the next series of elements was received and sampled for chemical verification of composition. Cladding tubing was shipped to Carpenter Technology, Inc. for ultrasonic testing and returned. Cutting oil was sampled for the presence of halogens, sulfur, and phosphorus and was found to be acceptable. These activities were performed in accordance with QA procedures.

Documentation associated with the fabrication of fuel elements under the QA program was reviewed,

and modification of the existing documentation was begun. The fuel element fabrication facility and personnel were audited by the QA Manager; however, no report of findings has been received.

## C. EBR-II Irradiation Testing

(J. O. Barner, J. F. Kerrisk, T. W. Latimer)

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-III to 463-X. 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-III  
SERIES K1, K2, AND K3 ENCAPSULATED CARBIDE EXPERIMENT

Expt. No.	Fuel Type <sup>a</sup>	Fuel Density, % Theo.	Bond and Diametral Gap, in.	Clad <sup>h</sup> Type	Clad O.D. x I.D., <sup>g</sup> in.	Max. Linear Power, Kw/ft	Maximum Centerline Temp., °C	Goal Burnup, at. %	Current Peak Burnup, at. %	Status
Series K1										
K-36B	(U <sub>0.8</sub> Pu <sub>0.2</sub> )C	90	Na-0.015	SA-316SS	0.300 x 0.280	27	1165	6	5.85*	Exam, b.8 <sup>e</sup>
K-37B	(U <sub>0.8</sub> Pu <sub>0.2</sub> )C	90	Na-0.015	SA-316SS	0.300 x 0.280	27	1165	6	2.9	Exam, a.7 <sup>b,e</sup>
K-38B	(U <sub>0.8</sub> Pu <sub>0.2</sub> )C	90	Na-0.015	SA-316SS	0.300 x 0.280	27	1165	6	5.8	Exam, a.7 <sup>o,e</sup>
K-39B	(U <sub>0.8</sub> Pu <sub>0.2</sub> )C	90	Na-0.015	SA-316SS	0.300 x 0.280	27	1165	10 <sup>i</sup>	5.8	EBR-II, Un-assigned
K-42B	(U <sub>0.8</sub> Pu <sub>0.2</sub> )C	90	Na-0.015	SA-316SS	0.300 x 0.280	27	1165	6	4.6	Completed <sup>d</sup>
Series K2										
K-49	(U <sub>0.8</sub> Pu <sub>0.2</sub> )C	95	Na-0.020	SA-316SS	0.300 x 0.280	41-46	1400	5	3.74*	Exam, b.8 <sup>e</sup>
K-50	(U <sub>0.8</sub> Pu <sub>0.2</sub> )C	95	Na-0.020	SA-316SS	0.300 x 0.280	41-46	1400	6.5	3.6	Exam, a.9 <sup>e</sup>
K-51	(U <sub>0.8</sub> Pu <sub>0.2</sub> )C	95	Na-0.020	SA-316SS	0.300 x 0.280	41-46	1400	8	3.5	Exam, a.9 <sup>e</sup>
Series K3										
K-43	(U <sub>0.8</sub> Pu <sub>0.2</sub> )C	94	Na-0.020	SA-316SS	0.300 x 0.280	27	1150	8	5.6	Exam, a.8 <sup>e</sup>
K-44	(U <sub>0.8</sub> Pu <sub>0.2</sub> )C	94	Na-0.020	SA-316SS	0.300 x 0.280	27	1150	8	7.1	EBR-II, X182
K-45	(U <sub>0.8</sub> Pu <sub>0.2</sub> )C	94	Na-0.020	SA-316SS	0.300 x 0.280	27	1150	5	2.7	Exam, b.6 <sup>e</sup>
K-46	(U <sub>0.8</sub> Pu <sub>0.2</sub> )C	94	Na-0.020	SA-316SS	0.300 x 0.280	27	1150	5	2.39*	Exam, b.8 <sup>e</sup>

<sup>a</sup>Series 1 and 3 experiments are 93% enriched in <sup>235</sup>U. Series 2 experiments are 97% enriched in <sup>233</sup>U. All fuel is single phase.

<sup>b</sup>K-37B was damaged during reconstitution of X152 to the extent that it cannot be irradiated further.

<sup>c</sup>K-38B was damaged during reconstitution of X152. Additional irradiation was completed.

<sup>d</sup>Reported in LA-4669-MS

<sup>e</sup>Element cladding failure indicated.

<sup>f</sup>Burnup values marked with \* were measured using the <sup>148</sup>Nd method. Remaining values were computed using an EBK-II power adjustment factor of 0.91.

<sup>g</sup>Dimensions are nominal.

<sup>h</sup>SA = solution annealed.

<sup>i</sup>Original goal burnup was 6 at.%. New AIP request for further irradiation in preparation.

<sup>j</sup>Corrected using an EBR-II power adjustment factor of 0.91.

#### 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 hot cell examinations are done by Project 401 personnel under the guidance of Project 463 personnel.

Table 463-III 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 27 Kw/ft). Three tests at very high power (> 40 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

TABLE 463-IV  
SERIES U1300 ENCAPSULATED CARBIDE EXPERIMENTS

Expt. No.	Fuel Type <sup>a</sup>	Fuel Density, % Theo.	Bond and Diametral Gap, in.	Clad <sup>d</sup> Type	Clad O.D. x I.D., in.	Max. Linear Power, Kw/ft <sup>f</sup>	Maximum Centerline Temp., °C	Goal Burnup, at. %	Current Peak Burnup, at. % <sup>e</sup>	Status
U93	MC+5% M <sub>2</sub> C <sub>3</sub>	84	He-0.004	SA-316SS	0.302 x 0.248	18.4	1750	11	9.64*	Exam, b.8
U94	MC+5% M <sub>2</sub> C <sub>3</sub>	83	He-0.007	SA-316SS	0.305 x 0.274	19.9	1680	11	9.42*	Exam, b.8
U97	MC+5% M <sub>2</sub> C <sub>3</sub>	84	He-0.004	SA-INC-800	0.299 x 0.245	16.4	1750	11	10.0	Exam, b.8
U98	MC+5% M <sub>2</sub> C <sub>3</sub>	84	He-0.007	SA-INC-800	0.299 x 0.269	19.9	1680	11	9.6	Exam, b.8 <sup>c</sup>
U105	MC+5% M <sub>2</sub> C <sub>3</sub>	76	He-0.008	SA-INC-800	0.300 x 0.246	13.7	1900	11	9.89*	Exam, b.8
U106	MC+5% M <sub>2</sub> C <sub>3</sub>	75	He-0.009	SA-INC-800	0.304 x 0.274	18.0	1825	11	9.9	Exam, a.8 <sup>c</sup>
U110	MC+10% M <sub>2</sub> C <sub>3</sub>	96 <sup>b</sup>	He-0.014	SA-INC-800	0.304 x 0.274	19.9	1960	10	9.2	Exam, a.8
U113	MC+10% M <sub>2</sub> C <sub>3</sub>	96 <sup>b</sup>	He-0.010	SA-INC-800	0.300 x 0.246	15.4	1880	11	10.0	Exam, a.8
U114	MC+10% M <sub>2</sub> C <sub>3</sub>	96 <sup>b</sup>	He-0.007	SA-INC-800	0.304 x 0.274	20.1	1575	10	9.5	Exam, a.8 <sup>c</sup>

<sup>a</sup>M = (U<sub>0.85</sub> Pu<sub>0.15</sub>)

<sup>b</sup>Cored pellet with nominal 0.080 in. diameter axial hole.

<sup>c</sup>Element cladding failure indicated.

<sup>d</sup>SA = Solution annealed

<sup>e</sup>Burnup values marked with \* were measured using the <sup>148</sup>Nd method. Remaining values were computed using an EBR-II power adjustment factor of 0.91.

<sup>f</sup>Corrected using an EBR-II power adjustment factor of 0.91.

these series (five in subassembly X119B, one from X142, and two from X152), using  $\gamma$ -scanning for <sup>133</sup>Xe. 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-IV 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 18 Kw/ft). Indications of element cladding

failure for three experiments were found at EBR-II using  $\gamma$ -scanning for <sup>133</sup>Xe. These element failures have been confirmed by  $\gamma$ -scanning for <sup>137</sup>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-V. 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 (11 to 14 Kw/ft). During interim examination at EBR-II after run 58, <sup>137</sup>Cs was detected by  $\gamma$ -scanning in the sodium reservoir of capsule U136. Release of fission gas from a breached helium-bonded element would be expected. However, no <sup>133</sup>Xe was detected in the capsule plenum. The lack of fission gas in the capsule and the presence of <sup>137</sup>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

TABLE 463-V  
SERIES U1950 ENCAPSULATED CARBIDE EXPERIMENTS

Expt. No.	Fuel Type <sup>a</sup>	Fuel Density % Thco.	Bond and Diametral Gap, in. <sup>e</sup>	Clad <sup>f</sup> Type	Clad O.D. x I.D., in.	Max. Linear Power, Kw/ft <sup>h</sup>	Maximum Centerline Temp., °C	Goal Burnup, at. %	Current Peak Burnup, at. % <sup>g</sup>	Status
U129	MC+5% M <sub>2</sub> C <sub>3</sub>	84	He-0.022	SA-316SS	0.303 x 0.260	11.6	1755	11	8.8	EBR-II, X055B
U130	MC+5% M <sub>2</sub> C <sub>3</sub>	75	He-0.022	SA-316SS	0.303 x 0.260	11.9	1500	11	8.8	EBR-II, X055B
U131	MC+5% M <sub>2</sub> C <sub>3</sub>	84	He-0.022	SA-316SS	0.303 x 0.260	11.6	1495	11	8.7	EBR-II, X055B
U132	MC+5% M <sub>2</sub> C <sub>3</sub>	84	He-0.022	SA-316SS	0.303 x 0.260	11.6	1495	11	8.6	EBR-II, X055B
U133	MC+5% M <sub>2</sub> C <sub>3</sub>	84	He-0.022	SA-316SS	0.303 x 0.260	11.6	1495	11	8.4	EBR-II, X055B
U134	MC+5% M <sub>2</sub> C <sub>3</sub>	84	He-0.022	SA-316SS	0.303 x 0.260	11.6	1495	11	8.4	EBR-II, X055B
U135	MC+5% M <sub>2</sub> C <sub>3</sub>	84	He-0.022	SA-INC-800	0.302 x 0.260	11.6	1475	11	8.7	EBR-II, X055B
U136 <sup>d</sup>	MC+5% M <sub>2</sub> C <sub>3</sub>	84	He-0.022	SA-INC-800	0.302 x 0.260	12.1	1475	11	8.2	EBR-II, X055B
U137	MC+10% M <sub>2</sub> C <sub>3</sub>	99	He-0.022	SA-316SS	0.303 x 0.260	12.2	1440	10	7.3	EBR-II, X055B
U138A <sup>b</sup>	MC+10% M <sub>2</sub> C <sub>3</sub>	99	He-0.022	SA-316SS	0.293 x 0.260	13.5	1440	8	4.2	EBR-II, X055B
U139	MC+10% M <sub>2</sub> C <sub>3</sub>	99	He-0.022	SA-INC-800	0.304 x 0.260	13.5	1440	10	7.4	EBR-II, X055B
U140	MC	93	He-0.022	SA-INC-800	0.302 x 0.260	12.6	1460	10	8.0	EBR-II, X055B
U141	MC	93	He-0.022	SA-316SS	0.303 x 0.260	13.0	1460	10	7.9	EBR-II, X055B
U142	MC	93	He-0.022	SA-316SS	0.304 x 0.260	13.2	1460	11	8.0	EBR-II, X055B
U143	MC+10% M <sub>2</sub> C <sub>3</sub>	99 <sup>c</sup>	He-0.022	SA-INC-800	0.302 x 0.260	11.6	1395	11	8.2	EBR-II, X055B
U144	MC+10% M <sub>2</sub> C <sub>3</sub>	99 <sup>o</sup>	He-0.022	SA-316SS	0.304 x 0.260	11.9	1395	11	8.3	EBR-II, X055B
U145	MC	93	Na-0.030	SA-304SS	0.305 x 0.270	12.2	820	10	7.8	EBR-II, X055B
U146A <sup>b</sup>	MC+10% M <sub>2</sub> C <sub>3</sub>	99	Na-0.030	SA-304SS	0.300 x 0.270	12.5	810	8	4.2	EBR-II, X055B
U147	MC+10% M <sub>2</sub> C <sub>3</sub>	99	Na-0.030	SA-INC-800	0.304 x 0.270	12.9	810	10	7.9	EBR-II, X055B

<sup>a</sup>M = (U<sub>0.85</sub>Pu<sub>0.15</sub>)

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

<sup>c</sup>Pellets cored with nominal 0.080-in.-diam axial hole.

<sup>d</sup>Possible element cladding failure indicated.

<sup>e</sup>All cladding I.D. and gap measurements are nominal.

<sup>f</sup>SA = Solution annealed.

<sup>g</sup>Computed using an EBR-II power adjustment factor of 0.91.

<sup>h</sup>Corrected using an EBR-II power adjustment factor of 0.91.

other capsules indicated fuel element failure during the examinations at EBR-II. All 19 capsules were reconstituted into subassembly X055B which is currently being irradiated.

The Series U1930 and U1960 experiments are described in Table 463-VI. Experimental parameters include fuel type, fuel density, bond type, and cladding type. The operating linear power ratings for the experiments are relatively high (27-31 Kw/ft). Nondestructive examination of the eleven experiments listed in part A of Table 463-VI 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 continuing.

The experiments listed in part B of Table 463-VI 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 464-VI

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. Non-destructive examination of the experiment is complete. The experiments listed in part D of Table 463-VI 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-VII 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  $\gamma$ -scanning for <sup>133</sup>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

TABLE VI

## SERIES U1930 AND U1960 ENCAPSULATED CARBIDE EXPERIMENTS

Expm't. No.	Fuel Type <sup>a</sup>	Fuel Density, % Theo.	Bond and Diametral <sup>f</sup> Gap, in.	Clad <sup>d</sup> Type	Clad O.D. x I.D., in.	Max. Linear Power, <sup>h</sup> Kw/ft	Maximum Centerline Temp., °C	Goal Burnup, at. %	Current Peak Burnup, at. % <sup>g</sup>	Status
U187	MC+5% M <sub>2</sub> C <sub>3</sub>	84	He-0.007	SA-316SS	0.304x0.264	27.3	1935	5	4.60*	Exam, b.12
U189	MC+5% M <sub>2</sub> C <sub>3</sub>	84	He-0.007	SA-INC-800	0.302x0.262	27.3	1935	5	4.80*	Exam, b.12
U191	MC	92	Na-0.030	SA-304SS	0.304x0.276	28.8	1148	5	4.50*	Exam, b.12
U192	MC	92	Na-0.030	SA-304SS	0.305x0.277	28.8	1148	5	4.30*	Exam, b.12
U194	MC+10% M <sub>2</sub> C <sub>3</sub>	98	Na-0.030	SA-304SS	0.305x0.277	30.1	1132	5	4.6	Exam, b.12 <sup>o</sup>
U195	MC+10% M <sub>2</sub> C <sub>3</sub>	98	Na-0.030	SA-304SS	0.305x0.276	30.1	1132	5	4.94*	Exam, b.12
U197	MC+10% M <sub>2</sub> C <sub>3</sub>	98	Na-0.030	SA-INC-800	0.305x0.277	30.4	1132	5	4.90*	Exam, b.12
U198	MC+10% M <sub>2</sub> C <sub>3</sub>	98	Na-0.030	SA-INC-800	0.305x0.277	30.4	1132	5	4.78*	Exam, b.12
U200	MC+5% M <sub>2</sub> C <sub>3</sub>	85	He-0.008	SA-304SS	0.288x0.260	28.0	2042	5	4.72*	Exam, b.12 <sup>o</sup>
U206	MC+5% M <sub>2</sub> C <sub>3</sub>	90	He-0.008	SA-316SS	0.288x0.252	28.7	2084	5	4.96*	Exam, b.12
U208	MC+10% M <sub>2</sub> C <sub>3</sub>	97 <sup>b</sup>	He-0.009	SA-316SS	0.293x0.257	29.0	1912	5	5.00*	Exam, b.12
B										
U188	MC+5% M <sub>2</sub> C <sub>3</sub>	84	He-0.007	SA-316SS	0.304x0.264	27.3	1935	11	9.6	EBR-II, X182
U190	MC+5% M <sub>2</sub> C <sub>3</sub>	84	He-0.007	SA-INC-800	0.302x0.262	27.3	1935	11	9.6	EBR-II, X182
U193	MC	92	Na-0.030	SA-304SS	0.305x0.277	28.8	1148	11	9.6	EBR-II, X182
U196	MC+10% M <sub>2</sub> C <sub>3</sub>	98	Na-0.030	SA-304SS	0.305x0.277	29.7	1132	11	9.5	EBR-II, X182
U199	MC+10% M <sub>2</sub> C <sub>3</sub>	98	Na-0.030	SA-INC-800	0.305x0.277	30.5	1132	11	6.9	EBR-II, X182
U201	MC+5% M <sub>2</sub> C <sub>3</sub>	85	He-0.008	SA-304SS	0.288x0.260	27.3	2042	11	6.5	EBR-II, X182
U207	MC+5% M <sub>2</sub> C <sub>3</sub>	90	He-0.008	SA-316SS	0.293x0.257	28.8	2088	11	6.6	EBR-II, X182
U209	MC+10% M <sub>2</sub> C <sub>3</sub>	97 <sup>b</sup>	He-0.009	SA-316SS	0.293x0.257	28.1	1909	11	6.6	EBR-II, X182
C										
U185	MC+10% M <sub>2</sub> C <sub>3</sub>	96	He-0.011	SA-316SS	0.304x0.264	27.3	2195	3	2.7	Exam, a.7
U186	MC+10% M <sub>2</sub> C <sub>3</sub>	96	He-0.011	SA-316SS	0.304x0.264	27.3	2195	3	2.7	Exam, a.7
U202	MC+5% M <sub>2</sub> C <sub>3</sub>	85	He-0	SA-316SS	0.269x0.251	28.8	1270	3	2.5	Exam, a.7
U203	MC+5% M <sub>2</sub> C <sub>3</sub>	85	He-0	SA-316SS	0.288x0.252	28.4	1260	3	2.5	Exam, a.7
U204	MC+10% M <sub>2</sub> C <sub>3</sub>	97 <sup>b</sup>	He-0	SA-316SS	0.266x0.248	29.3	1131	3	2.6	Exam, a.7
U205	MC+10% M <sub>2</sub> C <sub>3</sub>	97 <sup>b</sup>	0	SA-316SS	0.284x0.248	29.0	1124	3	2.6	Exam, a.7
D										
U260	MC+10% M <sub>2</sub> C <sub>3</sub>	98	He-0.015	20CW-316SS	0.298x0.264	31.0	2590	12	---	EBR-II, un- assigned
U261 <sup>e</sup>	MC+10% M <sub>2</sub> C <sub>3</sub>	98	He-0.015	SA-316SS	0.290x0.260	31.0	2590	12	---	
U262	MC+10% M <sub>2</sub> C <sub>3</sub>	97	He-0.015	SA-INC-800	0.290x0.260	31.0	2590	12	---	

<sup>a</sup>M = (U<sub>0.85</sub>Pu<sub>0.15</sub>)<sup>b</sup>Cored pellets with nominal 0.080-in.-diam axial hole.<sup>c</sup>Element cladding for 194 and 200 has failed.<sup>d</sup>SA = Solution annealed; 20CW = 20% cold-worked.<sup>e</sup>Eddy current test at EBR-II indicated capsule bond discontinuity.<sup>f</sup>All cladding I.D. and gap measurements are nominal.<sup>g</sup>Burnup values marked with \* were measured using the <sup>148</sup>Nd method. Remaining values were computed using an EBR-II power adjustment factor of 0.91.<sup>h</sup>Corrected using an EBR-II power adjustment factor of 0.91.

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 to 10%.

Table 463-VIII 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 (24-34 Kw/ft). Capsules B-1-4 and B-2-5 have been examined using  $\gamma$ -scanning techniques for the detection of <sup>137</sup>Cs, and both elements are apparently

TABLE 463-VII  
SERIES WF ENCAPSULATED CARBIDE EXPERIMENTS

Expm't. No.	Fuel Type <sup>a</sup>	Fuel Density, % Theo.	Bond and Diametral Gap, in.	Clad <sup>b</sup> Type	Clad O.D. x I.D., in.	Max. Linear Power, Kw/ft <sup>e</sup>	Maximum Centerline Temp., °C	Goal Burnup, at. %	Current Peak Burnup, at. % <sup>d</sup>	Status
W3F	MC+6-15% M <sub>2</sub> C <sub>3</sub>	91	Na-0.025	SA-316SS	0.300x0.276	24.6	1075	10 <sup>o</sup>	5.5	EBR-II, Un-assigned
W4F	MC+7-17% M <sub>2</sub> C <sub>3</sub>	93	Na-0.027	SA-316SS	0.300x0.276	25.5	1100	6	5.6	Exam, 1.7
W5F	MC+7-10% M <sub>2</sub> C <sub>3</sub>	89	Na-0.010	SA-316SS	0.250x0.230	18.2	975	10 <sup>o</sup>	4.3	EBR-II, X182
W6F	MC+9-10% M <sub>2</sub> C <sub>3</sub>	89	Na-0.011	SA-316SS	0.251x0.231	18.2	975	6	4.3	EBR-II, X182
W7F	MC+5% M <sub>2</sub> C <sub>3</sub>	89	Na-0.027	20CW-316SS	0.300x0.276	24.6	1075	10 <sup>o</sup>	5.5	EBR-II, Un-assigned
W8F	MC+3-6% M <sub>2</sub> C <sub>3</sub>	93	Na-0.025	20CW-316SS	0.300x0.276	25.5	1100	10 <sup>o</sup>	6.0	Exam, 2.1
W10F	MC+0-3% M <sub>2</sub> C <sub>3</sub>	88	Na-0.012	SA-316SS	0.251x0.231	18.2	975	10 <sup>o</sup>	5.4	EBR-II, Un-assigned
W12F	MC+19-24% M <sub>2</sub> C <sub>3</sub>	95	Na-0.013	SA-316SS	0.251x0.231	19.1	1000	10 <sup>o</sup>	4.3	EBR-II, X182

<sup>a</sup>M = U<sub>0.8</sub>Pu<sub>0.2</sub>

<sup>b</sup>SA = Solution annealed, 20 CW = 20% cold worked.

<sup>o</sup>Original goal burnup was 6 at.%. New AIP request for further irradiation in preparation.

<sup>d</sup>Computed using an EBR-II power adjustment factor of 0.91.

<sup>e</sup>Corrected using an EBR-II power adjustment factor of 0.91.

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  $\gamma$ -scanning for <sup>133</sup>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 is pending the preparation and approval of a request to the AEC to extend the burnup limit to 10 at.%. The failed experiment in this group has been nondestructively examined and destructive examination is under way.

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 <sup>133</sup>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-IX. 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 are currently being irradiated in subassembly X156. The first interim examination will be made at a burnup of 2.5 at.%.

The C-5 and O-N1 series of singly-clad experiments are described in Table 463-X. 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

TABLE 463-VIII  
 SERIES B-1, B-2 AND B-3 ENCAPSULATED NITRIDE EXPERIMENTS

Expt. No.	Fuel Type	Fuel Density, % Theo.	Bond and Diametral Gap, in.	Clad <sup>e</sup> Type	Clad O.D. x I.D., in.	Max. Linear Power, Kw/ft <sup>h</sup>	Maximum Centerline Temp., °C	Goal Burnup, at. %	Current Peak Burnup, at. % <sup>g</sup>	Status
Series B-1										
B-1-1	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	80	Na-0.019	SA-304SS	0.290x0.250	25.4	1125	5	5.9	Exam, b. 8 <sup>b</sup>
B-1-2	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	81	Na-0.018	SA-304SS	0.290x0.250	24.7	1125	9	5.7	Exam, a. 7 <sup>b</sup>
B-1-4	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	85	Na-0.012	SA-304SS	0.290x0.250	26.0	1150	10 <sup>f</sup>	2.7	Exam, a. 7 <sup>o</sup>
Series B-2										
B-2-1	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	82	Na-0.021	SA-316SS	0.316x0.275	29.8	1230	10 <sup>i</sup>	5.7	EBR-II, unassigned
B-2-2	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	82	Na-0.020	SA-316SS	0.316x0.275	29.6	1230	9	5.6	Exam, a. 8 <sup>b</sup>
B-2-3	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	81	Na-0.020	SA-316SS	0.315x0.275	29.5	1230	12	7.5	EBR-II, X182
B-2-5	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	76	Na-0.028	SA-316SS	0.315x0.284	29.5	1230	12	2.7	Exam, a. 7 <sup>o</sup>
B-2-6	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	82	Na-0.021	SA-316SS	0.316x0.295	33.3	1230	6	5.6	Exam, a. 7 <sup>b</sup>
B-2-7	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	82	Na-0.020	SA-316SS	0.316x0.295	33.4	1230	12	5.6	Exam, a. 7 <sup>b</sup>
Series B-3										
B-3-2	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	88	Na-0.009	SA-316SS	0.315x0.284	34.3	1250	9	2.78 <sup>*</sup>	Exam, b. 8 <sup>b</sup>
B-3-3	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	91	Na-0.010	SA-316SS	0.315x0.284	35.4	1280	12	2.8	Exam, a. 7 <sup>b, d</sup>
B-3-4	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	94	Na-0.013	SA-316SS	0.316x0.284	35.4	1280	12	2.70 <sup>*</sup>	Exam, b. 12 <sup>b</sup>
B-3-5	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	90	Na-0.010	SA-316SS	0.316x0.295	37.8	1310	6	2.8	Exam, b. 12 <sup>b</sup>
B-3-6	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	95 <sup>a</sup>	He-0.005	SA-316SS	0.315x0.275	31.1	1925	6	4.4	EBR-II, X182
B-3-7	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	89	He-0.005	SA-316SS	0.315x0.275	31.1	1925	6	4.5	EBR-II, X182
B-3-8	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	90 <sup>a</sup>	He-0.005	SA-316SS	0.315x0.275	29.5	1875	6	4.4	EBR-II, X182

<sup>a</sup> Pellets are annular with a 0.070-in.-diam axial hole.

<sup>b</sup> Element cladding failure indicated.

<sup>c</sup> Available for further irradiation.

<sup>d</sup> Nondestructive examination completed. Capsule stored for possible irradiation of failed element.

<sup>e</sup> Cladding is welded tubing. SA = Solution annealed.

<sup>f</sup> Original goal burnups were 3 to 5 at. %. New AIP request for further irradiation in preparation.

<sup>g</sup> Burnup values marked with \* were measured using the <sup>149</sup>Nd method. Remaining values were computed using an EBR-II power adjustment factor of 0.91.

<sup>h</sup> Corrected using an EBR-II power adjustment factor of 0.91.

elements have been made using the same equipment that will be used for the postirradiation examination. Eddy current examination of selected elements indicates that there are large sodium bond defects. All elements will be rebonded. 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<sub>0.8</sub>Pu<sub>0.2</sub>)N which is sodium bonded to 20% cold-worked Type 316 stainless steel cladding. Three elements have been rejected because of large fuel chips in the sodium annulus. The diameters of the elements have been measured on the same profilometer that will be used after irradiation. The status of the sodium bond in these experiments to EBR-II is the same as for the C-5 series.

It is planned that four of the fuel elements from Series O-N1 will be irradiated with selected elements 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

TABLE 463-IX  
SERIES U5100 SINGLY CLAD CARBIDE EXPERIMENTS

Expt. No.	Fuel Type <sup>a</sup>	Fuel Density, % Theo.	Bond and Diametral Gap, in. <sup>b</sup>	Clad Type <sup>c</sup>	Clad O.D. x I.D., in.	Max. Linear Power, Kw/ft <sup>e</sup>	Maximum Centerline Temp., °C	Goal Burnup, at. %	Current Peak Burnup, at. % <sup>d</sup>	Status
U241	MC	92	Na-0.018	SA-304SS	0.310x0.281	32.6	1175	6	1.8	EBR-II, X156
U242	MC	92	Na-0.017	SA-304SS	0.310x0.281	32.6	1175	9	1.8	EBR-II, X156
U243	MC	92	Na-0.031	SA-304SS	0.310x0.281	30.8	1150	6	1.7	EBR-II, X156
U244	MC	92	Na-0.017	SA-304SS	0.310x0.281	32.6	1175	9	1.8	EBR-II, X156
U245	MC	91	Na-0.032	SA-304SS	0.310x0.281	30.8	1150	12	1.7	EBR-II, X156
U246	MC	92	Na-0.017	SA-316SS	0.310x0.281	33.1	1190	6	1.9	EBR-II, X156
U247	MC	92	Na-0.032	SA-316SS	0.310x0.281	30.8	1150	6	1.7	EBR-II, X156
U248	MC	91	Na-0.032	SA-316SS	0.310x0.281	33.1	1140	12	1.9	EBR-II, X156
U249	MC	92	Na-0.017	SA-INC-800	0.309x0.281	33.1	1210	6	1.9	EBR-II, X156
U250	MC	91	Na-0.032	SA-INC-800	0.309x0.281	33.1	1145	6	1.9	EBR-II, X156
U251	MC	92	Na-0.031	SA-304SS	0.310x0.281	33.1	1145	12	1.9	EBR-II, X156
U252	MC	92	Na-0.024	SA-304SS	0.310x0.281	33.1	1140	12	1.9	EBR-II, X156
U253	MC	92	Na-0.024	SA-304SS	0.310x0.281	30.8	1145	12	1.7	EBR-II, X156
U254	MC	92	Na-0.024	SA-304SS	0.310x0.281	30.8	1140	12	1.7	EBR-II, X156
U256	MC+10% M <sub>2</sub> C <sub>3</sub>	95	Na-0.024	SA-304SS	0.309x0.281	30.9	1140	12	1.7	EBR-II, X156
U257	MC+10% M <sub>2</sub> C <sub>3</sub>	95	Na-0.024	SA-INC-800	0.309x0.281	30.5	1135	12	1.7	EBR-II, X156
U258	MC+10% M <sub>2</sub> C <sub>3</sub>	95	Na-0.024	SA-304SS	0.310x0.281	30.5	1145	6	1.7	EBR-II, X156
U259	MC+10% M <sub>2</sub> C <sub>3</sub>	95	Na-0.024	SA-INC-800	0.309x0.281	31.5	1150	12	1.8	EBR-II, X156

<sup>a</sup>MC = U<sub>0.85</sub>Pu<sub>0.15</sub> <sup>b</sup>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.

<sup>c</sup>SA = Solution Annealed <sup>d</sup>Computed using an EBR-II power adjustment factor of 0.91.

<sup>e</sup>Corrected using an EBR-II power adjustment factor of 0.91.

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.

Sufficient data are now available to calculate fission gas release for 16 elements that are currently under examination. The element data used included 1) a measurement of the number of moles and an isotopic analysis of the fuel element plenum gas and the capsule plenum gas, 2) a mass spectrometric burnup determination from a sample taken at a known axial location along the fuel element, 3) fabrication data which specifies the amount of fuel, the fuel column length, and the isotopic content

of the uranium and plutonium in the fuel, and 4) data specifying the element location in EBR-II and the reactor runs during which it was irradiated. Fission product yields were obtained from the 1972 compilation of Meek and Rider.<sup>1</sup> Relative fission rate data including axial and radial variation for <sup>235</sup>U, <sup>238</sup>U, <sup>239</sup>Pu, and <sup>240</sup>Pu were obtained from the EBR-II irradiation guide.<sup>2</sup> The fission rates of other fissionable isotopes were estimated as ratios to known fission rates:

$$^{233}\text{U}/^{235}\text{U} = 1.65,$$

$$^{241}\text{Pu}/^{239}\text{Pu} = 1.75, \text{ and}$$

$$^{242}\text{Pu}/^{239}\text{Pu} = 0.1.$$

The variation of fission rate data with changes in the EBR-II core configuration was also used. Since the information in the EBR-II irradiation guide is only approximate, the results of the calculations presented here must

TABLE 463-X  
 SERIES C-5 AND O-N1 SINGLY CLAD NITRIDE EXPERIMENTS

Expt. No.	Fuel Type	Fuel Density, % Theo.	Bond and Diametral Gap, in.	Clad <sup>a</sup> 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 <sub>0.8</sub> Pu <sub>0.2</sub> )N	93	Na-0.020	20CW-316SS	0.310x0.280	--	--	--	--	Reject <sup>b, o</sup>
C-5-2	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	93	Na-0.020	20CW-316SS	0.310x0.280	--	--	--	--	Reject <sup>b, c</sup>
C-5-3	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	94	Na-0.020	20CW-316SS	0.310x0.280	--	--	--	--	Reject <sup>b, o</sup>
C-5-4	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	95	Na-0.021	20CW-316SS	0.310x0.280	33.2	1166	12	0	At LASL <sup>d</sup>
C-5-5	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	95	Na-0.020	20CW-316SS	0.310x0.280	-----Spare-----		12	0	At LASL <sup>d</sup>
C-5-6	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	93	Na-0.021	20CW-316SS	0.310x0.280	33.3	1158	12	0	At LASL <sup>d</sup>
C-5-7	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	94	Na-0.020	20CW-316SS	0.310x0.280	33.7	1184	12	0	At LASL <sup>d</sup>
C-5-8	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	94	Na-0.030	20CW-316SS	0.310x0.280	32.7	1127	12	0	At LASL <sup>d</sup>
C-5-9	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	94	Na-0.020	20CW-316SS	0.310x0.280	33.5	1133	12	0	At LASL <sup>d</sup>
C-5-10	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	94	Na-0.020	20CW-316SS	0.310x0.280	32.6	1105	12	0	At LASL <sup>d</sup>
C-5-11	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	94	Na-0.020	20CW-316SS	0.310x0.280	33.4	1121	12	0	At LASL <sup>d</sup>
C-5-12	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	94	Na-0.030	20CW-316SS	0.310x0.280	32.5	1121	12	0	At LASL <sup>d</sup>
C-5-13	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	96	Na-0.030	20CW-316SS	0.310x0.280	32.1	1113	12	0	At LASL <sup>d</sup>
C-5-14	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	96	Na-0.030	20CW-316SS	0.310x0.280	32.0	1142	12	0	At LASL <sup>d</sup>
C-5-15	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	95	Na-0.030	20CW-316SS	0.310x0.280	32.1	1113	12	0	At LASL <sup>d</sup>
C-5-16	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	96	Na-0.030	20CW-316SS	0.310x0.280	-----Spare-----		12	0	At LASL <sup>d</sup>
C-5-17	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	96	Na-0.030	20CW-316SS	0.310x0.280	--	--	--	--	Reject <sup>c</sup>
C-5-18	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	94	Na-0.021	20CW-316SS	0.310x0.280	32.6	1115	12	0	At LASL <sup>d</sup>
C-5-19	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	94	Na-0.021	20CW-316SS	0.310x0.280	33.0	1123	12	0	At LASL <sup>d</sup>
C-5-20	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	95	Na-0.021	20CW-316SS	0.310x0.280	32.4	1107	12	0	At LASL <sup>d</sup>
Series O-N1										
O-N1-1	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	90	Na-0.020	20CW-316SS	0.310x0.280	32.9	1140	12	0	At LASL <sup>d</sup>
O-N1-2	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	90	Na-0.020	20CW-316SS	0.310x0.280	--	--	--	--	Reject <sup>b</sup>
O-N1-3	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	90	Na-0.020	20CW-316SS	0.310x0.280	32.6	1164	12	0	At LASL <sup>d</sup>
O-N1-4	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	90	Na-0.020	20CW-316SS	0.310x0.280	32.8	1164	12	0	At LASL <sup>d</sup>
O-N1-5	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	90	Na-0.020	20CW-316SS	0.310x0.280	--	--	--	--	Reject <sup>b</sup>
O-N1-6	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	89	Na-0.020	20CW-316SS	0.310x0.280	--	--	--	--	Reject <sup>b</sup>
O-N1-8	(U <sub>0.8</sub> Pu <sub>0.2</sub> )N	90	Na-0.020	20CW-316SS	0.310x0.280	33.1	1117	12	0	At LASL <sup>d</sup>

<sup>a</sup>20CW = 20% cold worked.    <sup>b</sup>Chips in bond.    <sup>c</sup>Air in plenum.    <sup>d</sup>QA evaluation in progress.

be considered as preliminary, until accurate run data are available.

Table 463-XI summarizes the fission gas release and burnup results. The peak burnup and axial average burnup were calculated from the measured burnup and its axial location. Two methods were used to calculate the amount of fission gas released from the fuel. In "Method A," the measured number of moles of gas in the fuel element plenum and in the capsule plenum, and the gas analyses, were used to calculate the amount of fission gas released from the fuel. This method uses direct experimental data without any subsidiary

assumptions, but it is subject to error if fission gas is lost during sampling or is trapped in sodium voids. "Method B" assumes that the element and capsule were loaded with only He and Ar during fabrication under local atmospheric pressure and at 25°C. The total number of moles of gas in each plenum can be calculated using this assumption, in addition to the measured plenum volume of the element and capsule, and the He and Ar content of the gas after irradiation. Thus the two methods differ only in the way the amount of fission gas is determined. An estimated uncertainty in the measured number of moles of gas is ± 20%. Considering this uncertainty, there is



TABLE 463-XI  
FISSION GAS RELEASE SUMMARY

Element	Burnup, at. %		% of Fission Gas Released <sup>a</sup>	
	Peak	Average	Method A	Method B
<u>He Bonded Carbides</u>				
U 187	4.80	4.15	22.1	20.4
U 189	4.80	4.33	20.6	18.7
U 200	4.72	4.26	15.9	18.1
U 206	4.96	4.48	9.8	9.8
U 208	5.00	4.51	10.5	10.3
<u>Na Bonded Carbides</u>				
K-36B	5.85	5.29	16.3	16.3
K-48	2.39	2.18	8.8	13.1
K-49	3.74	3.71	37.9	97.1
U 191	4.50	4.06	3.2	3.2
U 192	4.30	3.88	11.6	10.0
U 195	4.94	4.46	10.8	9.1
U 197	4.90	4.42	9.2	15.2
U 198	4.78	4.32	10.9	8.5
<u>Na Bonded Nitrides</u>				
B-3-2	2.78	2.65	8.9	7.1
B-3-4	2.70	2.57	5.8	11.5
B-3-5	2.33	2.21	8.4	8.8

<sup>a</sup> Calculation methods described in text.

good agreement between the two methods except for four elements, K-46, K-49, U 197, and B-3-4. For all four elements the calculation using the measured number of moles (Method A) is low, indicating a possible loss of gas. For two of these elements, a calculation of the fuel pressure using the measured number of moles and plenum volume results in a pressure below the initial loading pressure. This also points to a loss of gas. For these four elements the Method B result is considered the better estimate.

The five He bonded carbide elements were all irradiated in the same subassembly at approximately 30 Kw/ft. The variation in fission gas release correlates to some extent with fuel density since U 187 (84% TD), U 189 (84% TD), and U 200 (85% TD) show larger releases than U 206 (90% TD) and U 208 (97% TD cored pellet).

Of the 11 sodium bonded elements, K-49 requires special consideration. This element operated at approximately 45 Kw/ft and was a severe failure which showed evidence of fuel melting over much of the fuel column length. Thus the high gas release determined is not unexpected considering the operating history. The other 10 Na bonded elements showed fission gas releases from 3 to 16%. Sufficient data are not available at this time to attempt a correlation between fission gas release and

parameters such as linear power, fuel density and burnup.

Table 463-XII presents a detailed tabulation of the fission gas release results for the individual Kr and Xe isotopes. These results are from the "Method B" calculation. The <sup>85</sup>Kr (10.8 year half-life) results are not corrected for decay during or after irradiation. The systematic variation of the release fraction for some isotopes (<sup>83</sup>Kr and <sup>85</sup>Kr are always lower than <sup>84</sup>Kr and <sup>86</sup>Kr) is probably due to differences in actual fission yields from tabulated values. The tabulated yields used are not representative of the actual EBR-II neutron spectrum so that some differences can be expected. The data presented in Table 463-XI represent weighted averages of the data in Table 463-XII, with each release percentage being weighted by the amount of that isotope present.

D. TREAT Irradiation Testing  
(J. F. Kerrisk and D. C. Clifton)

The four Series UL TREAT tests (see Table 463-XIII), which were designed to determine the effect of irradiation on the behavior of helium and sodium bonded (U,Pu)C fueled elements, have completed the transient irradiations in the TREAT reactor. No further examinations have been performed on the capsules from tests UL-1 and UL-2.

TABLE 463-XII  
FISSION GAS RELEASE OF INDIVIDUAL ISOTOPES  
METHOD B CALCULATIONS

Element	% of Fission Gas Released							
	Kr			Xe				
	83	84	85 <sup>a</sup>	86	131	132	134	136
<u>He Bonded Carbides</u>								
U 187	19.1	20.9	18.7	21.0	19.3	20.2	21.0	20.8
U 189	18.0	19.7	15.7	19.8	17.6	18.4	19.1	18.8
U 200	16.1	17.5	14.1	17.9	17.3	17.8	18.9	18.3
U 208	8.8	9.3	7.58	9.5	9.1	9.5	9.9	9.7
U 208	9.3	10.1	8.2	10.3	9.8	10.3	10.7	10.5
<u>Na Bonded Carbides</u>								
K-36B	14.9	16.2	12.4	16.5	15.1	16.2	18.9	18.5
K-46	12.1	13.1	10.8	13.4	12.0	13.1	13.8	13.3
K-49	88.1	92.0	80.7	98.6	92.7	93.7	102.7	100.7
U 191	3.2	3.5	2.9	3.6	2.9	3.1	3.3	3.2
U 192	9.5	10.4	8.5	10.8	9.3	9.8	10.3	10.1
U 195	8.7	9.5	7.7	9.6	8.4	9.0	9.4	9.2
U 197	13.0	15.2	12.4	15.5	14.3	15.0	15.8	15.5
U 198	7.8	8.5	6.9	8.7	7.9	8.4	8.9	8.7
<u>Na Bonded Nitrides</u>								
B-3-2	6.8	7.4	6.1	7.6	6.0	7.0	7.4	7.4
B-3-4	10.9	11.7	9.7	11.9	10.3	11.4	11.9	11.6
B-3-5	8.2	8.9	7.3	9.0	7.9	8.5	8.9	8.8

<sup>a</sup> Not corrected for decay.

TABLE 463-XIII  
LASL SERIES UL TESTS

	TEST			
	LASL-UL-1	LASL-UL-2	LASL-UL-3	LASL-UL-4
Fuel Element <sup>a</sup>	263 (138 A)	264 (146 A)	265 (138)	266 (146)
Fuel Material <sup>b</sup>	90 vol% (U <sub>0.85</sub> Pu <sub>0.15</sub> )C + 10 vol% (U <sub>0.85</sub> Pu <sub>0.15</sub> ) <sub>2</sub> C <sub>3</sub>			
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 <sup>c</sup>	0	0	45,000	45,000
Test Objective	Fuel Melting	Fuel Melting	Same Transient as 263	Same Transient as 263

<sup>a</sup>Fuel element numbers reassigned by Gulf United. Old numbers shown in parentheses.

<sup>b</sup>Uranium enriched to 60% in <sup>235</sup>U.

<sup>c</sup>Irradiated in EBR-II at 10 to 15 Kw/ft in subassembly X055.

Assembly work on the capsules for tests UL-3 and UL-4 was completed by the Radiometallurgy Group of Hanford Engineering Development Laboratory (HEDL) and the capsules were shipped to TREAT in early July 1973. Both tests were performed in mid-July. A preliminary review of the capsule temperatures and reactor power indicates that the tests were performed as requested. Both capsules were shipped to the LASL hot cell facility in mid-September. Since these capsules contain elements preirradiated in EBR-II, all examination work will require the hot cell facility. The inner capsules were removed from the TREAT capsules and preparations for initial nondestructive examinations are in progress.

### III. QUALITY ASSURANCE (L. E. Lanham)

General: The quality assurance organization has reviewed Work Instructions and addendum changes to travelers and has conducted surveillance activities to

check for their inclusion in the basic traveler and their application by the operator to the appropriate procedure. Procurement packages have been reviewed. Shipping requests and shipping inspection reports have been reviewed and items released for shipment.

Fuel Preparation: An audit was conducted by the Quality Assurance Manager of the Fuel Element Preparation operation while prototypical fuel elements were being prepared. An audit report has been prepared. Additional surveillance of this operation was conducted by quality assurance and the document package is being reviewed. Some problems were encountered in the areas of complete and timely documentation and the use of calibrated instruments. The review point and quality control requirements used in the procedures are effective in controlling the quality of the product.

The required corrective actions are being taken which include informal on-the-job QA instructions

followed by a formal training session for all operators and supervisors.

Fuel Pin Fabrication: The inspection and over-checking of materials scheduled for use in Pin Fabrication has continued. An outside laboratory is being used for the ultrasonic examination of cladding material.

#### IV. REFERENCES

1. M. E. Meek and B. F. Rider, "Compilation of Fission Product Yields, Vallecitos Nuclear Center, 1972," General Electric Report NEDO-12154 (1972).
2. "Guide for Irradiation Experiments in EBR-II," Argonne National Laboratory, Chicago, Illinois, Appendix C.

PROJECT 472

FBR ANALYTICAL QUALITY ASSURANCE STANDARDS AND METHODS  
RESEARCH AND DEVELOPMENT

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

I. INTRODUCTION

Necessary to the development of high quality fuels, control rods, and other reactor components required by the FBR program are highly reliable analytical methods for the chemical characterization of the source materials and products, and for the measurement of burnup, O/M ratio, and various gases on irradiated fuels. Tasks for ensuring the production of these materials are: (1) the continual preparation and distribution of carefully characterized calibration materials and quality control samples for use by the vendors and purchasers and for the surveillance of vendors and purchasers during periods of production, (2) the preparation and guidance in the use of quality assurance programs for chemical specification sampling and analysis, (3) the development of improved methods of analysis, as required, (4) the preparation of continually updated compilations of analytical methods, and (5) the analysis, in a referee capacity, of samples in dispute between vendors and purchasers. For the near future, these tasks are dedicated to the FFTF. They will be extended, as appropriate, to the LMFBR demonstration and large production facilities.

Tasks concerned with irradiated FBR fuel examinations are: (1) the development of burnup methods based on conventional mass spectrometry, on chemical analyses using inexpensive chemical apparatus, and on spark source mass spectrometry for rapid, precise measurements, (2) the prooftesting of developed methods for burnup jointly with the Allied Chemical Corporation (Idaho), (3) the development of methods for the measurement of the O/M

ratio, and (4) the development of methods for the measurement of gases including techniques to measure the release rates of various gases as a function of temperature-time cycling.

As a high priority item, a program has been initiated to establish a quality assurance program and to develop analytical methods, as necessary, for the chemical characterization of low-friction, hard surfaces to be applied to various FFTF core components.

II. ANALYTICAL CHEMISTRY PROGRAM FOR LOW-FRICTION, HARD SURFACES

In August of this year, LASL joined HEDL in a cooperative effort to establish a program for the chemical characterization of hard surfaces that are to be applied to wear areas on FFTF components. The planned hard surface is  $\text{Cr}_3\text{C}_2$  applied as a molten blend of  $\text{Cr}_3\text{C}_2$  and nichrome powder binder.

In meetings at LASL and at HEDL, responsibilities and priorities have been delineated for analytical method development, round robin evaluations, and quality assurance aspects.

A. Status and Development of Analytical Methods  
(W. H. Ashley, J. E. Rein, G. R. Waterbury)

Methods used by a potential vendor for the chemical characterization of  $\text{Cr}_3\text{C}_2$  and nichrome powders and the hard surface product are being evaluated including laboratory tests when deemed advisable. In general, the vendor's wet chemical methods for the major components

of chromium, nickel, and carbon and for impurity silicon appear basically sound. Modifications are being added to improve throughput rates and measurement precisions. The potential vendor has been characterizing the two powders and the product for impurity metals iron, cobalt, and manganese. It is expected that specification limits will be established for a greater number of metal impurities and for oxygen and nitrogen, dependent on characterization data obtained on representative samples of the powders and hard surface product supplied by the potential vendor. In any case, analytical methods for such characterizations must be available.

The hard surface sample prepared for chemical specification overcheck analyses is planned to be a thin-wall metal cylinder that will have a relatively thick layer of hard surface applied during periods when reactor components are surfaced. The recommended metal for the cylinder is pure aluminum which does not interfere with the analytical methods used for chemical characterization and which is most unlikely to be in the list of specification impurities.

Until the end of this quarter, the only materials available for investigative purposes were one lot each of  $\text{Cr}_3\text{C}_2$  and nichrome powders. The information given in the following sections was obtained with these powders. At the end of the quarter, a limited quantity of about 6 g of hard surface was obtained.

#### 1. Dissolution of Samples

(R. D. Gardner, R. E. Perrin, G. C. Swanson)

The potential vendor dissolves samples of  $\text{Cr}_3\text{C}_2$  powder and hard surface by fusion with  $\text{Na}_2\text{O}_2$ . This operation requires close attention by a skilled analyst to achieve complete solubilization of the sample and quantitative recoveries of all components. Acidic dissolutions are being investigated under reflux conditions, and at elevated temperatures and pressures in sealed, fused-silica tubes<sup>1</sup> and in a Teflon-container apparatus.<sup>2</sup>

Complete dissolution of  $\text{Cr}_3\text{C}_2$  and recovery of chromium was effected under reflux conditions using  $\text{HClO}_4$  alone or with a small quantity of  $\text{HNO}_3$ . In the sealed fused-silica tube, 12M  $\text{H}_2\text{SO}_4$  at 623°K completely dissolved  $\text{Cr}_3\text{C}_2$  without loss of chromium. At higher

acidities and higher temperatures, the reaction was incomplete due to formation of insoluble  $\text{Cr}_2(\text{SO}_4)_3$ . At lower temperatures and acid strengths, the dissolution rate became impractically low. With the Teflon-container apparatus,  $\text{Cr}_3\text{C}_2$  powder dissolved completely with a  $\text{HClO}_4$ -HCl mixture at 548°K. Based on experiments using radioactive  $^{51}\text{Cr}$  tracer, there was no diffusion of any chromium through the Teflon.

It is expected that the hard surface, which is applied at temperatures above 2300°K, will have refractory characteristics and, hence, will be more difficult to dissolve than  $\text{Cr}_3\text{C}_2$  powder. Because complete sample dissolution is a requirement for accurate chemical analysis, a major and high priority effort will be testing of the dissolution procedures on the hard surface test material.

#### 2. Determination of Chromium

(R. E. Perrin, R. D. Gardner)

The direction of method development for determining chromium was guided by the premise that the hard surface sample would be dissolved by  $\text{HClO}_4$  under reflux (or elevated temperature-pressure) conditions. This oxidizing acid produced Cr(VI), the desired oxidation state for redox titration. However, free chlorine formed as a reaction product had to be removed to prevent a positive bias. This was accomplished by adding  $\text{AgNO}_3$  and boiling. This reaction partially reduced Cr(VI) which was reoxidized by adding  $(\text{NH}_4)_2\text{S}_2\text{O}_8$  and boiling. After cooling, a known quantity of Fe(II) was added in excess, and the excess was titrated with standard Ce(IV) solution using potentiometric end point detection. For nichrome samples and for hard surface samples, manganese oxidizes to Mn(VII) which interferes. Following the  $(\text{NH}_4)_2\text{S}_2\text{O}_8$  oxidation, the Mn(VII) is reduced to noninterfering Mn(II) by boiling with HCl.

#### 3. Determination of Nickel

(R. E. Perrin, R. D. Gardner)

The gravimetric method of the potential vendor, based on precipitation of nickel dimethylglyoximate, has been used successfully with no significant changes for the nichrome powder sample and a mixture of nichrome powder and  $\text{Cr}_3\text{C}_2$  powder.

4. Determination of Silicon  
(R. E. Perrin, R. D. Gardner)

Silicon is determined gravimetrically as  $\text{SiO}_2$  in the residue remaining from the acidic dissolution.

5. Determination of Carbon  
(R. E. Perrin, R. D. Gardner)

Two methods have been applied successfully to the determination of carbon in  $\text{Cr}_3\text{C}_2$ . The first uses customized equipment in which the sample is heated at  $1473^\circ\text{K}$  in a stream of oxygen, and the generated  $\text{CO}_2$  is measured gravimetrically after absorption in Ascarite. The second method uses a commercial combustion analyzer (LECO WE-12 apparatus). The two methods gave results on a batch of  $\text{Cr}_3\text{C}_2$  powder that were statistically indistinguishable. This fact and the fact that the method using the commercial combustion analyzer had been calibrated with NBS silicon carbide (SRM 112) indicate that both methods gave unbiased results. The prooftest for the methods will be the analysis of hard surface samples.

6. Determination of Oxygen  
(M. E. Smith, D. E. Vance)

A method<sup>3</sup> used for the determination of oxygen in  $\text{UC}_2$  samples was used on samples of  $\text{Cr}_3\text{C}_2$  powder and hard surface. The samples were heated to  $> 2773^\circ\text{K}$  in a graphite crucible. A flowing argon stream carried the generated CO through  $\text{I}_2\text{O}_5$ , and the  $\text{CO}_2$  formed was measured manometrically.

7. Determination of Nitride Nitrogen  
(R. E. Perrin)

Two methods<sup>4</sup> for the determination of nitrogen in  $\text{B}_4\text{C}$  will be evaluated for analyzing the hard surface material. Should the sample dissolve completely in  $\text{HClO}_4$  under reflux conditions, the simplest method is to add NaOH, distill the nitrogen as  $\text{NH}_3$  into a  $\text{H}_3\text{BO}_3$  solution, and titrate with standard acid. This same basic scheme can be used for samples dissolved at elevated temperatures and pressures. If the material is not dissolved in acid, the LiOH fusion method will be evaluated.

8. Determination of Metal Impurities  
(O. R. Simi, D. W. Steinhilber)

The potential vendor analyzes only for Fe, Co, and Mn by atomic absorption. For informational purposes and to guide the selection of additional metal impurities for specification purposes, a broader spectrum analytical

method is required. This method will be used for samples of the two powders and the hard surface product. Development of an emission spectrographic method has been initiated.

B. Analysis Summary of  $\text{Cr}_3\text{C}_2$  and Nichrome Powders

The lots of  $\text{Cr}_3\text{C}_2$  and nichrome powder have been analyzed for Cr, Ni, and C and for traces of Fe, Mn, O, and Si. The results are listed in the following table:

Element	$\text{Cr}_3\text{C}_2$ Powder	Nichrome Powder	$\text{Cr}_3\text{C}_2$ -Nichrome Powder Mixture
Chromium	88.63	20.18	78.96
Carbon	12.89	0.05	11.91
Nickel	0.0001 <sup>a</sup>	77.05	8.24
Iron	0.43	0.17	0.31
Manganese	0.03 <sup>a</sup>	0.61	0.11
Silicon	Negl.	0.76	0.04
Oxygen	0.1	0.47	b
Loss of Ignition	-	0.08	-
Sum	100.08	99.35	99.57

<sup>a</sup> Measured by semiquantitative emission spectroscopy.  
<sup>b</sup> Not measured.

C. Round Robin Evaluations of Analytical Capabilities  
(J. E. Rein, R. K. Zeigler, G. R. Waterbury)

Cooperatively with HEDL, plans have been formulated for a round robin evaluation of analytical methods with participating laboratories of the potential vendor, HEDL, and LASL. HEDL will obtain kilogram-plus quantities of a nichrome powder lot, a  $\text{Cr}_3\text{C}_2$  powder lot, and a hard surface lot to be used as the round robin test materials. The time goal for the start of the round robin is mid-December 1973.

D. Reference and Quality Control Materials

Based on discussions with QA and other HEDL personnel, a plan has been formulated for the characterization and use of reference and quality control materials. For the first two FFTF cores, appropriate NBS Standard Reference Materials have been selected and supplemented by designated high-purity materials. These materials, to be designated reference materials, will serve as the reference basis for the chemical analyses of the  $\text{Cr}_3\text{C}_2$  and nichrome powders by the vendor, HEDL, and LASL (if necessary for referee purposes). Use of quality control samples is not contemplated for the hard surface program for the first two FFTF cores.

The expected reference materials for long-term use will be the lots of  $\text{Cr}_3\text{C}_2$  powder, nichrome powder, and hard surface used for the round-robin evaluation provided they become sufficiently well characterized. Quality control samples are to be prepared from various lots of hard surface with varying proportions of the  $\text{Cr}_3\text{C}_2$  and nichrome powders.

### III. ANALYTICAL CHEMISTRY PROGRAM FOR BORON CARBIDE

#### A. Status of Analytical Methods and Qualification of Analytical Laboratories

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

The evaluation of the data reported by HEDL, LASL, and three potential vendor laboratories for the nitride nitrogen analytical method indicates that both the within-laboratory measurement precision and the between-laboratory precision are satisfactory for FFTF specification use. This completes the evaluation of analytical methods with the analytical measurements for the ten specified components all deemed satisfactory.

#### B. Preparation of Calibration Materials and Quality Control Samples

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

Because the pellet batches that were to be used as quality control samples for the nitride nitrogen determination had large between-pellet heterogeneity, they were pulverized to -200 mesh powders. Overcheck analyses of the powders verified homogeneity. Samples were packaged and sent to HEDL along with a detailed report that lists recommended control limits.

The calibration material previously specified for the total boron determination was NBS  $\text{H}_3\text{BO}_3$ , only for the purpose of standardizing the titrimetric reagent. Since a major source of error in the method is considered to be the solubilization of the sample (by a fusion with  $\text{Na}_2\text{CO}_3$ ), a preferred calibration material is a boron carbide pellet. Various batches of pellets were tested for consistent boron contents and one which had satisfactory homogeneity characteristics was selected as the calibration material. Pellets were packaged and sent to HEDL.

All quality assurance materials for analytical laboratory use, deemed adequate for the production of about 150,000 FFTF boron carbide pellets, have now been sent to HEDL.

#### C. Status of RDT Standards

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

A supplement to RDT Standard F2-8 "Qualification and Control of Analytical Chemistry Laboratories for Control Rod Absorber Material Analysis" was prepared to provide coverage for the determination of nitride nitrogen. A revision to the section dealing with the total boron determination was prepared to provide for the use of the boron carbide pellets as the calibration material. These writings have been reviewed by HEDL personnel and processing has started to incorporate them in F2-8.

Cooperatively with HEDL, the analytical method for nitride nitrogen has been written as a supplement to be added to RDT Standard F11-2 "Analytical Chemistry Methods for Control Rod Absorber Material."

#### D. Studies and Improvements of Analytical Methods

##### 1. Determination of Total Boron

(R. E. Perrin, R. D. Gardner, A. L. Henicksman, W. H. Ashley)

The use of a Plattner mortar to pulverize boron carbide pellets -- a necessary operation preceding the present technique of fusion with  $\text{Na}_2\text{CO}_3$  to solubilize the material -- is known to introduce as much as 0.6% by weight of iron contamination from the mortar.<sup>4</sup> Since the sample taken for fusion is a weighed fraction of the pulverized product, a negative bias is introduced proportional to the amount of iron contamination.

An alternate solubilization treatment has been developed in which pieces of boron carbide pellets are dissolved in sealed fused-silica tubes with 12M  $\text{H}_2\text{O}_4$  at 773°K. The maximum quantity of sample that can be processed with the routine 15-ml volume tubes is 300 mg. The RDT F11-2 method for total boron recommends that an amount of boron is determined equivalent to 60 mg of boron carbide sample. With this size sample, shorter fused-silica tubes can be used such that four fit into a single metal shell.

The solubilization treatment was tested with Carborundum Lot 3 Naval Standard boron carbide. The average

weight percent boron and standard deviation for six analyses was  $79.24 \pm 0.08$  which is not significantly different than the stated value of 79.32. It also was tested with the boron carbide pellet calibration material (see previous Section III-B). The average weight percent boron and standard deviation for six analyses was  $77.15 \pm 0.11$ , which is not significantly different than the value of  $77.00 \pm 0.16$  determined using the  $\text{Na}_2\text{CO}_3$  fusion.

Advantages of the sealed tube procedure are: (1) deletion of the pulverization operation and its low bias, (2) complete retention of the sample guaranteeing that boron is recovered fully, and (3) no introduction of salts. This latter advantage alludes to a subsequent treatment in the boron determination method in which impurity metals that interfere in the titrimetric measurement are separated on a cation exchange column. The high sodium levels from the  $\text{Na}_2\text{CO}_3$  fusion hinder the cation exchange separation.

2. Determination of Nitride Nitrogen  
(R. E. Perrin, A. Zerwekh, A. L. Henicksman,  
W. H. Ashley)

The method developed about 6 months ago<sup>5</sup> for this determination was modified slightly to give expanded applicability. In this method, the sample is dissolved with  $12\text{M}$   $\text{H}_2\text{SO}_4$  in a sealed fused-silica tube, and the solution is made basic with NaOH. The nitride nitrogen, converted to  $\text{NH}_3$ , is distilled, absorbed in a solution of  $\text{H}_3\text{BO}_3$ , and titrated with standard acid. The modification is an increase in the dissolution temperature from  $643^\circ\text{K}$  to  $773^\circ\text{K}$ . Some boron carbide materials did not completely dissolve at the lower temperature.

IV. ANALYTICAL CHEMISTRY PROGRAM FOR FBR MIXED OXIDE FUEL

A. Calibration Material and Quality Control Samples  
(J. V. Pena, H. J. Kavanaugh, L. A. Maestas,  
J. E. Rein)

Materials on hand are being packaged for quarterly shipments scheduled for December 1973 and April 1974. Because of higher priority tasks, HEDL has not completed the preparation of mixed oxide and plutonium oxide matrix materials that are required for the preparation of blends for these shipments. All operations that could be done

for this task have been done so that completion will be expedited upon receipt of the matrix materials.

B. Development of Burnup Method Using Conventional Low-Cost Apparatus  
(S. F. Marsh, M. R. Ortiz, J. E. Pena)

Development was continued on the method involving spectrophotometric measurements of uranium, plutonium, and total rare earths as the fission product monitor. Previously, a two-column ion exchange procedure was developed<sup>4</sup> which provided separated fractions of these three components. The dissolved fuel sample, fumed with  $\text{HClO}_4$  to oxidize plutonium (and uranium) to the +6 oxidation state, was passed through an anion exchange column in a  $12\text{M}$  HCl medium. The rare earths, trivalent actinides, and most other fission products that were not retained were fed to a pellicular cation exchange resin in an ethanol-HCl medium. The rare earths were sorbed while the trivalent actinides and extraneous fission products passed through. From the first column, plutonium was eluted with  $0.1\text{M}$  HI -  $12\text{M}$  HCl and then the uranium with  $0.1\text{M}$  HCl.

Spectrophotometric measurements of the three separated components are being investigated using Arsenazo III as the chromogenic reagent. Difficulty has been encountered with the measurement of the separated total rare earth fraction. An organic substance that bleeds from the pellicular resin has a broad absorbance band at the wavelength of the rare earth-Arsenazo complex peak. Fuming with  $\text{HNO}_3$  and  $\text{HClO}_4$  has reduced but not eliminated the interference. Other means of treatment are being investigated.

C. Determination of O/M Ratio in Solid Solution (U,Pu)O<sub>2</sub>  
(G. C. Swanson, G. R. Waterbury)

Two techniques are being investigated for calibrating the thermogravimetric method for the determination of the O/M ratio in (U,Pu)O<sub>2</sub> fuels. The first involves correlating the measured oxygen potential of a gas in equilibrium with solid solution (U,Pu)O<sub>2</sub>, cycled through a range of O/M ratios, with a model predicted from crystal defect theory. The second technique involves the preparation of (U,Pu)O<sub>2</sub> powders with known O/M ratios.

For the measurement of the gas oxygen potential, an oxygen electrode is being developed for operation close to



the sample surface in a Mettler thermobalance. To date, ten electrode configurations have been tested. Because none has given a usable response, an alternate technique is being evaluated in which oxygen potentials will be calculated from gas compositions measured by mass spectrometry.

A possible preparation of solid-solution (U,Pu)O<sub>2</sub> materials with known O/M ratios is to melt together the two pure metals to form an alloy which then is oxidized under controlled conditions. The O/M ratio is computed from the measured weight change. In preliminary experiments, 1 g of chemically polished uranium metal was melted in an alumina crucible in the thermobalance under a highly pure argon atmosphere. After the metal had cooled, an argon-oxygen atmosphere was introduced, the temperature was raised, and the uranium was oxidized to U<sub>3</sub>O<sub>8</sub>. The metal melted with no change in weight suggesting that U-Pu alloys can be prepared in the apparatus. Also promising was no discernible reaction between the melt and the container.

D. Development of Gas Measurement Techniques  
(R. M. Abernathy, J. E. Rein)

An apparatus is being assembled with an end capability to measure gas components on a real time basis as they are released from samples of fuel, boron carbide, or other materials as a function of temperature. The major apparatus components are a furnace with controllable atmospheres including vacuum, an induction heater, a gas processing train, and a mass spectrometer. All components have been received and final assembly of the apparatus is expected by November 1973.

V. QUALITY ASSURANCE  
(L. E. Lanham)

A quality assurance procedure for the preparation, packaging, and shipping of FBR calibration and quality control material has been prepared. This document, together with the procedures for the preparation, overcheck and verification of materials, has been reviewed by the Quality Assurance Manager.

VI. MEETINGS

M. Carlson and R. Stromatt met with LASL personnel at LASL on August 2, 1973, to discuss hard surfacing of reactor ducts.

J. Rein and G. Waterbury discussed all aspects of this program (07472) with HEDL personnel at HEDL on September 20 and 21, 1973.

VII. REFERENCES

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