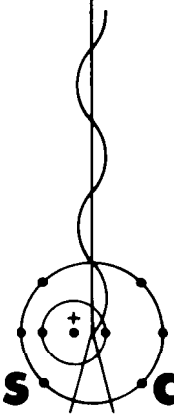
LA-5193-PR

PROGRESS REPORT

Quarterly Progress Report on the

Advanced Plutonium Fuels Program

October 1 to December 31, 1972



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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-4913-PR	LA-5067-PR
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Compiled by

R. D. Baker

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

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ABSTRACT

This is the 25th quarterly report on the Advanced Plutonium Fuels Program at the Los Alamos Scientific Laboratory.

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

PROJECT 401

EXAMINATION OF FAST REACTOR FUELS

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

I. INTRODUCTION

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

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

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

II. EQUIPMENT DEVELOPMENT

A. Inert Atmosphere Systems

(P. A. Mason, D. C. Maestas, E. O. Quintana, M. E. Lazarus)

Replacement shaft seals for the recirculating pump of the Disassembly Cell purifier were installed during the report period. Subsequent operation of the purifier indicated that sufficient oil from the recirculating pump had been adsorbed by the purifier chemical beds to significantly reduce their absorption capacity. Procurement of replacement chemicals has been initiated with delivery scheduled for January 1973. The Disassembly Cell has been maintained by a once-through argon purge in the interim.

The boost pump for the Disassembly Cell atmosphere sampling line also failed during the report period. A vacuum pump has been installed temporarily while a new boost pump is procured.

The Butyl Acetate Removal System for the Metallography Cells is presently being fabricated. Installation is anticipated about February 1, 1973. The Metallography Cells continue on a once-through argon purge pending successful operation of the removal system.

B. Manipulator Maintenance

(P. A. Mason, E. L. Mills, D. C. Maestas)

Frequent breakdowns have continued with the CRL Model "L" manipulators. Four manipulators have been converted to heavier tapes to increase the capacity, and

these have experienced a lower, but still unsatisfactory, breakdown rate. Central Research Laboratories has been consulted regarding the problems and are presently conducting tests to determine any recommended modifications. A decision was made to re-install the AMF Model "8" Manipulators whenever manipulator replacement was required. This should minimize downtime and delay to the metallography examination program.

The CRL Model "8" manipulators installed at DP-West have been giving symptoms of lack of lubrication in the bearings. This is believed to be due to the long storage period (since 1959) which permitted the evaporation of most of the bearing lubricant. Overhaul of these manipulators for relubrication of bearings is presently being performed. The slave hand assemblies are also being converted from the RCD to SRL type to provide a more secure grip.

C. Manipulator Boot Development
(P. A. Mason)

Development work on a polyurethane boot by the Plastics Section at LASL was terminated during the previous quarter. A boot design was submitted to Central Research Laboratories with an order for three boots for evaluation. The boots are similar in construction to the boots formerly procured for CRL Model "L" Manipulators but with some dimensional changes to make them compatible with both the Model "L" and AMF Standard Duty Manipulators and also to incorporate molded-in finger adapters. Two of the boots have been installed in the Metallography Cells for evaluation.

D. Alpha Containment Boxes
(P. A. Mason, C. D. Montgomery)

Two newly procured alpha boxes were received during the quarter. Several deviations from specifications were noted upon receipt. Correction of these discrepancies is to be performed by LASL personnel. An estimate of cost to perform the corrections was submitted to the vendor who has agreed to a reduction in contract cost for these boxes.

Minor redesign work was performed on the 7-in. and 18-in. Alpha Transfer Systems. Work Requests were prepared for fabrication of four units of each system and also for fabrication of other accessory equipment required for the alpha boxes.

E. Microsampling System
(P. A. Mason, C. D. Montgomery)

Development work was resumed during the report period with the design of a special mold for the metallography mount being completed. Fabrication of the mounting frame for the ultrasonic grinder and cross-feed indexing table was also completed. Procurement of the indexing table remote control unit remains to be completed. Preliminary testing of the assembly will commence after fabrication of the mold. Special drive gears for coupling the Slo-Syn motor to the stage drive were received.

F. Bulk Storage Alpha Seal Container
(C. D. Montgomery, P. P. Osvath, J. R. Trujillo)

A special alpha seal container was designed for remote use at DP-West. This container provides the maximum length and inside diameter available for bulk storage of such items as capsule cladding. One unit is being fabricated and should be ready for use by February 1.

G. Fuel Pin Handling System for Betatron Radiography
(C. D. Montgomery, J. C. Jaramillo, J. B. Deal, J. R. Trujillo)

The special radiography cask to accommodate 61-in. fuel pins was completed and delivered during November. A special mechanism for orienting and guiding of the fuel pin was installed in the cask.

Several improvements were made as a result of checking combined operations of the fuel pin handling cart with the radiography cask. However, the Slo-Syn drive with power supply and indexer did not give the desired reliability of performance and travel rate. The system has been changed to provide a simple, manually switched, motor drive with position detents actuating a microswitch. It is anticipated that these modifications will be completed and checked out, and that the system will be available for use during January 1973.

H. Trunnion Stand and Radiography Cask
(C. D. Montgomery, T. Romanik, J. R. Trujillo)

A special, ball-bearing, off-center trunnion stand was designed and built for handling the new radiography cask (Item G, above) in the hot cell corridor at DP-West. The cask will be transported with this stand by means of

a forklift. The stand will be sandblasted, painted, and ready for use by about February 15, 1973.

I. Alpha Tube for Betatron Pin Radiography
(C. D. Montgomery, J. R. Trujillo)

A new alpha seal tube for Betatron pin radiography was designed, fabricated, and tested. This container consolidates small imbedded rods of tungsten to act as fiducial markers for accurate referencing on the radiographs. It also has special machining for indexing with the radiography cask and handling mechanism.

J. Electric Shutter Adapter for Leitz Metallograph
C. D. Montgomery, J. H. Bender)

A special adapter system was designed and fabricated to permit replacement of the manual shutter on the Leitz metallograph with an Ilex Synchro-Electronic shutter to increase the output of the metallograph.

K. In-Cell Equipment
(M. E. Lazarus, T. Romanik, P. P. Osvath)

1. Mechanical Profilometer. Construction is now complete on the new Mechanical Profilometer unit. Checkout and installation should be completed in early February 1973.

2. Electro-Optical Profilometer.
(M. E. Lazarus, T. R. Wilson)

The new Electro-Optical unit (Optron) has arrived and is now being installed and checked out at DP-West. The Data Acquisition Computer Program has been modified and developed to accept data from the Optron system; sufficient data should be available in January to provide a basis for either accepting or rejecting the new system. This option had been provided in the purchase order.

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

A. Etching

A model of an ion gun etcher suitable for hot cell application has been designed, fabricated, and tested. Specific operating conditions are still being investigated.

B. Image Analyses

Image Analysis using the Quantimet 720 and the data reduction code IMAGE is now a routinely available examination capability for both irradiated and unirradiated materials.

Programming for a more generalized particle size distribution analysis has begun.

IV. ANALYTICAL CHEMISTRY

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

The two gamma scanning mechanisms were calibrated using a certified length standard as specified in the quality assurance program. The experimental length values and the certified length values for the Z-motion are compared in Table 401-I.

TABLE 401-I

EXPERIMENTAL AND CERTIFIED LENGTH MEASUREMENTS*

	Scanning Mechanism-1	Scanning Mechanism-2	Certified
#1 ^{60}Co	0.750 in.	0.750 in.	0.7482 in.
#2 ^{60}Co	0.775 in.	0.765 in.	0.7707 in.
Al Rod	9.542 in.	9.553 in.	9.5460 in.
Total	11.067 in.	11.068 in.	11.0649 in.

*The length standard consists of two precisely measured ^{60}Co rods separated by an aluminum rod of accurate length.

These data indicate that the error in length measurements is much less than the ± 0.010 in. that we claim.

The principal computer code, SURVEY, for processing the data stored on magnetic tapes has been expanded to permit the plotting of selected isotopic distributions. This improvement eliminates the need for separate plotting routines by incorporating all the data processing routines into one code. The new subroutine includes an option of self-scaling on the linear and logarithmic plots so that the magnitude of the data does not have to be an input parameter. This option eliminates one of the data processing steps and saves both computing and personnel time.

During this report period, 117 gross gamma and 24 multispectral scans were obtained on 27 irradiated fuel pins. This large volume of routine work severely limited the effort in development and improving gamma scanning methods.

2. Determination of Nitrogen in $(U,Pu)O_2$
(T. K. Marshall and C. H. Ward)

Nitrogen in $(U,Pu)O_2$ is determined in a LECO Nitrox-6 nitrogen analyzer by heating the sample inductively in a carbon crucible to a temperature of 2000° to 2500°C, removing O_2 by trapping as CO_2, and measuring the N_2 chromatographically. Replacement of the induction heater on the LECO unit with a Lepel heater was necessary because of the power loss caused by extending the leads of the analyzer into a gloved box. The power loss limited the attainable temperature to approximately 1700°C. With the new induction heater, temperatures in excess of 2500°C were attained. Blank determinations with the furnace temperature at 2300 to 2400°C averaged 20 ± 2 μg of nitrogen. This average was obtained over several days from 23 blank determinations taken randomly before and after sample analyses. For nine nitrogen standards each containing 57.2 μg of N_2, an average of 5.02 ± 0.31 counts per μg of N_2 was obtained. Analyses of a standard containing 13.1 μg of N_2 showed that the standard was contaminated, and new standards containing three different N_2 levels have been ordered.

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

V. REQUESTS FROM DRDT

A Examination of Irradiated Materials

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

Battelle Memorial Institute: Examinations made on material from BMI are being followed by representatives from LASL. Examinations performed on four irradiated fuel assemblies received on October 11, 1972 are tabulated in Table 401-II.

Gamma scanning was applied to the nondestructive examination of the four fuel pins listed in Table 401-III.

General Electric Company: Examinations performed on irradiated fuel assemblies received on October 11 and 12, 1972 are tabulated in Tables 401-IV and 401-V.

The nondestructive examination of each of three GE fuel pins (FOA, FOC, and E2R) included 4 gross and 1

TABLE 401-II

POSTIRRADIATION EXAMINATIONS OF CAPSULES FROM BMI

Examination	Capsule Identity, BMI
1. Visual Inspection	3-2, 3-3, 3-4, 3-5
2. Preliminary Measurements	Same as Item 1
3. Profilometry-Optical	Same as Item 1
4. Photography, Full Length	Same as Item 1
5. Photography, Incremental	Same as Item 1
6. Radiography	Same as Item 1
7. Gamma Scanning	Same as Item 1
8. Eddy Current	Same as Item 1

TABLE 401-III

GAMMA SCANNING OF BMI FUEL PINS

Fuel Pin No.	No. of Gamma Scans		No. of Isotopic Distributions Calculated
	Gross	Multispectral	
BMI-3-2	4	2	11
BMI-3-3	4	1	13
BMI-3-4	4	0	0
BMI-3-5	4	0	0

TABLE 401-IV

POSTIRRADIATION EXAMINATIONS OF CAPSULES FROM GE

Examination	Capsule Identity
1. Visual Inspection	E2R, FOA, FOC
2. Preliminary Measurements	E2R, FOA, FOC
3. Profilometry, Optical	FOA, FOC
4. Photography, Full Length	E2R, FOA, FOC
5. Photography, Incremental	E2R, FOA, FOC
6. Radiography	FOA, FOC
7. Gamma Scanning	FOA, FOC
8. Cover Gas Sampling	FOA, FOC
9. Na and NaK Removal	E2R, FOA, FOC
10. Clad Removal	E2R, FOA, FOC
11. Profilometry (on capsule cladding)	FOA, FOC
12. Temperature Measurements (on capsule cladding)	FOA, FOC
13. Sectioning (on capsule cladding)	FOA, FOC
a. 2-1/2 in. Samples for Density Measurements	FOA, FOC

TABLE 401-V

POSTIRRADIATION EXAMINATIONS
OF PINS FROM GE

Examination	Pin Identity
1. Visual Inspection	E2R, FOA, FOC
2. Preliminary Measurements	E2R, FOA, FOC
3. Profilometry, Optical	E2R, FOA, FOC
4. Radiography	E2R
5. Gamma Scanning	E2R
6. Photography (full length)w/wire	FOA, FOC
7. Photography (incremental) w/wire	FOA, FOC
8. Photography (full length) w/o wire	E2R, FOA, FOC
9. Photography (incremental) w/o wire	E2R, FOA, FOC
10. Wire Wrap Removal	FOA, FOC
11. Length Measurement	E2R, FOA, FOC
12. Fission Gas Sampling	FOA, FOC
13. Sectioning	E2R, FOA, FOC
14. Photography (sectioned faces)	E2R, FOA, FOC
15. Fuel Removal	FOA, FOC
16. Density Measurements	E2R, FOA, FOC

complete spectral gamma scans. The distributions of 6 and 14 isotopes in the pins were also determined.

Cross-section samples of fuel and clad were dissolved and analyzed for burnup on GE-F12Q and GE-F12P.

The cover gas and pin gas in GE-FOA and GE-FOC were analyzed mass spectrometrically.

Three clad samples from fuel pin GE-FOA and from fuel pin GE-FOC were cleaned and prepared for density measurement.

Microstructural examinations consisting of macrophotography, alpha and beta-gamma autoradiography, optical microscopy (including mosaics), and image analysis were carried out in an Ar atmosphere on specimens as listed in Table 401-VI. A sample was prepared for EMX examination.

Gulf United Nuclear Fuels Corporation: Examinations made on material from GUNC are being followed by representatives from LASL. Sixteen capsules were received on November 6, 1972 following irradiation in EBR-II. The examinations performed are tabulated in Tables 401-VII and 401-VIII.

TABLE 401-VI

MICROSTRUCTURAL ANALYSIS OF GE MATERIALS

		Special Analyses	
GE Pin No.	No. Samples	EMX	IMAGE ANALYSIS
GE-F12P	3	1	1
GE-F12Q	3		1
GE-FOA	2		2
GE-FOC	2		2

TABLE 401-VII

POSTIRRADIATION EXAMINATIONS
OF GU CAPSULES

Examination	Capsule Identity
1. Visual Inspection	UNC-88, -93, -94, -97, -98, -105, -106, -110, -113, -114, -185, -186, -202, -203, -204, -205
2. Preliminary Measurements	Same as Item 1
3. Profilometry, Optical	Same as Item 1
4. Photography, Full Length	UNC-88, -93, -94, -97, -98, -105, -106, -110, -113
5. Photography, Incremental	Same as Item 1
6. Radiography	Same as Item 1
7. Gamma Scanning	UNC-88, -93, -94, -97, -98, -105, -106, -110, -113, -114, -185, -186
8. Eddy Current	UNC-88, -93, -94, -97, -98, -105, -106, -110, -114, -185
9. Cover Gas Sampling	UNC-93, -94, -97, -105, -110

TABLE 401-VIII

POSTIRRADIATION EXAMINATIONS
OF GU PINS

Examination	Pin Identity
1. Sectioning	UNC-187, -189, -194, -206
2. Fuel Removal	UNC-187, -189, -194, -206
3. Na Distillation	UNC-194
4. Na Melting and Pressurizing	UNC-194

The fuel pins listed in Table 401-IX were examined using gamma scanning.

The shielded electron microprobe was used for examining cross-section sample (B) of UNC-194.

TABLE 401-IX

GAMMA SCANNING OF GU PINS

Fuel Pin No.	No. of Gamma Scans Gross	No. of Gamma Scans Multispectral	No. of Isotopic Distributions Calculated
UNC-88	4	0	0
UNC-93	4	0	0
UNC-94	4	1	9
UNC-97	4	0	0
UNC-98	4	1	4
UNC-105	4	1	4
UNC-106	4	1	4
UNC-110	4	1	4
UNC-113	4	1	4
UNC-114	4	1	4
UNC-185	4	0	0
UNC-186	4	0	0

A clad sample from UNC-194 was cleaned and prepared for density measurement.

Microstructural examinations consisting of macrophotography, alpha and beta-gamma autoradiography, and optical microscopy (including mosaics) were carried out in an Ar atmosphere on two samples from UNC-194.

Hanford Engineering Development Laboratory: Eaminations performed on HEDL irradiated fuel assemblies received on February 22, 1972 include the following: sectioning of pins P-17A-5, -29, -31, and -33; and determining atom percent burnup on pins P-17A-17 and -19.

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

Los Alamos Scientific Laboratory: Examinations performed on irradiated fuel assemblies on October 11 and November 6, 1972 are shown in Table 401-XI.

On November 1, 1972 two cladding sections from the fuel area of K-42B were shipped to ANL for examination on an image analyzer.

Six fuel pins were nondestructively examined by obtaining the gamma scans as shown in Table 401-XII.

TABLE 401-X

MICROSTRUCTURAL ANALYSIS OF HEDL MATERIALS

HEDL Pin No.	No. Samples Fuel and Clad	No. Samples Clad	EMX Prep.
P-17A-5	3	2	
P-17A-16	5		1
P-17A-17	5		
P-17A-19	5		
P-17A-20	3		1
P-17A-29	3		
P-17A-31	3		
P-17A-33	3		

TABLE 401-XI

EXAMINATION OF LASL FUEL PINS

Examination	Pin Identity
1. Visual Inspection	K-45, K-46, K-49, K-50, K-51, K-36-B
2. Preliminary Measurements	Same as Item 1
3. Profilometry - Optical	Same as Item 1
4. Photography (Full Length)	Same as Item 1
5. Photography (Incremental)	Same as Item 1
6. Radiography	Same as Item 1
7. Gamma Scanning	Same as Item 1
8. Eddy Current	Same as Item 1

TABLE 401-XII

GAMMA SCANS OF LASL FUEL PINS

Fuel Pin No.	No. of Gamma Scans Gross	No. of Gamma Scans Multispectral	No. of Isotopic Distributions Calculated
LASL-K-45	5	2	17
LASL-K-46	7	1	12
LASL-K-49	3	2	16
LASL-K-50	6	1	11
LASL-K-51	3	0	0
LASL-K-36B	4	0	0

Two cladding samples from LASL K-42-B were mounted and polished in preparation for ion probe work at ANL.

Oak Ridge National Laboratory: Examinations made on material from ORNL are being followed by representatives from LASL. Capsules ORNL-43-N1 and

43-120 were received on October 11, 1972, and the examinations performed are shown in Table 401-XIII.

It was agreed that the destructive examination of ORNL 43-120 would be done at ANL. Arrangements for shipping would be made during January 1973.

Eight gross and five complete spectral gamma scans, along with the calculation of the distributions of 20 isotopes, were obtained in the examination of ORNL-43-120, and 5 gross gamma scans were done on ORNL-43-N1.

Shipments of Materials from Experimenters.

Ten mixed-oxide fuel pins from HEDL are scheduled to arrive at LASL in late January 1973.

Nine mixed-oxide fuel pins (F-8 series) from GE will arrive at LASL about February 15, 1973 for the scheduled examinations.

Approximately nine pins of the advanced fuel type (mixed carbides and nitrides) are scheduled for shipment to LASL in February 1973.

TABLE 401-XIII

POSTIRRADIATION EXAMINATIONS OF ORNL MATERIAL

Examination	Capsule Identity
1. Visual Inspection	43-N1, 43-120
2. Preliminary Measurements	43-N1, 43-120
3. Photography (Full Length)	43-N1, 43-120
4. Photography (Incremental)	43-N1
5. Gamma Scanning	43-N1, 43-120
6. Radiography	43-N1, 43-120*

*In reviewing the radiographs, it was discovered that fuel pin numbered 43-120 is apparently breached. The experimenter and DRDT were notified.

VI. PUBLICATIONS AND TALKS

1. J. W. Dahlby, R. M. Abernathey, M. E. Smith, J. E. Rein, and A. Zerwekh, "Analyses of Irradiated Fuels and Measurements of Interstitials," presented at the 16th Conference on Analytical Chemistry in Nuclear Technology, Gatlinburg, TN, October 24-26, 1972.

2. C. S. MacDougall, M. E. Smith, and G. R. Waterbury, "Remotized Apparatus for Determining Oxygen in Irradiated Reactor Fuels and Cladding Materials," Proc. 20th Conference on Remote Systems Technology, 137-142 (1972).

3. J. R. Phillips, E. A. Hakkila, G. M. Matlack and J. Bubernak, "Special Instrumental Methods of Analysis," presented at the 16th Conference on Analytical Chemistry in Nuclear Technology, Gatlinburg, TN, October 24-26, 1972.

PROJECT 463

HIGH PERFORMANCE LMFBR FUEL MATERIALS

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

I. INTRODUCTION

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

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

II. IRRADIATION TESTING

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

A. Synthesis and Fabrication

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

1. Process Development. In anticipation of producing carbide for a new 19-pin subassembly, efforts were directed toward refining existing procedures and the development of new processes. The sintering conditions previously used called for 8 h at 1800°C followed by 2 h at 1400°C in a flowing Ar atmosphere. Most batches prepared under these conditions met a specification for theoretical density of 95 ± 2,however, a few batches were of low density. These batches have been resintered for 8 h at 1850°C and 2 h at 1400° and showed a marked increase in density. Data pertaining to the resintering of these batches are shown in Table 463-I.

TABLE 463-I

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

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

It can be seen that with but one exception the resintered material met the previously mentioned density specification. Metallographic examination of resintered pellets showed an increased grain size, and no indication of sesquicarbide formation or impurity contamination was observed. Chemical analyses verified these observations.

Routine spectrochemical analyses are capable of reporting Ta in quantities > 1000 ppm. To obtain higher sensitivities much larger samples are required. Two previously prepared batches of mixed carbide were selected at random and sampled for Ta. The reported analyses for each batch was < 100 ppm.

Developmental work was previously reported[1,2] on a carbothermic process for the preparation of high purity, single phase $U_{0.8}Pu_{0.2}C$. Analyses of a production-size batch are incomplete, but the material was found to be single phase and contained only 160 ppm O_2. Pellets produced by the carbothermic process were generally not as dense as pellets previously prepared from arc melted material. To assure that this was a function of the material rather than processing variables, equal masses of an arc melted ingot and a carbothermic reduction biscuit were processed in parallel through the grinding, hydrogen treatment, pressing and sintering procedures. The density of pellets from arc cast material was 94.5% TD as compared to 93.7% TD for pellets from carbothermically reduced material. On repeating the parallel preparation but pressing at a lower pressure, the densities were 92.8 and 91.4% TD respectively. All of the pellets from these preparations were found to be single phase. Additional experiments are needed to determine the effects of varying comminution, pressing and sintering conditions in the carbothermic production process.

2. Equipment Development. A new hydrogen treatment facility was previously described[2] which produced excessive heating of the atmosphere within the recirculating inert atmosphere glovebox. Incorporation of additional furnace insulation and a heat exchanger on the recirculator reduced the temperature within the enclosure but not to an acceptable level. The reaction tube was scaled down to permit using a smaller diameter furnace with less power output. This new equipment has been installed and will be tested in the near future. A similar facility was installed in the nitride preparation glovebox.

A new analytical balance was modified for immersion density measurements. This was installed in a glovebox which will be used exclusively for these measurements. The balance and weights have been calibrated to conform to Quality Assurance standards.

The tungsten mesh nitride sintering furnace was heated to 2000°C at 2×10^{-6} Torr using either manual controls or a programmed controller. Sintering crucibles were ordered and received and a new crucible support was fabricated and installed.

3. Quality Assurance Calibrations. A list was compiled of the various measuring devices associated with fuel preparation and fuel pin fabrication which may need calibration to conform to Quality Assurance standards. In cooperation with the Director of Surveys and Audits for the Measurement Standards Department of the Quality Assurance Department of Sandia Laboratories, Albuquerque, New Mexico, a program was initiated to bring the measuring and test equipment into compliance with the CMB-RDT Quality Assurance Program and RDT Standard F3-2T. The calibration program will involve three phases:

a. Calibrated measuring and test equipment will be obtained either as new items certified to recommended standards, as items calibrated against standards traceable to the U.S. Bureau of Standards in the Standards Laboratories

of LASL or Sandia, or as items calibrated within the project by Quality Assurance Approved procedures. These items will be either used directly or used to calibrate similar measuring and test equipment.

b. Certain pieces of measuring and test equipment require the development of new calibration procedures because of radioactive contamination considerations. These devices will be calibrated in situ with previously calibrated standards and test equipment.

c. Each item of calibrated measuring and test equipment will become a part of the master inventory recall and recalibration Quality Assurance system. Some 215 pieces of measuring and test equipment involving 33 types of calibrations are currently incorporated in the calibration program. Many calibrations are now complete and as standards and approved procedures become available the remaining devices will be calibrated.

B. EBR-II Irradiation Testing

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

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

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

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

Table 463-II describes the K1, K2, and K3 series tests. In these experiments single-phase, high-purity, uranium-plutonium monocarbide pellets are sodium bonded to Type 316 stainless steel cladding. In general, the operating linear power ratings of the capsules are relatively high (approximately 30 Kw/ft). Three tests at very high power (> 45 Kw/ft) were included to determine the effect of high thermal stresses and high fuel temperatures on fuel element behavior. Indications of element failure were found at EBR-II in several experiments from this group, five in subassembly X119B and one from X142, and two from X152, using γ-scanning for ^{133}Xe. Preliminary examinations of these experiments in the LASL hot cells indicate that the capsules are bowed as much as 3/8 in. Further nondestructive examination

TABLE 463-II

SERIES K1, K2, AND K3 CARBIDE EXPERIMENTS[a]

Experiment No.	Series No.	Fuel Type[b]	Fuel Density, % T.D.	Diametral Gap, in.	Max. Fuel Temp. at Startup, °C	Max. Linear Power, Kw/ft	Current Burnup at.%	Status
K-36B	1	$(U_{0.8}Pu_{0.2})C$	90	0.015	1165	30	6.8	NDT-1.7[f]
K-37B	1	$(U_{0.8}Pu_{0.2})C$	90	0.015	1165	30	3.2	NDT-1.7[c]
K-38B	1	$(U_{0.8}Pu_{0.2})C$	90	0.015	1165	30	6.4	NDT(EBR-II)[d,f]
K-39B	1	$(U_{0.8}Pu_{0.2})C$	90	0.015	1165	30	6.4	NDT(EBR-II)
K-40B	1	$(U_{0.8}Pu_{0.2})C$	95	0.020	1150	30	---	To be built
K-41B	1	$(U_{0.8}Pu_{0.2})C$	95	0.020	1150	30	---	To be built
K-42B	1	$(U_{0.8}Pu_{0.2})C$	90	0.015	1165	30	5.0	Completed[e]
K-43	3	$(U_{0.8}Pu_{0.2})C$	95	0.020	1150	30	6.1	NDT(EBR-II)[f]
K-44	3	$(U_{0.8}Pu_{0.2})C$	95	0.020	1150	30	6.1	NDT(EBR-II)
K-45	3	$(U_{0.8}Pu_{0.2})C$	95	0.020	1150	30	3.0	NDT-1.7[f]
K-46	3	$(U_{0.8}Pu_{0.2})C$	95	0.020	1150	30	2.9	NDT-1.7[f]
K-47	3	$(U_{0.8}Pu_{0.2})C$	95	0.020	1150	30	---	To be built
K-48	3	$(U_{0.8}Pu_{0.2})C$	95	0.020	1150	30	---	To be built
K-49	2	$(U_{0.8}Pu_{0.2})C$	95	0.020	1400	45 - 50	4.0	NDT-1.7[f]
K-50	2	$(U_{0.8}Pu_{0.2})C$	95	0.020	1400	45 - 50	4.0	NDT-1.7[f]
K-51	2	$(U_{0.8}Pu_{0.2})C$	95	0.020	1400	45 - 50	3.9	NDT-1.7[f]

[a] All elements are clad in 0.300 in o.d. x 0.280 in. i.d. Type 316SS. All are sodium bonded elements.

[b] The series 1 and 3 experiments are fully enriched in ^{235}U. The series 2 experiments contain 97% ^{233}U. All fuel is single phase.

[c] Capsule K-37B was damaged during reconstitution of X152 to such an extent that it cannot be irradiated further. Failure indicated by γ-scan.

[d] Capsule K-38B was damaged during reconstitution of X152 at EBR-II. Additional irradiation was completed after damage.

[e] Reported in LA-4669-MS.

[f] Failure indicated by γ-scan at EBR-II.

of the failed experiments is currently under way and destructive examination of several is planned for the next quarter.

Table 463-III describes the Series U1300 experiments. These experiments contain two-phase, uranium-plutonium carbide fuel pellets which are helium bonded to either Type 316 stainless steel or Incoloy 800 cladding. Two methods for the accommodation of fuel swelling were investigated in this series, i.e., the introduction of internal porosity by the use of either low-density solid fuel pellets or high-density cored pellets. These experiments reached their goal burnup of 10 at.% in subassembly X-142 after operation at moderate linear power ratings (approximately 20 Kw/ft). Indications of element failure for three experiments were found at EBR-II using γ-scanning for ^{133}Xe. These element failures have been confirmed by γ-scanning for ^{137}Cs at LASL. All of the capsules in the series are currently undergoing nondestructive or destructive examination in the LASL hot cells.

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

TABLE 463-III

SERIES U1300 CARBIDE EXPERIMENTS

Experiment No.	Fuel Type[a]	Fuel Density % T.D.	Clad Material	Clad Thickness, in.	Diametral Gap, in.	Fuel-to-Clad Bond	Max. Fuel Temp. at Startup, °C	Max. Linear Power Kw/ft	Current Burnup at.%	Status
U93	MC + 5 v/o M_2C_3	84	316SS	0.030	0.004	He	1750	18.0	11.1	Des. Exam-1.8
U94	MC + 5 v/o M_2C_3	84	316SS	0.015	0.007	He	1680	21.9	10.7	Des. Exam-1.8
U97	MC + 5 v/o M_2C_3	84	INC-800	0.030	0.004	He	1750	18.0	11.0	Des. Exam-1.8
U98	MC + 5 v/o M_2C_3	84	INC-800	0.015	0.007	He	1680	21.9	10.6	Des. Exam-1.8[c]
U105	MC + 5 v/o M_2C_3	84	INC-800	0.030	0.008	He	1900	15.1	11.5	Des. Exam-1.8
U106	MC + 5 v/o M_2C_3	84	INC-800	0.015	0.009	He	1825	19.8	10.9	NDT - 1.7[c]
U110	MC + 10 v/o M_2C_3	99[b]	INC-800	0.015	0.014	He	1960	21.9	10.1	NDT - 1.7
U113	MC + 10 v/o M_2C_3	99[b]	INC-800	0.030	0.010	He	1880	16.9	11.4	NDT - 1.7
U114	MC + 10 v/o M_2C_3	99[b]	INC-800	0.015	0.007	He	1575	22.1	10.4	NDT - 1.7[c]

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

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

[c] Failure indicated by γ-scan at EBR-II.

During interim examination at EBR-II after run 58, ^{137}Cs was detected by γ-scanning in the sodium reservoir of capsule U-136. Release of fission gas from a breached helium-bonded element would be expected. However, no ^{133}Xe was detected in the capsule plenum. The lack of fission gas in the capsule and the presence of ^{137}Cs in the capsule sodium present a contradictory picture and the failure of the element in capsule 136 can only be considered tentative and of a low degree. None of the other capsules indicated fuel element failure during the examinations at EBR-II. Reinsertion of all nineteen capsules is planned for EBR-II Run 61 so that they can be irradiated to their goal burnups of 10 at.%.

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

Destructive examination of U-187, -189, -192, -194, -195, -197, and -206 has started. Further examination of the remainder of these experiments is planned.

The experiments listed in part B of Table 463-V are awaiting further irradiation. U-188, -190, -193, and -196 were recently removed from subassembly X152. Interim examination, including γ-scanning for ^{133}Xe, indicate that the elements in these experiments have not failed. Further irradiation of the experiments is pending selection of companion capsules from the other high performance experiments to complete a subassembly.

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

TABLE 463-IV

SERIES U1950 CARBIDE EXPERIMENTS

Experiment No.	Fuel Type[a]	Fuel Density % T.D.	Clad Material	Clad Thickness, in.	Diametral Gap, in.	Fuel-To-Clad Bond	Max. Fuel Temp. at Startup, °C	Max. Linear Power Kw/ft	Current Burnup at.%	Status
U129	MC + 5 v/o M_2C_3	84	316SS	0.022	0.016	He	1755	12.8	8.3	X055B (interim)
U130	MC + 5 v/o M_2C_3	75	316SS	0.022	0.010	He	1500	13.1	8.3	X055B (interim)
U131	MC + 5 v/o M_2C_3	84	316SS	0.022	0.010	He	1495	13.1	8.2	X055B (interim)
U132	MC + 5 v/o M_2C_3	84	316SS	0.022	0.010	He	1495	12.8	8.1	X055B (interim)
U133	MC + 5 v/o M_2C_3	84	316SS	0.022	0.010	He	1495	12.8	7.9	X055B (interim)
U134	MC + 5 v/o M_2C_3	84	316SS	0.022	0.010	He	1495	12.8	7.9	X055B (interim)
U135	MC + 5 v/o M_2C_3	84	INC-800	0.022	0.010	He	1475	12.8	8.2	X055B (interim)
U136[d]	MC + 5 v/o M_2C_3	84	INC-800	0.022	0.010	He	1475	13.3	7.8	X055B (interim)
U137	MC + 10 v/o M_2C_3	99	316SS	0.022	0.010	He	1440	13.4	6.9	X055B (interim)
U138A[b]	MC + 10 v/o M_2C_3	99	316SS	0.022	0.010	He	1440	14.8	3.3	X055B (interim)
U139	MC + 10 v/o M_2C_3	99	INC-800	0.022	0.010	He	1440	14.8	7.0	X055B (interim)
U140	MC	93	INC-800	0.022	0.010	He	1460	13.9	7.5	X055B (interim)
U141	MC	93	316SS	0.022	0.010	He	1460	14.3	7.4	X055B (interim)
U142	MC	93	316SS	0.022	0.010	He	1460	14.5	7.5	X055B (interim)
U143	MC + 10 v/o M_2C_3	99[c]	INC-800	0.022	0.010	He	1395	12.8	7.7	X055B (interim)
U144	MC + 10 v/o M_2C_3	99[c]	316SS	0.022	0.010	He	1395	13.1	7.8	X055B (interim)
U145	MC	93	304SS	0.015	0.030	Na	820	13.4	7.3	X055B (interim)
U146A[b]	MC + 10 v/o M_2C_3	99	304SS	0.015	0.030	Na	810	13.7	3.3	X055B (interim)
U147	MC + 10 v/o M_2C_3	99	INC-800	0.015	0.030	Na	810	14.2	7.4	X055B (interim)

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

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

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

[d] Possible failure indicated from γ-scanning at EBR-II.

Table 463-VI describes the Series B-1 and B-2 experiments. These capsules are fueled with single-phase, uranium-plutonium mononitride. All the elements in this series are sodium-bonded and clad with either Type 304 or 316 welded stainless steel tubing. Operating linear power ratings for the experiments are relatively high (25-35 Kw/ft). Capsules B-1-4 and B-2-5 have been examined using γ-scanning techniques for the detection of ^{137}Cs, and both elements are apparently intact. Destructive examination of capsule B-2-8 gave no indication of element failure after 3.6 at.% burnup. The results of the latter examination are reported in BMI-1925.[3] Further irradiation of B-1-4 and B-2-5 is planned. The remaining experiments from this series were recently removed from subassembly X152. During the interim examination, capsules B-1-1, -1-2, -2-2, -2-6, and -2-7 were found to have failed as indicated by γ-scanning for ^{133}Xe at EBR-II. Elements in capsules B-2-1 and B-2-3 were found to have not failed. The failed experiments will be shipped to LASL for destructive examination.

The B-3 series of experiments are described in Table 463-VII. This nitride-fueled series is similar to the B-1 and B-2 series except that three helium bonded experiments are included and the average operating linear power ratings are slightly higher. Gamma-scans made at EBR-II for ^{133}Xe indicated that the four sodium-bonded elements have failed, while the three helium-bonded elements have not failed. Preliminary visual examination at LASL indicates that the four failed elements are bowed as much as 3/8 in. Nondestructive examination of the failed elements is currently under way.

The Series U5100 singly-clad experiments are described in Table 463-VIII. In this group, either single-phase or two-phase carbide fuel is sodium bonded to Type

TABLE 463-V

SERIES U1930 AND U1960 CARBIDE EXPERIMENTS

Experiment No.	Fuel Type[a]	Fuel Density % T.D.	Clad Material	Clad Thickness, in.	Diametral Gap, in.	Fuel-To-Clad Bond	Max. Fuel Temp. at Startup, °C	Max. Linear Power Kw/ft	Current Burnup at.%	Status
					A					
U187	MC + 5 v/o M_2C_3	84	316SS	0.020	0.007	He	1935	30.0	4.5	Destructive Exam-2.9
U189	MC + 5 v/o M_2C_3	84	INC-800	0.020	0.007	He	1935	30.0	4.5	Destructive Exam-2.9
U191	MC	93	304SS	0.015	0.030	Na	1148	31.7	4.7	Destructive Exam-2.6
U192	MC	93	304SS	0.015	0.030	Na	1148	31.7	4.7	Destructive Exam-2.9
U194	MC + 10 v/o M_2C_3	97	304SS	0.015	0.030	Na	1132	33.1	5.0	Destructive Exam-2.9[c]
U195	MC + 10 v/o M_2C_3	97	304SS	0.015	0.030	Na	1132	33.1	5.0	Destructive Exam-2.9
U197	MC + 10 v/o M_2C_3	97	INC-800	0.015	0.030	Na	1132	33.4	5.0	Destructive Exam-2.9
U198	MC + 10 v/o M_2C_3	97	INC-800	0.015	0.030	Na	1132	33.4	5.0	Destructive Exam-2.6
U200	MC + 5 v/o M_2C_3	84	304SS	0.015	0.008	He	2042	30.8	4.6	Destructive Exam-2.6[c]
U206	MC+ 5 v/o M_2C_3	90	316SS	0.020	0.008	He	2084	31.5	4.7	Destructive Exam-2.9
U208	MC + 10 v/o M_2C_3	97[b]	316SS	0.020	0.009	He	1912	31.9	4.8	Destructive Exam-2.6
					B					
U188	MC + 5 v/o M_2C_3	84	316SS	0.020	0.007	He	1935	30.0	8.7	NDT (interim)
U190	MC + 5 v/o M_2C_3	84	INC-800	0.020	0.007	He	1935	30.0	8.8	NDT (interim)
U193	MC	93	304SS	0.015	0.030	Na	1148	31.7	8.7	NDT (interim)
U196	MC + 10 v/o M_2C_3	97	304SS	0.015	0.030	Na	1132	32.6	8.6	NDT (interim)
U199	MC + 10 v/o M_2C_3	97	INC-800	0.015	0.030	Na	1132	33.5	5.0	Interim
U201	MC + 5 v/o M_2C_3	84	304SS	0.015	0.008	He	2042	30.0	4.5	Interim
U207	MC + 5 v/o M_2C_3	90	316SS	0.020	0.008	He	2088	31.7	4.7	Interim
U209	MC + 10 v/o M_2C_3	97[b]	316SS	0.020	0.009	He	1909	30.9	4.6	Interim
					C					
U185	MC + 10 v/o M_2C_3	96	316SS	0.020	0.011	He	2195	30.0	3.0	NDT-1.7
U186	MC + 10 v/o M_2C_3	96	316SS	0.020	0.011	He	2195	30.0	3.0	NDT-1.7
U202	MC + 5 v/o M_2C_3	84	316SS	0.010	zero	He	1270	31.7	2.8	NDT-1.7
U203	MC + 5 v/o M_2C_3	84	316SS	0.020	zero	He	1260	31.4	2.8	NDT-1.7
U204	MC + 10 v/o M_2C_3	97[b]	316SS	0.010	zero	He	1131	32.2	2.9	NDT-1.7
U205	MC + 10 v/o M_2C_3	97[b]	316SS	0.020	zero	He	1124	31.9	2.9	NDT-1.7
					D					
U260	MC + 10 v/o M_2C_3	98	316SS[d]	0.015	0.015	He	2590	34.1	---	At EBR-II
U261	MC + 10 v/o M_2C_3	98	316SS[e]	0.015	0.015	He	2590	34.1	---	At EBR-II
U262	MC + 10 v/o M_2C_3	98	INC-800	0.015	0.015	He	2590	34.1	---	At EBR-II

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

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

[c] Elements 194 and 200 have failed.

[d] 20% cold-worked.

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

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. Experiment U255 has been rejected due to a tungsten inclusion in the bottom weld. That experiment will be replaced with a stainless steel dummy element. Insertion of the experiments into the reactor is pending completion of the quality assurance data package by the Gulf United Nuclear Fuels Corporation, the original experimenter.

The C-5 series of singly-clad experiments is described in Table 463-IX. Single-phase nitride fuel is sodium bonded to 20% cold-worked Type 316 stainless steel cladding in all of the fuel elements in this group. Profilometry measurements of the C-5 series elements have been made using the same equipment that will be used for the postirradiation examination. Shipment

TABLE 463-VI

SERIES B-1 AND B-2 EBR-II NITRIDE EXPERIMENTS[a]

Experiment No.	Fuel Density, % T.D.	Smear Density, T.D.	Cladding Wall Thickness, in.	Max. Linear Power, Kw/ft	Current Burnup at.%	Status
B-1-1	94.1	80.2	0.020	27.9	6.5	NDT (EBR-II)
B-1-2	93.9	80.5	0.020	27.1	6.3	NDT (EBR-II)
B-1-4	93.8	84.9	0.020	28.6	3.0	NDT-1.7
B-2-1	94.7	81.5	0.020	32.7	6.3	NDT (Interim)
B-2-2	94.4	81.5	0.020	32.5	6.2	NDT (EBR-II)[b]
B-2-3	94.0	81.0	0.020	32.4	6.3	NDT (Interim)
B-2-5	94.2	75.5	0.015	32.4	3.0	NDT-1.7
B-2-6	94.0	81.8	0.010	36.6	6.2	NDT (EBR-II)[b]
B-2-7	93.9	81.7	0.010	36.7	6.1	NDT (EBR-II)[b]
B-2-8	93.8	81.6	0.010	36.5	3.6	Complete

[a] All capsules are sodium bonded. Capsule cladding is 0.375 in. o.d. x 0.335 in. i.d. Type 304 stainless steel.
Element cladding for the B-1 experiments is welded 0.290 in. o.d. Type 304 stainless steel.
Element cladding for the Series B-2 experiments is welded 0.315 in. o.d. Type 316 stainless steel.
The fuel is $(U_{0.8}Pu_{0.2})N$.

[b] Failure indicated by ^{133}Xe γ-scan at EBR-II.

TABLE 463-VII

SERIES B-3 EBR-II NITRIDE EXPERIMENTS[a]

Experiment No.	Fuel Density, % T.D.	Bond Type	Smear Density, % T.D.	Cladding Wall Thickness, in.	Max. Linear Power, Kw/ft	Current Burnup at.%	Status
B-3-2	88.4	Na	82.6	0.015	37.7	3.1	NDT-1.7[c]
B-3-3	91.3	Na	85.2	0.015	38.9	3.1	NDT-1.7[c]
B-3-4	93.9	Na	85.4	0.015	38.9	3.1	NDT-1.7[c]
B-3-5	90.4	Na	84.8	0.010	41.5	3.1	NDT-1.7[c]
B-3-6	94.8	He[b]	85.5	0.020	34.2	3.0	NDT (Interim)
B-3-7	88.5	He	87.9	0.020	34.2	3.0	NDT (Interim)
B-3-8	89.8	He[b]	85.5	0.020	32.4	2.9	NDT (Interim)

[a] Capsule claddings are type 304 stainless steel, 0.375 in. o.d. x 0.335 in. i.d. Element claddings are 0.315 in. o.d.

[b] Pellets are annular with a 0.070-in. diameter hole.

[c] Failed.

TABLE 463-VIII

SERIES U5100 CARBIDE EXPERIMENTS

Experiment No.	Fuel Type[a]	Fuel Density, % T.D.	Clad Material[b]	Shroud	Diametral Gap, in.	Max. Fuel Temp. at Startup, °C	Max. Linear Power, Kw/ft	Status
U241	MC	93	304SS	None	0.015	1175	35.8	At EBR-II Awaiting insertion into X156
U242	MC	93	304SS	None	0.015	1175	35.8	At EBR-II Awaiting insertion into X156
U243	MC	93	304SS	None	0.030	1150	33.8	At EBR-II Awaiting insertion into X156
U244	MC	93	304SS	None	0.015	1175	35.8	At EBR-II Awaiting insertion into X156
U245	MC	93	304SS	None	0.030	1150	33.8	At EBR-II Awaiting insertion into X156
U246	MC	93	316SS	None	0.015	1190	36.4	At EBR-II Awaiting insertion into X156
U247	MC	93	316SS	None	0.030	1150	33.8	At EBR-II Awaiting insertion into X156
U248	MC	93	316SS	None	0.030	1140	36.4	At EBR-II Awaiting insertion into X156
U249	MC	93	INC-800	None	0.015	1210	36.4	At EBR-II Awaiting insertion into X156
U250	MC	93	INC-800	None	0.030	1145	36.4	At EBR-II Awaiting insertion into X156
U251	MC	93	304SS	None	0.030	1145	36.4	At EBR-II Awaiting insertion into X156
U252	MC	93	304SS	Vanadium-slots	0.030	1140	36.4	At EBR-II Awaiting insertion into X156
U253	MC	93	304SS	Iron-slots	0.030	1145	33.8	At EBR-II Awaiting insertion into X156
U254	MC	93	304SS	304SS-slots	0.030	1140	33.8	At EBR-II Awaiting insertion into X156
U255	MC	93	304SS	304SS-holes	0.030			At EBR-II - Rejected W inclusion in bottom weld
U256	MC + 10 v/o M_2C_3	98	304SS	Vanadium-slots	0.030	1140	34.0	At EBR-II Awaiting insertion into X156
U257	MC + 10 v/o M_2C_3	98	INC-800	Tantalum-slots	0.030	1135	33.5	At EBR-II Awaiting insertion into X156
U258	MC + 10 v/o M_2C_3	98	304SS	304SS-slots	0.030	1145	33.5	At EBR-II Awaiting insertion into X156
U259	MC + 10 v/o M_2C_3	98	INC-800	304SS-slots	0.030	1150	34.6	At EBR-II Awaiting insertion into X156

of selected elements to EBR-II is pending receipt of these documents and a LASL review of the experiments from a quality assurance standpoint.

The O-N1 series of singly-clad experiments is described in Table 463-X. These experiments are similar to the C-5 series. The elements are fueled with $(U_{0.8}Pu_{0.2})N$ which is sodium bonded to 20% cold-worked Type 316 stainless steel cladding. Three elements have been rejected because of large fuel chips in the sodium annulus. The diameters of the elements have been measured on the same profilometer that will be used after irradiation. The status of shipment of these experiments to EBR-II is the same as for the C-5 series. Four of the fuel pins from Series O-N1 will be irradiated with fifteen pins from Series C-5.

The tentative description of the K4 series of experiments is shown in Table 463-XI. One of the primary purposes of the K4 Series is to provide data for a critical comparison of the overall irradiation behavior of carbide and nitride fuel elements which have been irradiated under conditions that are, as nearly as possible, identical. The test parameters include fuel type, cladding cold-work, smear density, linear power rate, operating temperature, fuel restraint, and burnup. The fuel used in the

TABLE 463-IX

SERIES C-5 EBR-II NITRIDE EXPERIMENTS[a]

Experiment No.	Fuel Density, % T.D.	Smear Density, % T.D.	Max. Fuel Temp. at Startup, °F (°C)	Max. Clad Temp., °F (°C)	Max. Linear Power, Kw/ft	Status
C-5-1	92.6	79.4	--	--	--	Reject; chips, air in plenum
C-5-2	93.3	79.4	--	--	--	Reject; chips, air in plenum
C-5-3	94.0	79.9	--	--	--	Reject; chips, air in plenum
C-5-4	95.1	80.9	2112 (1156)	1326 (719)	33.2	At LASL for NDT
C-5-5	95.4	81.6	----------SPARE----------			At LASL for NDT
C-5-6	93.3	79.3	2116 (1158)	1328 (720)	33.3	At LASL for NDT
C-5-7	94.4	80.5	2164 (1184)	1336 (724)	33.7	At LASL for NDT
C-5-8	94.3	74.4	2061 (1127)	1154 (623)	32.7	At LASL for NDT
C-5-9	94.2	80.2	2071 (1133)	1203 (651)	33.5	At LASL for NDT
C-5-10	94.1	80.2	2021 (1105)	1157 (625)	32.6	At LASL for NDT
C-5-11	93.9	80.6	2050 (1121)	1165 (629)	33.4	At LASL for NDT
C-5-12	94.2	74.1	2050 (1121)	1191 (644)	32.5	At LASL for NDT
C-5-13	95.5	75.2	2036 (1113)	1154 (623)	32.1	At LASL for NDT
C-5-14	95.7	75.4	2087 (1142)	1312 (711)	32.0	At LASL for NDT
C-5-15	95.2	75.4	2036 (1113)	1188 (642)	32.1	At LASL for NDT
C-5-16	95.6	75.4	----------SPARE----------			At LASL for NDT
C-5-17	95.7	75.5	--	--	--	Reject; leaked, air in plenum
C-5-18	93.9	79.6	2039 (1115)	1193 (645)	32.6	At LASL for NDT
C-5-19	93.6	78.9	2054 (1123)	1200 (649)	33.0	At LASL for NDT
C-5-20	94.7	80.6	2024 (1107)	1155 (624)	32.4	At LASL for NDT

[a] All elements are sodium bonded. The fuel is $(U_{0.8}Pu_{0.2})N$ pellets. The cladding is 20% cold-worked Type 316 stainless steel, 0.310 in. o.d. by 0.280 in. i.d.

TABLE 463-X

SERIES O-N1 EBR-II NITRIDE EXPERIMENTS [a]

Experiment No.	Fuel Density, % T.D.	Smear Density % T.D.	Temp. at Startup, °F (°C)	Max. Clad Temp., °F (°C)	Max. Linear Power, Kw/ft	Status
O-N1-1	89.9	77.7	2085 (1140)	1200 (649)	32.9	At LASL for NDT
O-N1-2	90.3	78.0	2043 (1117)	1325 (718)	33.1	At LASL for NDT[b]
O-N1-3	90.0	77.8	2128 (1164)	1323 (717)	32.6	At LASL for NDT
O-Ni-4	89.6	77.4	2133 (1167)	1323 (717)	32.8	At LASL for NDT
O-Ni-5	90.4	78.1	2068 (1131)	1160 (627)	32.9	At LASL for NDT[b]
O-N1-6	89.3	77.2	---------- SPARE ----------			At LASL for NDT[b]
O-N1-8	89.6	77.4	---------- SPARE ----------			At LASL for NDT

[a] All elements are sodium bonded. The fuel is $(U_{0.8}Pu_{0.2})N$ pellets. The cladding is 20% cold-worked. Type 316 stainless steel, 0.310 in. o.d. by 0.280 in. i.d.

[b] Elements O-N1-2, -5, and -6 have large fuel chips in the pellet-cladding annulus.

TABLE 463-XI

SERIES K4 CARBIDE EXPERIMENTS[a]

Experiment No.	Fuel Type[b]	Smear Density, %	Cladding[c]	Shroud	Peak Cladding Temp., °F (°C)	Approximate Peak Centerline Temp., °F (°C)	Goal Burnup, at.%)
52	C	82	SA	Yes	1060 (570)	1900 (1040)	7
53	C	82	SA	Yes	1170 (630)	2000 (1100)	11
54	C	82	SA	No	1170 (630)	2000 (1100)	11
55	C	82	SA	No	1060 (570)	1900 (1040)	7
56	C	82	SA	Yes	1160 (625)	2000 (1100)	11
57	C	82	CW	Yes	1050 (565)	1900 (1040)	11
58	C	82	CW	Yes	1160 (625)	2000 (1100)	11
59	C	82	CW	No	1050 (565)	1900 (1040)	11
60	C	85	SA	No	1050 (565)	1900 (1040)	7
61	C	85	CW	No	1060 (570)	1900 (1040)	11
62	N	82	SA	No	1050 (565)	2400 (1315)	7
63	N	82	CW	No	1170 (630)	2500 (1375)	11
64	N	82	SA	Yes	1170 (630)	2500 (1375)	11
65	N	82	CW	Yes	1180 (640)	2500 (1375)	7
66	N	82	CW	Yes	1070 (575)	2400 (1315)	11
67	N	82	SA	Yes	1080 (580)	2400 (1315)	11
68	N	82	SA	Yes	1050 (565)	2400 (1315)	11
69	N	85	SA	No	1070 (570)	2400 (1315)	11
70	N	85	CW	No	1050 (565)	2400 (1315)	7

[a] Heating rates will be in the range 38 to 40 Kw/ft. All elements sodium bonded.

[b] C = 95% dense $(U_{0.8}Pu_{0.2})C$, 93% enriched ^{235}U.
N = 95% dense $(U_{0.8}Pu_{0.2})N$, 93% enriched ^{235}U.

[c] SA = solution annealed Type 316 stainless steel. CW = 20% cold worked Type 316 stainless steel. Both types are 0.310 in. o.d. by 0.286 in. i.d.

fabrication of these elements will be 95% dense, single-phase $(U_{0.8}Pu_{0.2})C$ or $(U_{0.8}Pu_{0.2})N$. The carbide fuel will be fabricated at LASL from material synthesized using the arc-melting process. Material prepared by the carbothermic reduction process may also be used. The nitride fuel for the initial loadings will be supplied by Battelle Memorial Institute and will be prepared using the hydride-nitride process. BMI has produced 15 batches of fuel of which 4 batches have been received. The cladding tubing will be Type 316 stainless steel 0.310 in. o.d. with a wall thickness of 0.012 in. The cladding was purchased through HEDL from Superior Tube Co. Both solution annealed and 20% cold worked tubing were purchased. The tubing complies, as nearly as possible, to the fuel element cladding specification RDT-E13-8. End plug material has been received from HEDL. The 20% cold-worked, Type 316 stainless steel for the end plugs is a portion of the batch of bar stock fabricated for the FFTF control rod structural components and complies to RDT-M-7-23T. Hold-down springs will be made from 0.031 in. diam, Type 302 stainless steel, FFTF driver fuel element spacer wire material.

Shroud tubes with perforated slots are planned for use as a fuel fragment restraint mechanism in about 10 of

the Series K-4 fuel elements. The actual number of elements that will use shrouds will depend on the analysis of the results for the sodium-bonded elements from the encapsulated experiments currently being examined. The suggested shroud tube geometry was selected after considering several wire restraint systems, tubular restraint systems, and packing restraint systems. The suggested shroud tube is:

1. perforated with slots,
2. slot axial pitch, 0.125 in.,
3. slot length, 0.100 in.,
4. slot width, 0.020 in.,
5. twenty-four slots per tube revolution,
6. adjacent circumferential rows of slots staggered axially by 1/3 the axial pitch,
7. circumferential ligament between slots, 0.015 in.,
8. axial ligament between slots, 0.025.,
9. tube dimensions, 0.2675 in. x 0.2615 in.,
10. slot area, 43.8%,
11. tube material, 25% cold-worked Type 316 stainless steel,
12. method of slot fabrication, Electro Discharge Machine (EDM).

In addition to the experiments described above, two nitride fueled thermal irradiation experiments from ORNL (43N1 and 43N2) and eight carbide fueled EBR-II experiments from Westinghouse will be examined. Results and status will be reported in future reports pending receipt of experiment description and history.

Engineering Test Plans have been prepared in order to satisfy the Quality Assurance requirements for the irradiation and examination for the experiments in the K1, K2, K3, U1300, U1950, U1930, U1960, U5100, B-1, B-2, B-3, C-5 and O-N1 series tests. A major effort is currently being expended in the overall evaluation of all of the high performance irradiation experiments under LASL direction. All available data on each experiment is being cataloged and evaluated with respect to both technical characterization and quality control completeness.

C. TREAT Irradiation Testing

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

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

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

a. Test LASL-UL-1 and LASL-UL-2. The TREAT capsules for these tests were shipped to the TREAT reactor in early October. The capsule for test LASL-UL-1 was loaded into the reactor and a calibration transient (TREAT transient number 1476) was run on November 15. The object of the calibration transient was to experimentally determine a figure of merit for these tests by the heat balance method. The figure of merit is defined as the ratio of the power density in the fuel to the reactor power. This quantity is calculated from the total reactor energy deposited in

TABLE 463-XII

LASL SERIES UL TESTS

	TEST			
	LASL-UL-1	LASL-UL-2	LASL-UL-3	LASL-UL-4
Fuel Element[a]	263 (138 A)	264 (146 A)	265 (136)	266 (146)
Fuel Material[b]	90 vol% $(U_{0.85}Pu_{0.15})C$ + 10 vol% $(U_{0.85}Pu_{0.15})_2C_3$			
Fuel Pellet o.d., in.	0.246	0.240	0.246	0.240
Bond Material	He	Na	He	Na
Bond Thickness (Radial), in.	0.005	0.015	0.005	0.015
Clad Material	316SS	304SS	316SS	304SS
Clad Thickness, in.	0.022	0.015	0.022	0.015
Smear Density, % Theoretical	90	77	90	77
Fuel Column Length, in.	13.75 ± 0.125			
Burnup, MWD/MTM[c]	0	0	45,000	45,000
Test Objective	Fuel Melting	Fuel Melting	Same Transient as 263	Same Transient as 264

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

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

[c] Irradiated in EBR-II at 10 to 15 Kw/ft in subassembly X-055.

the fuel and inner capsule as determined by the temperature rise of the inner capsule. The TREAT capsule-inner capsule system is designed so that the inner capsule loses little heat over a time period required to equilibrate temperatures radially in the inner capsule over the fuel column length.

The inner capsule thermocouples indicated spurious signals during this calibration transient. These thermocouples have ungrounded junctions and these signals were traced to the fact that these thermocouples had not been externally grounded prior to the transient.[4] The thermocouples were grounded and a second calibration transient (TREAT transient number 1477) was run. This calibration transient generated 82 MW-sec of reactor energy using a computer controlled transient. Figure 463-1 shows a plot of reactor power as a function of time for this transient. The temperature rise of the inner capsule, adjusting for heat losses, was 152°F. The energy deposited in the inner capsule was calculated from the heat capacity of the inner capsule components as 467.5 cal/cm of length. If all this energy were generated in the fuel the figure of merit would be 7.75×10^{-5} W/(cc fuel) (W reactor power). This estimate would ignore γ heating in the nickel heat sink and stainless steel capsule components which must be considered to accurately estimate the figure of merit due to fission heating in the fuel. Using an estimate of γ heating as 2.51×10^{-6} W/(cc) (W reactor power)^5, the figure of merit due to fission heating in the fuel was calculated as 7.34×10^{-5} W/(cc fuel) (W reactor power). This is 20% below the calculated value of 9.17×10^{-5} reported previously.[2]

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

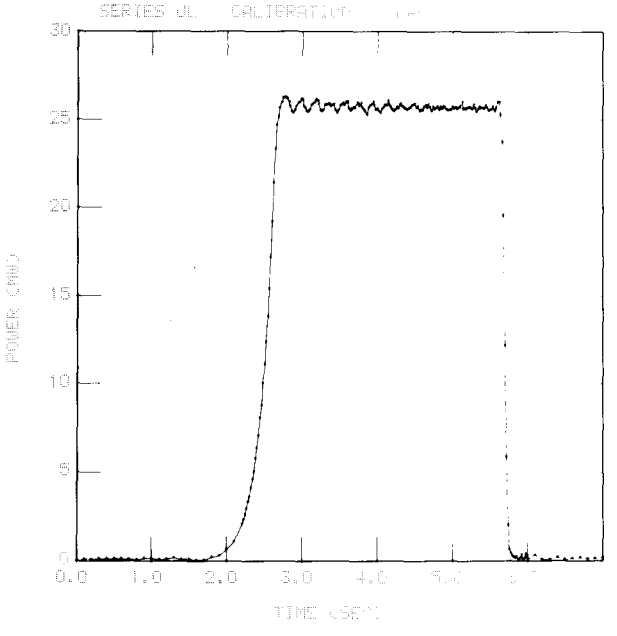

Fig. 463-1. TREAT reactor power as a function of time for calibration transient for Test UL-1.

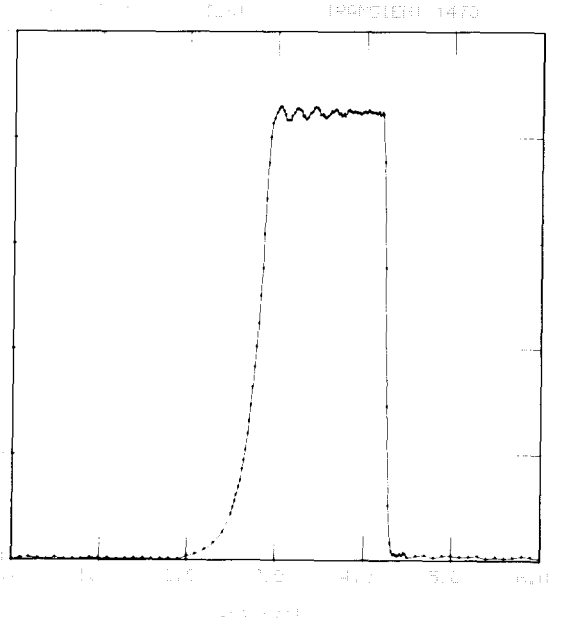

Fig. 463-2. TREAT reactor power as a function of time for test transient for Test UL-1.

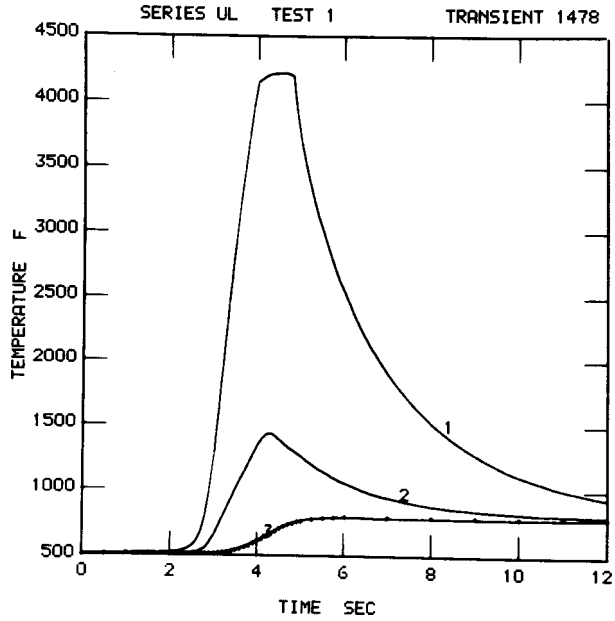

Fig. 463-3. Temperature as a function of time for Test UL-1.
Legend: Curve 1 = calculated fuel center, Curve 2 = calculated clad inside surface, Curve 3 = calculated thermocouple location, * = measured inner thermocouple temperature.

to the start of fuel melting and the end of fuel melting (complete resolidification) at the fuel center. The maximum clad temperature of 1435°F occurs at 4.3 sec.

Figure 463-4 shows an expanded plot of the calculated and measured thermocouple temperatures. Since the

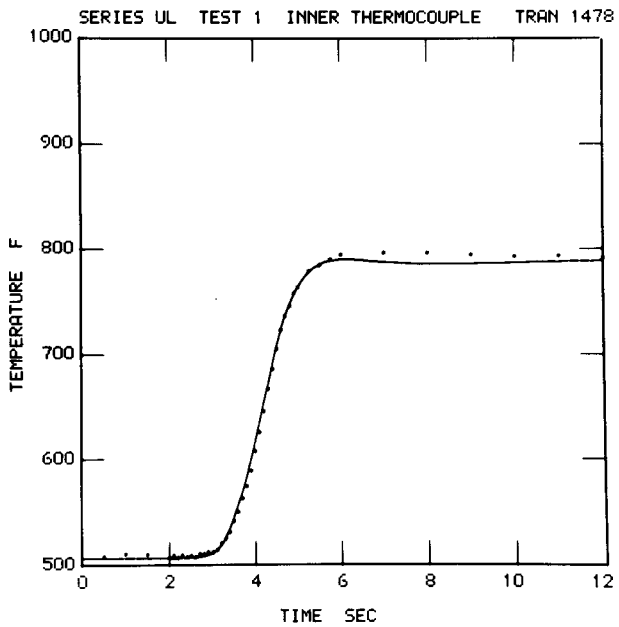

Fig. 463-4. Inner thermocouple temperature as a function of time for test UL-1
Legend: Curve = calculated temperature, * = measured temperature.

thermocouples in the inner capsule have ungrounded junctions, the actual location of the thermocouple junction relative to the end of the thermocouple sheath and the response time of the thermocouple junction must be considered. Thermocouple structure (junction location, junction size, sheath thickness, insulation thickness, and wire thickness) was determined from radiographs of the actual thermocouples used. The calculated thermocouple junction temperatures shown in Figs. 463-3 and 463-4 were obtained from numerical heat transfer calculations on a model of the thermocouple using dimensions and properties of the sheath, insulation, and wires. Sheath surface temperatures were assigned as the temperature of the surrounding medium (NaK in the inner annulus or nickel in the heat sink) at that radial location. The time constant of the model thermocouple was calculated as approximately 300 μsec which is in good agreement with measurements on similar thermocouples.[6] Spare thermocouples are being obtained from Gulf United so that actual measurements of the thermocouple response time may be made.

Neutron radiographs of the TREAT capsule taken before and after test UL-1 have been compared. No significant change in the fuel can be seen after the test. This TREAT capsule is awaiting shipment back to LASL for further examination.

The TREAT capsule for test UL-2 was loaded into the reactor on November 17 but the test was not run owing to unfavorable weather conditions through November 27. At that time the capsule was unloaded from the reactor and placed in storage because of higher priority tests at TREAT.

b. Tests LASL-UL-3 and LASL-UL-4. The assembly of the inner capsules for tests 3 and 4 of the UL Series was done by GUNFC up to the point of insertion of the fuel elements into the inner capsules. Since the fuel elements were pre-irradiated in EBR-II, the completion of the assembly of the inner capsules and the assembly of the inner capsules into the TREAT capsules requires hot cell facilities. LASL, in conjunction with RDT, has arranged for the completion of the assembly by the Radiometallurgy Group of Hanford Engineering Development Laboratory. LASL is continuing a discussion of the assembly of these tests with HEDL. Welding samples for use at HEDL during the assembly of the capsules have been fabricated.

2. Series 1 Tests. A group of eight tests using LASL fabricated fuel elements has been designated LASL Series 1 tests. The tests are designed to determine if any significant safety related behavioral problems exist for sodium bonded, stainless steel clad (U,Pu)C and (U,Pu)N fuels by defining failure thresholds and the types of failure experienced by these fuels. Table 463-XIII summarizes the test parameters and objectives. Approval-in-principle has been received from the AEC for this series of tests. Design work for the modification of TREAT capsules to incorporate a thermal neutron filter and calculations for a preliminary safety analysis are continuing. A preliminary Engineering Test Plan for the Series 1 test has been prepared.

3. Post-Irradiation Disassembly Facilities. Facilities for a post-irradiation disassembly of TREAT tests are being prepared. These facilities will allow the disassembly of TREAT capsules and inner capsules containing fuel elements with no other irradiation except the TREAT irradiation. Two existing evacuable glove boxes will be modified to allow the transfer of failed capsules between boxes and disassembly through sectioning of the fuel element in preparation for metallography. Drawings for the modifications of the two boxes are in preparation.

TABLE 463-XIII

LASL SERIES 1 EXPERIMENTS

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

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

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

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

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

III. FUEL PROPERTY MEASUREMENTS

Differential Thermal Analysis

(J. G. Reavis, R. Basinger)

A sample of irradiated UO_2-25% PuO_2 supplied by G.E. -Sunnyvale was examined recently by differential thermal analysis techniques. The sample was described by G.E. as having an O/M ratio of 1.97 and a burnup of 7.6 atomic percent. It was sealed in a tungsten DTA capsule by TIG welding, using a 0.004-in.-thick tantalum shim between the lid and capsule wall to reduce grain growth during welding.

Thermal arrests observed for this sample are listed in Table 463-XIV. The capsule apparently remained intact through three partial or total melting cycles, but ruptured after the fourth melting. The arrests indicate major sample melting begins on heating through 2700°C and freezing begins on cooling through 2760°. The degree of accuracy of these results has not been established. The temperatures were obtained by applying the same correction curve as has been used during the past two years. A recent observation of an Ir standard sample has indicated

TABLE 463-XIV

THERMAL ARREST TEMPERATURES OBSERVED FOR IRRADIATED UO_2-25% PuO_2 SAMPLE E1H-29A

Cycle No.	Max. Temp, °C	Arrest Temp., °C	
		Heating	Cooling
5	2745	2705	> 2745[(a)]
6	2880	2730	2770
7	2835	2685	2755
8	2785	2685	(b)
Avg.	--	2700	2760

(a) The thermal analysis curves indicated melting was not completed at the maximum temperature reached (2745°) in Cycle 5.

(b) The capsule apparently leaked and the furnace arced at 2785° so that no cooling curve was obtained.

The corrections used to obtain these temperatures should be about 20°higher, i.e., the best average values in Table 463-XIV should be 2720°C and 2780°C. Additional calibration standards will be observed to check the correction curve.

IV. ANALYTICAL CHEMISTRY

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

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

Chemical characterization of stainless steels for FFTF Program applications requires reliable measurements of several specified impurities including Ta. Modifications were made to an existing solvent extraction method for separation of Ta from stainless steel[7] to permit final measurement of Ta by x-ray fluorescence. This separation involves extraction of Ta into hexone from a 6M HCl-0.8M HF mixture, followed by a second extraction from 6M H_2SO_4-0.4M HF. Relative standard deviations ranged between 6 and 29 percent for the determination of 110 to 13 μg of Ta in 250-mg samples of stainless steel. When separations were attempted using 500-mg samples of stainless steel to improve sensitivity, recoveries of Ta were erratic because of coextraction of Fe. To minimize coextraction 5M H_2SO_4-HF systems[8] were investigated for both extractions. A study of the effect of HF concentration during the first extraction on the recovery of Ta showed that optimum recovery was obtained at approximately 1M HF. (Table 463-XV).

TABLE 463-XV

EFFECT OF HF CONCENTRATION ON EXTRACTION OF Ta FROM STAINLESS STEEL (50-mg SAMPLES)

HF, M	Ta, μg	Ta Recovery, %	Rel. Std. Dev., %
0.4	500	90	5
1.0	500	100	4
2.0	500	95	6
4.0	500	86	15

At lower HF concentrations the fluoride ion is apparently complexed by Fe and Cr and reduced in concentration below the level needed to extract Ta quantitatively. At higher HF concentrations, appreciable amounts of Fe and Cr are coextracted with the Ta and interfere with the final x-ray measurements. For these reasons, the extraction procedure was modified to use 5 M H_2SO_4-1M HF in the first extraction and 5 M H_2SO_4-0.4M HF in the second extraction.

The relative standard deviation of the measurement was determined for amounts of Ta between 500 and 1000 μg to be 4.6%. The data indicate that as little as 5 or 10 μg of Ta can be measured. The precision at lower Ta concentrations and the effects of possible interfering elements are being determined.

2. Analytical Chemistry Quality Assurance Program

(J. W. Dahlby)

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

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

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PROJECT 472

ANALYTICAL STANDARDS FOR FAST BREEDER REACTOR OXIDE FUEL

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I. INTRODUCTION

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

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

II. ANALYTICAL CHEMISTRY PROGRAM FOR BORON CARBIDE

A program equivalent to that established for the production of FFTF mixed oxide fuel is being set up for the production of FFTF boron carbide pellets.

A. Status of Analytical Methods and Qualification of Analytical Laboratories

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

The second round robin with vendor participation for the chemical analysis of boron carbide was completed. Participating laboratories were three industrial companies (Carborundum Company, Eagle-Pitcher Industries, Inc., and GTE Sylvania, Inc.) and two AEC contractors (HEDL and LASL). Analytical measurements were evaluated for seven specifications: (1) total boron, (2) hydrochloric acid soluble boron, (3) nitric acid soluble boron, (4) soluble carbon, (5) chloride, (6) fluoride, and (7) metallic impurities.

A detailed report covering the statistical evaluation of the results and technical discussions of the analytical methods was issued November 13, 1972. The general conclusion was that the within-laboratory measurement standard deviations for the seven measurements were satisfactory for FFTF specification use. It is expected that the between-laboratory differences will decrease to acceptable levels by the use of common calibration materials (see next section) in the vendor and HEDL analytical laboratories.

The first round robin with vendor participation indicated that the measurements for total carbon and the $^{10}B/^{11}B$ isotopic ratio were acceptable. As analyses for the additional seven specifications listed above are now satisfactory, all chemical measurements for the specification analysis of FFTF boron carbide are considered adequate. Further round robins are not planned at this time.

B. Preparation of Calibration Materials and Quality Control Samples

(J. V. Pena, O. R. Simi, H. J. Kavanaugh, J. E. Rein)

The preparation of these materials, in amounts considered adequate for use by vendor and HEDL laboratories for the production of 150,000 FFTF boron carbide pellets, was completed. Chemical analyses verified makeup values and homogeneity of all materials. All the materials were sent to HEDL accompanied by a detailed report that listed concentration for all materials and the recommended control value limits for quality control materials.

C. Status of RDT Standards

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

The draft of RDT Standard F11-2 "Analytical Chemistry Methods for Control Rod Absorber Material" was reviewed by RDT with relatively minor editorial changes requested. HEDL submitted RDT Standard F2-8 "Qualification and Control of Analytical Chemistry Laboratories for Control Rod Absorber Material Analysis" for RDT review.

D. Studies and Improvements of Analytical Methods

(W. H. Ashley, R. G. Bryan, R. D. Gardner, A. L. Henicksman, and C. H. Ward)

1. Determination of F and Cl. The pyrohydrolysis of B_4C and subsequent analysis of the distillate for F by specific ion electrode and for Cl by spectrophotometric measurement were investigated to determine the effect of adding H_3BO_3 to the H_2O in the pyrohydrolytic receiver. The H_3BO_3 did not affect the determination of Cl, but it caused a negative bias of 40 to 50% in the measurement of F. This negative bias was much larger than the -14% bias that resulted when known amounts of F were pyrohydrolyzed from U_3O_8 into H_2O and measured relative to solutions containing known amounts of F added directly to H_2O. If the H_3BO_3 was then added to saturate the solution in the receiver, the results for F were high by approximately 80% when measured relative to saturated H_3BO_3 solutions to which known amounts of HF were added directly. Addition of H_3BO_3 to the distillate did not affect the measurement of Cl.

Pyrohydrolysis of known amounts of F and Cl from U_3O_8 plus 1 g of H_3BO_3 into H_2O also did not produce satisfactory results for F. Analyses of the distillates for F was low by approximately 48%. In similar experiments it was found that known amounts of F and Cl pyrohydrolyzed from U_3O_8 plus 1 g of B_4C were recovered more quantitatively. The F measurement had a negative bias of only 19%, and the Cl recovery was 108%.

These results indicated that F can be separated satisfactorily by pyrohydrolysis from either U_3O_8 or U_3O_8 plus B_4C, but not from U_3O_8 plus H_3BO_3, and can be measured with reasonable accuracy in H_2O receiver solutions. These results also indicate that the F specie that pyrohydrolyzes from B_4C may not form H_3BO_3 in the receiver and may differ from the specie that pyrohydrolyzes from H_3BO_3.

2. Soluble Carbon. The uncombined C in B_4C is oxidized by H_2CrO_4 to CO_2 and swept by N_2 carrier gas to the measurement section of the apparatus. It was found that differences were as great as ± 40% in the measured values when the configuration of the reaction vessel was changed. The differences were apparently caused by variations in the stirring produced by the N_2 stream. The LASL observations are being checked by HEDL, and also at LASL.

3. Nitrogen. The recent interest in N_2 in B_4C has led to initiation of investigations of analysis methods at HEDL and LASL. At LASL, the alkali fusion method, Dumas method, and the use of a LECO Nitrox-6 analyzer are being tried first. Although the Dumas method seems to be satisfactory in initial tests, the analysis requires a very prolonged treatment to release the nitrogen. For this reason, fusion with NaOH in a LECO induction furnace and titration of the NH_3 produced with standard acid seems to offer more promise. A 3-min fusion seems sufficient, and a 100-mg sample yields enough NH_3 for reliable measurement. More work will be done, particularly in establishing the recovery, using BN as a reference material.

Initial tests with a LECO Nitrox-6 show that N_2 is liberated from B_4C. Tests will be continued using BN as a reference material.

III. ANALYTICAL CHEMISTRY PROGRAM FOR FBR MIXED OXIDE FUEL

A. Qualification of Analytical Laboratories

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

HEDL reports that the two vendor laboratories and its own laboratory have essentially completed qualification tests.

B. Calibration Materials and Quality Control Samples

(J. V. Pena, H. J. Kavanaugh, L. A. Maestas, C. J. Martell, W. M. Myers, C. E. Collier, J. E. Rein)

A series of specially packaged mixed oxide pellets was sent to HEDL as quality control samples for the O/M ratio measurement. The pellets were individually sealed in glass ampoules at a very low air pressure. Sealing was accomplished with a specially fabricated resistance heated apparatus.

The preparation of additional batches of calibration materials and quality control samples at an accelerated rate is under way. The reasons for the increase were presented in the previous report.[1]

C. Status of RDT Standards

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

HEDL submitted RDT Standard F2-6 "Qualification and Control of Analytical Chemistry Laboratories for Mixed Oxide Fuel Analysis" to RDT for review.

D. Development of Analytical Methods

1. Determination of Burnup

(S. F. Marsh, J. E. Rein)

a. Improvement in Mass Spectrometry Method.

The chemical separation procedure previously developed[2] to obtain separated fractions of Nd, U, and Pu for mass spectrometric analysis involves two sequential ion exchange columns. The composition of the 0.0078M HNO_3-90% methanol elutriant for Nd must be maintained within very close limits in this method. The high volatility of methanol leads to an unacceptable concentration of the HNO_3 unless this elutriant is metered out in many small increments during the 2-h elution of Nd.

The neodymium-elution step of the procedure has been modified to provide an automated continuous addition of the elutriant in which a constant liquid-head is maintained in the column reservoir. This, in turn, provides a constant flow rate of the elutriant. Evaporation of methanol is prevented by operating the columns in an enclosure with a methanol-saturated atmosphere. The modifications described have resulted in 2 h of automatic operation, freeing the analyst for other work during this period.

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

In preliminary experiments in separating rare earths from trivalent actinides, ethanol-HCl solutions of Eu or Am tracers were batch-contacted with cation-exchange resin. Erratic results were obtained. These poor results were attributed to (1) variable moisture content of the air-dried resin, (2) unattained equilibrium of the ions between the solution and resin phases even though the reaction tubes were manually shaken periodically for periods up to 1 wk, and (3) insufficient fraction of the total activity remaining in solution after the contact to provide acceptable counting precision. Experimental conditions were revised to correct these problems as follows: A Kahn-type mechanical shaker was purchased and used for 18- to 20-h contact periods. Macroporous cation-exchange resin was used in place of conventional resin to obtain rapid equilibrium with even highly nonaqueous solvents. Higher levels of ion activity, and a corresponding improvement in counting precision,were obtained by using one-tenth as much resin in the contact experiments. The macroporous resin was dried before use at 65°C for 3 days in a well-ventilated oven to achieve constant weight, which corresponded to 55% loss of the initial weight. The dried resin, when exposed to laboratory air of ~ 55% relative humidity, regained about 20% of the lost weight. Apparently, the as-received resin contains 45 wt% "unbound" water.

The revised batch-contact experiments yielded acceptable agreement. Separation factors (Eu relative to Am) ranged from 50 to 115 over a wide range of ethanol percentages and HCl concentrations. These factors are about tenfold higher than previously reported for acid-alcohol mixtures. Experiments are planned with columns to select the HCl-ethanol composition that provides the most suitable combination of separation factor and

kinetic effects. Dried resin will not be required for column experiments as macroporous resin is reported to equilibrate rapidly with mixed solvent.

Arsenazo III has been tentatively selected as the chromogen to be used for the spectrophotometric determination of the separated rare earth group. A major advantage of Arsenazo III is the nearly equal molar absorptivities for different rare earth elements. Average values at 656 nm are:

La	31,700	Sm	35,200
Ce	32,400	Eu	33,600
Pr	33,000	Gd	35,000
Nd	34,300	Y	30,500

The proposed chemical separation consists of fuming the HF-$HClO_4$ solution of the sample to volatilize RuO_4. The residue is dissolved in ethanol-HCl and the rare earths are adsorbed on a cation exchange resin column. The rare earths are eluted later with HCl. The actinides pass into a coupled anion exchange resin column where they are retained. The Am and Cm are subsequently eluted with 12M HCl, the Pu with 12M HCl-0.1M HI, and the U with 0.1M HCl. The total rare earths, U, and Pu are measured spectrophotometrically as the respective Arsenazo III complexes.

A solution of mixed rare earths and Y has been prepared in proportions which simulate fission products from FBR U-Pu mixed oxide. This mixture will be used for initial evaluations of the procedure. If satisfactory data are obtained, dissolved fuel samples will be processed next.

2. Determination of O/M Atom Ratios
(G. C. Swanson and G. R. Waterbury)

Thermogravimetric methods for determination of the oxygen-to-heavy metal atom ratio (O/M) for mixed oxide fuels are capable of great sensitivity and precision. The principle of these methods is to weigh the oxide, subject it to conditions "known" to produce stoichiometry (O/M = 2,000), reweigh, and calculate the initial stoichiometry. Highly pure U_3O_8 and PuO_2 prepared by careful ignition of the respective metals have been used at LASL to determine the conditions producing stoichiometry for UO_2, PuO_2, and mechanical mixtures of the two. These calibration data may not apply directly to solid-solution $(U,Pu)O_2$ because of expected differences in chemical potentials of the mechanical mixtures and the true solid solutions.

The crystal defect structure theory of Kröger[3] as modified by Tripp, Vest, and Graham[4] provides a basis for determination of the conditions of stoichiometry for solid solution mixed oxides without knowledge of initial material stoichiometry. The theoretical development results in the relationship:

$$\left(\frac{\partial \Delta W}{\partial \ell n P_{O_2}}\right)_T = 0 \text{ at stoichiometry.}$$

Apparatus has been developed and installed to allow the measurement of sample weight (Δ w) at constant temperature while varying O_2 pressure greater than four orders of magnitude. A Mettler recording thermoanalyzer with high vacuum system and DTA amplifier, an automatic digital data logging system, and a gas handling manifold in which O_2 potential is controlled by coulometric generation of O_2 from a high-temperature, solid-state electrochemical cell, have been installed and some components calibrated. Development work is continuing on a high-temperature, solid-electrolyte O_2 concentration cell to allow confirmatory O_2 potential measurement of the oxide sample at temperature.

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