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Quarterly Report

Advanced Plutonium Fuels Program

January 1 through March 31, 1974

Compiled by

R. D. Baker

LOS ALAMOS NATIONAL LABORATORY
3 9338 00377 3339

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

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

LA-5284-PR	**LA-5477-PR**
LA-5390-PR	**LA-5582-PR**

In the interest of prompt distribution, this progress report was not edited by the Technical Information staff.

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ABSTRACT

This is the 30th 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, J. M. Ledbetter, M. E. Lazarus, P. A. Mason, O. Serna, W. T. Wood)

1. Fuel Pin Sectioning Jig

A newly designed jig is being fabricated for more accurate positioning of the saw used in sectioning fuel pins. The jig incorporates a micrometer indicator to locate the sectioning cuts with an error less than ± 0.025 mm (0.001 in.). The present requirement of referencing all cuts to the lower welded seam of the fuel clad infers that this position must be established visually which increases the error to ± 0.254 mm (0.010 in.) for the saw cuts relative to the actual clad-end adapter interface.

2. Sodium Distillation Furnace

Recent receipt of a UHV power conductor feedthrough has permitted completion of the furnace. Thermocouples were attached to both the heating plate and a "dummy" uranium carbide fuel sample mounted in the usual metallography grinding gage and cup. A temperature of 305°C was attained by the "dummy" fuel sample while maintaining the heat plate at 450°C. To permit heating the fuel samples to the desired 450°C during distillation,

the "dummy" sample will be included in the assembly to provide a true test of the furnace capability.

Figure 401-1 is a photo of the furnace after a distillation operation. Note the deposition of sodium on the concave surface of the liquid N_2 cold trap. The "dummy" fuel sample with attached thermocouple is also visible on the remotely replaceable heating plate assembly.

3. Fuel Pin Striping Fixture

A fixture to permit striping fuel pins with a solid paint line at 0° and a dashed line at 30° has been designed. The assembly is presently being fabricated. This design will provide improved retention of the pin orientation.

4. Fuel Pin Scribing Fixture

A fixture to permit scribing GE fuel pins with marks along the length of the pin at 0°, 90°, and 135°, has been designed and fabricated. The scribe marks are easily visible on the macro-photographs and provide a check on the orientation of the transverse metallography samples.

5. Chamfer Tool for Metallography Samples

A tool for chamfering one end of sectioned fuel samples has been designed and fabricated. The chamfer distinguishes top from bottom on longitudinal metallograph samples.

6. Balance Point Fixture

New fixtures were installed to provide a more accurate determination of the balance point of capsules and pins.

7. Data-Logger

A Data-Logger has been ordered which will automatically record thermocouple (TC) output in $^\circ C$. The unit has a capacity of 40 points. Presently, measurements are made by measuring the TC output on a potentiometer and then calculating the temperature using suitable tables.

The Data-Logger will be extremely useful in fuel element length measurements, where the temperature must be measured every 5 cm (2 in.) along the fuel element. The Length Gage is shown in Figs. 401-2 and 401-3. Note the holes, spaced at 5-cm (2-in.) intervals, in the aluminum fuel element holders where the TC measurements are made. Measurements are presently made by a single TC inserted in one of the holes until equilibrium is established. The measurement is then repeated in each of the remaining holes. When the Data-Logger is ready for use, TC Assemblies, up to a 30 TC assembly for a 150-cm (60-in.) pin, will be used. Measurements can be printed by the Logger on paper at a rate of 2 measurements/sec. The Data-Logger, besides saving time, will eliminate possible errors that can be made when

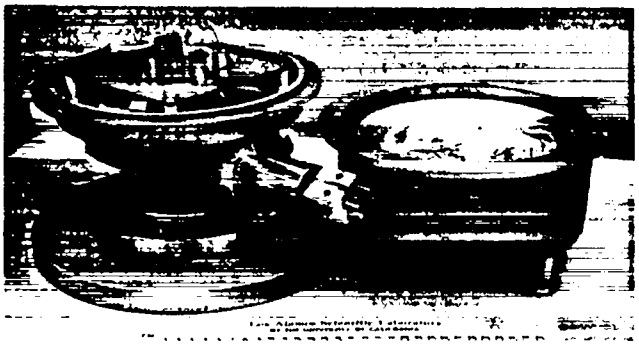

Fig. 401-1. Sodium Distillation Furnace

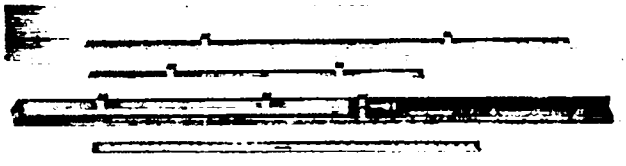

Fig. 401-2. Length Gauge

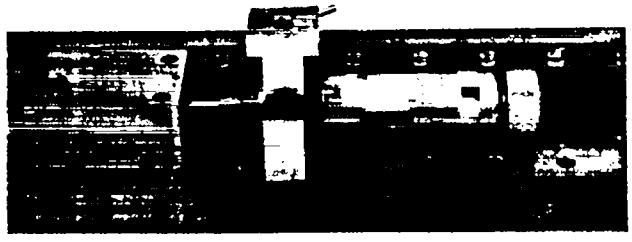

Fig. 401-3. Length Gauge - Closeup (of Readout Unit)

adjusting and reading the potentiometer and when calculating temperatures.

8. Electro-Optical Profilometer

The data-reduction computer code has been changed to provide HEDL with both corrected "raw" data and reduced data on a single magnetic tape.

GE has requested spiral profilometry traces. A study has been initiated to determine what equipment changes and computer code changes are necessary to provide information in this manner.

9. Macro-Photography System

Modifications have been completed on the Macro-Photography System for photographic purposes. The power supply for the xenon flash tube has been automated to provide the correct voltage repeatedly.

10. Packaging of Fuel Sections

Sections which have been removed from an irradiated fuel pin are placed in a stainless steel sleeve for either storage or inter-cell transfer. The identity of the specimen is scribed on the sleeve. The ends of the sleeve are secured with socket heat set screws. For storage the loaded sleeves are first placed in a stainless steel tube which is sealed in an inert atmosphere with solder joints. Inter-cell transfer is effected with the usual transfer devices.

11. Sectioning Attachment

A fixture has been designed and fabricated for use with the cut-off saw to remove capsule cladding from the end hardware which is attached to the fuel pin. The device is clamped in the saw vise and slowly rotates the capsule as the saw blade cuts through the cladding without damage to the fuel pin.

B. Inert Atmosphere Systems

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

1. With the installation in the Disassembly Cell of manipulators incorporating the new design of through-seal packages, the air leakage due to diffusion through the booting material has been eliminated. This reduced the overall air leak rate into the cell sufficiently to permit operating the recirculating purifier. Levels of 3 to 10 ppm O_2 and < 1 ppm H_2O are being currently maintained for the Disassembly Cell atmosphere.

Figure 401-4 displays both the internal seal package and the external inflatable seal package.

2. A certified bottle of Ar with 10 ppm O_2 was obtained to allow a verification of the accuracy of the built-in calibrator unit of the Delphi Oxygen Analyzers. The two methods agreed well within the certified accuracy of 10 ± 0.05 ppm for the standard gas. The analyzers in service will be recalibrated on a monthly basis with the standardized gas, in addition to the weekly calibration performed with the built-in calibration unit.

3. In an effort to reduce the operating costs of maintaining the inert atmosphere cells, N_2 gas has been substituted for the Ar gas previously used. The N_2 gas contains approximately 0.7 ppm O_2 and < 1 ppm H_2O. The carbide fuels will continue to be stored in an Ar atmosphere as requested.

4. Installation of manipulators incorporating the new through-tube seal package in the Metallography Cells has been delayed due to a shortage of manipulator boots. A shipment of boots received recently, contained a design change unsuitable for our use. The boots were returned to the factory. A replacement shipment is expected shortly.

C. Manipulator Maintenance

(W. R. Carter, E. L. Mills, P. A. Mason)

Fig. 401-4. AMF Manipulator Through-Tube Seal Assemblies

Ten manipulators have been modified with the new seal packages for use in the inert atmosphere cells.

D. Shipping Casks

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

Modifications have been made to both casks (designated as DOT 5885) to permit handling at the EBR-II facilities, HFEF-North and HFEF-South. The required tests were performed at HFEF-South to demonstrate compatibility. ANL personnel adapted a lifting fixture to permit retention of the LASL cask inserts (approximating 2R Containers). Shipments of fuel pins have already been made using a threaded pipe insert in the modified casks. Use of the LASL insert (capacity for 19 pins, 40 in. long) will be made during the early part of the next Quarter.

III. ANALYTICAL CHEMISTRY

A. Gamma Scanning

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

Precision gamma scanning was used for the quantitative determination of ^{106}Ru, ^{137}Cs, and ^{144}Pr in three irradiated (U,Pu) carbide fuel materials having undergone 10.5 at.% burnup. This nondestructive technique involves the extensive calibration of the gamma scanning system including the experimental measurement of the effective collimating slit areas and the surface response functions for the detector assembly. These calibration data are combined with the source geometry factors including fuel composition, fuel density, radial distributions of fission products, and other source parameters as data input for a computer code to calculate the quantities of specific fission products in the fuel pin.

After the gamma scanning measurements of the three isotopes, the fuel sections were dissolved and analyzed using well-proven radiochemical methods. The gamma scanning results and the radiochemical results differed by only 3.1% for ^{106}Ru, 3.2% for ^{137}Cs, and 2.8% for ^{144}Pr.

These data show that precision gamma scanning can be successfully used for the nondestructive quantitative determination of selected fission products in irradiated (U,Pu) carbide fuel materials. This nondestructive technique is being applied presently to the examination of (U,Pu) oxide fuel materials.

B. Sealed Tube-Reflux Dissolution System

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

The sealed tube-reflux dissolution system[1] was simplified by incorporating a new sealing technique to contain the pressure in the dissolution tube. In this system, the sample plus 2 ml of 12M HCl, 1 drop of 15M HNO_3, and 1 drop of 0.41M HF are placed in a fused-silica or borosilicate glass tube which is flame sealed. Only the end of the tube containing the sample and solvent is heated, and the top of the tube serves as a reflux column.

The pressure developed in the sealed tube-reflux system by the acid mixture was measured over the temperature range of 30 to 165°C. At the recommended operating temperature of 150°C, the pressure was 620 KPa (90 psi) in a tube 8 mm i.d. by 380 mm long and 1030 KPa (150 psi) in a tube 8 mm i.d. by 200 mm long. To contain these relatively low pressures, the apparatus was simplified by using a rubber stopper, held in place by a spring-loaded clamp to seal the tube. This modification eliminated the need to flame seal the tube. The temperature at the seal was only 40°C during the dissolution and did not adversely affect the rubber stopper. This new seal has greatly simplified the dissolution system and increased sample throughput.

As impurities being leached from the borosilicate glass or fused-silica dissolution tubes might interfere in subsequent analyses, acid mixtures which were maintained at 150°C and 620 KPa for one week in the two types of tubes were analyzed. The impurity levels for 34 elements in both solutions were quite low (Table 401-I), with the solution in the fused-silica tube containing lesser amounts of impurities.

This new dissolution technique was successfully tested on approximately 20 irradiated fuel-clad samples, having up to 12 at.% burnup, and was demonstrated to be simple, rapid, and effective. This technique also was successfully used to dissolve numerous PuO_2 samples. Application to dissolution of other materials by varying the type and concentration of the acids is being investigated.

IV. MICROSTRUCTURAL ANALYSIS

(J. H. Bender, D. D. Jeffries, K. A. Johnson, J. L. Lehmann, L. N. Sanchez)

TABLE 401-I

IMPURITIES LEACHED FROM BOROSILICATE GLASS AND FUSED SILICA TUBES

Impurity	Impurities, μg, in solvent* Borosilicate Glass	Fused Silica
Li	4	< 0.2
Be	< 0.02	< 0.02
B	120	0.07
Na	300	1.5
Mg	10	1
Al	200	1
Si	120	70
P	< 6	< 5
K	10	< 3
Ca	20	2
Ti	4	< 0.2
V	< 0.4	< 0.3
Cr	0.4	< 0.2
Mn	0.4	< 0.2
Fe	10	0.3
Co	< 0.4	< 0.3
Ni	< 0.6	< 0.5
Cu	1.5	0.2
Zn	10	2
Ga	0.2	< 0.2
Ge	< 0.4	< 0.3
Sr	1	0.02
Zr	30	0.3
Nb	< 0.6	< 0.5
Mo	< 0.2	< 0.2
Ag	0.08	0.07
Cd	< 0.6	< 0.5
In	< 0.6	< 0.5
Sn	< 0.6	< 0.5
Sb	< 0.6	< 0.5
Ba	0.8	0.05
W	< 6	< 5
Pb	1	< 0.5
Bi	< 0.2	< 0.2

*Solvent: 2 ml 12M HCl, 1 drop each of 15M HNO_3 and 0.41M HF

The photo-darkroom trailer is now fully installed. Films, print papers, and processes are being tested and studied for the conversion of most of our photomicroscopy to roll film.

V. REQUESTS FROM RRD

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

During the Third Quarter of FY 1974, thirty-five irradiated fuel capsules were received. The distribution is as follows: GE, 21; HEDL, 13; LASL, 1.

Argonne National Laboratory: The scanning electron microscope was used in examining one sample of a driver fuel pin from EBR-II.

General Electric Company: Examinations performed on 54 irradiated fuel capsules received between March 21, 1973, and March 14, 1974, are listed in Table 401-II.

Other examinations --- density measurements were made on 22 unirradiated archive cladding samples. At the request of ANL, fission gas samples from 4 irradiated fuel pins were sent to ANL-West for isotope analysis of "tagged Xe".

Gulf General Atomic: Five samples were analyzed for tritium content.

Hanford Engineering Development Laboratory: Examinations were performed on thirteen irradiated fuel capsules received between November 12, 1973, and March 7, 1974, as listed in Table 401-III.

Los Alamos Scientific Laboratory: This section discusses 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 performed on two BMI irradiated fuel capsules received on February 16, 1973, and March 7, 1974, are shown in Table 401-IV.

2. LASL Experiments -- Examinations performed on one LASL irradiated fuel capsule received on October 11, 1972, are shown in Table 401-V.

3. UNC Experiments -- Two capsules were examined by gamma scanning which included 8 gross gamma-scans, 4 complete spectral scans, and determination of 20 isotopic distributions of fission and activation products.

VI. QUALITY ASSURANCE
(L. E. Lanham)

A. General

A meeting was held with HEDL personnel to discuss quality assurance and the HEDL requirements for post-irradiation examinations. The report of this meeting was an agreement stating the actions that will be taken by

TABLE 401-II

POSTIRRADIATION EXAMINATIONS OF CAPSULES AND PINS FROM GE

	Examination	No. of Capsules	No. of Pins
1.	Visual Inspection	21	3
2.	Preliminary Measurements	21	8
3.	Profilometry, Optical	--	6
4.	Profilometry, Mechanical	--	1
5.	Radiography	2	--
6.	Gamma Scan[a]	4	--
7.	Gas Sampling and Analysis	5	12
8.	Na Removal	2	--
9.	Clad Removal	3	--
10.	Photography, Full Length	--	2
11.	Photography, Maximum Bow	8	--
12.	Photography, Incremental	--	3
13.	Wire Wrap Removal	--	3
14.	Eddy Current	--	3
15.	Weight	21	--
16.	Balance Point	21	--
17.	Diameter Measurement, Micrometer	--	1
18.	Sectioning	--	15
19.	Density Measurement	--	13 (34 samples)
20.	Atom % Burnup	--	11 (16 samples)
21.	Shielded Microprobe	--	3 (4 samples)
22.	Oxygen Analysis	--	5 (10 samples
23.	Radiochemistry (^{134}Cs)		1 (4 samples)
24.	Radiochemistry (^{137}Cs)		1 (4 samples)
25.	Total Cs	--	1 (4 samples)
26.	Sodium	--	1
27.	Clad Plenum Leach	--	1 (4 samples)
28.	Microstructural Analysis[b]	--	14 (36 samples)

[a] 16 Gross gamma scans, 6 complete spectral scans, and 50 isotopic distributions of fission and activation products were determined.

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

TABLE 401-III

POSTIRRADIATION EXAMINATIONS OF CAPSULES AND PINS FROM HEDL

	Examination	No. of Capsules	No. of Pins
1.	Visual Inspection	2	2
2.	Preliminary Measurements	9	11
3.	Profilometry, Optical	—	2
4.	Radiography	9	2
5.	Photography, Full Length	—	1
6.	Photography, Incremental	—	1
7.	Gas Sampling and Analysis	2	1
8.	Na Removal	2	--
9.	Clad Removal	2	—
10.	Atom % Burnup	--	1
11.	Shielded Microprobe	—	1 (2 samples)
12.	Gamma Scanning[a]	16	—
13.	Microstructural Analysis (Optical Microscopy)	—	3 (6 samples)

[a] Sixty-four gross gamma scans, 15 complete spectral scans, 4 TWODIM's, and 105 isotopic distributions of fission and activation products were determined.

TABLE 401-IV

POSTIRRADIATION EXAMINATIONS ON CAPSULES AND PINS FROM BMI

	Examination	No. of Capsules	No. of Pins
1.	Gas Sampling and Analysis	--	1
2.	Density Measurements	--	1 (1 sample)

TABLE 401-V

POSTIRRADIATION EXAMINATIONS ON CAPSULES AND PINS FROM LASL

	Examination	No. of Capsules	No. of Samples
1.	Density Measurements	1	1
2.	Microstructural Analysis (Optical Microscopy)	1	3
3.	Shielded Microprobe	1	1

both HEDL and LASL to provide the examination, the examination results, and the check list verification requested by HEDL.

In response to requests received from AEC, actions have been initiated to provide a more detailed independent quality assurance overcheck and surveillance function for postirradiation examinations by increasing the staff of the quality assurance organization. These actions will be implemented as soon as qualified personnel can be obtained.

B. Hot Cell Examinations

A revised procedure has been prepared for the sectioning operation. Included in this revised procedure is a check list to be used by the operator performing the sectioning operation and by the person performing the overcheck. Each examination is being reviewed and check lists are being prepared for those examinations where a formal verification-overcheck is needed to give the added assurance in a document that the examination results meet the sponsors' requested technical requirements.

An audit was conducted by the Quality Assurance Manager of the Hot Cell Examinations and an audit report was prepared.

Incident LASL-2 for a bent fuel element is being investigated. Corrective actions have been taken by initiating special handling procedures and specific operator instructions. These procedures have been used successfully with the same type of fuel rod and cask in an unloading operation. Design changes are in progress on the remotized handling equipment.

C. Microstructural Analysis

A new numbering system has been established for photography. In addition to the mount identification number each photograph is assigned a sequential number which is recorded on a document log and on the traveler. A check list has been prepared and will be used to perform overchecks as required by the CMB-RRD Quality Assurance Manual.

An audit was conducted to evaluate the effectiveness of the overcheck system in assuring that data meets the sponsors' technical requirements. The overcheck system is effective and is being used. An audit report has been prepared.

D. Chemical Analysis

An addendum has been added to the chemical analysis Quality Assurance Plan to define the overcheck procedure and its application to each chemical analysis. A change has been made in the distribution of the travelers which are used as work instructions. This gives a better control of the documentation and notifies the Quality Assurance Manager of the examination at the time it is performed.

VII. REFERENCE

1. R. D. Baker, "Quarterly Report on Advanced Plutonium Fuels Program, October 1 to December 31, 1973," Los Alamos Scientific Laboratory report LA-5582-PR, October 1 to December 31, 1973.

VIII. PUBLICATIONS

1. J. R. Phillips, G. R. Waterbury, N. E. Vanderborgh, "Distributions of ^{134}Cs and ^{137}Cs in the Axial UO_2 Blankets of Irradiated $(U,Pu)O_2$ Fuel Pins," J. Inorg. Nucl. Chem., 36, pp. 17-23 (January 1974).

2. J. R. Phillips, N. E. Vanderborgh, G. R. Waterbury, "Application of Modern Gamma Scanning Techniques." Invited paper to be presented at the 76th Annual Meeting of the American Ceramic Society, Chicago, Ill. May 1, 1974.

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 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, D. W. Kelley, J. Mascarenas)

1. Carbide Fuel Production

Carbide fuel pellets previously synthesized at LASL have been high-purity single-phase $U_{0.80}Pu_{0.20}C_{0.97}$ having densities $\geq$ 93% of theoretical. New irradiation experiments require fuels of comparable purity having sesquicarbide concentrations which vary from 1 to 17 vol% and having minimum densities of 98% of theoretical. One means of achieving this higher density is by the use of nickel sintering aid. The carbothermic process was

designated as the process to be used for this synthesis because of the commercial attractiveness of the process and because of the experience gained from previous scoping experiments.[1] The steps of this process are:

1. Blend and comminute UO_2, PuO_2, and C powders in a ball mill.
2. Press the blended powder into pellets.
3. Heat the pellets in vacuum to remove CO.
4. Comminute in a Spex mill.
5. Add Ni powder, blend and comminute in a ball mill.
6. Press into pellets.
7. Sinter.
8. Characterize.

To establish optimum processing parameters, a 225-g batch of $U_{0.8}Pu_{0.2}C_{1+x}$ was carried through step 4 above. Sufficient carbon was added initially to give a product which contained 13 vol% sesquicarbide. The powder was divided into four equal batches and blended with 0, 0.10, 0.15, and 0.20 wt% Ni powder by ball milling for 16 h. The Ni-containing batches were sintered at two different temperatures. Shown in Table 463-I are the sintering results. The amount of Ni found in the sintered pellets was generally about 0.05 wt% less than the amount added. This loss was very possibly due to the preferential sticking of the Ni powder to the surfaces of new process equipment. Comparison of the magnitude of the Ni loss at different temperatures and the vapor pressures of Ni at different temperatures precludes vaporization as a significant mechanism for the decreased Ni content of the product. Density values listed in Table 463-I show essentially no variation due to changes in the Ni concentration. Pellets sintered at 1800°C were essentially the same density as pellets of similar composition which were sintered at 1550°C.

TABLE 463-I

DENSITIES OF SINTERED $U_{0.8}Pu_{0.2}C_{1+x}$ PELLETS CONTAINING Ni ADDITIONS

Batch No.	Ni Added, wt.%	Sinter Time, h	Sinter Temp, °C	Ni Conc. By Anal, wt.%	Density %TD[a]	vol% M_2C_3[b]
1	0.10	1	1550	0.06	98.4	13
2	0.10	1	1800	0.03	98.6	15
3	0.15	1	1550	0.10	98.7	17
4	0.15	1	1800	0.12	—	--
5	0.20	1	1550	0.16	98.2	13
6	0.20	1	1800	0.14	98.0	13
7	0.00	8	1850	---	92.7	> 6

[a] Based on theoretical density of 13.45 g/cm^3 for MC and 12.72 g/cm^3 for M_2C_3

[b] Values by image analysis of photomicrographs

Pellet batch No. 7, which was fabricated without Ni sintering aid, had a significantly lower pellet density. In this batch, the measured vol% M_2C_3 was not representative of the actual M_2C_3 concentration because of the state of aggregation of the second phase. In the absence of Ni, much of the M_2C_3 exists as platelets and the image analysis apparatus did not respond to the narrow grain boundary network. The presence of Ni appears to promote agglomeration of the M_2C_3 into more nearly equiaxed grains which are more readily measured by the apparatus.

2. Preparation of Single-Phase UC Insulator Pellets

The LASL specification for UC insulator pellets requires that, at a 500X magnification of the microstructure, no sesquicarbide or higher carbides be detectable. A review of the microstructure of previous UC preparations indicated that this stringent microstructural requirement necessitated additional developmental work. Although arc-cast material is capable of meeting this specification, it is possible that the presence of higher carbides is disguised by an extremely fine state of subdivision, leaving the high carbon activity associated with these carbides.

Accordingly, ingots with initial compositions of $UC_{0.98}$, $UC_{1.00}$ and $UC_{1.02}$ were prepared and subjected to a variety of processing conditions. The process was similar to the LASL arc-melting process used for the preparation of single phase $U_{0.80}Pu_{0.20}C_{0.97}$. Process variables which were investigated included time of H_2 treatment, comminution cycles, powder aging, pre-pressing and pressing pressures, sintering time and temperature. The process which evolved from this investigation and which is capable of producing pellets which meet all UC specifications is as follows:

1. Arc melt U and graphite chunks combined to give a C/U atomic ratio of 0.98, using a graphite electrode and copper hearth.

2. Solution treat the ingots at 1600°C for 16 h.
3. Grind the ingots in a Spex mill to -250 mesh.
4. Pass H_2 over shallow trays (~ 6 mm deep) of powder for 65 h at 800°C.
5. Press the powder at 210 000 kPa without prepressing.
6. Sinter 8 h at 1800°C under flowing Ar, using about 2 h to heat to 1800°C, cooling at about 25°C/min to 1400°C, holding 2 h, and cooling at about 25°C/min to room temperature.

These conditions are not necessarily optimum and may be altered in the future.

3. Preparation of UN Insulator Pellets

A 225-g batch of UN pellets was synthesized according to Quality Assurance procedures. The product pellets were sampled and the results of the analyses are not yet complete.

4. Equipment Development

The centerless grinder was modified to provide more precise control of the advance mechanism and thus a smoother grinding operation. In addition, a dust collection system was adapted to the apparatus in order to reduce powder scattering, minimize cross contamination, and facilitate material accountability. A pellet feed and collection system was also fabricated and installed on the apparatus. The apparatus was installed in a recirculating inert atmosphere glovebox and used to successfully grind pellets of UN, UC, and (U,Pu)C.

A new recirculating inert atmosphere glovebox system is being placed in operation. As soon as a pure atmosphere is established the glovebox will be used for examination and sampling of pellets.

B. Fuel Element Fabrication

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

1. Weld Development and Qualification

A series of welds of end plugs to clad tubing were made with a 150-amp programable welder and a hand-held torch to establish preliminary welding parameters. In a subsequent series of developmental welds on xenon-tag-gas sample containers, the torch and work pieces were oriented in a fixture. It was found that the sensitivity of the controls was not sufficient to prevent an abrupt tail-off which caused the formation of small craters in the weld. In addition, the welding fixture did not reproducibly orient the work pieces and electrode. A new 50-amp programable welder was installed to provide the control sensitivity required for the operation. Gauges were fabricated to facilitate reproducible positioning of the work pieces and electrode. Further qualification welds are being made.

2. Xenon Tagging

A second calibrated volume was added to the xenon manifold to provide a temporary reservoir for frozen xenon. This was necessary in order to recover the excess xenon which remained in the manifold after a single tagging operation. Xenon in the manifold cannot be conveniently returned directly to the supply tank. Two encapsulated samples of xenon which were fabricated during the developmental welding operations were analyzed for xenon and found to contain 1.64 cm^3 and 1.33 cm^3 (STP) of xenon. These specimens meet EBR-II requirements. Additional tag samples are being prepared.

3. Sodium Bonding

The procedure for bonding sodium to the fuel and clad begins with a heat treatment of the loaded fuel element at approximately 600°C to ensure wetting of the interior components. Temperature gradients in the bonding furnace were measured for both the heating and cooling modes as a function of power settings with the system evacuated or gas filled, and with various times of opening and closing the furnace. These measurements were made to establish an operational mode which would ensure the proper directionality of sodium melting and freezing. A minor modification is being made to the bonding furnace to facilitate introduction and removal of the fuel elements.

After pretreatment, the element is heated to 350°C and centrifuged to remove gas bubbles between the fuel and cladding. A system was devised so that the temperature along the exterior surface of the centrifuge tube could be measured during centrifugation. A sodium-bonded (U Pu)N fuel element with thermocouples strung along the length was placed in the centrifuge tube and a correlation was established between interior and exterior temperature profiles with the centrifuge in a stationary position. The interior thermocouples were removed from the element

and the centrifuge tube cooling rates were measured with the centrifuge operating at 144 rpm. It was observed that the cooling rate increased 44% and the necessary cooling directionality was maintained during centrifugation. A photoelectric counting device and interval timer were installed to determine rpm. This system was found to be capable of measuring the rate of centrifugation to within 1 rpm.

4. Associated Activities

Developmental work was continued in the areas of wire wrapping, component cleaning and shroud tube fabrication.

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-II to 463-IX. In order to better define the status of those experiments which are undergoing postirradiation examination, the following steps are referenced in the tables:

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

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-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 85 kW/m). Three tests at very high power (> 125 kW/m) were included to determine the effect of high thermal stresses and high fuel temperatures on fuel element behavior. Indications of element cladding failure were found at EBR-II in several experiments from these series (five in subassembly X119B, one from X142, and two from X152), using γ-scanning for ^{133}Xe. Examinations of these experiments in the LASL hot cells confirmed the failures. Complete postirradiation examination of the failed experiments is continuing.

One unfailed experiment, K-44, completed irradiation in subassembly X182 after reaching a maximum burnup of 7.1 at.%. A second unfailed experiment, K-39B, is at EBR-II awaiting further irradiation.

Table 463-III describes the Series U1300 experiments. These experiments contain two-phase, uranium-plutonium

TABLE 463-II

SERIES K1, K2, AND K3 ENCAPSULATED CARBIDE EXPERIMENT

Expmt. No.	Fuel Type[a]	Fuel Density, % Theo.	Bond and Diametral Gap, mm.	Clad Type[b]	Clad O.D. x I.D. mm.	Max. Linear Power, kW/m[g]	Maximum Centerline Temp., °C[g]	Goal Burnup, at.%[c]	Maximum Current Burnup at.%	Status
				Series K1						
K-36B	MC	90	Na-0.38	SA-316SS	7.62 x 7.11	82†	990†	6	5.85*	b.12[d]
K-37B	MC	90	Na-0.38	SA-316SS	7.62 x 7.11	85	1000	6	2.9	a.7[d,e]
K-38B	MC	90	Na-0.38	SA-316SS	7.62 x 7.11	85	1000	6	5.8	a.7[d,e]
K-39B	MC	90	Na-0.38	SA-316SS	7.62 x 7.11	85	1000	10	5.8	EBR-II, Unassigned
K-42B	MC	90	Na-0.38	SA-316SS	7.62 x 7.11	85†	1000†	6	4.46*	Completed[f]
				Series K2						
K-49	MC	95	Na-0.51	SA-316SS	7.62 x 7.11	130†	1270†	5	3.74*	b.12[d]
K-50	MC	95	Na-0.51	SA-316SS	7.62 x 7.11	130	1300	6.5	3.6	a.9[d]
K-51	MC	95	Na-0.51	SA-316SS	7.62 x 7.11	130	1300	8	3.5	a.9[d]
				Series K3						
K-43	MC	94	Na-0.51	SA-316SS	7.62 x 7.11	80	1000	8	5.6	b.3[d]
K-44	MC	94	Na-0.51	SA-316SS	7.62 x 7.11	80	1000	8	7.1	EBR-II Exam
K-45	MC	94	Na-0.51	SA-316SS	7.62 x 7.11	80	1000	5	2.37*	b.9[d]
K-46	MC	94	Na-0.51	SA-316SS	7.62 x 7.11	80†	965†	5	2.39*	b.12[d]

[a] $M = (U_{0.8}Pu_{0.2})$
Series 1 and 3 experiments are 93% enriched in ^{235}U
Series 2 experiments are 97% enriched in ^{238}U.

[b] SA = Solution annealed

[c] Burnup values marked with * were measured using the ^{148}Nd method. Remaining values were computed from EBR-II data.

[d] Element cladding failure indicated.

[e] Damaged during reconstitution of X152.

[f] Reported in LA-4669-MS

[g] Linear power & centerline temperature marked with † are beginning-of-life values computed using measured burnup results. Remaining values based on EBR-II power adjustment factor of 0.91.

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 56 kW/m). Indications of element cladding failure for three experiments were found at EBR-II using γ-scanning for ^{133}Xe. These element failures have been confirmed by γ-scanning for ^{137}Cs at LASL. Three of these experiments have completed postirradiation examination and evaluation of the results is continuing. Destructive examination of the remaining experiments is awaiting examination of higher priority experiments.

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 77 to 99% theoretical. Most of these experiments are currently within 1.5 at.% of their goal burnup after operation at low linear power (38 to 44 kW/m). During interim examination at EBR-II after run 58, ^{137}Cs was detected by γ-scanning in the sodium reservoir of capsule U136. Release of fission gas from a breached

TABLE 463-III

SERIES U1300 ENCAPSULATED CARBIDE EXPERIMENTS

Expmt. No.	Fuel Type[a]	Fuel Density, % Theo.[b]	Bond and Diametral Gap, mm.	Clad Type[d]	Clad O.D. x I.D. mm.	Max. Linear Power, kW/m[g]	Maximum Centerline Temp., °C	Goal Burnup, at.%[e]	Maximum Current Burnup at.%	Status
U93	MC+5%M_2C_3	86	He-0.10	SA-316SS	7.66 x 6.10	59†	1750	11	9.64*	b.12
U94	MC	85	He-0.18	SA-316SS	7.73 x 6.86	70†	1680	11	9.42*	b.12
U97	MC+5%M_2C_3	86	He-0.10	SA-INC-800	7.60 x 6.10	64	1750	11	10.0	b.5
U98	MC+5%M_2C_3	86	He-0.18	SA-INC-800	7.60 x 6.86	65	1680	11	9.6	a.7[f]
U105	MC+5%M_2C_3	77	He-0.20	SA-INC-800	7.61 x 6.10	52†	1900	11	9.89*	b.12
U106	MC+5%M_2C_3	77	He-0.20	SA-INC-800	7.72 x 6.86	59	1820	11	9.9	a.7[f]
U110	MC+10%M_2C_3	99[c]	He-0.33	SA-INC-800	7.73 x 6.86	65	1960	10	9.2	a.7
U113	MC+10%M_2C_3	98[c]	He-0.28	SA-INC-800	7.61 x 6.10	50	1880	11	10.2	a.7
U114	MC+15%M_2C_3	98[c]	He-0.18	SA-INC-800	7.72 x 6.86	66	1570	10	9.5	a.7[f]

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

[b] Theoretical density of MC = 13.45 g/cm^3
Theoretical density of M_2C_3 = 12.72 g/cm^3

[c] Cored pellet with nominal 2.0 mm diameter axial hole

[d] SA = Solution annealed

[e] Burnup values marked with * were measured using the ^{148}Nd method. Remaining values were computed from EBR-II data.

[f] Element cladding failure indicated.

[g] Linear powers marked with † are beginning-of life values computed using measured burnup results. Remaining values based on EBR-II power adjustment factor of 0.91.

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 U136 can only be considered tentative and of a low degree. None of the other capsules indicated fuel element failure during the examinations at EBR-II. All 19 capsules were reconstituted into subassembly X055B which is currently being irradiated.

The Series U1930 and U1960 experiments are described in Table 463-V. Experimental parameters include fuel type, fuel density, bond type, and cladding type. The operating linear power ratings for the experiments are relatively high (73-100 kW/m). Nondestructive examination of the eleven experiments listed in part A of Table 463-V was completed several months ago. The results of these examinations showed that fuel elements U194 and U200 had failed. Destructive examination of this group of experiments has been completed. Data reduction and interpretation are continuing.

The experiments listed in part B of Table 463-V completed irradiation in subassembly X182. Prior to the irradiation in X182, elements U188, U190, U193, and U196 had not failed after maximum burnups of 8.5 at.% and elements U199, U201, U207, and U209 had not failed after maximum burnups of 5.5 at.%. After an accumulation of approximately an additional 2 at.% burnup, elements U190 and U199 have not failed. However, U188, U193, U196, U201, U207, and U209 have failed as evidenced by γ-scanning at EBR-II. Five of the capsules - U188, U190, U193, U199, and U209 - were accidentally damaged at EBR-II while being transferred between the air cell and the argon cell in FEF-S. The capsules were dented and bent 3-5° approximately 12 inches from the bottom ends. A sixth capsule, U207, sustained only slight damage. All of the experiments in this group will be returned to LASL for examination.

The experiments listed in part C of Table 463-V were used as replacement capsules in order to allow the

TABLE 463-IV

SERIES U1950 ENCAPSULATED CARBIDE EXPERIMENTS

Expmt. No.	Fuel Type[b]	Fuel Density, % Theo.[c]	Bond and Diametral Gap, mm.	Clad Type[e]	Clad O.D. x I.D. mm.	Max. Linear Power, kW/m[g]	Maximum Centerline Temp., °C	Goal Burnup, at.%	Maximum Current Burnup at.%[g]	Status
U129	MC	86	He-0.41	SA-316SS	7.70 x 6.60	38	1750	11	10.2	EBR-II, X055B
U130	MC	77	He-0.25	SA-316SS	7.70 x 6.60	39	1500	11	10.3	EBR-II, X055B
U131	MC	85	He-0.36	SA-316SS	7.70 x 6.60	38	1490	11	10.0	EBR-II, X055B
U132	MC	85	He-0.36	SA-316SS	7.70 x 6.60	38	1490	11	9.9	EBR-II, X055B
U133	MC	85	He-0.36	SA-316SS	7.70 x 6.60	38	1490	11	9.7	EBR-II, X055B
U134	MC	85	He-0.36	SA-316SS	7.70 x 6.60	38	1490	11	9.7	EBR-II, X055B
U135	MC	85	He-0.36	SA-INC-800	7.67 x 6.60	38	1470	11	10.0	EBR-II, X055B
U136	MC	85	He-0.36	SA-INC-800	7.67 x 6.60	40	1470	11	9.6	EBR-II, X055B[f]
U137	MC+20% M_2C_3	97	He-0.36	SA-316SS	7.70 x 6.60	40	1440	10	8.6	EBR-II, X055B
U138A[a]	MC+20%M_2C_3	98	He-0.36	SA-316SS	7.44 x 6.60	44	1440	8	5.6	EBR-II, X055B
U139	MC+20%M_2C_3	97	He-0.36	SA-INC-800	7.72 x 6.60	44	1440	10	8.8	EBR-II, X055B
U140	MC	90	He-0.36	SA-INC-800	7.72 x 6.60	41	1460	10	9.4	EBR-II, X055B
U141	MC	91	He-0.25	SA-316SS	7.70 x 6.60	43	1460	10	9.2	EBR-II, X055B
U142	MC	91	He-0.25	SA-316SS	7.72 x 6.60	43	1460	11	9.3	EBR-II, X055B
U143	MC+20%M_2C_3	96[d]	He-0.36	SA-INC-800	7.67 x 6.60	38	1390	11	9.5	EBR-II, X055B
U144	MC+20%M_2C_3	96[d]	He-0.36	SA-316SS	7.72 x 6.60	39	1390	11	9.6	EBR-II, X055B
U145	MC	90	Na-0.76	SA-304SS	7.75 x 6.86	40	820	10	9.1	EBR-II, X055B
U146A[a]	MC+20%M_2C_3	99	Na-0.76	SA-304SS	7.62 x 6.86	41	810	8	5.6	EBR-II, X055B
U147	MC+20%M_2C_3	97	Na-0.76	SA-INC-800	7.72 x 6.86	42	810	10	9.2	EBR-II, X055B

[a]Capsules 138 and 146 were removed at 4.5 at.% burnup for TREAT testing. Duplicates replaced the originals.

[b]M = $(U_{0.85}Pu_{0.15})$; U is 60% enriched in ^{235}U.

[c]Theoretical density of MC = 13.49 g/cm^3
Theoretical density of M_2C_3 = 12.76 g/cm^3

[d]Cored pellet with nominal 2.0 mm diameter axial hole.

[e]SA = Solution annealed.

[f]Possible element cladding failure indicated.

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

Table 463-VI describes the Series WF experiments. These sodium-bonded, carbide capsules were designed to evaluate the effects of (1) various amounts of sesquicarbide in the fuel, (2) linear power rating, and (3) cladding cold work on element performance. The amount of sesquicarbide reported to be in the fuel varies from 0 to 24 vol%. One experiment, W6F, was determined to have an element cladding failure after a maximum burnup of 5.4 at.%. This was the first failure in this series and and was determined by γ-scanning at EBR-II. W6F will be returned to LASL for examination. Two other experiments, W4F and W8F, have completed postirradiation examination. Irradiation of the remaining five capsules will continue to a goal burnup of 10 at.%.

Table 463-VII describes the Series B-1, B-2, and B-3 experiments. These capsules are fueled with single-phase, uranium-plutonium mononitride. All the elements in Series B-1 and B-2 are sodium bonded and clad with

TABLE 463-V

SERIES U1930 AND U1960 ENCAPSULATED CARBIDE EXPERIMENTS

Expmt. No.	Fuel Type[a]	Fuel Density, % Theo.[b]	Bond and Diametral Gap, mm.	Clad Type[d]	Clad O.D. x I.D. mm.	Max. Linear Power, kW/m[h]	Maximum Centerline Temp., °C[h]	Goal Burnup, at.%	Maximum Current Burnup at.%[g]	Status
					A					
U187	MC+5%M_2C_3	86	He-0.18	SA-316SS	7.72 x 6.60	77†	1930	5	4.60*	b.12
U189	MC+5%M_2C_3	85	He-0.25	SA-INC-800	7.67 x 6.60	81†	1930	5	4.80*	b.12
U191	MC	92	Na-0.76	SA-304SS	7.72 x 6.91	84†	1010†	5	4.50*	b.12
U192	MC	92	Na-0.76	SA-304SS	7.75 x 6.91	73†	940†	5	4.3	b.12
U194	MC+10%M_2C_3	98	Na-0.84	SA-304SS	7.75 x 6.91	81†	980†	5	4.64*	b.12[e]
U195	MC+10%M_2C_3	98	Na-0.84	SA-304SS	7.75 x 6.91	83†	990†	5	4.94*	b.12
U197	MC+10%M_2C_3	98	Na-0.84	SA-INC-800	7.75 x 6.91	80†	1020†	5	4.90*	b.12
U198	MC+10%M_2C_3	98	Na-0.84	SA-INC-800	7.75 x 6.91	84†	1000†	5	4.78*	b.12
U200	MC+5%M_2C_3	86	He-0.23	SA-304SS	7.32 x 6.60	83†	2040	5	4.72*	b.12[e]
U206	MC+5%M_2C_3	90	He-0.25	SA-316SS	7.44 x 6.58	90†	2080	5	4.96*	b.12
U208	MC+10%M_2C_3	97[c]	He-0.30	SA-316SS	7.44 x 6.58	87†	1910	5	5.00*	b.12
					B					
U188	MC+5%M_2C_3	85	He-0.18	SA-316SS	7.72 x 6.60	90	1930	11	11.0	EBR-II, Exam,[e,i]
U190	MC+5%M_2C_3	85	He-0.28	SA-INC-800	7.67 x 6.60	90	1930	11	11.0	EBR-II, Exam,[i]
U193	MC	92	Na-0.79	SA-304SS	7.75 x 6.91	94	1000	11	11.0	EBR-II, Exam,[e,i]
U196	MC+15% M_2C_3	98	Na-0.79	SA-304SS	7.75 x 6.91	97	1000	11	10.9	EBR-II, Exam,[e]
U199	MC+10%M_2C_3	98	Na-0.79	SA-INC-800	7.75 x 6.91	100	1000	11	8.2	EBR-II, Exam,[i]
U201	MC+5%M_2C_3	85	He-0.23	SA-304SS	7.32 x 6.60	90	2040	11	7.8	EBR-II, Exam,[e]
U207	MC+5%M_2C_3	90	He-0.25	SA-316SS	7.44 x 6.58	94	2090	11	7.9	EBR-II, Exam,[e,i]
U209	MC+10%M_2C_3	97[c]	He-0.30	SA-316SS	7.44 x 6.58	92	1910	11	7.9	EBR-II, Exam,[e,i]
					C					
U185	MC+10%M_2C_3	97	He-0.28	SA-316SS	7.72 x 6.60	90	2190	3	2.7	a.7
U186	MC+10%M_2C_3	97	He-0.28	SA-316SS	7.72 x 6.60	90	2190	3	2.7	a.7
U202	MC+5%M_2C_3	85	He-0	SA-316SS	6.83 x 6.38	94	1270	3	2.5	a.7
U203	MC+5%M_2C_3	85	He-0.05	SA-316SS	7.32 x 6.43	93	1260	3	2.5	a.7
U204	MC+10%M_2C_3	97[c]	He-0.05	SA-316SS	6.76 x 6.32	96	1130	3	2.6	a.7
U205	MC+10%M_2C_3	97[c]	He-0.08	SA-316SS	7.21 x 6.35	95	1120	3	2.6	a.7
					D					
U260	MC+10%M_2C_3	98	He-0.48	20CW-316SS	7.57 x 6.71	102	2590	12	0	EBR-II, unassigned
U261	MC+10%M_2C_3	98	He-0.38	SA-316SS	7.49 x 6.60	102	2590	12	0	EBR-II, unassigned[f]
U262	MC+10%M_2C_3	96	He-0.38	SA-INC-800	7.34 x 6.60	102	2590	12	0	EBR-II, unassigned

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

[b] Theoretical density of MC = 13.45 g/cm^3
Theoretical density of M_2C_3 = 12.72 g/cm^3

[c] Cored pellet with nominal 2.0 mm diametral axial hole.

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

[e] Element cladding failure.

[f] EBR-II eddy current test indicates capsule bond discontinuity.

[g] Burnup values marked with * were measured using the ^{148}Nd method. Remaining values were computed using an EBR-II power adjustment factor of 0.91.

[h] Linear power and centerline temperature marked with † are beginning-of-life values computed using the measured burnup results. Remaining values are based in EBR-II power adjustment factor of 0.91.

[i] Damaged at EBR-II

either Type 304 or 316 welded stainless steel tubing. Operating linear power ratings for these series are relatively high (79-110 kW/m). Capsules B-1-4, B-2-1, B-2-3 and B-2-5 have been examined using γ-scanning techniques for the detection of ^{137}Cs, and the elements are apparently intact. Further irradiation of B-1-4, B-2-1, and B-2-5 is planned. During the interim examination, capsules B-1-1, B-1-2, B-2-2, B-2-6,

TABLE 463-VI

SERIES WF ENCAPSULATED CARBIDE EXPERIMENTS

Expmt. No.	Fuel Type[a]	Fuel Density, % Theo.[b]	Bond and Diametral Gap, mm.	Clad Type[c]	Clad O.D. x I.D. mm.	Max. Linear Power, kW/m[f]	Maximum Centerline Temp., °C	Goal Burnup, at.%	Maximum Current Burnup at.%[e]	Status
W3F	MC+15%M_2C_3	93	Na-0.64	SA-316SS	7.62 x 7.01	81	1000	10	5.5	EBR-II, unassigned
W4F	MC+10%M_2C_3	95	Na-0.69	SA-316SS	7.62 x 7.01	82†	950	6	5.32*	b.12
W5F	MC+10%M_2C_3	91	Na-0.25	SA-316SS	6.35 x 5.84	60	925	10	5.4	EBR-II, Exam.
W6F	MC+10%M_2C_3	90	Na-0.28	SA-316SS	6.38 x 5.87	60	925	6	5.4	EBR-II, Exam.[d]
W7F	MC+5%M_2C_3	90	Na-0.69	20CW-316SS	7.62 x 7.01	81	1000	10	5.5	EBR-II, unassigned
W8F	MC+5%M_2C_3	94	Na-0.64	20CW-316SS	7.62 x 7.01	83†	950	6	5.8.2*	b.12
W10F	MC	89	Na-0.30	SA-316SS	6.38 x 5.87	60	925	10	5.4	EBR-II, unassigned
W12F	MC+25%M_2C_3	97	Na-0.33	SA-316SS	6.38 x 5.87	63	975	10	5.4	EBR-II, Exam.

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

[b] Theoretical density of MC = 13.45 g/cm^3
Theoretical density of M_2C_3 = 12.72 g/cm^3

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

[d] Element cladding failure indicated by gamma scanning.

[e] Burnup values marked with * were measured using the ^{148}Nd method. Remaining values were computed using EBR-II power adjustment factor of 0.91.

[f] Linear powers marked with † are beginning-of-life values computed using the measured burnup results. Remaining values are based on EBR-II power adjustment factor of 0.91.

and B-2-7, were found to have failed as indicated by γ-scanning for ^{133}Xe at EBR-II. Destructive examination of B-1-1 and B-2-2 is under way. Examination of the remaining experiments that are available is pending examination of higher priority capsules.

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. The three helium-bonded elements have not failed at a maximum burnup of 6 at.%. Helium-bonded experiment B-3-7 will be returned to LASL for destructive examination. The other two helium-bonded experiments will continue irradiation.

Three of the failed sodium-bonded elements in the Series B-3 have completed postirradiation examination. The fourth experiment of this type is awaiting examination pending examination of higher priority experiments.

The Series U5100 singly-clad experiments are described in Table 463-VIII. In this group, either single-phase or two-phase carbide fuel is sodium bonded to Type 304 or 316 stainless steel or to Incoloy 800. In seven of the elements, a perforated 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. An element failure was indicated in this subassembly after a maximum burnup of 3 at.%. The interpretation of results from EBR-II reactor monitoring systems was that the size of the failure was very small. The subassembly was removed from the reactor on January 1, 1974. Examination of these elements at EBR-II indicated that one element, U248, had failed. The failure was detected by means of a weight loss of 4 g. All other elements had no weight losses. Visual examination of U248 revealed no obvious failure point. After neutron radiography at ANL West, this element will be returned to LASL for destructive examination. It is planned to continue irradiation of the remaining elements.

TABLE 463-VII

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

Expmt. No.	Fuel Type[a]	Fuel Density, % Theo.[b]	Bond and Diametral Gap, mm.	Clad Type[d]	Clad O.D. x I.D. mm.	Max. Linear Power, kW/m[h]	Maximum Centerline Temp., °C[h]	Goal Burnup, at.%	Maximum Current Burnup at.%[e]	Status
					Series B-1					
B-1-1	MN	94	Na-0.48	SA-304SS	7.37 x 6.35	79†	1045†	5	5.70*	b.12[f]
B-1-2	MN	94	Na-0.46	SA-304SS	7.37 x 6.35	81	1100	9	5.7	a.7
B-1-4	MN	94	Na-0.30	SA-304SS	7.37 x 6.35	85	1100	10	2.7	LASL, unassigned[g]
					Series B-2					
B-2-1	MN	95	Na-0.53	SA-316SS	8.02 x 6.99	98	1230	10	5.7	EBR-II, unassigned
B-2-2	MN	94	Na-0.51	SA-316SS	8.01 x 6.98	93†	1230	9	5.48*	b.12[f]
B-2-3	MN	94	Na-0.51	SA-316SS	8.00 x 6.99	97	1230	12	8.5	EBR-II, Exam.
B-2-5	MN	94	Na-0.71	SA-316SS	8.00 x 7.21	97	1230	12	2.7	LASL, unassigned[g]
B-2-6	MN	94	Na-0.53	SA-316SS	8.03 x 7.49	109	1230	6	5.6	a.7[f]
B-2-7	MN	94	Na-0.51	SA-316SS	8.03 x 7.49	110	1230	12	5.6	a.7[f]
					Series B-3					
B-3-2	MN	88	Na-0.23	SA-316SS	8.01 x 7.22	110†	1235†	9	2.78*	b.12[f]
B-3-3	MN	91	Na-0.25	SA-316SS	8.01 x 7.22	116	1280	12	2.8	a.7[f]
B-3-4	MN	94	Na-0.33	SA-316SS	8.01 x 7.22	108†	1200†	12	2.70*	b.12[f]
B-3-5	MN	90	Na-0.25	SA-316SS	8.01 x 7.48	118†	1255†	6	2.74*	b.12[f]
B-3-6	MN	95[c]	He-0.13	SA-316SS	8.00 x 6.99	102	1920	10	6.0	EBR-II, Exam.
B-3-7	MN	89	He-0.13	SA-316SS	8.00 x 6.99	102	1920	6	6.0	EBR-II, Exam.
B-3-8	MN	90[c]	He-0.13	SA-316SS	8.00 x 6.99	97	1870	10	6.0	EBR-II, Exam.

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

[b] Theoretical density of MN = 14.17 g/cm^3

[c] Cored pellet with nominal 1.78 mm. diameter axial hole.

[d] Cladding is welded tubing
SA = Solution annealed

[e] Burnup values marked with * were measured using the ^{148}Nd method. Remaining values were computed using an EBR-II power adjustment factor of 0.91.

[f] Element cladding failure indicated.

[g] Available for further irradiation.

[h] Linear power and centerline temperature marked with † are beginning-of-life values computed using measured burnup results. Remaining values based on EBR-II power adjustment factor of 0.91.

The C-5 and O-N1 series of singly-clad experiments are described in Table 463-IX. Single-phase nitride fuel is sodium bonded to 20% cold-worked Type 316 stainless steel cladding in all of the fuel elements in this group. Profilometry measurements of the C-5 series elements have been made using the same equipment that will be used for the postirradiation examination. Shipment of selected elements to EBR-II is pending LASL review of the experiments from a quality assurance standpoint.

The O-N1 series of singly-clad experiments is similar to the C-5 series. The elements are fueled with $(U_{0.8}Pu_{0.2})N$ which is sodium bonded to 20% cold-worked Type 316 stainless steel cladding. Three elements have been rejected because of large fuel chips in the sodium annulus. The diameters of the elements have been measured on the same profilometer that will be used after irradiation. Eddy current examination of these elements indicates multiple sodium bond defects. All elements will be rebonded. It is planned that four of the fuel elements from Series O-N1 will be irradiated with selected elements from Series C-5.

In addition to the experiments described above, two nitride-fueled thermal irradiation experiments from

TABLE 463-VII

SERIES U5100 SINGLY CLAD CARBIDE EXPERIMENTS

Expmt. No.	Fuel Type[a]	Fuel Density, % Theo.[b]	Bond and Diametral Gap, mm.[c]	Clad Type[d]	Clad O.D. x I.D. mm.	Max. Linear Power, kW/m[f]	Maximum Centerline Temp., °C	Goal Burnup, at.%	Maximum Current Burnup at.%[f]	Status[e]
U241	MC	94	Na-0.46	SA-304SS	7.87 x 7.14	107	1170	6	3.0	EBR-II, X156
U242	MC	93	Na-0.44	SA-304SS	7.87 x 7.14	107	1170	9	3.0	EBR-II, X156
U243	MC	93	Na-0.80	SA-304SS	7.87 x 7.14	101	1150	6	2.9	EBR-II, X156
U244	MC	93	Na-0.44	SA-304SS	7.87 x 7.14	107	1170	9	3.0	EBR-II, X156
U245	MC	93	Na-0.81	SA-304SS	7.87 x 7.14	101	1150	12	2.9	EBR-II, X156
U246	MC	93	Na-0.43	SA-316SS	7.87 x 7.14	109	1190	6	3.1	EBR-II, X156
U247	MC	93	Na-0.81	SA-316SS	7.87 x 7.14	101	1150	6	2.9	EBR-II, X156
U248	MC	93	Na-0.81	SA-316SS	7.87 x 7.14	109	1140	12	3.1	EBR-II, X156
U249	MC	93	Na-0.43	SA-INC-800	7.85 x 7.14	109	1210	6	3.1	EBR-II, X156
U250	MC	93	Na-0.81	SA-INC-800	7.85 x 7.14	109	1140	6	3.1	EBR-II, X156
U251	MC	93	Na-0.81	SA-304SS	7.87 x 7.14	109	1140	12	3.1	EBR-II, X156
U252	MC	93	Na-0.64	SA-304SS	7.87 x 7.14	109	1140	12	3.1	EBR-II, X156
U253	MC	93	Na-0.66	SA-304SS	7.87 x 7.14	101	1140	12	2.9	EBR-II, X156
U254	MC	93	Na-0.66	SA-304SS	7.87 x 7.14	101	1140	12	2.9	EBR-II, X156
U256	MC+15%M_2C_3	96	Na-0.61	SA-304SS	7.85 x 7.14	101	1140	12	2.9	EBR-II, X156
U257	MC+15%M_2C_3	96	Na-0.58	SA-INC-800	7.85 x 7.14	100	1130	12	2.9	EBR-II, X156
U258	MC+15%M_2C_3	96	Na-0.58	SA-304SS	7.87 x 7.14	100	1140	6	2.9	EBR-II, X156
U259	MC+15%M_2C_3	96	Na-0.58	SA-INC-800	7.85 x 7.14	103	1150	12	3.0	EBR-II, X156

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

[b] Theoretical density of MC = 13.45 g/cm^3
Theoretical density of M_2C_3 = 12.72 g/cm^3

[c] Elements U252 through U259 contain slotted shroud tubes ~ 0.09 mm thick, made of V, Fe, 304SS, V, Ta, 304SS, and 304SS, respectively. The shroud thickness is not included in the bond gap value.

[d] SA = Solution annealed

[e] Element cladding failure indicated in subassembly

[f] Computed using an EBR-II power adjustment factor of 0.91.

ORNL (43N1 and 43N2) will be examined. Results and status will be reported in future reports.

A series of singly-clad elements is currently being designed to irradiate sodium-bonded carbide and nitride fuels under, as nearly as possible, identical conditions.

2. Postirradiation Examination Results

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

3. Fission Gas Release and Distribution

Sufficient postirradiation examination data are now available to calculate the fission gas release for 25 elements that are currently under examination. Of the 25 elements, data on 16 were reported in a previous progress report.[2] In addition, fission gas release results have been recalculated and reported here for three elements that had previously completed examination and been reported: U101[3], U109[3], and K-42B.[4] These data are used for comparative purposes.

The element data used to calculate fission gas release included 1) a measurement of the number of moles and an

TABLE 463-IX

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

Expmt. No.	Fuel Type[a]	Fuel Density, % Theo.[b]	Bond and Diametral Gap, mm.	Clad Type[c]	Clad O.D. x I.D. mm.	Max. Linear Power, kW/m	Maximum Centerline Temp., °C	Goal Burnup, at.%	Maximum Current Burnup at.%	Status
C-5-1	MN	93	Na-0.51	20CW-316SS	7.87 x 7.11	---	---	---	--	Reject[d,e]
C-5-2	MN	94	Na-0.51	20CW-316SS	7.87 x 7.11	---	---	---	--	Reject[d,e]
C-5-3	MN	95	Na-0.51	20CW-316SS	7.87 x 7.11	---	---	---	--	Reject[d,e]
C-5-4	MN	96	Na-0.53	20CW-316SS	7.87 x 7.11	109	1160	12	0	At LASL[g]
C-5-5	MN	96	Na-0.51	20CW-316SS	7.87 x 7.11	---	---	---	--	Reject[f]
C-5-6	MN	94	Na-0.53	20CW-316SS	7.87 x 7.11	109	1160	12	0	At LASL[g]
C-5-7	MN	95	Na-0.51	20CW-316SS	7.87 x 7.11	111	1180	12	0	At LASL[g]
C-5-8	MN	95	Na-0.76	20CW-316SS	7.87 x 7.11	107	1130	12	0	At LASL[g]
C-5-9	MN	95	Na-0.51	20CW-316SS	7.87 x 7.11	110	1130	12	0	At LASL[g]
C-5-10	MN	95	Na-0.51	20CW-316SS	7.87 x 7.11	107	1100	12	0	At LASL[g]
C-5-11	MN	95	Na-0.51	20CW-316SS	7.87 x 7.11	110	1120	12	0	At LASL[g]
C-5-12	MN	95	Na-0.76	20CW-316SS	7.87 x 7.11	107	1120	12	0	At LASL[g]
C-5-13	MN	96	Na-0.76	20CW-316SS	7.87 x 7.11	105	1110	12	0	At LASL[g]
C-5-14	MN	97	Na-0.76	20CW-316SS	7.87 x 7.11	105	1140	12	0	At LASL[g]
C-5-15	MN	96	Na-0.76	20CW-316SS	7.87 x 7.11	105	1110	12	0	At LASL[g]
C-5-16	MN	96	Na-0.76	20CW-316SS	7.87 x 7.11	107	1120	12	0	At LASL[g]
C-5-17	MN	97	Na-0.76	20CW-316SS	7.87 x 7.11	---	---	---	--	Reject[e]
C-5-18	MN	95	Na-0.53	20CW-316SS	7.87 x 7.11	107	1110	12	0	At LASL[g]
C-5-19	MN	94	Na-0.53	20CW-316SS	7.87 x 7.11	108	1120	12	0	At LASL[g]
C-5-20	MN	96	Na-0.53	20CW-316SS	7.87 x 7.11	106	1110	12	0	At LASL[g]
O-N1-1	MN	91	Na-0.51	20CW-316SS	7.87 x 7.11	108	1140	12	0	At LASL[g]
O-N1-2	MN	91	Na-0.51	20CW-316SS	7.87 x 7.11	---	---	---	--	Reject[d]
O-N1-3	MN	91	Na-0.51	20CW-316SS	7.87 x 7.11	107	1160	12	0	At LASL[g]
O-N1-4	MN	90	Na-0.51	20CW-316SS	7.87 x 7.11	108	1160	12	0	At LASL[g]
O-N1-5	MN	91	Na-0.51	20CW-316SS	7.87 x 7.11	---	---	---	--	Reject[d]
O-N1-6	MN	90	Na-0.51	20CW-316SS	7.87 x 7.11	---	---	---	--	Reject[d]
O-N1-8	MN	91	Na-0.51	20CW-316SS	7.87 x 7.11	109	1120	12	0	At LASL[g]

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

[b] Theoretical density of MN = 14.17 g/cm^3

[c] 20CW = 20% cold worked

[d] Fuel chips in bond

[e] Air in plenum

[f] Possible impurities present

[g] QA evaluation in progress

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.[5] Relative fission rate data including axial and radial variation for ^{235}U, ^{238}U, ^{239}Pu, and ^{240}Pu were obtained from the EBR-II irradiation guide.[6] The fission rates of other fissionable isotopes were estimated as ratios to known fission rates:

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

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

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

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

Table 463-X lists the measured Xe and Kr content of the fuel element plenum and capsule plenum, the measured isotopic content of Xe and Kr, and the total moles of gas in each plenum. Table 463-XI summarizes the fission gas release and burnup results. The peak burnup and axial average burnup shown in Table 463-XI 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. The number of moles of plenum gas shown in Table 463-X is based on the Method B calculation. A comparison of fission gas release calculated by the two methods (see Table 463-XI) shows that there is either fair agreement or the Method A calculation gives a substantially lower result. In a number of cases where the Method A fission gas release is low, a calculation of the fuel element plenum pressure using the measured number of moles and the plenum volume results in a pressure below the initial loading pressure. This indicates a loss of plenum gas. In general, the Method B fission gas release is considered a better estimate.

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 ^{85}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 (^{83}Kr and ^{85}Kr are always lower than ^{84}Kr and ^{86}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.

The results from all He-bonded carbide elements irradiated in EBR-II are being examined in order to correlate these fission gas release values with linear power, burnup, fuel density, and smear density.

An examination of the Xe isotopic compositions found in the capsules of failed Na-bonded carbide and nitride elements (see Table 463-X) indicated that the plenum gas in a number of capsules had a Xe isotopic composition that was grossly different from that in the element plenum (K-36B, K-45, K-46, U194, and B-1-1). No variations of this nature have been found in failed He-bonded elements. This observation indicates that the distribution of fission gas between the element and capsule is probably being influenced by the fuel element Na bond. In order to more completely examine the fission gas distribution between the element and capsule, a number of parameters have been calculated from the data of Table 463-X. Table 463-XIII lists three groups of calculated parameters, 1) the ratio of total capsule fission gas to total element fission gas, 2) the ratio of total Kr to total Xe for both the element and capsule, and 3) the ratio of Xe isotopic content in the capsule to that in the element. All three parameters show similar variations for four of the five elements noted above (K-36B, K-45, K-46, and B-1-1), indicating a common mechanism may be the cause.

TABLE 463-X

XENON AND KRYPTON COMPOSITIONS AND TOTAL MOLES OF PLENUM GAS

Element No. He Bonded	Plenum[a] Carbides	Total Moles of Gas[b]	Xe Content (Mole %)	Xe Isotopic Content (Mole %)[c] ^{131}Xe	^{132}Xe	^{134}Xe	^{136}Xe	Kr Content (Mole %)	Kr Isotopic Content (Mole %) ^{83}Kr	^{84}Kr	^{85}Kr	^{86}Kr
U 93[d]	E	1.92×10^{-3}	71.3	14.99	21.91	34.29	28.76	11.2	15.6	27.8	6.08	50.5
	C	2.55×10^{-4}	-	-	-	-	-	-	-	-	-	-
U 94[d]	E	4.65×10^{-3}	75.7	14.99	21.87	34.33	28.76	11.8	15.6	27.9	6.08	50.4
	C	1.90×10^{-4}	-	-	-	-	-	-	-	-	-	-
U 105[d]	E	3.20×10^{-3}	78.5	14.94	21.89	34.36	28.76	12.2	15.66	27.89	6.05	50.40
	C	2.08×10^{-4}	-	-	-	-	-	-	-	-	-	-
U 187[d]	E	1.35×10^{-3}	66.4	15.14	21.82	34.25	28.73	10.87	15.62	27.86	6.84	49.68
	C	2.27×10^{-4}	-	-	-	-	-	-	-	-	-	-
U 189[d]	E	1.29×10^{-3}	65.7	15.12	21.83	34.24	28.77	11.16	15.61	27.78	6.82	49.79
	C	2.36×10^{-4}	-	-	-	-	-	-	-	-	-	-
U 200	E	1.27×10^{-3}	65.6	15.2	21.6	34.5	28.7	10.2	15.6	27.5	6.82	50.1
	C	3.84×10^{-4}	6.4	15.1	21.2	34.7	29.0	1.0	16.0	27.7	6.8	49.5
U 206[d]	E	8.63×10^{-4}	57.0	15.22	21.84	34.21	28.71	8.93	15.64	27.60	6.86	49.90
	C	2.97×10^{-4}	-	-	-	-	-	-	-	-	-	-
U 208[d]	E	9.33×10^{-4}	54.9	15.15	21.79	34.34	28.68	8.63	15.58	27.56	6.88	49.99
	C	2.85×10^{-4}	-	-	-	-	-	-	-	-	-	-
U 101[f]	E	8.97×10^{-4}	55.0	15.26	21.77	34.30	28.67	8.92	15.58	27.47	7.05	49.89
	C	4.57×10^{-4}	28.6	14.99	21.63	34.66	28.72	4.64	15.48	27.56	7.02	49.95
U109[f]	E	8.10×10^{-4}	49.9	15.03	21.84	34.27	28.86	7.54	15.52	27.72	6.37	50.40
	C	4.20×10^{-4}	29.0	14.93	21.98	34.28	28.86	4.61	15.60	27.51	6.85	50.04
Na Bonded Carbides												
K-36B	E	1.81×10^{-3}	57.7	14.99	22.00	34.16	28.82	8.84	15.71	27.77	6.49	50.03
	C	1.77×10^{-4}	0.09	89	8	3	<1	≤0.002	(e)	(e)	(e)	(e)
K-42B[g]	E	8.70×10^{-4}	42.2	15.17	22.04	33.89	28.91	6.62	15.77	27.47	7.11	49.65
	C	1.62×10^{-4}	-	-	-	-	-	-	-	-	-	-
K-45	E	7.79×10^{-4}	20.9	14.75	21.88	34.40	28.96	3.28	15.7	27.6	6.74	50.0
	C	1.77×10^{-4}	0.23	73	11	11	4	<0.002	(e)	(e)	(e)	(e)
K-46	E	1.25×10^{-3}	27.4	14.74	22.01	34.33	28.90	4.28	15.61	27.44	6.94	50.00
	C	1.75×10^{-4}	0.08	86	8	5	1	<0.001	(e)	(e)	(e)	(e)
K-49[b]	E	1.14×10^{-3}	35.6	15.94	23.37	29.60	31.05	9.01	17.4	28.1	7.58	46.9
	C	4.31×10^{-3}	32.3	17.80	22.67	29.00	30.47	7.44	17.2	27.9	7.72	47.1
U 191[d]	E	4.84×10^{-4}	31.0	14.78	21.71	34.45	28.91	5.56	15.56	27.70	6.90	48.92
	C	1.97×10^{-4}	-	-	-	-	-	-	-	-	-	-
U 192[d]	E	7.49×10^{-4}	55.0	14.90	21.62	34.53	28.91	9.33	15.54	27.61	6.89	49.96
	C	2.07×10^{-4}	-	-	-	-	-	-	-	-	-	-
U 194	E	1.57×10^{-3}	63.0	15.2	21.7	34.4	28.6	9.85	15.5	27.3	6.88	50.3
	C	1.95×10^{-4}	0.45	26.4	53.0	13.2	7.4	0.07	(e)	(e)	(e)	(e)
U 195[d]	E	7.65×10^{-4}	55.7	14.82	21.76	34.45	28.93	9.35	15.62	27.64	6.87	49.87
	C	2.56×10^{-4}	-	-	-	-	-	-	-	-	-	-
U 197[d]	E	1.14×10^{-3}	65.3	14.97	21.74	34.36	28.89	10.5	15.62	27.52	6.90	49.96
	C	2.45×10^{-4}	-	-	-	-	-	-	-	-	-	-
U 198[d]	E	6.95×10^{-4}	58.6	14.73	21.72	34.55	28.96	9.43	15.63	27.61	6.87	49.89
	C	2.38×10^{-4}	-	-	-	-	-	-	-	-	-	-
W 4F[d]	E	9.03×10^{-4}	37.1	14.75	21.93	34.34	28.97	5.83	15.60	27.67	6.65	50.07
	C	1.27×10^{-4}	-	-	-	-	-	-	-	-	-	-
W8F[d]	E	8.52×10^{-4}	34.7	14.96	21.94	34.19	28.87	5.42	15.73	27.77	6.35	50.15
	C	1.40×10^{-4}	-	-	-	-	-	-	-	-	-	-
Na Bonded Nitrides												
B 1-1	E	3.72×10^{-4}	12.6	14.33	22.12	34.63	28.89	2.25	15.55	27.92	6.48	50.05
	C	4.18×10^{-4}	0.04	91	5	4	(e)	(e)	(e)	(e)	(e)	(e)
B 2-2	E	8.46×10^{-4}	25.70	14.73	21.82	34.35	29.08	4.0	15.6	27.5	6.75	50.1
	C	2.22×10^{-4}	3.86	17.25	22.31	33.51	26.89	0.58	17.1	27.8	6.75	48.3

TABLE 463-X (continued)

B-3-2	E	7.80×10^{-4}	22.8	12.84	21.67	34.82	30.66	3.78	15.2	27.2	7.02	50.3
	C	2.01×10^{-4}	7.54	23.0	24.2	33.5	19.3	1.00	20.8	30.6	6.3	42.3
B-3-4	E	8.54×10^{-4}	31.4	14.35	21.97	34.57	29.11	5.15	15.84	27.47	6.99	49.70
	C	3.94×10^{-4}	11.25	15.63	22.04	33.92	28.40	1.52	15.72	28.50	6.74	49.05
B-3-5	E	5.83×10^{-4}	14.8	12.83	22.03	35.36	29.76	2.87	15.6	27.7	6.97	49.8
	C	4.61×10^{-4}	28.5	15.93	21.77	33.71	28.57	3.96	15.9	27.4	6.97	49.7

(a) E = Element Plenum; C = Capsule Plenum.
(b) Based on Method B Calculation.
(c) Small quantities of other Xe fission product isotopes not reported.
(d) Unfailed element - no fission gas in capsule plenum.
(e) Insufficient fission gas for an analysis.
(f) Failed elements reported previously by Gulf United Nuclear Fuels Corp. (Ref. 2)
(g) Unfailed element reported previously by Los Alamos Scientific Laboratory (Ref. 3)
(h) Element contained ^{233}U.

TABLE 463-XI

FISSION GAS RELEASE SUMMARY

Element	Burnup, at.%		% Fission Gas Released[a]	
	Peak	Average	Method A	Method B
He Bonded Carbides				
U 93	9.64	8.68	12.5	16.3
U 94	9.42	8.48	1.7	34.9
U 105	9.89	8.91	8.2	34.0
U 187	4.60	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	9.6	9.6
U 208	5.00	4.51	10.5	10.3
U 101[b]	5.22	4.67	8.8	10.9
U 109[b]	7.04	6.31	4.1	7.2
Na Bonded Carbides				
K-36B	5.85	5.29	16.3	16.3
K-42B[c]	4.46	4.02	7.6	7.6
K-45	2.37	2.17	5.8	6.3
K-46	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 194	4.64	4.19	15.1	21.3
U 195	4.94	4.46	10.6	9.1
U 197	4.90	4.42	9.2	15.2
U 198	4.78	4.32	10.9	8.5
W 4F	5.32	5.09	8.5	8.1
W 8F	5.82	5.53	7.1	6.5
Na Bonded Nitrides				
B-1-1	5.70	5.16	0.72	0.86
B-2-2	5.48	5.21	4.1	4.5
B-3-2	2.78	2.65	6.9	7.1
B-3-4	2.70	2.57	5.8	11.5
B-3-5	2.33	2.21	8.4	8.6

[a] Calculation methods described in text.

[b] Elements reported previously by Gulf United Nuclear Fuels Corp. (Ref. 2).

[c] Element reported previously by Los Alamos Scientific Laboratory (Ref. 3).

TABLE 463-XII

FISSION GAS RELEASE OF INDIVIDUAL ISOTOPES
(Method B Calculation)

Element	% of Fission Gas Released							
	Kr				Xe			
	83	84	85[a]	86	131	132	134	136
He Bonded Carbides								
U 93	14.7	16.1	11.4	16.4	15.4	16.3	16.9	16.6
U 94	31.1	34.2	24.3	34.7	32.9	34.8	36.2	35.4
U 105	30.4	33.3	23.5	33.8	31.9	34.0	35.3	34.6
U 187	19.1	20.9	16.7	21.0	19.3	20.2	21.0	20.6
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 206	8.6	9.3	7.6	9.5	9.1	9.5	9.9	9.7
U 208[b]	9.3	10.1	8.2	10.3	9.8	10.3	10.7	10.5
U 101[b]	10.0	10.9	9.1	11.1	10.4	10.7	11.3	11.0
U 109[b]	6.3	6.9	5.3	7.0	6.8	7.2	7.4	7.3
Na Bonded Carbides								
K-36B[c]	14.9	16.2	12.4	16.5	15.1	16.2	16.9	16.5
K-42B[c]	7.1	7.6	6.5	7.8	7.1	7.5	7.7	7.6
K-45	5.8	6.3	5.0	6.4	5.8	6.2	6.5	6.4
K-46	12.1	13.1	10.8	13.4	12.0	13.1	13.6	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	10.6	9.3	9.8	10.3	10.1
U 194	19.0	20.5	16.9	21.2	20.4	21.1	22.2	21.5
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
W 4F	7.4	8.1	6.4	8.3	7.4	8.0	8.4	8.2
W 8F	6.1	6.6	4.9	6.7	6.1	6.5	6.8	6.6
Na Bonded Nitrides								
B-1-1	0.87	0.96	0.73	0.97	0.77	0.85	0.88	0.85
B-2-2	4.1	4.5	3.6	4.6	4.1	4.5	4.7	4.6
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

[a] Not corrected for decay.

[b] Elements reported previously in Gulf United Nuclear Fuels Corp. (Ref. 2).

[c] Element reported previously in Los Alamos Scientific Laboratory (Ref. 3).

TABLE 463-XIII

FISSION GAS DISTRIBUTION PARAMETERS

Element No.	Ratio of Capsule Fission Gas to Element Fission Gas[f]	Ratio of Total Kr/Total Xe		Ratio of Xe Isotopic Content in Capsule to Content in Element			
		Element	Capsule	^{131}Xe	^{132}Xe	^{134}Xe	^{136}Xe
	He Bonded Carbides						
U 93[a]	0	0.157	-	-	-	-	-
U 94[a]	0	0.156	-	-	-	-	-
U 105[a]	0	0.155	-	-	-	-	-
U 187[a]	0	0.164	-	-	-	-	-
U 189[a]	0	0.170	-	-	-	-	-
U 200	0.029	0.155	0.156	0.99	0.98	1.01	1.01
U 206[a]	0	0.157	-	-	-	-	-
U 208[a]	0	0.157	-	-	-	-	-
U 101[c]	0.27	0.162	0.162	0.98	0.99	1.01	1.00
U 109[c]	0.30	0.151	0.159	0.99	1.01	1.00	1.00
	Na Bonded Carbides						
K-36B[d]	9.5×10^{-5}	0.153	0.02	5.9	0.36	0.088	0.035
K-42B	0	0.157	-	-	-	-	-
K-45	1.9×10^{-3}	0.157	<0.01	5.0	0.50	0.32	0.14
K-46	2.5×10^{-4}	0.156	0.01	5.8	0.36	0.15	0.035
K-49[e]	3.4	0.253	0.230	1.12	0.97	0.98	0.98
U 191[a]	0	0.179	-	-	-	-	-
U 192[a]	0	0.170	-	-	-	-	-
U 194	3.5×10^{-4}	0.156	0.156	1.7	2.4	0.38	0.26
U 195[a]	0	0.168	-	-	-	-	-
U 197[a]	0	0.161	-	-	-	-	-
U 198[a]	0	0.161	-	-	-	-	-
W 4F[a]	0	0.157	-	-	-	-	-
W 8F[a]	0	0.156	-	-	-	-	-
	Na Bonded Nitrides						
B-1-1	2.7×10^{-3}	0.176	(b)	6.4	0.23	0.12	(b)
B-2-2	0.027	0.156	0.150	1.17	1.00	0.98	0.93
B-3-2	0.083	0.166	0.133	1.79	1.12	0.98	0.83
B-3-4	0.16	0.164	0.135	1.09	1.00	0.98	0.98
B-3-5	1.5	0.194	0.139	1.24	0.99	0.95	0.96

(a) Unfailed element - no fission gas found in capsule
(b) Insufficient fission gas for complete analysis.
(c) Failed elements reported previously by Gulf United Nuclear Fuels Corp. (Ref. 2).
(d) Unfailed element reported previously by Los Alamos Scientific Laboratory (Ref. 3).
(e) Element contains ^{233}U.
(f) Method B Calculation.

The ratio of total capsule fission gas to total element fission gas is an indication of the magnitude of the cladding failure. Elements K-36B, K-45, K-46, U 194, and B-1-1 have very small values for this ratio. The total quantity of fission gas in the capsules of these elements is less than 1×10^{-6} moles in each case. This small release of fission products is in line with the classification of these elements as slight failures based on visual and radiographic examinations. (An element classed as a slight failure is one in which the cladding breach that allowed fission products to escape cannot be found.) Thus all five elements have the common trait of releasing only a very small quantity of fission products.

The ratio of total Kr to total Xe should approximate the ratio of yields of these gases. This is the case for the element plenum data and some capsule plenum data, but four of the five elements noted above (K-36B, K-45, K-46, and B-1-1) have very low values for this ratio (see Table 463-XIII). In addition to these large differences, the ratio of total Kr to total Xe in the capsules of Na-bonded elements is always equal to or less than the ratio in the element. Thus the same mechanism which produced abnormally low values in a few elements may be in operation in all Na-bonded elements.

The ratio of the Xe isotopic content in the capsule to that in the element (see Table 463-XIII) is a parameter

which is sensitive to the variation that was first observed, i.e. the grossly different Xe isotopic contents of some elements and capsules. Four of the five elements noted above (K-36B, K-45, K-46, and B 1-1) were highly enriched in ^{131}Xe and depleted in the other Xe isotopes in the capsule. The fifth (U 194) was enriched in ^{131}Xe and ^{132}Xe and depleted in the others. A closer examination of all failed, Na-bonded elements indicated that all capsules were enriched to some extent in ^{131}Xe and depleted in ^{134}Xe and ^{136}Xe. This was even true when the capsule contained more total fission gas than the element (K-49 and B-3-5). The data for failed, He-bonded elements showed that the elements and capsules had essentially identical Xe isotopic compositions. A similar statement can be made here to that made for the total Kr to total Xe ratio data, i.e. a single mechanism may be operative in all Na-bonded elements which affects the distribution of fission gas between the element and capsule. In a few cases, this mechanism predominates and results in grossly different distributions.

A calculation of the ratio of the Kr isotopic content in the capsule to that in the element is not shown. This was omitted because many of the elements that would show significant variation did not have enough Kr in the capsule to allow an isotopic analysis (see Table 463-X).

Some feeling for mechanisms which could explain the observations of Table 463-XIII can be obtained from an examination of the calculated isotopic concentrations of fission product Xe and Kr and their precursors as a function of irradiation time. These calculations were performed using the RIBD code[7] and the fuel composition of U 194. (Small variations in the calculated fission product compositions would result from the different U and Pu compositions of different fuel elements but these differences would not change the conclusions.) Figures 463-1 through 463-6 show the isotopic concentration of fission product Xe, I, Te, Kr, Br, and Se as a function of irradiation time.

One possible mechanism, the release of newly produced fission gas from the element to the capsule early in life or shortly after a prolonged shutdown, can be eliminated immediately by examining Fig. 463-1. Newly

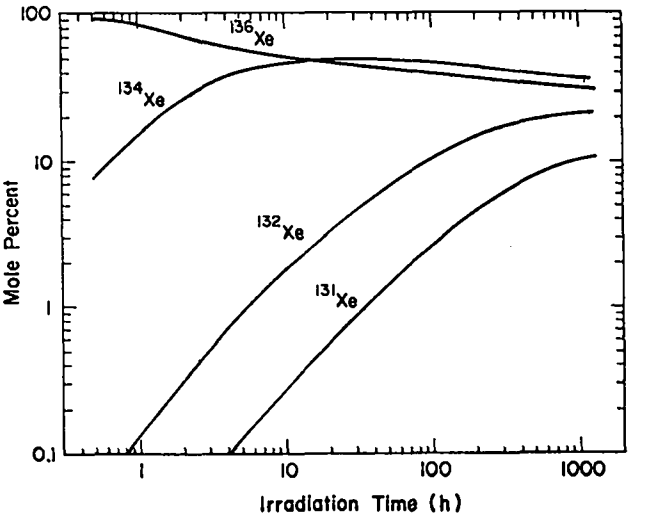

Fig. 463-1. Isotopic concentration of xenon fission products vs irradiation time.

produced Xe is enriched in ^{136}Xe and depleted in ^{131}Xe. This is the opposite of what was observed.

Considering only the four elements which showed similar anomalous behavior (K-36B, K-45, K-46, and B 1-1), a mechanism involving precursors can qualitatively explain the observations. In the range of 30 to 100 hours irradiation time, the isotopic concentration of I precursors approximates the capsule Xe isotopic

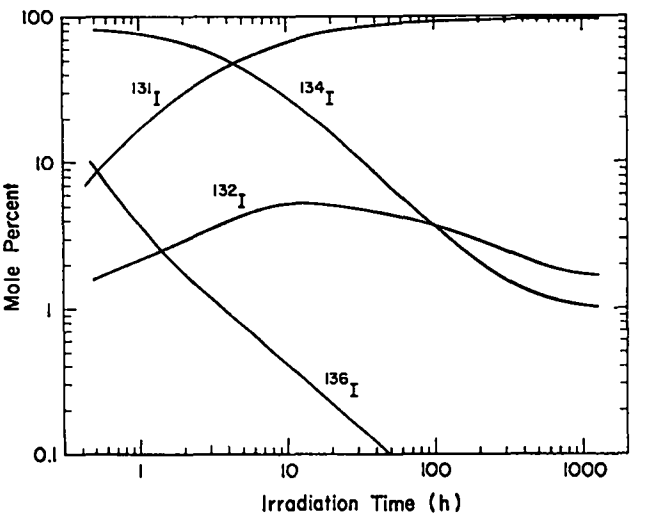

Fig. 463-2. Isotopic concentration of iodine fission products vs irradiation time.

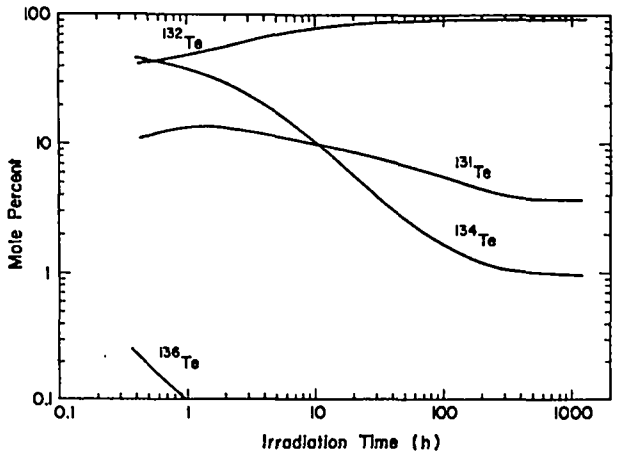

Fig. 463-3. Isotopic concentration of tellurium fission products vs irradiation time.

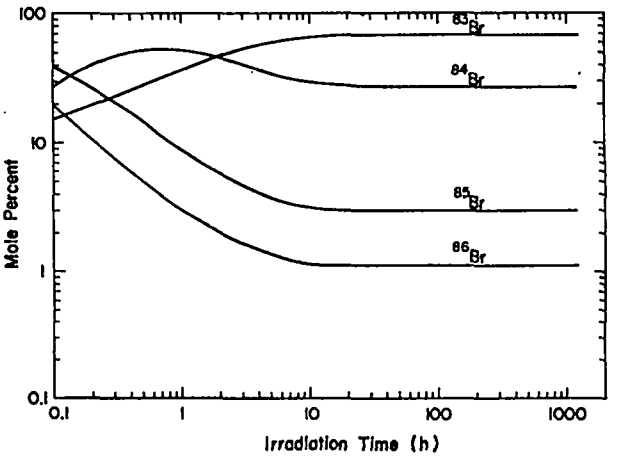

Fig. 463-5. Isotopic concentration of bromine fission products vs irradiation time.

concentrations observed for these elements (see Fig. 463-2 and Table 463-X). Thus the release of fission products from the elements could be a transfer of element bond sodium containing dissolved I (as NaI) from the element to the capsule. This I subsequently decays to Xe and is released to the capsule plenum. There is evidence that a sodium transfer in this direction occurs. The element sodium level of a number of fuel elements classed as slight failures has been observed to be lower after irradiation than as fabricated.[1] Estimates of the quantity of sodium transferred from the element to the capsule are in the range of 1/3 to 2 cm^3. The solubility of I (as NaI) in sodium was estimated to be 4×10^{-6} moles/cm^3 at 500°C.[8] The quantity of Xe in the capsules of these four elements ranged from 1×10^{-7} to 4×10^{-7} moles. Thus sufficient sodium was transferred into the capsule to account for all the Xe found in the capsules.

This same mechanism can also explain the low values of the total Kr to total Xe found in the capsules of these four elements. Figure 463-7 shows a plot of the

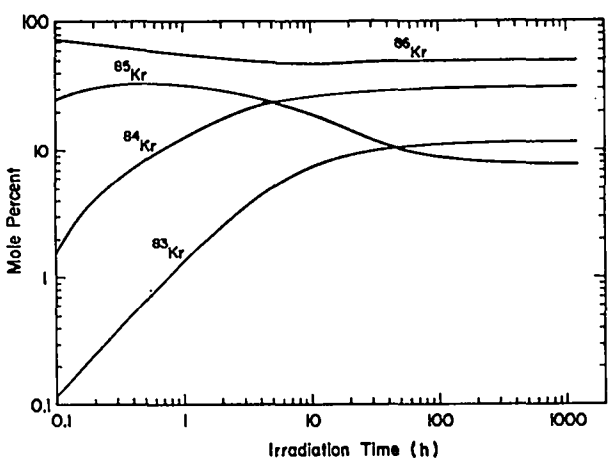

Fig. 463-4. Isotopic concentration of krypton fission products vs irradiation time.

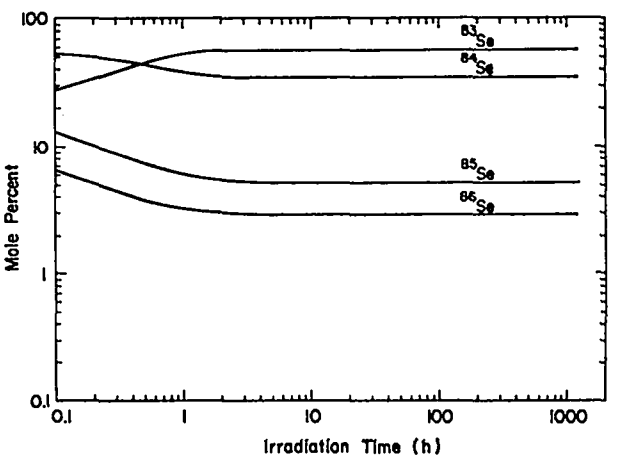

Fig. 463-6. Isotopic concentration of selenium fission products vs irradiation time.

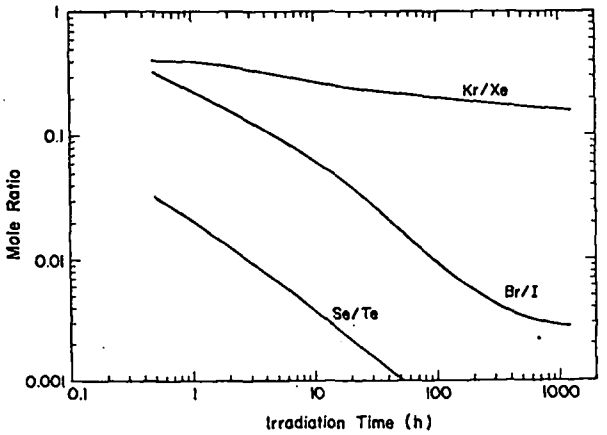

Fig. 463-7. Kr/Xe, Br/I, and Se/Te fission products ratios vs irradiation time.

Kr/Xe, Br/I, and Se/Te ratios as a function of irradiation time. These data came from the same calculations used to produce Figs. 463-1 to 463-6. The Kr/Xe ratio decreases monotonically with time, so that a low value would not result from the transfer of newly produced gas. If it is assumed that the Xe in the capsules of these four elements comes mainly from the decay of I precursors, a similar assumption can be made for the Kr; i.e. it comes mainly from the decay of Br precursors. Thus the final Kr/Xe ratio measured would be controlled by the Br/I ratio in the sodium transferred into the capsule. In the same range of irradiation times where I precursor isotopic concentrations matched the Xe isotopic concentrations observed (30 to 100 h), the Br/I ratio matches the Kr/Xe ratio observed in the capsules of the four elements. Thus the observations for these four elements in Table 463-XIII are consistent with the hypothesis that the fission gas in the capsules was transferred mainly as halogen precursors dissolved in the element bond sodium.

The observed Xe isotopic content and the total Kr/Xe ratio in the capsule of U 194 are not in agreement with this mechanism. The capsule fission product Xe is more highly enriched in ^{132}Xe than ^{131}Xe. The release of neither Xe nor I would result in a higher ^{132}Xe enrichment than ^{131}Xe (see Figs. 463-1 and 463-2), but the preferential release of Te could result in a high ^{132}Xe enrichment after decay (see Fig. 463-3). Also the total Kr/Xe ratio in the capsule is identical to that in the element. There does not appear to be any simple mechanism which can account for these two observations on U 194. This anomaly will continue to be studied.

The observation that all Na-bonded elements show some enrichment in ^{131}Xe and depletion in ^{134}Xe and ^{136}Xe in the capsule fission gas can be explained as a combination of the release of fission gas (Xe and Kr) directly from the element to the capsule through cladding cracks, and the transfer of some bond sodium between the element and capsule. This bond sodium would contain dissolved precursors of Xe and Kr which would skew the capsule fission gas isotopic compositions to some extent.

Of the 18 Na-bonded elements, K-49 requires special consideration. This element operated at a very high linear power (130 kW/m) and was a severe failure which showed evidence of fuel melting over much of the fuel column length. Thus the high gas release (see Table 463-XI) is not unexpected, considering the operating history. Two of the Na bonded nitride elements (B 3-4 and B 3-5) were also severe failures. Although no fuel melting was observed, the fuel temperatures were higher than normal due to some gas blanketing of the element. Thus the fission gas release of these elements may be higher than normally expected for Na-bonded nitrides.

If the fission gas release of Na-bonded nitride elements is compared with Na-bonded carbides (excluding the three elements noted above, K-49, B 3-4, and B 3-5),the nitride fueled elements show a lower release (0.86% to 7.1%) than the carbide elements (3.2% to 21.3%). It is not certain at this time whether this is a significant trend.

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

The four Series UL TREAT tests (see Table 463-XIV), 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-XIV

LASL SERIES UL TESTS

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

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

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

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

The inner capsules from tests UL-3 and UL-4 have been x-radiographed and gamma scanned at LASL. An examination of the radiographs of UL-3 (the He-bonded element) indicates cladding cracks in a number of areas. An examination of the radiographs of UL-4 (the Na-bonded element) indicates a number of areas which may have cladding cracks. There is no evidence of fuel melting in either radiograph. An analysis of the gamma scanning results is still in progress but a preliminary review of the results indicates that the He-bonded element (UL-3) has failed since ^{137}Cs was detected outside the element cladding. There was no evidence of fission products outside the cladding of the Na-bonded element (UL-4).

III. QUALITY ASSURANCE
(L. E. Lanham)

Fuel Preparation

The Engineering Test Plan for the development of fuel fabrication has been amended to extend its use to the preparation of nitride fuels. Several development plans have been written and reviewed by the Quality Assurance Manager. Each has been written and documented using the quality assurance requirements of the development Engineering Test Plan. Independent quality assurance surveillance has been conducted during these developmental activities.

Fuel preparation documentation consisting of an Engineering Test Plan, fuel material specifications, fuel preparation parameters, work instructions, travelers, and reference procedures has been prepared for the production of fuel insulator pellets for the LASL Series K-4 fuel elements. Work has been started on this fuel preparation and those operations in progress were audited as a part

of the audit of the Fuel Preparation operation. An audit report has been written.

Fuel Pin Fabrication

Additional procedures for Fuel Pin Fabrication have been written and approved. Equipment and systems are being checked and calibrated. The cladding to be used for the preparation of weld samples has been reviewed and approved for fabrication. The fabrication has been completed and each piece has been inspected. Independent quality assurance surveillance has been made on a continuing basis for all operations.

IV. REFERENCES

1. "Quarterly Report, Advanced Plutonium Fuels Program, April 1 through June 30, 1973 and Seventh Annual Report, FY 1973," Los Alamos Scientific Laboratory report LA-5390-PR (Sept. 1973).

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4. J. O. Barner, "Behavior of Sodium-Bonded (U,Pu)C Fuel Elements After Moderate Burnup," Los Alamos Scientific Laboratory report LA-4669-MS (April 1971).

5. M. E. Meek and B. F. Rider, "Compilation of Fission Product Yields, Vallecitos Nuclear Center, 1972," General Electric report NEDO-12154 (1972).

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7. R. O. Gumprecht, "Mathematical Basis of Computer Code RDBD," Douglas United Nuclear, Inc. report DUN-4136 (June 1968).

8. M. A. Bredig et al., "Miscibility of Liquid Metals with Salts, I. The Sodium-Sodium Halide System," J. Am. Chem. Soc. 77, 307 (1955).

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 analysis 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. Also initiated is a task to prepare a manual of analytical methods for the chemical characterization of metallic core components for issuance as an RDT Standard.

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

In August of 1973, LASL began a cooperative effort with HEDL to establish a program for the chemical characterization of low-friction hard surfaces to be applied to contacting components of the FFTF reactor. The hard surface is to be chromium carbide applied as a molten blend of Cr_3C_2 and nichrome powders.

A. Round Robin Evaluations of Analytical Capabilities

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

HEDL has reported that the batches of nichrome powder, chromium carbide powder, and hard surface that are to serve as round robin materials were received in late March. The hard surface material has to be converted to homogeneous powder by HEDL before it can be used.

B. Analytical Method Manual

Drafts of eight methods for (1) General Information on Sampling and Analysis, (2) Dissolution of Samples for Subsequent Chemical Determination of Chromium, Nickel, and Aluminum, (3) Chromium, (4) Nickel, (5) Aluminum, (6) Carbon, (7) Nitrogen, and (8) Oxygen were sent to HEDL for review. Based on the HEDL review, final writeups are being prepared.

HEDL has provided drafts of four methods for (1) Carbon in Chromium Carbide and Hardfacing, (2) Carbon in Nichrome, (3) Iron, Cobalt, and Nickel in Nichrome, and Iron in Chromium Carbide, and (4) Silicon in Nichrome Powder that are being reviewed.

III. ANALYTICAL CHEMISTRY PROGRAM FOR METALLIC CORE COMPONENTS

(W. H. Ashley, E. A. Hakkila, M. E. Smith, J. E. Rein, G. R. Waterbury)

Jointly with HEDL, analytical methods for RDT Standard F11-3 "Analytical Chemistry Methods for Metallic Core Components" are being prepared. Rough drafts of methods are essentially complete and final writeups have been started.

IV. ANALYTICAL CHEMISTRY PROGRAM FOR BORON CARBIDE

The status of all tasks in this program is essentially the same as reported last quarter.[1] No significant further effort is anticipated until contracts are let by HEDL and production of FFTF boron carbide pellets is started.

V. ANALYTICAL CHEMISTRY PROGRAM FOR FBR MIXED OXIDE FUEL

A. Calibration Materials and Quality Control Samples

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

Packaging has been completed of calibration materials and quality control samples presently available for shipments C and D, nominally shipped by March 1974. Shipment C materials are being sent minus those specific items for which the supply has been depleted. Packaging of Shipment E materials is under way.

HEDL has supplied batches of plutonium oxide and mixed oxide powders intended as matrix material for additional calibration and quality control blends. These powders are being analyzed for the various metal and nonmetal impurities to establish whether the levels are satisfactorily low to permit their use.

B. Reference Analysis

(R. G. Bryan, R. K. Zeigler, G. R. Waterbury)

A series of mixed oxide pellets, representative of shipments of fuel by a vendor to HEDL, is being analyzed under a statistical design to provide plutonium content referee values. Analyses will be completed during April, 1974.

C. Development of Analytical Methods

1. Determination of Burnup

a. Mass Spectrometric Procedure

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

A LASL report that describes the modified chemical separation procedure for uranium plutonium, and neodymium described last quarter[1] is undergoing final editing.

b. Method Using Conventional Low-Cost Apparatus

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

Development was continued on a method incorporating the ion exchange separation of uranium, plutonium, and total rare earths (as the fission product monitor) followed by a spectrophotometric measurement of each. The separation procedure involves fuming the sample with $HClO_4$ to oxidize Pu to Pu^{+6} prior to retention of uranium and plutonium on a macroporous anion exchange column from 12M HCl. The effluent which contains trivalent actinides, rare earths, and most other fission products, is converted to an ethanol - HCl medium and transferred to a pellicular cation exchange resin column. The rare earths are sorbed while trivalent actinides (Am and Cm) and extraneous fission products pass through. The rare earths then are eluted with 5M HCl. Plutonium and uranium are sequentially eluted from the first (anion exchange) column with 0.1M HI - 12M HCl, followed by 0.1M HCl.

A mixture of rare earths plus yttrium, in proportions simulating fission products from a fast reactor, mixed uranium-plutonium fuel, has been processed repeatedly through the separation procedure and colorimetrically measured as the arsenazo-III complex. Frequently, the measured absorbance has been low compared to equal

amounts of the mixture not processed through the separation procedure. Part of this lower absorbance has been traced to a wavelength shift of the absorbance peak caused by a higher salt content of the separated fraction. This higher salt content is produced when the 5M HCl effluent is neutralized with NaOH, necessary to develop the rare earth-arsenazo-III colored complex. This difference is eliminated by evaporating effluents to remove the HCl prior to pH adjustment.

The cause for the remainder of the lower absorbance seems to be associated with a degradation product of the pellicular cation exchange resin released during elution of the rare earths. Experiments with radiotracers of lanthanum and europium, covering the range of rare earths in fission products, and with yttrium have shown that there is no leak-through nor delayed elution of the rare earths. Other investigators have reported degradation of these resins by strong acid. We speculate that such degradation products complex rare earths thereby competing with the rare earth - arsenazo II complex formation.

Pellionex pellicular resin (distributed by Reeve - Angel) has been used for most of this investigation. A du Pont product, Zipax pellicular resin, was tested. The separation of rare earths from americium was not satisfactory.

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

Gravimetric methods for determining the O/M ratio in UO_2, PuO_2, and $(U,Pu)O_2$ are being investigated in a thermobalance as a function of oxygen potential and temperature.

A gas manifold was constructed that provides controlled generation of gases with very low oxygen potentials in thermobalance. The oxygen potential is controlled by the ratio of water vapor to hydrogen in an argon-carrier stream. The source of hydrogen either is the highly purified gas itself or a pure 0.1% H_2-Ar mixture. The water is produced from H_2 and O_2 at a heated platinum catalyst located just prior to introduction into the thermobalance. The H_2 and O_2 in a 2/1 ratio are produced by electrolysis of water. A Molecular Sieve trap is located between the electrolysis cell and the heated platinum catalyst to remove water vapor carried from the cell. The rate of electrolysis is controlled to produce an oxygen potential range of -292 to -569 kJ $mole^{-1}$ (-70 to -135 kcal/mol) at 800°C and -234 to -611 kJ mol^{-1} (-56 to -146 kcal/mol) at 1200°C.

Highly pure uranium metal and plutonium are the starting materials so that all weight changes of formed oxides can be computed in terms of oxygen-to-metal (O/M) ratios. Separate studies of uranium and of plutonium are in progress. Preparation of a U-Pu metal alloy and study of its oxide is scheduled for next quarter.

3. Development of Gas Measurement Techniques
(R. M. Abernathey, J.E. Rein)

Electrical power has been installed necessary to operate the apparatus that is designed to measure gas components as they are released from samples of fuel, boron carbide, and other FBR materials as a function of temperature. Initial testing is scheduled for next quarter.

VI. QUALITY ASSURANCE
(L. E. Lanham)

A possible problem has been brought to the attention of LASL in the identification of quality control samples. An evaluation was made by the Quality Assurance Manager of the procedures and documentation as they apply to the specific problem. The assurance of identification and traceability of samples is provided by the document signatures certifying to the work performed and the overcheck. Procedures for sample preparation and overchecking are complete.

VII. REFERENCE

1. R. D. Baker, "Quarterly Report-Advanced Plutonium Fuels Program October 1 to December 31, 1973," Los Alamos Scientific Laboratory report LA-5582-PR (October 1 - December, 1973).

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