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First Annual Report, FY 1975

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Compiled by

R. D. Baker

Los Alamos
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ABSTRACT

This is the first annual report on the Advanced Fast Reactor 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 548 (A1696)

FABRICATION OF ADVANCED LMFBR FUELS

Person in Charge: R. D. Baker

Principal Investigators: J. L. Green and K. W. R. Johnson

I. INTRODUCTION

An essential portion of the National Advanced LMFBR Fuels Program is preparation of fuel and fabrication of experiments for testing under steady-state and transient irradiation conditions. To assure the quality of the product, all fuel is prepared and experiments are fabricated in accordance with RDT Quality Assurance Standard F2-2.

The primary effort at LASL is devoted to fabrication of fuel and elements for steady-state irradiation in the EBR-II reactor. These elements are fabricated to specifications provided by the Irradiation Task Group. A significant fabrication effort is also devoted to design and fabrication of experimental, instrumented assemblies for irradiation in the TREAC reactor, in response to experimental concepts and requirements provided by ANL. Additional fabrication efforts include providing various fuel forms for property measurements at LASL and ANL, and supplying other laboratories in the Advanced LMFBR Fuels Program with fuel process technology and fuel for development of characterization capabilities.

Because current experiments use a variety of fuel densities, chemistries, and diameters, developmental experiments are performed to establish fabrication parameters for each new fuel form. At the same time, the procedures for preparation of existing fuel forms are continually examined to improve process control and efficiency. A similar effort is devoted to development and improvement of element fabrication capabilities.

Existing fabrication facilities are also continually upgraded and a new fuel preparation pilot line is being installed to increase the fuel preparation capacity.

Associated with this project is the assigned responsibility for coordinating the fabrication activities of all laboratories in the Advanced LMFBR Fuels Program. The initial effort involves implementation of a program for certification of other fabricators. Additional coordination responsibilities include the dissemination of information pertaining to all of the fabrication laboratories and serving as technical consultant to these laboratories in their efforts to establish a production capability.

II. FABRICATOR CERTIFICATION AND LIAISON
(O. A. Lee)

LASL was requested by ERDA/RDD to coordinate the establishment of several other laboratories as qualified fabricators of advanced fuel and elements for irradiation experiments (uranium-plutonium carbides and nitrides). The Advanced LMFBR Fuels Program requires that each fabricator establish the technical capability for fuel and element production and establish a quality assurance system as defined in RDT Quality Assurance Standard F2-2, to be implemented during all phases of process development and production. Simultaneously, a high level of attention must be afforded to safety and safeguards; especially in anticipating and recognizing accident and malfunction scenarios. The Advanced LMFBR Fuels Program requirements are described in appropriate sections of RDD manuals, NRC guides, ERDA manual chapters,

10 CFR of the Federal Register, etc. Because direct references to the Advanced LMFBR Fuels Program are not made, some interpretation is required.

Atomics International, Battelle Columbus Laboratories, and Westinghouse Advanced Reactor Division are participants with LASL in the Advanced Fuels Program as fabricators of fuel and elements. Those laboratories had previous experience in advanced fuels work, but the facilities had been decommissioned for several years. During that earlier period, basic fuel and fuel element fabrication processes were developed at LASL. AI, BCL, and W-ARD have been asked to transfer those processes to their laboratories, with process development as necessary for adjustment of parameters. Guidance documents (Ref. LASL internal report, Sept. 17, 1974) were formulated describing a sequence through the development of technology and quality assurance, ultimately leading to "certification" as RDD-qualified fuel fabricators. In addition, LASL coordinated the development of a target specification for advanced fuels produced by all fabricators. The intent of the specification is to minimize the degree to which irradiation test results are fabricator-dependent.

The process by which certification is achieved involves a series of RDD/LASL team reviews of the quality assurance system developed and implemented by each fabricator for fuel and element production, the state of safety and safeguards, and product characterization capability. Each aspect is addressed twice: During early development of technology and quality assurance, and later, when the fabricator demonstrates the capability to independently produce fuel and fuel elements meeting RDD program requirements. The certification process includes the following phases.

Preliminary: An RDD/LASL team visits each laboratory for an evaluation of facilities, safety and safeguards, and to develop a mutual understanding of the certification sequence.

Phase I: A review is made of the safety and safeguards, facilities, and procedural and quality assurance documentation systems under development by each fabricator in support of process development plans.

Phase II: An in-plant review is made by the RDD/LASL team to determine the correlation of the fabricator's quality assurance system with actual fabrication process development operations. The fabricators are required to undertake fabrication process development work under a complete QA system.

Phase III: An exchange is made between LASL and each fabricator of chemical, metallographic, and radiographic samples of fuel and fuel element materials to certify the fabricator's capability to characterize the products per RDD program requirements.

Phase IV: An in-plant review is made, similar to Phase II, except that the RDD/LASL team evaluates the fabricator's quality assurance, fabrication, and product characterization functions in producing a preproduction demonstration quantity of usable fuel and elements. Upon successful completion of Phase IV, LASL will recommend to RDD that the fabricator be considered certifiable for independent participation in the program.

Throughout the certification phases, critical evaluations are made by the certification review team. In areas of lesser concern, recommendations are made directly to the fabricator; where concern is greater, as in safety-related items, recommendations are made to RDD to identify action items. Extensive liaison with the fabricators has been required to sustain progress in the certification process. LASL has provided technical specialists in areas of analytical chemistry, microstructural analysis, fabrication technology, quality assurance, non-destructive analysis, and management and coordination. It is expected that all of the fabricators will be certified during FY 1976. At the end of FY 1975, the overall effort among all fabricators was approximately half completed, as shown in Table 548-I.

III. NEW FACILITIES PREPARATION

(J. G. Reavis)

The LASL Advanced Fuels Fabrication glovebox line, consisting of a room in the basement of Bldg. 150, TA-21 (with nine gloveboxes, and associated equipment, and introductory boxes) was planned, designed, and con-

TABLE 548-I
SUMMARY OF CERTIFICATION^a STATUS

I. Atomics International						
	<u>Pre-Certification</u>					
	<u>Review</u>	<u>Phase I</u>	<u>Phase II</u>	<u>Phase III</u>	<u>Phase IV</u>	
Fuel	Held-September '74	Complete	Held-May	Initiate-Aug.	Oct.	
Elements	Held-September '74	Complete	Mid-Oct.	In Progress	Dec.	
II. Battelle Columbus Laboratories						
	<u>Pre-Certification</u>					
	<u>Review</u>	<u>Phase I</u>	<u>Phase II</u>	<u>Phase III</u>	<u>Phase IV</u>	
Fuel	Held-September '74	Complete	Held-Mar.	In Progress	Mid-Aug.	
Elements	Held-September '74	Complete	Mid-Aug.	Complete	Oct.	
III. Westinghouse Advanced Reactors Division						
	<u>Pre-Certification</u>					
	<u>Review</u>	<u>Phase I</u>	<u>Phase II</u>	<u>Phase III</u>	<u>Phase IV</u>	
Fuel	Held-December '74	In Progress	Early-Sept.	Initiate-July	Jan. '76	
Elements	Held-December '74	In Progress	Oct.	Initiate-July	Feb. '76	

^a Fabrication certification sequence as described in a LASL internal document, dated Sept. 17, 1974.

structed. The installation was largely completed by the end of FY 1975. Exceptions are two large high-temperature furnaces, which were not delivered by the promised date, and an automatic press which is now being used for development work outside the glovebox. A hydrogen treatment system was also deferred. Leak checking and shakedown operations were about 50% completed. The facility is expected to become fully operational during FY 1976.

The chronology of the installation is listed in Table 548-II. The effort indicated by Table 548-II was broken down into three categories: (1) planning and preliminary sketches, (2) studies and construction drawing preparation, and (3) construction. Item (1) required 3 months, item (2) required about 5 months, and item (3) required about 4 months. It appears that the minimum time for establishment of such a facility is about one year. The essential factor in expeditious construction of the facil-

ity was timely ordering and delivery of major equipment items. Table 548-III lists the dates of completion of specifications, the date of placement of the order, the shipping date promised by the manufacturer, and the actual date received at LASL.

It can be seen from this tabulation that procurement (the time span between "Specification" and "Ordered") was in some cases lengthy, and that after placement of the order, the performance of suppliers was variable. The gloveboxes presented a special problem in that the first order was placed in July 1974, with a shipping date of November 1974. An inspection trip to the manufacturer's (mfg. "A") plant in January 1975 indicated that delivery could not occur before March, and was not certain even then. A rush order was placed with an alternate manufacturer (mfg. "B") who delivered all the gloveboxes by March 1975.

TABLE 548-II
CHRONOLOGY OF THE INSTALLATION OF THE
ADVANCED FUEL FABRICATION GLOVEBOX LINE OPERATION

<u>Date</u>	<u>Operation</u>
July-August 1974	Preliminary planning and preparation of sketches of the room and glovebox arrangement
August 1974	Formal request to the Engineering Dept. for a scoping study
November 1974	Engineering Dept. scoping study completed
November 1974	Scoping study revision completed
December 1974	Lab Job Folder given to Engineering Design Section
December 1974	Construction drawings approved
January 1975	Final drawings approved
January 1975	Detailed cost estimate completed
February 1975	Preliminary extension of electrical, water and gas lines
March 1975	Craftsmen begin connecting utility services to boxes
March 1975	Delivery of gloveboxes completed
June 1975	Installation completed except for high-temperature furnaces, hydrogen system, automatic press, and leak checking

TABLE 548-III
HISTORY OF ORDERING AND DELIVERY OF
MAJOR EQUIPMENT ITEMS

<u>Item</u>	<u>Specification Completed</u>	<u>Date Ordered</u>	<u>Promised Ship Date</u>	<u>Received at LASL</u>
Gloveboxes (mfg. "A")	4/74	7/74	11/74	Defaulted
Gloveboxes (mfg. "B")	4/74	1/75	3/75	3/75
Atmosphere Purifiers	4/74	7/74	1/75	1/75
Automatic Press	3/74	7/74	3/75	4/75
Sintering Furnaces	11/74	2/75	5/75	Pending
Oxygen Analyzers	2/75	3/75	5/75	6/75
Moisture Monitor	3/75	3/75	4/75	4/75

Many new design features were incorporated into the new facility and minor reworking was frequently required to produce usable items. Among items in this category were airlock doors, alpha transfer receptacles, recirculating-atmosphere purification systems, oxygen- and moisture-monitoring systems, service panels, electrical feed-throughs, and power and control systems. The new systems are expected to give safer, more dependable operation, but resulted in expenditure of greater time and effort during construction and shakedown.

IV. PRODUCTION

A. The Fuel Production Process (R. J. Herbst, R. L. Gutierrez)

The production of advanced LMFBR fuel and insulator pellets by carbothermic reduction of oxide starting materials was proved viable. Sufficient fuel and insulator pellets were supplied for several subassemblies to be irradiated in EBR-II. Uranium-plutonium carbide fuel was fabricated, along with the necessary amount of uranium carbide insulator material. In addition, uranium nitride insulator pellets were prepared for use in elements containing uranium-plutonium nitride fuel previously prepared by Battelle Columbus Laboratories. All fabrication activities were carried out in strict accord with RDD Quality Assurance requirements.

A carbothermic vacuum reduction procedure was used to synthesize single phase $(U, Pu)C$ and two-phase $(U, Pu)C + (U, Pu)_2C_3$ powders from mixtures of the respective oxides and carbon. The general fuel and insulator fabrication process is shown schematically in Fig. 548-1. In addition, a small quantity of single-phase uranium carbide was synthesized by arc-melting uranium metal with carbon, although that method was not found amenable to large-scale use. Carbide fuel and insulator pellets were prepared from the carbide powders by conventional cold compaction and sintering. Nondestructive examination and a series of characterization operations, including analyses for microstructure, chemical composition, physical defects, density, and dimensions, were required to verify that the pellets met stringent fuel and insulator specifications. Most carbide fuel and insulator

Fig. 548-1. Carbothermic Process for Uranium-Plutonium Fuel Pellet Production

types prepared for irradiation testing complied with the generalized technical requirements summarized in Table 548-IV. Several different fuel types, varying in dimensions, sesquicarbide content, and pellet density were fabricated. The ratios of oxide and carbon starting materials, and powder processing, pelletizing, and sintering parameters were adjusted to provide the desired fuel characteristics. Generally, the development of processing parameters for each fuel type was the responsibility of a "development" subgroup, although in-process parameter adjustment was occasionally required to alleviate problems.

Uranium-plutonium nitride pellets, fabricated by Battelle Columbus Laboratories, were assigned to LASL for loading into fuel elements. Nitride pellets sufficient for eight fuel elements were centerless-ground to the required diameter and certified by LASL for use in the K4 subassembly. Uranium nitride insulator pellets for these elements were fabricated at LASL. Depleted uranium metal nibblents were converted to metal powder by a hydrogenation-dehydrogenation process. The metal

TABLE 548-IV

**GENERAL TECHNICAL REQUIREMENTS
FOR CARBIDE LMFBR FUEL AND INSULATOR PELLETS**

<u>Characteristic</u>	<u>Requirement</u>	
	<u>Fuel</u>	<u>Insulator</u>
<u>Chemistry</u>		
U, wt%	75.5 ± 1	95 ± 1
Pu, wt%	19.0 ± 1	± 0.2
N, ppm	< 1000	< 1000
O, ppm	< 1000	< 1000
Be, ppm	< 10	< 10
B, ppm	< 10	< 10
Si, ppm	< 50	< 50
Fe, ppm	< 250	< 250
Cd, ppm	< 50	< 50
<u>Isotopic</u>		
^{235}U , wt% U	93.0 ± 0.5 wt%	not applicable
$^{239}\text{Pu} + ^{241}\text{Pu}$, wt% Pu	94.5 ± 0.5 wt%	not applicable
^{238}U , wt%	not applicable	> 99
<u>Phase Composition</u>		
Free U or Pu metal	none detectable at 500x	none detectable at 500x
<u>Physical Characteristics</u>		
Cracks	none at 2x	none at 1x
Diameter, cm ^a	± 0.0025	+ 0.0025 - 0.0125
Length, cm ^a	± 0.030	± 0.125

^aTolerance about nominal.

powder was then nitrided to form UN powder. Pelletizing and sintering operations were similar to those for the carbide process.

The fuel and insulator types required for each subassembly were fabricated in campaigns. The specific fuel fabrication campaigns are individually described as follows.

1. Fuel and Insulator Materials for the K4 Irradiation Test Subassembly

Fuel and insulator pellet types and quantities fabricated for the K4 subassembly are summarized in Table 548-V. Each fuel element required approximately fifty-four fuel pellets and three insulator pellets. In addition to pellets produced for the actual elements,

TABLE 548-V

**CARBIDE AND NITRIDE FUEL INSULATOR PELLET TYPES FABRICATED
FOR THE K4 IRRADIATION TEST SUBASSEMBLY**

<u>Characteristic</u>	<u>Carbide Type I</u>	<u>Carbide Type II</u>	<u>Nitride (BCL)</u>	<u>Carbide Insulator</u>	<u>Nitride Insulator</u>
<u>Phase Composition</u>					
M_2C_3 , vol. %	1-5	11-17	none ^a	none ^a	none ^a
<u>Ni Sintering Aid Used</u>	Yes	Yes	No	Yes	No
<u>Physical</u>					
<u>Theoretical Density (T. D.)</u>	> 98% T. D.	> 98% T. D.	13.5 g/cm ³	> 12 g/cm ³	> 12 g/cm ³
<u>Quantity</u>	4 elements	4 elements	8 elements	24 pellets	24 pellets

^a None detectable at 500x.

about 25% additional pellets for destructive testing and archival samples were required. Current fuel production capability is limited by the capacity of the sintering furnaces which defines a fuel pellet "batch" for quality assurance and sampling purposes. Presently, a fuel batch contains sufficient pellets to load one fuel element and provide the necessary 25% overage.

The characteristics of all the carbide fuel batches (two fuel types) prepared for the K4 subassembly are summarized in Table 548-VI, relative to the specification requirements. The principal problem encountered in 98% dense, two-phase carbide fuel preparation for the K4 subassembly was phase composition control. The problem was approached initially by establishing an empirical correlation between the ratio of the oxide and carbon starting materials and the resulting phase composition of the pellets as determined by Quantitative Television Microscopy (QTM). Pellets made per the empirical correlation suffered excessive batch-to-batch phase content variation. A reaction equation hypothetically describing the carbothermic formation of (U, Pu)-carbide from a mixture of stoichiometric oxides and carbon was developed as follows:

$$(1-a) UO_2 + a PuO_2 + (3 + c)C \rightarrow (1-2c) (U_{1-a}, Pu_a)_C + c(U_{1-a}, Pu_a)_2 C_3 + 2CO.$$

The results of QTM phase analysis of the empirically prepared fuel correlated well

**TABLE 548-VI
CHARACTERISTICS OF CARBIDE FUEL PRODUCED FOR
THE K4 SUBASSEMBLY**

THE K4 SUBASSEMBLY		Fuel Type ^a	
Characteristic	Specification	I	II
<u>Chemistry</u>			
U, wt%	75.5 ± 1.0	75.4	75.2
Pu, wt%	19.0 ± 1.0	19.23	19.02
C, wt%	---	4.87	5.12
N, ppm	< 1000	230	240
O, ppm	< 1000	380	220
Be, ppm	< 10	< 1	< 1
B, ppm	< 10	< 1	< 1
Cd, ppm	< 50	< 10	< 10
Fe, ppm	< 250	96	85
Si, ppm	< 50	25	20
<u>Microstructure</u>			
M ₂ C ₃ , vol%	(Type I)	3.8	---
	(Type II)	---	15.2
<u>Physical</u>			
Density	> 98% T. D.	> 98% T. D.	> 98% T. D.

^a The fuel types differ in vol% M_2C_3 per Table V.

TABLE 548-VII
FUEL AND INSULATOR TYPES AND QUANTITIES FOR
THE K6-A AND -B SUBASSEMBLIES

Characteristic	FUEL			
	Type I	Type II	Type IIIa	Type IIIb
<u>Phase Composition</u>				
M ₂ C ₃ vol%	(a)	10	10	10
<u>Physical Properties</u>				
Density, % T. D.	87	87	81	81
Diameter, cm	0.660	0.660	0.660	0.673
<u>Quantity</u>				
K6-A	10 elements	12 elements	---	---
K6-B	---	---	12 elements	10 elements

^a None detectable at 500x.

Phase Composition	INSULATORS	
	Type I	Type II
U ₂ C ₃ + UC ₂ , vol%	< .1%	0-10%
<u>Physical Properties</u>		
Density	> 12 g/cm ³	> 12 g/cm ³
Diameter, cm	0.660	0.660
<u>Quantity</u>		
K6-A	0	66 pellets
K6-B	30 pellets	36 pellets

with the predicted phase composition, provided the pellet nitrogen and oxygen atoms were assumed to substitute for carbon on a one-for-one basis (i. e., "equivalent carbon").

Two additional fuel batches were prepared assuming behavior according to the carbothermic reduction reaction. The target pellet phase composition for the batches was 14 vol% M₂C₃. They were determined to contain 12.2 vol% and 13.6 vol% M₂C₃ by chemical analysis and 13.2 vol% and 14.1 vol% by QTM. The same procedure was used to formulate subsequent K4 carbide fuel batches, significantly improving control of the phase composi-

tion. Sufficient carbide fuel was prepared for the eight carbide elements in the K4 subassembly. Single-phase UC insulator pellets for the K4 subassembly were fabricated by the arc-melting technique which was subsequently abandoned in favor of the carbothermic technique. No significant problems were encountered in insulator preparation.

2. Fuel and Insulator Materials for the K6-A and K6-B Irradiation Test Subassemblies

Fuel pellet fabrication for the K6-A and K6-B subassemblies is progressing. The four required fuel types and sizes and the two insulator types are described in Table 548-VII. Preparation of the 87% T. D., two-

TABLE 548-VIII
CHARACTERISTICS OF 87% DENSE,
TWO-PHASE FUEL FOR THE K6-A SUBASSEMBLY

<u>Characteristic</u>	<u>Specification</u>	<u>Measured Value</u>
<u>Chemistry</u>		
U, wt. %	75.5 ± 1.0	76.1
Pu, wt. %	19.0 ± 1.0	18.64
C, wt. %	-----	4.94
N, ppm	< 1000	156
C, ppm	< 1000	180
Be, ppm	< 10	< 1
B, ppm	< 10	< 1
Cd, ppm	< 50	< 10
Fe, ppm	< 250	12
Si, ppm	< 50	< 10
<u>Microstructure</u>		
M ₂ C ₃ , vol %	10 ± 5	6.2 to 8.5
<u>Physical Density, % T. D.</u>	87 $\begin{smallmatrix} +2 \\ -1 \end{smallmatrix}$	87.6

phase fuel was completed, and the 87% T. D. single-phase fuel is in process. Production of the 81% T. D. fuel has not yet been initiated.

To prepare the 87% T. D. fuel, the powder grinding cycle was reduced from that used in fabricating the 98% T. D. fuel for the K4 subassembly and the nickel sintering aid was deleted. The sintering temperature was substantially increased.

The characteristics of the 87% T. D., two-phase fuel produced are summarized in Table 548-VIII. Pellets sufficient to fabricate the required number of elements of that fuel type were prepared. However, production problems were encountered in phase composition and density control. Pellet phase composition was calculated from results of chemical analysis for total carbon, nitrogen, and oxygen content because QTM proved biased and imprecise at the 87% T. D. level. The ratio of the oxide and carbon starting materials was adjusted early in the processing history to try to place the mean sesquicarbide content near the middle of the tolerance allowed by the

specification (10 vol% ± 5%). This shifted the sesquicarbide content from 6.2 to 8.5%.

A problem in density control for the 87% T. D., two-phase fuel was also experienced. Unexplained batch-to-batch variations occurred resulting in a reject rate of about 25%. To attempt to reduce the reject rate, a pilot sintering procedure was implemented in which a fraction of each fuel batch was pelletized and sintered before the bulk of the batch. This procedure allowed process adjustment for batch-to-batch variations in behavior. Additional process development directed toward reducing batch-to-batch variability in densification behavior and obviating the need for pilot processing has been requested.

Ten 87% T. D., single-phase, carbide fuel elements were required for the K6-B subassembly. Production of that fuel type is progressing. Several technical problems have extended the effort. As with the two-phase, 87% T. D. fuel, the densification behavior proved variable. Pilot sintering was implemented, but has not mitigated

the problem. Several batches, which had acceptable pilot pellets, sintered to unacceptably high density. In addition, an air leak was found in one of the hydrogen-treatment units used to reduce the excess carbon content of the powder to essentially zero. The air leak caused rejection of several fuel batches because of nitrogen contamination.

The phase composition of the 87% T. D., single-phase fuel also proved difficult to control within the limit of the specification. The difficulty was compounded when infrequent platelets seen in the microstructure of sintered fuel were interpreted as evidence of sesquicarbide in excess of the specification. That discovery resulted in rejection of two batches of fuel and ultimately necessitated revision of the specification to permit acceptance of up to 2 vol% sesquicarbide. In spite of these problems, about 50% of the required amount of 87% T. D., single-phase fuel was completed.

The insulator pellets required for the K6-A and K6-B subassemblies were all specified to be >88% T. D. (12 g/cm^3). Single-phase insulators were designated for the elements containing single-phase fuel, and two-phase insulators were required where two-phase fuel was indicated. Because of the >88% T. D. density requirement as opposed to a tolerance band, density control was not a problem. Little difficulty was experienced in meeting phase composition requirements because the single-phase insulator pellets were fabricated after the allowed volume percent of sesquicarbide was broadened to 2 vol% from essentially zero. Some difficulty was experienced in plutonium contamination in excess of the specification (2000 ppm). Greater attention to glovebox cleanliness and limitation of some equipment to exclusive use for insulators alleviated that problem.

3. Special Preparations. Fuel materials, in special sizes and compositions, were supplied to other laboratories involved in the Advanced LMFBR Fuels Program for use in characterization capability development and certification. In addition, requested fuel types were provided to other LASL groups for special projects.

B. The Fuel Element Production Process

(D. G. Clifton, G. E. Meadows, J. R. Ruhe)

1. C-5/ON-1 Series Fuel Elements. The C-5/ON-1 elements were fabricated by Battelle Columbus Laboratories and Oak Ridge National Laboratory under an earlier program and assigned to LASL for storage. Upon reactivation of the Advanced LMFBR Fuels Program, these elements were designated for irradiation testing at EBR-II. Some reprocessing was required to enable the elements to conform to current specifications. Although the C-5/ON-1 series fuel elements had earlier passed one set of sodium-bond defect criteria, new criteria were proposed, necessitating rebonding of most of the elements. Sixteen elements (plus one spare) were selected for irradiation in EBR-II. The elements were successfully processed through the following operations:

- (a) Sodium rebonding and eddy current testing for bond defects.
- (b) X-radiography for verification of internal component location, fuel pellet integrity, and examination for fuel chips in the bond.
- (c) Autoradiography to assure that all pellets in the fuel stack contained fissile material.
- (d) Element bow measurement and straightening, if necessary.
- (e) Element length and weight measurement.
- (f) Element surface examination.
- (g) Element profilometry.
- (h) Wire wrapping and diametral inspection.
- (i) Cleaning and packaging into shipping tubes in the DOT-6M shipping container.
- (j) Shipment to EBR-II.

2. K4 Series Fuel Elements. Sixteen elements were required for this series. Each element had the following hardware components: bottom end plug, cladding tube, spacer, shroud tube, shroud tube plug, hold-down spring, and top end plug. Raw materials from which the components were manufactured were received from HEDL, and chemically overchecked, certified, and dimensionally and metallographically verified by LASL. Archive samples from each piece of raw material were taken. The archive samples were cleaned, weighed,

packaged, and stored. Sets of components for approximately twenty-five K4 fuel elements were fabricated in the LASL shops, inspected, cleaned, weighed, packaged, and stored. A set of 25 bottom end plugs and clad tubes were welded together. The welds were leak checked, visually and dimensionally inspected, machined to the diameter required, leak checked a second time, and x-radiographed. Upon passing those tests, the assemblies were used in elements. Fuel and insulator pellets needed for the K4 series elements were received from the fuel fabrication group. Simulated (stainless steel) fuel and insulator pellets for six demonstration fuel elements were also made. Six demonstration fuel elements were built using stainless steel simulated pellets and all the QA procedures for the actual element fabrication operations. Some procedural changes were made because of difficulties that were exposed by that exercise. Actual assembly of the K4 series elements followed. The elements were processed through the following assembly operations:

- (a) Sodium, fuel, and insulator pellets were loaded into the elements.
- (b) Hold-down springs were introduced, and the elements were engaged in the tag gas welder.
- (c) The elements were xenon tagged and the top end plugs were welded in place.
- (d) The welds were leak checked, inspected visually and dimensionally, machined to the required diameter, leak checked a second time, and x-radiographed.
- (e) The elements were processed through the sodium-bonding operations (repeating centrifugation and heat treating until eddy current tests indicated that bond integrity met the specification).
- (f) X-radiography verified the location of the internal components and the integrity of the fuel pellets.
- (g) Autoradiography was performed to assure that all pellets in the fuel stack were actually fuel.
- (h) Bow measurements were made and the ele-

ment straightened, where necessary.

- (i) The element lengths and weights were measured.
- (j) The element surfaces were examined.
- (k) The elements were profilometered.
- (l) The elements were wire wrapped and passed through a ring gauge to check diametrical requirements.
- (m) The elements were cleaned and stored in individual shipping tubes in a DOT-6M shipping container.
- (n) The elements were shipped to EBR-II.

3. K6 Series Fuel Elements. Bar stock and cladding were received from HEDL and end plugs and cladding weld specimens were fabricated for weld development. Twenty-five empty tag gas capsules and 120 xenon-filled gas capsules for the K6 series were also received. A tag gas capsule penetrator-actuator was designed, built, and tested on empty tag gas capsules fabricated at HEDL. A quality assurance qualification program will be written for use of this equipment. Additional hardware associated with the K6 series elements was fabricated: loading funnels to load elements that have only 0.127 mm diametral gap (pellet-to-cladding), archive sample capsules and lids, and loading funnels for smaller size fuel pellets. The hardware was cleaned, packaged, and stored.

4. Special Preparations. Several encapsulations of special fuel samples were prepared and sent to other laboratories involved in the Advanced Fuels Program.

- (a) Three shipments, comprising a total of seven capsules of carbide fuel pellets, were shipped to ANL.
- (b) Three capsules of carbide fuel pellets were shipped to BCL.
- (c) Three capsules of carbide fuel pellets were shipped to W-ARD.
- (d) One capsule of carbide fuel pellets was shipped to AI.

Two lengths of shroud tubing and two fabricated shroud tubes were sent to AI.

Fig. 548-2. Carbothermic Process Development

V. DEVELOPMENT

A. Fuel Process Development

(P. D. Shalek, R. L. Gutierrez)

Developmental experiments were run to establish fabrication parameters for five different fuel and insulator types: 81% and 87% T. D. $(\text{U, Pu})\text{C} + 10 \text{ vol}\% \text{M}_2\text{C}_3$ fuels, 87% T. D. single-phase $(\text{U, Pu})\text{C}$ fuel, >88% T. D. $\text{UC} + \text{U}_2\text{C}_3$ insulators, and >88% T. D. UC insulators. A general outline of the process used is given in Fig. 548-2. All development work was carried out in accordance with the quality assurance requirements of RDT F2-2.

An initial consideration was the proper carbon addition to the oxide powders to give the desired phase composition after carbothermic reduction. Initially, carbon additions were based on the carbothermic reduction of stoichiometric uranium and plutonium oxides as was done during early production of 98% dense fuel for the K4 subassembly (see Sec. IV). Subsequently, the carbon addition was empirically adjusted to compensate for the net effect of such modifying factors as volatiles and hyperstoichiometry in the oxides, volatiles in the

carbon, partial CO_2 formation during reduction, and incompleteness of reduction. As an example, while calculations based on stoichiometric reaction called for 11.96 wt% carbon addition to give 8.5 vol% M_2C_3 , an empirically established addition of 12.03 wt% actually was required to give this M_2C_3 content for both the 81% and 87% T. D. fuels. At the other extreme, calculations based on full reduction of the hyperstoichiometric oxides used ($\sim \text{MO}_{2.04}$) call for a 12.10 wt% C addition to get 8.5 vol% M_2C_3 .

For sinter phase fuel and insulator materials, the carbon additions were chosen to produce 2-4 vol% M_2C_3 in the carbothermic reduction product. This product was then reduced at 800°C for 0.3-0.45 h/g in flowing hydrogen at 0.023 m³/h to obtain the single-phase product.

The carbon addition for $\text{UC} + \text{U}_2\text{C}_3$ insulator material was established in the same empirical manner as for fuel. An addition of 11.90 wt% resulted in 2-5 vol% U_2C_3 in the sintered pellets. Assuming the simple, stoichiometric reaction, this addition should have given about 8 vol% U_2C_3 .

The other main area of concern was development of process parameters to give the lower 81% and 87% T. D. required for fuels and the >88% T. D. required for insulators. This work is summarized in Table 548-IX, where the parameters previously established for making two-phase 98% T. D. fuel are given on the top line for reference.

The initial changes made in the established process were the elimination of the nickel sintering aid with associated 16-hr ball milling, and the elevation of the sintering temperature to 1830°C to promote thermal stability at the lower densities. The only further changes made during low-density-fuel development were in the comminution and pressing parameters.

The first attempt at making 87% T. D. fuel is shown in Table 548-IX, as experiment DC-5. Sub-batch DC-5B, which retained the comminution parameters from the 98% T. D. process, gave densities in the required 86-89% T. D. range with an indicated low sensitivity to pelletizing pressure, although the presence of

TABLE 548-IX
SUMMARY OF CARBIDE FUEL ($U_{0.8}Pu_{0.1}Zr_{0.1}$) AND
INSULATOR ($UC_{1.5}$) FABRICATION DEVELOPMENT

Experiment No.	Nominal Pellet Type Attempted % T.D. %M ₂ C ₂	Grind Cycle (min.)	Sieve Size (mesh)	Prepress Ball Mill (hrs.)	Sintering Temp. (°C)	Sintering Time (hrs.)	Sintered % T.D. ^b for given pelletizing pressure					Remarks
							35 x 10 ⁶ Pa (2.5 ksi)	70 x 10 ⁶ Pa (5.0 ksi)	140 x 10 ⁶ Pa (10 ksi)	210 x 10 ⁶ Pa (15 ksi)	280 x 10 ⁶ Pa (25 ksi)	
FUEL												
Production Process	88	10	20	250	16(Ni)	1550	2			88.2		Established production process (0.2% Ni addition)
DC-5A	87	10	10	250	0	1830	8	82.6	82.8	84.4	85.3	Internal cracks
-5B	87	10	20	250	0	1830	8	86.6	86.8	87.7	87.7	End Caps at > 140 x 10 ⁶ Pa
DC-6 ^a	87	10	20	250	0	1830	8	86.3				No cracks or caps
DC-11	87	10	20	250	0	1830	8	87.8 ^b		90.2		Control for 81% process
DC-10 ^a	87	0	20	250	0	1830	8	88.3				H ₂ -reduced before pressing
DC-9	81	10	10	250	0	1830	8			85.6		Reduced grind time on oversize
	81	10	5	400	0	1830	8		85.7	87.3		
DC-11	81	10	5	170	0	1830	8	83.5		85.7		
	81	10	2.5	170	0	1830	8	80.6		83.8		
	81	10	2.5	35	0	1830	8	77.3		81.0		
	81	10	2	100	0	1805	8	78.6				
DC-13 ^a	81	10	2	100	0	1830	8	78.2		78.8 ^b		
	81	10	2	100	0	1855	8	78.9				
INSULATORS												
DC-7(B) ^a	>88	0	20	250	0	(1830 A+ (20000	8) A 2)			82.0 85.5		H ₂ -reduced before pressing
DC-7B	>88	5	20	250	0	(1830 A+ (20000	8) A 2)			81.2 87.8		
DC-8A ^a	>88	5	20	250	10	2000	3			82.1		
-8B	>88	5	20	250	16(Ni)	1550	3			88.1		0.2% Ni addition

^aNominal process transferred to production.

^bGeometric densities given for DC-11 and DC-13; rest are immersion densities.

internal flaws may have masked any such tendency. The density was significantly lower in DC-5A, where the grind cycle was reduced to 10 min. In experiment DC-6, a scale-up of DC-5B, a new die was used and sound pellets were obtained when pressed at 70×10^6 Pa (5.0 ksi). With only minor changes, the DC-6 process was used in production, where occasional refiring was required for density adjustment. In retrospect, it is felt that higher pelletizing pressures are needed to stabilize the process.

Nominally 87% T.D., single-phase fuel was successfully produced (experiment DC-10, Table 548-IX), using the process developed for the 87% T.D. two-phase fuel plus the hydrogen-reduction cycle described

earlier. The single-phase material appeared to be slightly more sinterable than the two-phase material (88.2% vs 86.3% T.D. final pellet density, all other parameters being constant).

The approach adopted for lowering the fuel density to 81% T.D. was to coarsen and narrow the particle-size distribution of the pressed fuel by varying the comminution parameters (grind cycle time, sieve size). Experiment DC-9, Table 548-IX, incorporated only a minor change from the DC-5A comminution cycle and did not lower the density. In experiment DC-11, the grind time was further reduced while the sieve opening size was increased. Satisfactory densities were obtained at both

70×10^6 Pa (5.0 tsi) and 210×10^6 Pa (15 tsi) pelletizing pressures, the higher being preferred. The coarser powders had much less tendency than the finer powder used for the 87% T. D. fuel to give internal flaws such as cap cracks when pelletized at the higher pressure. Because the coarse sieve used in the optimum process (2-1/2 min. grind time, -35 mesh) allowed relatively large, dense particulates to occur throughout the microstructure, compromise comminution parameters (2 min. grind time, -100 mesh sieve) were used in DC-13, the process accepted for production of 81% T. D. fuel.

Second-phase sesquicarbide occurred in both the 81% and 87% T. D. pellet microstructures as irregularly sized and shaped areas, in sharp contrast to the more uniform, equiaxed M_2C_3 particles found in the 98% T. D. fuel. The M_2C_3 content of the 98% T. D. fuel, as determined by quantitative television microscopy (QTM), agreed very well with that predicted by the chemical carbon equivalent content (oxygen + nitrogen + carbon), while the QTM values for M_2C_3 in the lower density fuels were significantly lower than the chemical values.

The 87% T. D. control material, run in DC-11 using exactly the DC-6 processing parameters, gave significantly higher immersion density than in the earlier experiment (89.4% vs 86.3% T. D. at 70×10^6 Pa (5 tsi) pressing pressure). The difference is believed due to a change in UO_2 feed stock, and indicates that the carbothermic reduction step cannot be relied upon to neutralize such changes.

UC insulator material produced by the carbothermic process was found to be considerably less sinterable than carbide fuel. When fabricated in experiment DC-7B using the 87% T. D. fuel process, a density of only 81.2% T. D. was obtained as compared to 87.7% T. D. for the fuel. Because of the greater length-to-diameter ratio in the insulator pellet, there was a tendency for cap cracking to occur. To minimize this tendency, the sintering temperature was raised to 2000°C , rather than alter the comminution or pressing parameters to achieve densities of $\geq 88\%$ T. D. As with the fuel, the single-phase insulator material was more sinterable than the two-phase material.

In experiment DC-8, two other methods were used to increase insulator pellet density. Ball-milling for 16 hours before pressing enabled higher density to be achieved at lower pelletizing pressure, and the DC-8A process was adopted for production of two-phase insulators. Comparing with DC-7B, it appears that the ball-milling step increased insulator pellet density about 5% T. D. Using nickel sintering aid in DC-8C enabled very high insulator pellet densities to be attained at 1550°C .

Data collected during development work indicate that the immersion-density technique may require some modification when applied to low-density pellets. Immersion densities ran 0.5 to 1% higher than geometric densities for 87-88% T. D. pellets. The difference increased to about 1.5% for the 81% T. D. fuel. The problem is believed to be variable absorption of the coating media by pellet surface porosity. The best value is felt to lie between the extremes and an attempt is being made to relate the two measurements.

An experiment was performed to determine if carbothermically reduced powders could be stored without affecting the pressing characteristics of the powder. It was found that after tumbling the powder for five minutes each day before pressing, the product pellets were dimensionally consistent. It is suspected that inherent glovebox vibration may cause particle-size segregation during even short-term storage.

B. Element Fabrication Development

(D.G. Clifton, G.E. Meadows, J.R. Ruhe)

A significant portion of the element fabrication effort was devoted to designing, installing, and testing equipment used in the fabrication operations and non-destructive testing of EBR-II experimental fuel elements.

1. Xenon Tagging for the K4 Series Elements. The xenon tag for the K4 elements consisted of a minimum of one ml STP of a prescribed isotopic composition of gas freely mixed with the ~ 1 atm of helium present in the plenum in each element. The tag had to be added to the element and kept in place while the top end plug was seated and welded. A xenon tagging apparatus was developed and tested. The principle used was to evacuate the loaded ele-

ment, measure the available plenum volume by expansion of a known quantity of helium into the plenum, measure the pressure, and calculate the plenum volume. The necessary partial pressure of xenon tag gas, p_{xe} , to ensure a minimum of one STP ml of tag in the plenum was calculated. The element was re-evacuated and the xenon tag gas introduced to p_{xe} . Helium was then introduced to ambient, the top end plug was inserted, and the closure weld made.

The gas apparatus was designed to fit the welding fixture in a manner making a gas-tight seal to the element and having the capability of inserting the end plug without breaking the gas-tight seal. The system was tested by tagging seven prototype plenum volumes with natural xenon and then analyzing samples for the correct volumetric content of xenon. All samples tested met the EBR-II requirement of ≥ 1 ml of tag at STP, and EBR-II staff approval of the tagging procedures was obtained. The procedure was used successfully to fill all K4 series elements plus sample tag gas capsules that were required by EBR-II.

2. Welding Development for the K4 Series Elements. A GTA welding fixture, joined to the xenon tagging device and mounted in a helium-atmosphere glovebox, was fabricated. The fixture incorporated a horizontally rotating fuel element with the GTA welding torch held mechanically on the weld joint. Welding torch position was adjustable by x, y, and z micrometers, and could be fixed at any angle. Welding power was supplied by a fully programmable 50-ampere source. Current and voltage profiles were recorded on a strip chart recorder. During development of the welding apparatus, trial welds were made on K4 series end plugs (a simple butt joint to the cladding) to establish welding parameters. The parameters were used to make a set of qualification welds that qualified the equipment and two operators.

3. Wire Wrap Machine. A machine to make a helical wrap of wire with a 15.24-cm pitch around a 101.6-cm-long, 0.7874-cm-diameter fuel element was designed, built, and installed. Test wraps were successfully made on dummy K4 fuel element geometries. Attendant to wire wrap development was a welding qualification program for

welds fixing the wire to the end plugs. A manual weld procedure was established and two operators were qualified.

4. Sodium and Sodium-Bonding. A sodium vacuum distillation apparatus powered by an induction heater was designed, built, and installed. High-purity sodium produced by the equipment was used in developing an oxygen-in-sodium analysis capability. This work was performed in conjunction with the analytical chemistry group.

The sodium preparation apparatus was put into operation and 24 identical, high-purity, extruded sodium samples were submitted for analysis at HEDL and LASL. The samples verified that sodium quality conformed to program specifications for the K4 series elements.

The sodium bonding equipment (centrifuge, bonding furnace, and eddy current testing equipment) was qualified for performing bonding operations for the K4 series elements. Development work to establish proper bonding procedures was conducted on a C-5 nitride element that was rejected for use because of chemical contamination. Development involved establishing the proper temperature profiles in the furnace and centrifuge to provide directional melting and solidification of the sodium, avoiding formation of bubbles in the bond.

5. Shipping. DOT shipping container requirements were changed, requiring that three DOT-6M shipping containers with inserts to hold 12 fuel-element arrays be constructed and mounted on shipping pallets. Approximately 40 aluminum shipping tubes to contain individual elements were prepared.

6. Welding Development for the K6 Series Elements. In preparation for weld development for the K6 series, mock-up end plugs were made with various sizes of notches and grooves cut into the section of the plug that lies immediately below the plug-to-cladding weld joint. At the time, it was understood that an absolute indicator of 100% weld penetration was required to meet EBR-II radiographic standards. The resolution of LASL radiography was inadequate to measure 0.0508 mm to 0.0762 mm of unpenetrated joint (a reject condition at EBR-II). However, by providing a radiographically visible notch immediately below the weld joint, 100% penetration could be

assured when the notch was obliterated by the weld. Considerable developmental effort was expended in attempting to identify a process to weld a joint with an underlying notch. With a vee-notch, a major problem arose in gas-bubble entrapment in the weld. Changing from a vee-notch to a recess about 0.0762 mm deep by approximately 2.54 mm long below the weld joint appeared to solve the bubble entrapment problem but caused excessive weld subsidence as molten metal flowed into the recess. It is probable that some weldable shape of radiographic indicator exists; however, conference with EBR-II staff on the problem provided a more direct solution. EBR-II staff agreed to accept the successful result of destructive analysis of a defined number of welds. Radiography was thus limited to the function of gross flaw detection, and weld penetration was qualified metallographically. The weld joint geometry was accordingly revised to the simple butt joint used on the K4 Series, which gave minimum difficulty in welding. Welding of the actual K6 hardware will take place in FY 1976.

7. Xenon Tagging for the K6 Series Elements.

The xenon tag method for the K6 Series elements differs from the K4 series in that a sealed metal capsule of the prescribed tag gas is inserted into each element and the element is welded closed. The tag capsule contains a metal penetrator which is driven through the capsule by a magnetic field imposed by a solenoid, and the tag gas is released into the element plenum. A solenoid apparatus to pierce the tag gas capsule was fabricated and tested. Successful piercing of the capsules was confirmed by radiography.

C. TREAT Assembly Development

(H. E. Martinez)

In the designation of near-term responsibilities for the TREAT program, LASL serves primarily as a fabricator for test assemblies and ANL determines the types of experiments to be conducted. Through meetings and discussions between representatives of the various laboratories involved in the Advanced Fuels Program, a near-term test matrix was proposed and submitted to ERDA/RDD. Four tests, designated HC-1, HC-4, HC-5 and SC-1, were selected for fabrication and irradiation

in FY 1976. In addition, a TREAT mock-up incorporating the features of an actual assembly, except for nuclear fuel, will be fabricated for permanent use at EBR-II.

The HC-1 test will be the first test of the series for off-normal testing of advanced fuel. The test is intended to serve as the calibration test for subsequent carbide fuel element tests. The HC-1 fuel element will contain helium bonded carbide, and will be instrumented with a centerline thermocouple at the midplane of the fuel element.

The HC-4 and HC-5 tests will consist of two helium-bonded carbide fuel elements with prior irradiation history in EBR-II. A pressure transducer will be incorporated externally to the fuel elements in an attempt to determine the instant of fuel element rupture.

The SC-1 test will consist of a sodium-bonded carbide fuel element. A pressure transducer will be incorporated to continuously measure fuel element pressures during the test.

The initial TREAT development efforts at LASL consisted of indoctrination of personnel, familiarization with TREAT requirements in general, as well as tests previously conducted by LASL, and development and implementation of the necessary quality assurance system for fabrication of the planned tests.

Fabrication design and drafting work was largely completed on the mock-up and HC-1 assemblies. Partial fabrication design was also accomplished for the HC-4, HC-5, and SC-1 tests. Much of the required instrumentation was designed and procured. Fabrication support equipment such as welding and brazing fixtures, NaK bonding apparatus, etc., was obtained. The extensive quality assurance plan for much of the overall test matrix was developed, along with many of the essential operational and quality verification documents. Actual fabrication of test assemblies will be initiated early in FY 1976. It appears that many on-hand materials and components from earlier TREAT tests can be applied to the new test matrix.

VI. PROGRAM SUPPORT

A. Analytical Chemistry

(G. R. Waterbury)

1. Determination of Carbon in (U, Pu)C. The precision in the determination of total carbon in (U, Pu)C fuels by combusting the sample in O_2 and weighing the evolved CO_2 was improved by increasing the sizes of the sample and CO_2 collector, by reducing the rate of the combustion, and by inserting quartz wool plugs in the combustion apparatus to hold all of the sample in the combustion zone. The samples were previously burned rapidly in an oxygen atmosphere. Testing showed that under certain conditions, cinders of uranium-plutonium oxide could form around portions of the (U, Pu)C, preventing oxidation of some of the carbide. Also, during this rapid burning, sparking of the sample resulted in loss of unburned (U, Pu)C out of the hot zone of the furnace. These two conditions have been eliminated by burning the sample in an atmosphere of four parts argon and one part oxygen at $1000^\circ C$. Small particles emitted from the sample are trapped by the quartz wool plugs in the combustion furnace and quantitatively burned to (U, Pu) O_2 . By increasing the sample size to one gram and making the other described modifications, the relative standard deviation was improved from 0.8% to 0.3%. On a mixed carbide containing 5 wt% carbon, the absolute precision was $\pm 0.015\%$.

2. The Determination of Manganese and Molybdenum in Stainless Steel by Atomic Absorption. A capability was developed and evaluated to determine molybdenum and manganese, each at about 2 wt% in stainless steel fuel element materials by atomic absorption. Manganese and molybdenum are determined in the same solution, and separations are not necessary. The analysis of six samples of NBS SRM 160, which is certified for both chemical elements, showed no bias in determining either element. The relative standard deviations were 0.8% for Mo and 0.5% for Mn. A quality assurance procedure for this method was written and approved.

3. Determination of P, S, Cl, F, and C in (U, Pu)C Fuels. Four RDT Standard Methods for analyzing mixed oxide fuels were tested for measurements of P, S, Cl, and F in carbide fuels. This was done by adding known quan-

ties of these four elements to weighed portions of the (U, Pu)C fuel and then determining the elements using the existing methods without modification. The bias in measuring each element was less than 5 relative percent of the amount added, indicating that the carbide fuels did not interfere with these determinations. The lower limits of detection were 1 ppm for F and 10 ppm for Cl in a 1-g sample and 5 ppm for P and S in 100-mg and 400-mg samples, respectively.

4. Determination of Oxygen in Sodium Used for Bonding. A cursory evaluation was made of a new technique for the determination of oxygen in sodium in which the sodium is dissolved in anhydrous methanol and the water formed from the contained oxygen is determined by a modified coulometric Karl Fischer titration. A reported standard deviation of 0.015 μg of water in the range 0.05 to 20 μg of water indicates that 1 ppm of oxygen could be detected in as little as 100 mg of sodium. Experimental verification of the applicability of this technique was started.

A vacuum distillation flame photometric method was set up to determine oxygen in sodium. In this method, the sodium in the vacuum distillation residue is measured, and the oxygen content is calculated assuming that all of the sodium is present as Na_2O . The distillation was done at the sodium loading facility. The still operated without apparent difficulties.

The equipment for the flame photometric measurement of sodium in the distillation residues also was set and tested satisfactorily. Sodium reference solutions were prepared, a calibration curve was determined, and repeated analyses on solutions containing known amounts of sodium showed that the standard deviation of the flame photometric measurements was 5% or less. Quality assurance methods were written and approved for the distillation of the sodium in the distillation residues.

Initial tests of the entire system in analyzing loop sodium for oxygen revealed contamination of the sample prior to the distillation. A set of test samples was sent to HEDL for oxygen determination by the amalgamation method and a duplicate set was distilled and analyzed at LASL. The LASL results were somewhat higher than the

the results reported by HEDL. Modifications were made to the inert atmosphere enclosure in which samples were taken and distilled. Results for oxygen were then much lower. Another set of samples will be analyzed at LASL and at HEDL and the results compared.

5. Evaluation of Other Methods for Analyzing Sodium. In anticipation of the analysis of sodium for various trace impurities, either the methods from RDT Standard F3-40T or other methods developed at LASL will be set up and evaluated. The methods now considered operative measure carbon, cadmium, zinc, and the impurities normally measured spectrographically.

6. Vendor Liaison. As part of the LASL fabricator certification team, visits were made to Atomics International, Battelle Columbus Laboratories, and Westinghouse Advanced Reactor Division to assist in implementation of RDD Quality Assurance Programs. Analyses were made on one set of stainless steel samples from BCI, and two sets from AI.

B. Microstructural Analysis

(K. A. Johnson)

High-density (>98% T. D.), two-phase (MC and M_2C_3) fuels with Ni sintering aid to be used in experimental fuel elements were given specified limits on the volume of second-phase (M_2C_3). An investigation was made to characterize both hand and automatic methods of measuring the second-phase content on prepared metallographic cross sections. Both methods were evaluated from the viewpoint of effort required and resulting precision. The study did confirm that the automatic method was faster and more precise than the hand method. The study was extended to develop a technique of setting the statistically significant number of photomicrographs needed to characterize a batch of pellets. In addition, an extensive cross correlation was made with analytical chemistry carbon analyses which showed that analytical chemistry could also be used to measure the amount of second-phase.

Eight persons performed the hand count of volume of M_2C_3 using the Systematic Point Count (SPC) technique. This technique is generally accepted as the best hand technique. Nine photomicrographs from each of three pellets were counted. Results of the eight operators showed a

mean ($\bar{x}$) = 5.152 vol%, standard deviation of each measurement (s_x) = 0.978 vol%, and the standard deviation of the mean ($s_{\bar{x}}$) = 0.3457 vol%. The figures from two automatic measurement sets on the same photomicrographs were $\bar{x}$ = 4.067 vol%, s_x = .0424 vol% and $s_{\bar{x}}$ = .03 vol%. Not only were the automatic technique results much more precise, and presumably more accurate but the analysis time was one-fifth of the SPC analysis time.

A similar set of comparison analyses was made on a nominal 20 vol% M_2C_3 group of pellets. The difference in precision was about the same, but at this vol% level, the automatic analysis time was less than one-tenth of the SPC analysis time. The most important conclusion of the analyses was that the automatic technique was much more precise and faster than the hand count technique.

To determine the number of photomicrographs required to obtain a M_2C_3 content determination within a designated degree of certainty, multiple replicate sets of photomicrographs were taken on the same specimens. It was found, using analysis of variance calculation, that pellet inhomogeneity was a relatively local phenomenon at 500X and that areas for analyses could not be selected on the basis of the operator judging them to be typical. Areas were selected based on precalculated stage-driven micrometer readings.

Several theoretical formulations of the sampling standard deviation^{*} with respect to quantitative microscopy were proposed. A LASL modification to that of Gladman and Woodhead¹ seemed to apply best to present analyses. The equation is as follows: $\sigma_{est.} = f \left(\frac{1-f}{n} \right)^{\frac{1}{2}}$,

where f is the measured volume fraction, n is the number of features detected, and σ is the estimated standard deviation. This requires that the number of features be counted as well as the feature area fraction.

Given a desired confidence level and a preliminary estimate of the number of features per photomicrograph, the number of photomicrographs required from a pellet

* The standard deviation which would be expected using replicate analyses on the same pellet if the material were uniformly distributed.

can then be easily determined. The number of pellets which should be used to characterize a batch can be judged best through the use of preliminary data and the calculation of a standard deviation of the theoretical sample mean, $\bar{x}$ corrected for sampling from a limited population², i. e., $\sigma_x = \frac{\sigma_{\text{measured}}}{n^{\frac{1}{2}}} \left(1 - \frac{n}{N}\right)^{\frac{1}{2}}$, where

$n^{\frac{1}{2}}$ is the number of pellets and N is the size of the batch. It was clear that a significant decrease (ca. a factor of two) in σ_x would require essentially n^2 samples. Three pellets in a batch size of 80 to 180 were found to be statistically adequate. Less than three pellets would currently be unacceptable in terms of the possibility of unknown variables. With a more extended fabrication history, this may be cut to two pellets.

VII. QUALITY ASSURANCE

A. Quality Assurance

(L. E. Lanham, R. C. Minor)

The staffing of the quality assurance organization (QA) supporting the Advanced Program reached its planned objective. All personnel have been trained and are performing their assigned duties to provide an independent verification of quality achievement per RDD program requirements. Two people perform the inspection, checking, and surveillance activities for the Advanced Fuels Program. One is assigned to each of the fuel and element fabrication areas. Additional quality assurance technicians are temporarily assigned to various areas when required. A quality assurance engineer has been assigned the responsibility for the quality assurance activities in the Advanced Fuels Program. In addition to the QA Manager, a second person has been assigned to the program for certification of QA aspects of the other fabricators involved in fuel and element fabrication. He has assisted in the review of documents and is being trained to perform fabricator evaluations as required.

Internal audits are being conducted by a three-member team. One team member has been assigned the responsibility of preparing the audit plan and selecting or preparing audit check lists. The second member is the quality assurance person responsible for the area being

audited and the third is from the quality assurance organization but has no direct responsibility for the work being audited. In the future, variations of the audit team may be used, for example, technically qualified people outside the quality assurance organization may be used to meet difficult audit scheduling requirements to place proper emphasis on a specific area or problem.

An internal audit was conducted prior to final preparations for K4 series element production. After corrective actions were taken on all findings, six simulated elements were fabricated. This fabrication campaign was conducted to complete the training and certification of operators, and to make a final check of procedures. During this fabrication campaign, a second audit was conducted. An audit was also conducted of the wire wrap and bonding operations in the K4 series of element production. An audit report was prepared.

The Quality Assurance Specialist assigned to the computer-programmed instrument-calibration-recall system attended a class for KRONOS magnetic tape usage. In addition to the recalibration recall system, a current printout of calibrated instruments will be used as a checklist for audits. Future plans are to use a computer program for quality assurance control of the current revision and status of all documents.

Discussions were held at LASL with the Quality Assurance Manager of each of the three additional fabricators to be certified for fuel and element fabrication in the Advanced Fuels Program. In each case, discussions involved the quality assurance requirements for certification reviews as described in LASL guidance documents and RDT F2-2. In-plant visits were made by the LASL QA Manager (and other LASL specialists), to *Atomics International*, *Battelle Columbus Laboratories*, and *Westinghouse Advanced Reactor Division* to evaluate their QA systems relative to future certification. The quality assurance assessments were documented in *Recommendations*, and *Agreements and Commitments*. Checklists based on the RDD program requirements were developed at LASL and used in evaluating the quality assurance programs of other fabricators. The checklist will be updated and completed as the evaluation proceeds through the various phases.

B. Fuel Production Quality Assurance

(R. J. Herbst, R. L. Gutierrez)

The quality assurance system associated with fuel and insulator pellet development and production was extensively modified. Approximately 100 documents were prepared by the fuel production and development group. Key elements of the system are the Inspection Plan, Material Specification, Preparation Procedure, Data Form, Certification/Verification Form, Data Record, and Data Package. The Inspection Plan identifies the operations and documents to be exercised and their sequence, Material Specifications and Preparation Procedures to be referenced, and the Data Records for documenting compliance with procedures. The Material Specifications describe the minimum technical requirements for starting materials. Compliance with the specifications is recorded on Certification/Verification Form. Preparation Procedures describe the operations to be performed at each stage of preparation. Data Forms include instructions unique to each particular fuel type.

Data Records are used to document compliance with the process requirements of the Data Forms and Preparation Procedures. A comprehensive Data Package is assembled for each fuel or insulator batch. The Data Package includes all the information recorded in the course of preparing and characterizing a fuel or insulator batch and is traceable to the element into which the pellets are loaded. Final acceptability of a fuel or insulator batch is determined by comprehensive review of the Data Package. The fuel and insulator Data Package, together with the element fabrication Data Package, is ultimately supplied to the experimenter when the completed subassembly is shipped from LASL. A copy is

also retained by LASL. Data Packages are also compiled for special fuel preparations. Standardization of the various documents and the Data Package format has improved the maintenance of process records.

A QA audit was conducted on the fuel production facilities and operations used for production of the K6 series. An audit report was prepared and approved. Corrective action was taken on all findings of the audit.

C. Fuel Element Production Quality Assurance

(D. G. Clifton)

A total of 45 QA documents were prepared or extensively revised in a continuing effort to improve the QA system associated with fuel element fabrication.

D. Analytical Chemistry Quality Assurance

(G. R. Waterbury)

The analytical chemistry quality assurance system was improved by writing computer programs to aid in keeping records for the following items: (1) Calibration of equipment, (2) Preventive maintenance of equipment, (3) Current index of all analytical chemistry QA procedures, and (4) QA procedures for which each analyst is certified. The computer notifies the QA Representative on a monthly basis of actions needed on the four listed items to keep the system updated and current. As the required actions are completed, these are entered into the computer for storage and later recall.

VIII. REFERENCES

1. T. Gladman and J. H. Woodhead, J. Iron Steel Inst. (London) 194, 189 (1960).
2. C. A. Bennett and N. L. Franklin, Statistical Analysis in Chemistry and the Chemical Industry, (John Wiley and Sons, Inc., New York, 1954), p. 60.

PROJECT 463 (L002)

HIGH-PERFORMANCE LMFBR FUEL MATERIALS

Person in Charge: R. D. Baker
Principal Investigators: J. L. Green and J. O. Barner

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 nitride and carbide fuel elements under conditions that appropriately exploit the potential of these materials to operate to high burnup at moderate-to-high linear powers. 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 carbide and nitride fuel elements. These experiments are designed to investigate fuel swelling, interactions between the fuel and cladding 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, off-normal experiments are being considered which would allow the study of the effects of rapid, overpower, reactor transients and loss of sodium bond on carbide and nitride fuel assemblies.

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.

II. STEADY-STATE (EBR-II) IRRADIATION TESTING
(J. O. Barner, N. S. DeMuth, G. R. Harry,
D. E. Hill, J. F. Kerrisk, T. W. Latimer,
B. D. McLaughlin, R. L. Petty and
P. E. Rexroth)

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.

A. Experiment Description and Status

Sixteen series of experiments have been originated. The description and status of these series are summarized in Tables 463-I to 463-X. In order to define the status of those experiments which are undergoing post-irradiation 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 Decapsulation

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-I describes the Series K1, K2, and K3 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.

The hot cell examinations were completed on two experiments in these series, unfailed element K-44 and failed element K-51. Another unfailed experiment, K-39B, has been reconstituted into EBR-II subassembly X233 for additional irradiation. The four remaining elements in these series which have not completed their postirradiation examinations were placed in long-term storage pending the completion of the examination of higher priority experiments.

Table 463-II describes the Series U1300 experiments. These experiments contain two-phase, uranium-plutonium carbide fuel pellets which are helium bonded to either Type 316 stainless steel or Incoloy 800 cladding. Two methods for the accommodation of fuel swelling were investigated in this series, viz., 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 (52-70 kW/m). Four of these experiments (all unfailed) have completed postirradiation examination, and report preparation is in progress as indicated in Table 463-II.

Indications of element cladding failure for three experiments (U98, U106, U114) were found at HFEF using γ -scanning for ^{137}Cs in the capsule sodium; these results were confirmed at LASL. The γ -scanning technique at HFEF did not reveal any ^{133}Xe in the capsule cover gas. No fission gases were found at LASL in the capsule plenum regions of experiments U98 and U106. No obvious failure points in the cladding of elements U98 and U106 were found, either visually or by helium pressurization tests. This discrepancy has not been satisfactorily resolved. It is anticipated that the remaining three elements in this series will be destructively examined during FY 1976.

The Series U1950 experiments are described in Table 463-III. 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. This series of experiments completed irradiation in

TABLE 463-I

SERIES K1, K2, AND K3 ENCAPSULATED-CARBIDE EXPERIMENTS

Exptl. 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 ^h	Max Centerline Temp., °C ^h	Goal Burnup at, % ^d	Max. Current Burnup at, % ^d	Status
Series K1										
K-36B	MC	90	Na-0.38	SA-316SS	7.62 x 7.11	82 [□]	990 [□]	6	5.85*	b.12 ^e
K-37B	MC	90	Na-0.38	SA-316SS	7.62 x 7.11	85	1000	6	2.9	a.7 ^{e, f, i}
K-38B	MC	90	Na-0.38	SA-316SS	7.62 x 7.11	85	1000	6	5.8	a.7 ^{e, f, i}
K-39B	MC	90	Na-0.38	SA-316SS	7.62 x 7.11	85	1000	10	5.8	EBR-II, X233
K-42B	MC	90	Na-0.38	SA-316SS	7.62 x 7.11	85 [□]	1000 [□]	6	4.46*	Completed ^g
Series K2										
K-49	MC	95	Na-0.51	SA-316SS	7.62 x 7.11	130 [□]	1270 [□]	5	3.74*	b.12 ^e
K-50	MC	95	Na-0.51	SA-316SS	7.62 x 7.11	130	1300	6.5	3.6	a.9 ^{e, i}
K-51	MC	95	Na-0.51	SA-316SS	7.62 x 7.11	130	1300	8	3.5	b.12 ^e
Series K3										
K-43	MC	94	Na-0.51	SA-316SS	7.62 x 7.11	80	1000	8	5.6	b.3 ^{e, i}
K-44	MC	94	Na-0.51	SA-316SS	7.62 x 7.11	80	1000	8	8.44*	b.12
K-45	MC	94	Na-0.51	SA-316SS	7.62 x 7.11	80	1000	5	2.37*	b.12 ^e
K-46	MC	94	Na-0.51	SA-316SS	7.62 x 7.11	80 [□]	965 [□]	5	2.39*	b.12 ^e

^aM = (U_{0.8} Pu_{0.2}).

Series 1 and 3 experiments are 93% enriched in ²³⁵U.

Series 2 experiments are 97% enriched in ²³³U.

^bTheoretical density of MC = 13.45 Mg/m³.

^cSA = Solution annealed.

^dBurnup values marked with * were measured using the ¹⁴⁸Nd method. Remaining values were computed from EBR-II data. The value shown for K-39B is that prior to irradiation in X233.

^eElement cladding failure indicated.

^fDamaged during reconstitution of X152.

^gReported in LA-4669-MS.

^hLinear 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.

ⁱStorage.

TABLE 463-II

SERIES U1300 ENCAPSULATED-CARBIDE EXPERIMENTS

Expt. No.	Fuel Type ^a	Fuel Density % Theo. ^b	Bond and Diametral Gap, in.	Clad ^d Type	Clad O. D. x I. D., in.	Max Linear Power, kW/m ^g	Max Centerline Temp., °C	Goal Burnup, at. %	Current Peak Burnup, at. % ^e	Status
U93	MC+5% M ₂ C ₃	86	He-0.10	SA-316SS	7.66x6.10	59 [□]	1750	11	9.64*	b.13
U94	MC	85	He-0.18	SA-316SS	7.73x6.86	79 [□]	1680	11	9.42*	b.13
U97	MC+5% M ₂ C ₃	86	He-0.10	SA-INC-800	7.60x6.10	54	1750	11	9.60*	b.13
U98	MC+5% M ₂ C ₃	86	He-0.18	SA-INC-800	7.60x6.86	65	1680	11	9.33*	b.8 ^f
U105	MC+5% M ₂ C ₃	77	He-0.20	SA-INC-800	7.61x6.10	52 [□]	1900	11	9.89*	b.13
U106	MC+5% M ₂ C ₃	77	He-0.20	SA-INC-800	7.72x6.86	59	1820	11	9.53*	b.8 ^f
U110	MC+10% M ₂ C ₃	99 ^c	He-0.33	SA-INC-800	7.73x6.86	65	1960	10	9.2	a.7
U113	MC+10% M ₂ C ₃	98 ^c	He-0.28	SA-INC-800	7.61x6.10	50	1880	11	10.2	a.7
U114	MC+15% M ₂ C ₃	98 ^c	He-0.18	SA-INC-800	7.72x6.86	66	1570	10	9.5	a.7 ^f

^aM = (U_{0.85} Pu_{0.15}).

^bTheoretical density of MC = 13.45 Mg/m³.
Theoretical density of M₂C₃ = 12.72 Mg/m³.

^cCored pellet with nominal 2.0-mm-diameter axial hole.

^dSA = Solution annealed.

^eBurnup values marked with * were measured using the ¹⁴⁸Nd method.
Remaining values were computed from EBR-II data.

^fElement cladding failure indicated by γ-scan.

^gLinear power marked with □ are beginning-of-life values computed using measured burnup results.
Remaining values based on EBR-II power adjustment factor of 0.91.

December 1974 in EBR-II subassembly X055 B after operation at relatively low linear power (38 to 44 kW/m).

The γ-scanning technique at EBR-II detected the presence of ¹³³Xe and/or ¹³⁷Cs in the capsule plenum or sodium regions of the failed elements. This evidence of element cladding failure was confirmed by γ-scanning measurements at LASL. Those nine elements with indicated cladding failures are identified in the last column of Table 463-III.

All ten unfailed elements and two failed elements, U131 and U132, have been decapsulated and are undergoing destructive examination. Elements U133 and U134 are being held for incorporation into a subsequent TREAT test. More detailed discussions of the elements

in this series are found in Section II-B.

The Series U1930 and U1960 experiments are described in Table 463-IV. 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-IV showed that fuel elements U194 and U200 had failed. Destructive examination of this group of experiments has been completed. Data reduction, interpretation, and report preparation are continuing.

The experiments listed in part B of Table 463-IV completed irradiation in subassembly X182. These

TABLE 463-III

SERIES U1950 ENCAPSULATED-CARBIDE EXPERIMENTS

Expm't. No.	Fuel Type ^b	Fuel Density % Theo. ^c	Bond and Diametral Gap, mm	Clad ^e Type	Clad O. D. x I. D., mm	Max Linear Power, kW/m ^g	Max Centerline Temp., °C	Goal Burnup, at. %	Max Current Burnup at. % ^g	Status
U129	MC	86	He-0.41	SA-316SS	7.70x6.60	38	1750	11	11.7	a, 8 ^f
U130	MC	77	He-0.25	SA-316SS	7.70x6.60	39	1500	11	11.8	b, 8
U131	MC	85	He-0.36	SA-316SS	7.70x6.60	38	1490	11	11.5	b, 7 ^f
U132	MC	85	He-0.36	SA-316SS	7.70x6.60	38	1490	11	11.4	b, 7 ^f
U133	MC	85	He-0.36	SA-316SS	7.70x6.60	38	1490	11	11.1	b, 4 ^h
U134	MC	85	He-0.36	SA-316SS	7.70x6.60	38	1490	11	11.2	b, 4 ^h
U135	MC	85	He-0.36	SA-INC-800	7.67x6.60	38	1470	11	11.5	a, 8 ^f
U136	MC	85	He-0.36	SA INC-800	7.67x6.60	40	1470	11	11.1	a, 9 ^f
U137	MC+20% M ₂ C ₃	97	He-0.36	SA-316SS	7.70x6.60	40	1440	10	10.0	a, 8 ^f
U138A ^a	MC+20% M ₂ C ₃	98	He-0.36	SA-316SS	7.44x6.60	44	1440	8	7.1	b, 8
U139	MC+20% M ₂ C ₃	97	He-0.36	SA-INC-800	7.72x6.60	44	1440	10	10.2	a, 8 ^f
U140	MC	90	He-0.36	SA-JNC-800	7.72x6.60	41	1460	10	10.8	b, 5
U141	MC	91	He-0.25	SA-316SS	7.70x6.60	43	1460	10	10.6	a, 8 ^f
U142	MC	91	He-0.25	SA-316SS	7.72x6.60	43	1460	11	10.7	b, 7
U143	MC+20% M ₂ C ₃	96 ^d	He-0.36	SA-INC-800	7.67x6.60	38	1390	11	10.9	b, 5
U144	MC+20% M ₂ C ₃	96 ^d	He-0.36	SA-316SS	7.72x6.60	39	1390	11	11.1	b, 5
U145	MC	90	Na-0.76	SA-304SS	7.75x6.86	40	820	10	10.5	a, 8 ^f
U146A ^a	MC+20% M ₂ C ₃	99	Na-0.76	SA-304SS	7.62x6.86	41	810	8	7.0	b, 5
U147	MC+20% M ₂ C ₃	97	Na-0.76	SA-INC-800	7.72x6.86	42	810	10	10.6	b, 5

^a Capsules 138 and 146 were removed at 4.0 at. % burnup for TREAT testing.
Duplicates replaced the originals.

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

^c Theoretical density of MC = 13.49 Mg/m³.
Theoretical density of M₂C₃ = 12.76 Mg/m³.

^d Cored pellet with nominal 2.0-mm-diameter axial hole.

^e SA = Solution annealed.

^f Element cladding failure indicated.

^g Computed using EBR-II power adjustment factor of 0.91.

^h Scheduled for TREAT testing.

eight elements were the only ones from these series under active hot-cell postirradiation examination during FY 1975. Elements U188, U196, U201, U207, and U209 have been decapsulated and revealed obvious cladding breaches (discussed in Section II. B. 1d).

The capsules which were damaged at HFEEF (see Table 463-IV) required special decapsulation procedures in order to recover the element in a condition which will allow nearly normal examination procedures to be used. Damaged elements U188, U207, and U209

TABLE 463-IV
SERIES U1930 AND U1960 ENCAPSULATED-CARBIDE EXPERIMENTS

Exptl. No.	Fuel Type ^a	Fuel Density % Theo. ^b	Bond and Diametral Gap, mm	Clad Type	Clad O. D. x I. D., mm	Max. Linear Power, kW/m ^h	Max. Centerline Temp., °C ^h	Clad Burnup at %	Core Burnup at %	Status
U187	MC+5% M ₂ C ₃	96	He-0.18	SA-316SS	7.72 x 6.60	77	1900	5	4.60 ^e	b, 13
U189	MC+5% M ₂ C ₃	85	He-0.25	SA-INC-800	7.67 x 6.60	81	1900	5	4.60 ^e	b, 13
U191	MC	92	Na-0.76	SA-304SS	7.72 x 6.91	84	1010 ^c	5	4.50 ^e	b, 12
U192	MC	92	Na-0.76	SA-304SS	7.75 x 6.91	73	910 ^c	5	4.3	b, 12
U194	MC+10% M ₂ C ₃	98	Na-0.84	SA-304SS	7.75 x 6.91	81	900 ^c	5	4.61 ^e	b, 12 ^f
U195	MC+10% M ₂ C ₃	98	Na-0.84	SA-304SS	7.75 x 6.91	83	990 ^c	5	4.9 ^e	b, 12
U197	MC+10% M ₂ C ₃	98	Na-0.84	SA-INC-800	7.75 x 6.91	80	1020 ^c	5	4.90 ^e	b, 12
U198	MC+10% M ₂ C ₃	98	Na-0.84	SA-INC-800	7.75 x 6.91	84	1090 ^c	5	4.75 ^e	b, 12
U200	MC+5% M ₂ C ₃	86	He-0.23	SA-304SS	7.32 x 6.60	83	2040	5	4.72 ^e	b, 13 ^f
U206	MC+5% M ₂ C ₃	96	He-0.25	SA-316SS	7.44 x 6.58	90	2080	5	4.80 ^e	b, 13
U208	MC+10% M ₂ C ₃	97 ^c	He-0.30	SA-316SS	7.44 x 6.58	87	1910	5	4.80 ^e	b, 12
U188	MC+5% M ₂ C ₃	85	He-0.18	SA-316SS	7.72 x 6.60	90	1900	11	11.0	a, 9 ^g
U190	MC+5% M ₂ C ₃	85	He-0.28	SA-INC-800	7.67 x 6.60	90	1920	11	11.6	a, 4 ^g
U193	MC	92	Na-0.79	SA-304SS	7.73 x 6.91	91	1090	11	11.1	a, 9 ^g
U196	MC+15% M ₂ C ₃	95	Na-0.79	SA-304SS	7.75 x 6.91	97	1060	11	10.43 ^e	b, 9 ^g
U199	MC+10% M ₂ C ₃	95	Na-0.79	SA-INC-800	7.75 x 6.91	100	1360	11	8.2	a, 5 ^g
U201	MC+5% M ₂ C ₃	95	He-0.23	SA-304SS	7.32 x 6.60	90	2040	11	7.30 ^e	b, 7 ^g
U207	MC+5% M ₂ C ₃	90	He-0.25	SA-316SS	7.44 x 6.58	94	2000	11	7.8	b, 7 ^g
U209	MC+10% M ₂ C ₃	97 ^c	He-0.30	SA-316SS	7.44 x 6.58	92	1910	11	7.9	b, 7 ^g
U185	MC+10% M ₂ C ₃	97	He-0.28	SA-316SS	7.72 x 6.60	90	2150	3	2.7	a, 7 ^g
U186	MC+10% M ₂ C ₃	97	He-0.28	SA-316SS	7.72 x 6.60	93	2150	3	2.7	a, 7 ^g
U202	MC+5% M ₂ C ₃	85	He-0.01	SA-316SS	6.83 x 3.35	94	1270	3	2.5	a, 7 ^g
U203	MC+5% M ₂ C ₃	85	He-0.03	SA-316SS	7.32 x 6.43	95	1260	3	2.6	EBR-II, X233
U204	MC+10% M ₂ C ₃	97 ^c	He-0.05	SA-316SS	6.76 x 6.32	96	1130	3	2.6	a, 7 ^g
U205	MC+10% M ₂ C ₃	97 ^c	He-0.08	SA-316SS	7.21 x 6.35	95	1120	3	2.6	a, 7 ^g
U260	MC+10% M ₂ C ₃	98	He-0.48	20CW-316SS	7.57 x 3.71	90	2400	0	0	EBR-II, X233
U261	MC+10% M ₂ C ₃	98	He-0.38	SA-316SS	7.49 x 6.30	--	--	-	0	Refect ⁱ
U262	MC+10% M ₂ C ₃	96	He-0.38	SA-INC-800	7.34 x 6.60	90	2260	3	0	EBR-II, X233

^aM = (U_{0.65}Pu_{0.15}).

^bTheoretical density of MC = 13.45 Mg/m³. Theoretical density of M₂C₃ = 12.72 Mg/m³.

^cCored pellet with nominal 2.0-mm-diametral axial hole.

^dSA = Solution annealed; 20 CW = 20% cold worked.

^eElement cladding failure.

^fEBR-II eddy current test indicates capsule bond discontinuity.

^gBurnup values marked with * were measured using the ¹⁴⁸Nd method. Remaining values were computed using an EBR-II power adjustment factor of 0.91. The values shown for the elements in X233 are prior to irradiation in X233.

^hLinear powers and centerline temperatures 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.

ⁱDamaged at EBR-II.

^jStorage.

TABLE 463-V
SERIES WF ENCAPSULATED-CARBIDE EXPERIMENTS

Exptl. No.	Fuel Type ^a	Fuel Density % Theo. ^b	Bond and Diametral Gap, mm.	Clad Type	Clad O. D., I. D., mm	Max Linear Power, kW/m ²	Max Centerline Temp., °C	Goal Burnup at, %	Max Current Burnup at, % ^c	Status
W3F	MC-15% M ₂ C ₃	83	Na-0.64	SA-316SS	7.62x7.01	81	1000	10	5.5	EBR-II, X233
W4F	MC-10% M ₂ C ₃	95	Na-0.69	SA-316SS	7.62x7.01	82 ^f	950	6	5.32*	b, 12
W5F	MC-10% M ₂ C ₃	91	Na-0.25	SA-316SS	6.35x5.94	60	925	10	5.4	EBR-II, X233
W6F	MC-10% M ₂ C ₃	90	Na-0.28	SA-316SS	6.35x5.97	60	925	6	5.26*	b, 12 ^d
W7F	MC-5% M ₂ C ₃	90	Na-0.69	20CW-316SS	7.62x7.01	81	1000	10	5.5	EBR-II, X233
W8F	MC-5% M ₂ C ₃	94	Na-0.64	20CW-316SS	6.35x5.97	83 ^f	950	6	5.52*	b, 12
W10F	MC	89	Na-0.30	SA-316SS	6.35x5.97	60	925	10	5.4	EBR-II, X233
W12F	MC-25% M ₂ C ₃	87	Na-0.33	SA-316SS	6.35x5.97	63	975	10	5.4	EBR-II, X233

^aM = (C_{0.8} Pu_{0.2}).

^bTheoretical density of MC = 13.45 Mg/m³. Theoretical density of M₂C₃ = 12.72 Mg/m³.

^cSA = Solution annealed, 20 CW = 20% cold worked.

^dElement cladding failure.

^eBurnup values marked with * were measured using the ¹⁴⁸Nd method. Remaining values were computed using EBR-II power adjustment factor of 0.91. The values shown for the elements in X233 are those prior to irradiation in X233.

^fLinear power marked with ² are beginning-of-life values computed using the measured burnup results. Remaining values are based on EBR-II power adjustment factor of 0.91.

were successfully removed with only minor additional damage; damaged elements U190, U193, and U199 will be decapsulated in FY 1976.

The experiments listed in part C of Table 463-IV were used as replacement capsules in order to allow irradiation to be continued to the desired burnup in lead experiments from other series. A cursory examination was performed on these elements, and the five indicated elements were then placed in long-term storage. The sixth element in this group, U203, along with elements U260 and U262 from part D of Table 463-IV, was reconstituted into EBR-II subassembly X233 for additional irradiation.

Table 463-V describes the Series WF experiments. These sodium-bonded carbide elements 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%. The only failed experiment in this series, W6F, has completed postirradiation examination at LASL. The other experiments, W4F and W8F, have completed postirradiation examination. The remaining five capsules have been reconstituted in EBR-II subassembly X233 for continued irradiation.

Table 463-VI describes the Series B-1, B-2, and B-3 experiments. These capsules are fueled with single-phase, uranium-plutonium mononitride. All the elements in Series B-1 and B-2 are sodium bonded and clad with either Type 304 or 316 welded stainless steel tubing. Operating linear power ratings for the experiments are relatively high (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 ¹³⁷Cs; the fuel elements were apparently intact. Element B-2-3 was returned to LASL, and its normal postirradiation examination

TABLE 463-VI

SERIES B-1, B-2 AND B-3 ENC. INSULATED-NITRIDE EXPERIMENTS

Expm't. 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	Max Centerline Temp., °C ^g	Goal Burnup at. %	Max Current Burnup at. % ^e	Status
Series B-1										
B-1-1	MN	94	Na-0.48	SA-304SS	7.37x6.35	79 [□]	1045 [□]	5	5.70*	b.12 ^f
B-1-2	MN	94	Na-0.46	SA-304SS	7.37x6.35	81	1100	9	5.7	a.7 ^{f, h}
B-1-4	MN	94	Na-0.30	SA-304SS	7.37x6.35	85	1100	10	2.7	EBR-II, X233
Series B-2										
B-2-1	MN	95	Na-0.53	SA-316SS	8.02x6.99	98	1230	10	5.7	EBR-II, X233
B-2-2	MN	94	Na-0.51	SA-316SS	8.01x6.98	93 [□]	1230	9	5.48*	b.12 ^f
B-2-3	MN	94	Na-0.51	SA-316SS	8.00x6.99	97	1230	12	8.23*	b.12
B-2-5	MN	94	Na-0.71	SA-316SS	8.00x7.21	97	1230	12	2.7	EBR-II, X233
B-2-6	MN	94	Na-0.53	SA-316SS	8.03x7.49	109	1230	6	5.6	a.7 ^{f, h}
B-2-7	MN	94	Na-0.51	SA-316SS	8.03x7.49	110	1230	12	5.6	a.7 ^{f, h}
Series B-3										
B-3-2	MN	88	Na-0.23	SA-316SS	8.01x7.22	110 [□]	1235 [□]	9	2.78*	b.12 ^f
B-3-3	MN	91	Na-0.25	SA-316SS	8.01x7.22	116	1280	12	2.77*	b.12 ^f
B-3-4	MN	94	Na-0.33	SA-316SS	8.01x7.22	108 [□]	1200 [□]	12	2.70*	b.12 ^f
B-3-5	MN	90	Na-0.25	SA-316SS	8.01x7.48	118 [□]	1255 [□]	6	2.74*	b.12 ^f
B-3-6	MN	95 ^c	He-0.13	SA-316SS	8.00x6.99	102	1920	10	6.0	EBR-II, X233
B-3-7	MN	89	He-0.13	SA-316SS	8.00x6.99	102	1920	6	5.50*	b.12
B-3-8	MN	90 ^c	He-0.13	SA-316SS	8.00x6.99	97	1870	10	6.0	EBR-II, X233

^a M = (U_{0.8} Pu_{0.2}).^b Theoretical density of MN = 14.17 Mg/m³.^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 ¹⁴⁸Nd method. Remaining values were computed using an EBR-II power adjustment factor of 0.91. The values shown for the elements in X233 are those prior to irradiation in X233.^f Element cladding failure indicated.^g 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.^h Storage

sequence has been completed. The other three elements, B-1-4, B-2-1, and B-2-5 have been reconstituted in EBR-II subassembly X233 for further irradiation. Failed elements B-1-2, B-2-6, and B-2-7 were placed in long-term storage pending the completion of higher priority examinations.

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 had not failed at a maximum burnup of 6 at. %. All of the failed sodium bonded elements in the Series B-3 have completed postirradiation examination. One helium bonded experiment, B-3-7, has been returned to LASL, and its normal postirradiation examination sequence has been completed. The other two helium-bonded nitride experiments, B-3-6 and B-3-8, have been reconstituted in EBR-II subassembly X233 for further irradiation.

TABLE 463-VII
SERIES U5100 SINGLY-CLAD CARBIDE EXPERIMENTS

Exptmt. 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	Max. Centerline Temp., °C	Goal Burnup, at. %	Max Current Burnup at. %	Status
U241	MC	94	Na-0.46	SA-304SS	7.87x7.14	107	1170	6	3.0	a.4 ^g
U242	MC	93	Na-0.44	SA-304SS	7.87x7.14	107	1170	9	3.0	a.4 ^g
U243	MC	93	Na-0.80	SA-304SS	7.87x7.14	101	1150	6	2.9	a.4 ^g
U244	MC	93	Na-0.44	SA-304SS	7.87x7.14	107	1170	9	3.0	a.4 ^g
U245	MC	93	Na-0.81	SA-304SS	7.87x7.14	101	1150	12	2.9	a.4 ^g
U246	MC	93	Na-0.43	SA-316SS	7.87x7.14	109	1190	6	3.02*	b.12
U247	MC	93	Na-0.81	SA-316SS	7.87x7.14	101	1150	6	2.9	a.4 ^g
U248	MC	93	Na-0.81	SA-316SS	7.87x7.14	109	1140	12	3.16*	b.11 ^e
U249	MC	93	Na-0.43	SA-INC-800	7.85x7.14	109	1210	6	3.1	a.4 ^g
U250	MC	93	Na-0.81	SA-INC-800	7.85x7.14	109	1140	6	3.1	a.4 ^g
U251	MC	93	Na-0.81	SA-304SS	7.87x7.14	109	1140	12	3.1	a.4 ^g
U252	MC	93	Na-0.64	SA-304SS	7.87x7.14	109	1140	12	3.1	EBR-II, X246
U253	MC	93	Na-0.66	SA-304SS	7.87x7.14	101	1140	12	2.9	EBR-II, X246
U254	MC	93	Na-0.66	SA-304SS	7.87x7.14	101	1140	12	2.9	EBR-II, X246
U256	MC+15% M ₂ C ₃	96	Na-0.61	SA-304SS	7.85x7.14	101	1140	12	2.9	EBR-II, X246
U257	MC+15% M ₂ C ₃	96	Na-0.58	SA-INC-800	7.85x7.14	100	1130	12	2.9	EBR-II, X246
U258	MC+15% M ₂ C ₃	96	Na-0.58	SA-304SS	7.87x7.14	100	1140	6	2.9	EBR-II, X246
U259	MC+15% M ₂ C ₃	96	Na-0.58	SA-INC-800	7.85x7.14	103	1150	12	3.0	EBR-II, X246

^a M = (U_{0.85}Pu_{0.15}).

^b Theoretical density of MC = 13.45 Mg/m³.
Theoretical density of M₂C₃ = 12.72 Mg/m³.

^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.

^f Computed using an EBR-II power adjustment factor of 0.91. Burnup values marked with * were measured using the ¹⁴⁸Nd method.

^g Storage

The Series U5100 singly-clad experiments are described in Table 463-VII. 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.

Failed element U248 and element U246, which had displaced fuel pellets, were returned to LASL, and both have essentially completed the postirradiation examination sequences. The failure point in U248, a tiny longitudinal crack, was located by a helium pressurization test. The flow region was examined metallographically using a sequential grinding technique (see discussion of details in Section II. B. 2e).

The remainder of the unshrouded elements, all unfilled (U241 through U251, with the exception of U246 and U248) were returned to LASL, given a cursory examination, and then were placed in long-term storage. The seven shrouded elements, U252 through U259, were reconstituted as part of EBR-II subassembly X246 for additional irradiation to begin in mid-July 1975.

The C-5 and O-N1 series of singly-clad experiments are described in Table 463-VIII. 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 in the C-5 series elements have been made using the same equipment that will be used for the postirradiation examination. Twelve of the elements have been shipped to EBR-II. Irradiation in subassembly X163 is to start in mid-July 1975.

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. Four of these elements were shipped to EBR-II, and irradiation in subassembly X163 is to start in mid-July 1975.

The K4 series experiments are described in Table 463-IX. The K4 series is comprised of eight carbide-fueled elements and eight nitride-fueled elements. All of the K4 elements are sodium bonded and contain fuel fragment restraint devices (shrouds) which are thin-walled, perforated, metal tubes. The shroud tubes are 25% cold-worked, Type 316 stainless steel, 0.076 mm (0.003 in.) thick. The K4 series, designed and fabricated at LASL, is intended to test the fuel-shroud concept and to compare the behavior of sodium-bonded carbide and nitride elements under as nearly identical conditions as possible. Twelve of the K4 elements, six carbide and six nitride, are in EBR-II subassembly X246 with seven U5100 elements. Three of the K4 elements, one carbide and two nitride,

are in EBR-II subassembly X163 with the C-5 and O-N1 elements. These fifteen K4 elements are planned to begin irradiation in EBR-II in mid-July 1975.

The K6 series elements are described in Table 463-X. The K6 series will be irradiated in two A-19B subassemblies presently designated X258 (K6A) and X259 (K6B). The primary objective of the K6 series tests is to provide initial, statistical, behavioral data for helium-bonded carbide-fueled elements for FTR testing. Approval-in-Principle was granted September 18, 1974, and the "Experiment Description and Hazards Analysis - K6 Series Helium-Bonded, Carbide-Fueled Elements" was completed on June 17, 1975, and will be forwarded to the Experiment Manager, EBR-II Project, ANL, early in July 1975.

Since subassembly X233 comprises elements for which the descriptive data are in four separate tables, Table 463-XI is included for convenience and describes the elements being irradiated for an additional 3 at.% burnup in X233.

B. Postirradiation Examination Results

As indicated in Tables 463-I through 463-VII, most of the experiments which have completed irradiation are in various stages of their postirradiation examination. The final results for those elements which have completed the examination sequences will be reported in topical reports, either singly or in groups. This section will report on significant examination results, although complete results may not be available for any given element. This is being done to avoid piecemeal reporting and to prevent undue delays in reporting before the final topical reports can be issued. In this sense, any trends indicated in these discussions should be considered as preliminary, since the complete examination results can alter the initial interpretations.

During FY 1975, a total of 49 experiments were processed in some degree through the various post-irradiation examinations. These examinations ranged from a cursory examination of nine experiments (Series U5100), through partial examination of 32 experiments as dictated by time and hot cell availability, to the complete examination sequence of 8 experiments.

TABLE 463-VIII
SERIES C-5 AND ON-1 SINGLY-CLAD NITRIDE EXPERIMENTS

Expt. No.	Fuel Type ^a	Fuel Density % Theo.	Bond and Diametral Gap, mm	Clad Type	Clad O. D. x I. D. mm	Max Linear Power, kW/m	Max Centerline Temp., °C	Goal Burnup, at. %	Max Current Burnup at. %	Status
C-5-1	MN	93	Na-0.51	20CW-316SS	7.87x7.11	---	----	--	--	Reject ^{d, e}
C-5-2	MN	94	Na-0.51	20CW-316SS	7.87x7.11	---	----	--	--	Reject ^{d, e}
C-5-3	MN	95	Na-0.51	20CW-316SS	7.87x7.11	---	----	--	--	Reject ^{d, e}
C-5-4	MN	96	Na-0.53	20CW-316SS	7.87x7.11	---	----	--	--	Reject ^g
C-5-5	MN	96	Na-0.51	20CW-316SS	7.87x7.11	---	----	--	--	Reject ^f
C-5-6	MN	94	Na-0.53	20CW-316SS	7.87x7.11	82	1050	12	0	EBR-II, X163
C-5-7	MN	95	Na-0.51	20CW-316SS	7.87x7.11	84	1060	12	0	EBR-II, X163
C-5-8	MN	95	Na-0.76	20CW-316SS	7.87x7.11	82	960	12	0	EBR-II, X163
C-5-9	MN	95	Na-0.51	20CW-316SS	7.87x7.11	84	980	12	0	EBR-II, X163
C-5-10	MN	95	Na-0.51	20CW-316SS	7.87x7.11	83	970	12	0	EBR-II, X163
C-5-11	MN	95	Na-0.51	20CW-316SS	7.87x7.11	83	970	12	0	EBR-II, X163
C-5-12	MN	95	Na-0.76	20CW-316SS	7.87x7.11	---	----	--	--	Reject ^d
C-5-13	MN	96	Na-0.76	20CW-316SS	7.87x7.11	82	950	12	0	EBR-II, X163
C-5-14	MN	97	Na-0.76	20CW-316SS	7.87x7.11	81	1030	12	0	EBR-II, X163
C-5-15	MN	96	Na-0.76	20CW-316SS	7.87x7.11	82	970	12	0	EBR-II, X163
C-5-16	MN	96	Na-0.76	20CW-316SS	7.87x7.11	80	1030	12	0	EBR-II, X163
C-5-17	MN	97	Na-0.76	20CW-316SS	7.87x7.11	---	----	--	--	Reject ^e
C-5-18	MN	95	Na-0.53	20CW-316SS	7.87x7.11	---	----	--	--	Reject ^d
C-5-19	MN	94	Na-0.53	20CW-316SS	7.87x7.11	82	980	12	0	EBR-II, X163
C-5-20	MN	96	Na-0.53	20CW-316SS	7.87x7.11	84	1050	12	0	EBR-II, X163
O-N1-1	MN	91	Na-0.51	20CW-316SS	7.87x7.11	80	990	12	0	EBR-II, X163
O-N1-2	MN	91	Na-0.51	20CW-316SS	7.87x7.11	---	----	--	--	Reject ^d
O-N1-3	MN	91	Na-0.51	20CW-316SS	7.87x7.11	80	1060	12	0	EBR-II, X163
O-N1-4	MN	90	Na-0.51	20CW-316SS	7.87x7.11	80	1060	12	0	EBR-II, X163
O-N1-5	MN	91	Na-0.51	20CW-316SS	7.87x7.11	---	----	--	--	Reject ^d
O-N1-6	MN	90	Na-0.51	20CW-316SS	7.87x7.11	---	----	--	--	Reject ^d
O-N1-8	MN	91	Na-0.51	20CW-316SS	7.87x7.11	81	980	12	0	EBR-II, X163

^aM = (U_{0.8} Pu_{0.2}).

^fPossible impurities present.

^bTheoretical density of MN = 14.17 Mg/m³.

^gDamaged.

^c20CW = 20% cold worked.

^dFuel chips in bond.

^eAir in plenum.

TABLE 463-IX
SERIES K4 SINGLY-CLAD CARBIDE AND NITRIDE EXPERIMENTS

Exptl. No.	Fuel Type ^a	Fuel Density ^b % Theo.	Bond and Diametral Gap, mm	Clad ^d Type	Clad O. D. x I. D. mm	Max Linear Power kW/m	Max Centerline Temp., °C	Goal Burnup at, %	Max Current Burnup at, %	Status
K4-1	MC+3% M_2C_3	99	Na-0.69	20CW-316SS	7.87x7.26	88	940	12	0	EBR-II, X246
K4-2	MC+2% M_2C_3	99	Na-0.69	20CW-316SS	7.87x7.26	88	940	12	0	EBR-II, X246
K4-3	MC+4% M_2C_3	99	Na-0.69	20CW-316SS	7.87x7.26	88	940	12	0	EBR-II, X246
K4-4	MC+12% M_2C_3	99	Na-0.69	20CW-316SS	7.87x7.26	86	930	12	0	EBR-II, X246
K4-5	MC+14% M_2C_3	99	Na-0.69	20CW-316SS	7.87x7.26	80	890	12	0	EBR-II, X246
K4-6	MC+17% M_2C_3	99	Na-0.69	20CW-316SS	7.87x7.26	79	880	12	0	EBR-II, X246
K4-8	MN	97	Na-0.69	20CW-316SS	7.87x7.26	85	980	12	0	EBR-II, X246
K4-9	MN	97	Na-0.69	20CW-316SS	7.87x7.26	84	980	12	0	EBR-II, X246
K4-10	MN	97	Na-0.69	20CW-316SS	7.87x7.26	89	1060	12	0	EBR-II, X246
K4-11	MN	97	Na-0.69	20CW-316SS	7.87x7.26	88	1050	12	0	EBR-II, X246
K4-12	MN	97	Na-0.69	20CW-316SS	7.87x7.26	87	1040	12	0	EBR-II, X246
K4-13	MC+6% M_2C_3	99	Na-0.69	20CW-316SS	7.87x7.26	--	---	--	0	LASL- unassigned ^e
K4-14	MC+18% M_2C_3	99	Na-0.69	20CW-316SS	7.87x7.26	86	1010	12	0	EBR-II, X163
K4-15	MN	97	Na-0.69	20CW-316SS	7.87x7.26	85	970	12	0	EBR-II, X163
K4-16	MN	97	Na-0.69	20CW-316SS	7.87x7.26	86	1010	12	0	EBR-II, X163
K4-17	MN	97	Na-0.69	20CW-316SS	7.87x7.26	87	1000	12	0	EBR-II, X246

^aM = (U_{0.8}Pu_{0.2}).

^bTheoretical density of MC = 13.45 Mg/m³, M_2C_3 = 12.72 Mg/m³, and MN = 14.17 Mg/m³.

^cThe diametral gap shown is the difference between the fuel o. d. and the cladding i. d. The thickness of the perforated shroud tube has not been subtracted.

^d20 CW = 20% cold worked.

^eReplacement element.

Three of the 32 partially completed sequences involved the completion of examination requirements on older experiments whose examination sequence had been initiated in prior years. Of the 49 processed experiments, 44 were new experiments which began their postirradiation examination sequence during FY 1975.

Since the examination sequences involved one or more experiments from each of the series as described in Tables 463-I through 463-VII, the ensuing discussion will summarize the extent to which each examination sequence was involved in the overall program. Significant findings will be presented on an individual experiment basis.

1. Examination Sequence

a. Gamma Scan

Gross gamma scans were performed on 28 experiments during the year. Seven of these experiments were from subassembly X182 (K-44, B-2-3, B-3-7, W6F, U196, U201, and U207). The gamma-scan examinations for five other elements in this subassembly which were severely damaged at HFEF were waived because straight-line orientations for the slit system could not be maintained through the fueled regions of these elements. Multispectral gamma scans were also performed on two of the experiments, U196 and

TABLE 463-X
 SERIES K6 SINGLY-CLAD CARBIDE EXPERIMENTS
 (Nominal values)

Exptl. No.	Fuel Type ^a	Fuel Density ^b % Theo.	Bond and Diametral Gap, mm	Clad ^{c, d} Type	Clad O. D. x I. D. mm	Max Linear Power kW/m	Max Centerline Temp., °C	Goal Burnup at, %	Max Current Burnup at, %	Status
K6A-52	MC+10% M ₂ C ₃	81	He-0.13	20CW-316SS	7.87x6.86	73	1460	12	0	Fabrication
K6A-53	MC+10% M ₂ C ₃	81	He-0.13	20CW-316SS	7.87x6.86	73	1470	12	0	Fabrication
K6A-54	MC+10% M ₂ C ₃	81	He-0.13	20CW-316SS	7.87x6.86	72	1450	12	0	Fabrication
K6A-55	MC+10% M ₂ C ₃	81	He-0.13	20CW-316SS	7.87x6.86	73	1460	12	0	Fabrication
K6A-56	MC+10% M ₂ C ₃	81	He-0.13	20CW-316SS	7.87x6.86	73	1460	12	0	Fabrication
K6A-57	MC+10% M ₂ C ₃	81	He-0.13	20CW-316SS	7.87x6.86	71	1440	12	0	Fabrication
K6A-58	MC+10% M ₂ C ₃	81	He-0.13	20CW-316SS	7.87x6.86	73	1460	12	0	Fabrication
K6A-59	MC+10% M ₂ C ₃	81	He-0.13	20CW-316SS	7.87x6.86	72	1450	12	0	Fabrication
K6A-60	MC+10% M ₂ C ₃	81	He-0.13	20CW-316SS	7.87x6.86	72	1450	12	0	Fabrication
K6A-61	MC+10% M ₂ C ₃	81	He-0.25	20CW-316SS	7.87x6.86	73	1870	12	0	Fabrication
K6A-62	MC+10% M ₂ C ₃	81	He-0.25	20CW-316SS	7.87x6.86	73	1870	12	0	Fabrication
K6A-63	MC+10% M ₂ C ₃	81	He-0.25	20CW-316SS	7.87x6.86	71	1850	12	0	Fabrication
K6A-64	MC+10% M ₂ C ₃	81	He-0.25	20CW-316SS	7.87x6.86	73	1880	12	0	Fabrication
K6A-65	MC+10% M ₂ C ₃	81	He-0.25	20CW-316SS	7.87x6.86	72	1860	12	0	Fabrication
K6A-66	MC+10% M ₂ C ₃	81	He-0.25	20CW-316SS	7.87x6.86	72	1860	12	0	Fabrication
K6A-67	MC+10% M ₂ C ₃	81	He-0.25	20CW-316SS	7.87x6.86	72	1870	12	0	Fabrication
K6A-68	MC+10% M ₂ C ₃	81	He-0.25	20CW-316SS	7.87x6.86	71	1850	12	0	Fabrication
K6A-69	MC+10% M ₂ C ₃	81	He-0.25	20CW-316SS	7.87x6.86	71	1850	12	0	Fabrication
K6A-70	MC+10% M ₂ C ₃	81	He-0.25	20CW-316SS	7.87x6.86	72	1860	12	0	Fabrication
K6A-71	MC+10% M ₂ C ₃	81	He-0.25	20CW-316SS	7.87x6.86	----	----	--	0	Fabrication ^e
K6A-72	MC+10% M ₂ C ₃	81	He-0.25	20CW-316SS	7.87x6.86	----	----	--	0	Fabrication ^e
K6A-73	MC+10% M ₂ C ₃	81	He-0.25	20CW-316SS	7.87x6.86	----	----	--	0	Fabrication ^e
K6B-74	MC	87	He-0.25	20CW-316SS	7.87x6.86	70	1800	12	0	Fabrication
K6B-75	MC	87	He-0.25	20CW-316SS	7.87x6.86	68	1770	12	0	Fabrication
K6B-76	MC	87	He-0.25	20CW-316SS	7.87x6.86	71	1810	12	0	Fabrication
K6B-77	MC	87	He-0.25	20CW-316SS	7.87x6.86	67	1750	12	0	Fabrication
K6B-78	MC	87	He-0.25	20CW-316SS	7.87x6.86	70	1790	12	0	Fabrication
K6B-79	MC	87	He-0.25	20CW-316SS	7.87x6.86	68	1770	12	0	Fabrication
K6B-80	MC	87	He-0.25	20CW-316SS	7.87x6.86	71	1800	12	0	Fabrication
K6B-81	MC	87	He-0.25	20CW-316SS	7.87x6.86	69	1780	12	0	Fabrication
K6B-82	MC	87	He-0.25	20CW-316SS	7.87x6.86	72	1820	12	0	Fabrication
K6B-83	MC+10% M ₂ C ₃	87	He-0.25	20CW-316SS	7.87x6.86	73	1830	12	0	Fabrication
K6B-84	MC+10% M ₂ C ₃	87	He-0.25	20CW-316SS	7.87x6.86	75	1870	12	0	Fabrication
K6B-85	MC+10% M ₂ C ₃	87	He-0.25	20CW-316SS	7.87x6.86	74	1840	12	0	Fabrication
K6B-86	MC+10% M ₂ C ₃	87	He-0.25	20CW-316SS	7.87x6.86	77	1880	12	0	Fabrication

TABLE 463-X (continued)

Expmnt. No.	Fuel Type ^a	Fuel Density ^b % Theo.	Bond and Diametral Gap, mm	Clad ^{c, d} Type	Clad O. D. x I. D. mm	Max Linear Power kW/m	Max Centerline Temp., °C	Goal Burnup at, %	Max Current Burnup at, %	Status
K6B-87	MC+10% M ₂ C ₃	87	He-0.25	20CW-316SS	7.87x6.86	72	1820	12	0	Fabrication
K6B-88	MC+10% M ₂ C ₃	87	He-0.25	20CW-316SS	7.87x6.86	75	1860	12	0	Fabrication
K6B-89	MC+10% M ₂ C ₃	87	He-0.25	20CW-316SS	7.87x6.86	78	1900	12	0	Fabrication
K6B-90	MC+10% M ₂ C ₃	87	He-0.25	20CW-316SS	7.87x6.86	73	1840	12	0	Fabrication
K6B-91	MC+10% M ₂ C ₃	87	He-0.25	20CW-316SS	7.87x6.86	76	1870	12	0	Fabrication
K6B-92	MC+10% M ₂ C ₃	87	He-0.25	20CW-316SS	7.87x6.86	74	1850	12	0	Fabrication
K6B-93	MC+10% M ₂ C ₃	87	He-0.25	20CW-316SS	7.87x6.86	----	----	--	0	Fabrication ^e
K6B-94	MC+10% M ₂ C ₃	87	He-0.25	20CW-316SS	7.87x6.86	----	----	--	0	Fabrication ^e
K6B-95	MC	87	He-0.25	20CW-316SS	7.87x6.86	----	----	--	0	Fabrication ^e

^a M = (U_{0.8} Pu_{0.2}).

^b Theoretical density of MC = 13.45 Mg/m³ and of M₂C₃ = 12.72 Mg/m³.

^c 20CW = 20% cold worked.

^d FFTF first core material, M-3-28.

^e Replacement element.

B-3-7. Both gross and multispectral gamma scans were performed on two elements in the U5100 series, U246 and U248. Finally, gross gamma scans were performed on all 19 elements in the U1950 series (Table 463-III). In addition, multispectral gamma scans were performed on 14 of these elements; three of these scans covered the fuel stack only, six were made in the capsule sodium region only, and the remaining five covered both regions.

The gross gamma activity over the fuel stack showed a normal distribution for most of the examined experiments, but there were a few exceptions. In addition, abnormal distributions of various fission product or activation product isotopes were found in the multispectral gamma scans of some experiments. These abnormalities will be discussed in Section II, B. 2.

b. Profilometry

Optical profilometry measurements were made on the 12 capsules from subassembly X182 and the 19 capsules from subassembly X055B. Optical profilometry measurements were also made on unfailed elements K-44, B-2-3, B-3-7, U246, and the ten unfailed

elements from subassembly X055B (see Table 463-III).

Mechanical profilometry measurements were made on ten failed elements: U98 and U106 from the U1300 series; W6F, U196, U201, U207, and U209 from subassembly X182; U248 from the U5100 series; U131 and U132 from the U1950 series. The results of the fuel element profilometry measurements are summarized in Table 463-XII.

c. Decapsulation

Twenty-five experiments were decapsulated during the year. Of these, 11 were classified as failed, and the other 14 were unfailed. The two TREAT experiments, UL-3 and UL-4, required special decapsulation procedures because of their instrumented capsules. Most of the 12 experiments which were decapsulated from subassembly X055B (U1950 series) required some extra effort because of the multiple bowing of the element within the capsule. This effect consisted of element bowing in variable directions over relatively short axial distances. In these cases, the capsule cladding was cut off above and below the element. After melting the sodium, varying degrees of tapping from the top were required in order

TABLE 463-XI

EBR-II SUBASSEMBLY X233 ENCAPSULATED ADVANCED FUELS

Exptmt. 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	Max Centerline Temp., °C	Goal Burnup, at. %	Max Current Burnup at. % ^d
K39B	MC	90	Na-0.38	SA-316SS	7.62x7.11	104	1190	9	5.8
W3F	MC+15% M ₂ C ₃	93	Na-0.64	SA-316SS	7.62x7.01	89	1050	8	5.5
W5F	MC+10% M ₂ C ₃	91	Na-0.25	SA-316SS	6.35x5.84	68	930	8	5.4
W7F	MC+5% M ₂ C ₃	90	Na-0.69	20CW-316SS	7.62x7.01	88	1050	8	5.5
W10F	MC	89	Na-0.30	SA-316SS	6.38x5.87	66	920	8	5.4
W12F	MC+25% M ₂ C ₃	97	Na-0.33	SA-316SS	6.38x5.87	72	920	8	5.4
B-1-4	MN	94	Na-0.30	SA-304SS	7.37x6.35	91	1170	6	2.7
B-2-1	MN	95	Na-0.53	SA-316SS	9.02x6.99	105	1230	9	5.7
B-2-5	MN	94	Na-0.71	SA-316SS	8.00x7.21	103	1220	6	2.7
B-3-6	MN	95	He-0.13	SA-316SS	8.00x6.99	110	1740	9	6.0
B-3-8	MN	90	He-0.13	SA-316SS	8.00x6.99	101	1700	9	6.0
U203	MC+5% M ₂ C ₃	85	He-0.05	SA-316SS	7.32x6.43	81	1250	5	2.6
U260	MC+10% M ₂ C ₃	98	He-0.48	20CW-316SS	7.57x6.71	86	2400	3	0
U262	MC+10% M ₂ C ₃	96	He-0.38	SA-INC-800	7.34x6.60	87	2270	3	0

^aM = (U_{0.8}Pu_{0.2}) except for U203, U260, and U262, where M = (U_{0.85}Pu_{0.15}).

^bTheoretical density of MC = 13.45 Mg/m³, of M₂C₃ = 12.72 Mg/m³, and of MN = 14.17 Mg/m³.

^cSA = solution annealed, 20CW = 20% cold worked, INC-800 = Incoloy 800.

^dThe burnup values shown are those prior to the present irradiation in X233.

to move the multiply bowed sections of the element through the open lower end of the capsule. A special procedure was also developed for decapsulation of the experiments which had been damaged at HFEF. One of these experiments, U209, was successfully decapsulated by making a cut through the capsule cladding both above and below the dented section. After removal of the upper and lower capsule sections, the dented portion was worked off or slit lengthwise for removal. The remainder of the experiments, both failed and unfailed, were decapsulated in normal fashion.

d. Failure Point Detection

Twelve experiments that had been classified as having cladding failures were processed during the year. The failure classification had been assigned as a result of the detection of ¹³⁷Cs in the capsule sodium by gamma-scanning techniques at HFEF or at LASL.

Element U248, a singly-clad experiment, was classified as failed upon detection of xenon tag gas at EBR-II and subsequent weighing at HFEF.

Within the group of failed elements, several failure points were quite obvious, others required special techniques for their detection, and an unresolved anomaly was encountered on two of the experiments. The obvious failure points occurred in the following elements: (1) UL-3 (U265), a TREAT experiment, one large rupture which extended from 58 to 132 mm from the bottom of the element, plus two smaller longitudinal cracks centered at about 38 mm and 190 mm, respectively, from the bottom of the element; (2) K-51, a large longitudinal crack which extended from 100 to 175 mm from the bottom of the element; (3) W6F, a transverse crack which completely encircled the element at about 57 mm from the bottom (the element was decapsulated in two

TABLE 463-XII
SUMMARY OF PROFILOMETRY MEASUREMENTS

<u>Experiment No.</u>	<u>Maximum Average Postirradiation Diameter, ^a mm</u>	<u>Initial Element Diameter, mm</u>	<u>Element Max $\Delta D/D$, %</u>
K-44	7.905	7.640	3.46
U98 (f)	7.935	7.734	2.59
U106 (f)	7.813	7.734	1.08
U130	7.899	7.724	2.24
U131 (f)	7.945	7.737	2.69
U132 (f)	7.889	7.696	2.51
U133	7.899	7.727	2.24
U134	7.892	7.722	2.20
U138A	7.526	7.455	0.95
U140	7.869	7.681	2.45
U142	7.927	7.719	2.70
U143	7.851	7.686	2.15
U144	7.854	7.717	1.78
U146A	7.656	7.625	0.40
U147	7.869	7.742	1.64
U196 (f)	7.836	7.755	1.05
U201 (f)	7.422	7.336	1.21
U207 (f)	7.534	7.442	1.23
U209 (f)	7.582	7.432	2.02
U246	7.894	7.882	0.16
U248 (f)	7.884	7.872	0.16
W6F (f)	6.381	6.348	0.52
B-2-3	8.049	8.026	0.28
B-3-7	8.108	8.021	1.08

^aThe maximum average diameter is the maximum value of a curve derived from averaging four profilometry traces which were taken at 45° intervals.

(f) Failed elements.

sections); (4) U196, a longitudinal crack centered at about 38 mm from the bottom of the element; (5) U201, two longitudinal straight line cracks in line with each other which appeared similar to seam splits, one extending from 73 to 102 mm and the other extending from 194 to 216 mm from the bottom of the element; (6) U207, a longitudinal crack which extended from 111 to 127 mm from the bottom of the element; and (7) U209, a transverse crack with a possibility of associated melting located at 159 mm from the bottom of the element.

Three failed elements required the use of an internal helium pressurization test for the detection of the failure points. Element U248, from Series U5100, had a small longitudinal crack approximately 1.5 mm in length and located at 287 mm from the bottom weld of the element. The crack had not been detected visually, but it could be seen under appropriate magnification after its position had been pinpointed by the stream of bubbles in

the helium pressurization test. The bubbles were not detected until an internal pressure of 2.41 MPa (350 psig) had been reached. Crude pressure-drop measurements with time allowed the calculation of an approximate leak rate and a corresponding hole size of about one μm^2 .

The helium pressurization tests indicated very small failure points for elements U131 and U132 from Series U1950 (subassembly X055B). At internal helium pressures of 3.45 MPa (500 psig), tiny bubbles, at the rate of 1 or 2 per second, could be seen coming from specific points on the elements: at 136 mm and at 188 mm from the bottom of element U131 and at 160 mm from the bottom of element U132. The capsule cover gases from both elements contained appreciable quantities of xenon and krypton. It is probable that the obviously tiny failure points could have been larger at reactor operating temperatures.

Experiments U98 and U106 from Series U1300 provided the anomalous failure data. Cesium was detected in the capsule sodium by gamma-scanning techniques at HFEF and on the upper capsule end plug by a similar technique at LASL. No fission gases were found in the capsule cover gas of either element. The presence of cesium, determined by gamma scanning, on the upper capsule end plug was confirmed by a subsequent repeat measurement. For element U106, the presence of ^{134}Cs and ^{137}Cs was further verified by chemical analysis after leaching the upper capsule end plug. No failure points could be detected in the cladding of either element by normal techniques, including a helium pressurization test to a maximum pressure of 3.45 MPa (500 psig). No satisfactory mechanism has been advanced to explain how cesium could have been released from the pin without the release of xenon and krypton.

Small cladding samples from the gross failure regions were taken and submitted for a fractographic study of the crack surfaces on the scanning electron microscope (SEM). These SEM samples were from elements UL-3 (U265), U196, U201, U207, U209, W6F, and K-51.

e. Metallography

Seventy-seven samples were submitted for metallographic examination from 22 sectioned elements. Examination sequences were completed on 35 of these samples from 11 elements. In addition, microprobe studies were made on eight of the completed samples. One sample, U248-F, which contained the previously described failure point, was subjected to a sequential grinding operation in order to follow the development of the crack, and this required at least one partial cladding mosaic at each grinding step. For every other sample, the normal procedure required 10x autoradiographs (both α and β - γ), one 50x mosaic of the sample cross-section in the as-polished condition, one 300x diametral strip, and selected areas both in the as-polished and in an etched condition, and one 500x etched cladding strip. The completed samples were from the following elements: UL-3 (U265), UL-4 (U266), K-44, K-51, W6F,

B-2-3, B-3-3, B-3-7, U97, U246, and U248. The microprobe samples were from elements UL-3 (U265), K-44, K-51, B-3-7 (2), U98, U246, and U248. The uncompleted samples were from elements U98, U106, U196, U201, U207, U209, U130, U131, U132, U138A, and U142.

f. Fission-Gas Release

Fission-gas-release calculations have been made for all elements for which fission-gas data are available. Final burnup values are not yet available for 8 of the total 22 elements; release calculations for these 8 were based on estimated burnup values. The percentage release values are presented in Table 463-XIII.

2. Preliminary Interpretations

The primary examination sequences have been completed for 11 of the processed elements and are still in progress for the remainder. The completed elements are in the final data reduction stages and topical reports will be issued. Preliminary interpretations have been made for some of the elements in both categories. Final conclusions, however, will be dependent on the outcome of the data reduction and completion of all examinations.

a. Series K1, K2, and K3

Two elements were examined from these series, K-44 and K-51. Experiment K-44 was operated at relatively high power, 80 kW/m, and was intact at 8.29 at. % burnup. The fission-gas-release value of 18.5% was normal for this type of fuel element. The profilometry data gave a $\Delta D/D$ value of 3.46% for this element which is significantly greater than for any other sodium-bonded carbide element examined thus far. Significant portions of the increase can be attributed to stainless steel swelling. Metallographic samples for this element revealed the possibility of some fuel-cladding chemical interaction and a possible intergranular second phase in the fuel. Preliminary results from the microprobe examination showed a number of fuel particles bonded to the cladding as a result of a reaction. This reaction zone penetrated the cladding to a maximum depth of 15 μm and was composed mainly

TABLE 463-XIII
FISSION-GAS-RELEASE SUMMARY

Element No.	Density % Theo.	Bond Type	Peak Burnup, at. %	% FG Release
<u>Carbide Fuel Elements</u>				
K44	94	Na	8.29	18.5
K51	95	Na	3.5	21.0 ^e
U97	86	He	9.60	13.4
U98	86	He	9.33	12.2
U106	77	He	9.53	25.1
U130	77	He	11.8	42.5 ^e
U131	85	He	11.5	28.2 ^e
U132	85	He	11.4	29.6 ^e
U138A	98	He	7.1	5.4 ^e
U142	91	He	10.7	7.0 ^e
U196	98	Na	10.43	37.1
U201	85	He	7.30	19.2
U207	90	He	7.9	14.4 ^e
U209	97 ^c	He	7.9	14.7 ^e
U246	93	Na	3.02	18.2
U248	93	Na	3.16	16.3
UL-3	99	He	3.56	18.8
UL-4	99	Na	3.62	10.4
<u>Nitride Fuel Elements</u>				
B-2-3	94	Na	8.23	3.8
B-3-3	91	Na	2.77	7.4
B-3-7	89	He	5.50	22.5

c - cored fuel pellets.

e - Used estimated burnup values.

of Pu, U, Ni, and Fe in varying amounts accompanied by traces of fission products Ce and Pd. A phase with a metallic appearance was observed between fuel grains near the outer surface of the fuel and might be evidence of a possible shift toward higher fissile metal activity as burnup accumulates. Partial analysis of this phase showed that it contained approximately 30% Pu and 4% Ru with small amounts (~1%) of Nd, Tc, and Pr.

Experiment K51 operated at high linear power, 130 kW/m, and was failed at approximately 3.5 at. % burnup. This element incurred severe element-

capsule heat-transfer interaction because of gas blanket-ing and was not typical of normal operation. The element had a cladding failure consisting of a massive rupture which extended from 100 to 175 mm from the bottom of the element, plus several smaller cracks. Preliminary microprobe results from a sample in the vicinity of the cladding rupture showed evidence of cladding constituents scattered throughout the fuel with marked concentrations of plutonium, fission products, and cladding constituents at isolated positions in existing fuel cracks.

b. Series U1300

The conflicting failure data for the two processed elements in this series, U98 and U106, have been discussed in Section II.B.1d. The profilometry measurements and fission-gas-release calculations for these two elements show some differences. Both elements had approximately the same burnup. Element U98 (86% fuel density) had a maximum average $\Delta D/D$ of 2.59% and a fission-gas release of 12.2%; element U106 (77% fuel density) had a maximum average $\Delta D/D$ of 1.08% and a fission-gas release of 25.1%.

The metallographic examination of samples from U98 and U106 have shown similar characteristics of bonding between the fuel and the incoloy 800 cladding over most of the circumferential contact area in all samples. A microprobe study of one sample from element U98 showed concentrations of plutonium and nickel in the bonding zone.

c. Series U1950

The postirradiation examination of the elements in this series (irradiation completed as EBR-II subassembly X055B) was begun during the third quarter of FY 1975. It is noteworthy that of the nineteen elements that attained burnups in the range 7 to 8 at. % only one possible failure (U136) occurred. Thus, eighteen elements achieved a very meaningful burnup for current commercial LMFBR designs. Subsequent operation to burnups in the 10-12 at. % range resulted in nine cladding breaches. The ten unfailed elements (see Table 463-III), as well as two failed elements (U131 and U132), have been decapsulated, and the nondestructive examinations have been completed. Five elements (U130, U131, U132,

U138A, and U142) have been sectioned, and appropriate samples from each were submitted for metallographic examination. Elements U133 and U134 have been set aside for a proposed off-normal TREAT test after completion of their nondestructive examination sequences.

Gross gamma scans were performed on all elements in this series. These scans tended to show minor abnormalities, and, in particular, tendencies toward higher gamma activity in the lower portions of the fuel stack of the helium bonded elements. Multispectral gamma scans were performed in the capsule sodium region for those elements which had shown evidence of element cladding failures at HFEF. In each of these nine cases, both ^{134}Cs and ^{137}Cs were detected in the capsule sodium at LASL, and the ^{137}Cs gamma activity levels were comparable to those found at HFEF. Conversely, no unusual gamma activity was found in the capsule sodium or capsule plenum regions for the ten unfailed elements.

Multispectral gamma scans were also taken over the fueled lengths of U129, U130, U133, U134, U141, U144, and U145. The scans were made on U133 and U134 for a better characterization and a later comparison of possible fission product movement during the proposed off-normal TREAT transient tests. The other gamma scans were taken on representative elements for a study of possible variations in behavior due to fuel density or other design differences. The fission product distributions were normal with the exception of cesium, which showed abnormal distributions over the fuel stacks in some elements. This study is not yet complete.

The profilometry measurements for the elements in this series are shown in Table 463-XII. The available fission-gas-release values are presented in Table 463-XIII. The metallographic examination of samples from this series has not reached a stage where definitive behavioral interpretations can be made.

d. Series U1930 and U1960

Four elements were processed from these series, but they have not completed their examination

sequences. The failure points were described in Section II, B.1.d for these four elements: U196, U201, U207, and U209. Element U196 is in the final stages of metallographic examination and some preliminary interpretations are possible. This element exhibited an uncharacteristic linear increase in gross gamma activity from the top of the fuel to approximately 60% of the way down the fuel stack; the activity decreased almost linearly over the remainder of the fuel. A similar distribution was noted in the ^{95}Zr - ^{95}Nb spectral scan. Activity spikes for ^{134}Cs and ^{137}Cs were located at approximately the same position as the maxima in the other activity curves. These maxima and spikes did not correlate with the failure point (a small longitudinal crack) which was located about 75 mm farther down the fuel stack. As-polished metallographic samples from U196 (sodium-bonded carbide fuel containing 15 vol% sesquicarbide) have shown evidence of a marked carburization zone in the solution-annealed Type 304 stainless steel cladding.

e. Series U5100

Two elements in this series have received extensive examinations during the year. Element U246 had four fuel pellets which were displaced above their normal positions. Element U248 had a failure point in the cladding in the upper portion of the fuel stack as described in Section II, B.1d.

The uppermost displaced fuel pellet in U246 was located approximately 131 mm above its normal location (there was no axial fuel restraint device). This pellet was submitted for metallographic and electron microprobe studies. The second displaced pellet, which was approximately 38 mm above its normal position, was submitted for burnup analysis and a samarium isotopic analysis. Additional burnup samples were taken near the top of the fuel stack (remaining undisplaced pellets) and near the center of the fuel stack. These studies, plus multispectral gamma-scan studies, were made in an attempt to deduce when the displacement event might have occurred. The evidence was inconclusive but suggested that the event probably occurred late in the irradiation period.

The metallographic sections from U246 indicated a large amount of fuel swelling with only small gaps remaining between the fuel and cladding. Since the bonding sodium had apparently been displaced upwards out of the bond region during some portion of the operation of the element resulting in subsequent element operation as a gas bonded element, high fuel temperatures with resultant large amounts of swelling were not unexpected. The metallographic sample which contained the uppermost displaced fuel pellet showed some possibly metallic material between grains near the fuel periphery and another phase scattered throughout the fuel. A final report on a microprobe study of these phases has not been issued.

The multispectral gamma scan on element U248 indicated a depletion of ^{137}Cs at and above the suspect crack area. This area had been determined by an internal pressurization test, and was confirmed visually, by eddy current tests, and by ultrasonic examinations. During sectioning of this element, no sodium was observed in the fueled region, and the fuel pellets were difficult to maintain in their normal positions. This gave rise to the speculation that there may have been an additional failure in or near the lower element weld. The lower weld was subjected to a 60-hour liquid dye penetrant test; the result was negative. The loss of bond sodium below the failure point, which was near the top of the fuel stack, has not been satisfactorily explained.

The failure point region from U248 was isolated in a 13-mm-long specimen. A sequential grinding and polishing operation with corresponding metallographic examination of each step was performed from approximately 6 mm above the external crack to about the same distance below the crack. The sequential grinding steps were made at intervals of 0.63 mm (0.025 in.) or less. The major crack, along with several minor ones, commenced on the inside of the cladding; i. e., the length of the major crack extended for some axial distance both above and below the length of the external crack. Etching at various steps in the grinding operation revealed that the major fracture

was predominantly transgranular. A normal gap existed between the fuel and cladding at all grinding levels. There was no evidence of any interaction between the fuel and cladding at any level.

A second metallographic sample from U248, which was taken from the region above the failure point, showed apparently 100% dense grain regions (no fission-gas bubbles) near the center of the fuel. However, large fission-gas bubbles were observed in the grain boundaries. A second-phase material was also found between grains in the same region. An electron microprobe examination, including the taking of one-dimensional distributions with a low-current, fine-diameter electron beam to give increased spatial resolution, did not reveal any significant differences in composition between the dense fuel grains and the remainder of the fuel. The grain-boundary phase was found to be slightly lower in uranium content and slightly higher in plutonium content than the original (U, Pu)C fuel; this change was accompanied by the presence of the noble fission products, Ru, Rh, and Tc, in the grain-boundary phase. The fuel-cladding gap was about one-half filled. The unusual microstructure and fuel swelling are indicative of high operating temperatures for at least some portion of the approximately 30 hours of operation after initial cladding failure, i. e., the fuel was partially or totally unbonded for a short period of time.

f. Series WF

The distinctive feature concerning element W6F, which was the only experiment examined from this series during FY 1975, was the transverse cladding crack which completely encircled the element. An SEM fractographic study of the crack surface has not been completed. The normal metallographic samples did not show any unusual effects.

g. Series B-1, B-2, and B-3.

Elements B-2-3, B-3-3 (both sodium-bonded nitrides), and B-3-7 (helium-bonded nitride) were examined during the fiscal year. Element B-2-3 was unfailed at 8.23 at. % burnup, and element B-3-3 was failed at 2.77 at. % burnup. The metallographic samples from both sodium-bonded nitride elements

were characterized by the presence of small cracks on the internal cladding surface. The metallographic samples from the helium-bonded nitride element, B-3-7, were free of cladding cracks in the examined sections. A metallographic sample that included the top UN insulator pellet and the uppermost fuel pellets was taken from element B-3-7. A second phase was found along the bottom surface and along crack surfaces in the UN insulator pellet. Uranium was the only element detected in this phase by electron microprobe analysis. Nitrogen was not analyzed because of the high radioactivity background of the specimen. Point counts across unreacted portions of the UN pellet into the unknown phase showed a 2-3% decrease in the amount of uranium in the unknown phase. From these observations, it was concluded that the phase is probably U_2N_3 . Because of the relatively small amount of U_2N_3 formed, it was speculated that a small amount of $(U, Pu)_2N_3$ in the $(U, Pu)N$ fuel may have provided the source of nitrogen for the U_2N_3 formation, or, most probably, that gaseous nitrogen was generated as burnup accumulated. The Type 316 stainless steel cladding was not attacked in these areas. A small metallic inclusion in the uppermost fuel pellet was composed mainly of the fission products, Pd, Mo, Ru, Rh, and Tc. A transverse microprobe sample from another region of element B-3-7 showed rings of fission products at mid-radius in the fuel. The rings are commonly found in carbide-fueled, helium-bonded elements that have accumulated significant burnup. Apparently, they are also typical of similar nitride-fueled elements.

C. Discussion of Postirradiation Results

1. Effect of Design Parameters on the Performance of Helium-Bonded Carbide Fuel Elements

The effect of selected design parameters on the failure rates of all helium bonded fuel elements containing pelletized (U, Pu) carbide fuels that have been irradiated in EBR-II was studied. This study was based on a Weibull analysis¹ of 66 helium-bonded carbide fuel elements. Some uncertainties in the results of this analysis were introduced because all the helium bonded experiments tested thus far were

encapsulated; therefore, the exact burnup levels at which failures occurred are not known. In order to at least partially compensate for these uncertainties in the burnup level at which failures occurred, the lines in the Weibull plots were drawn through or to the left (lower burnup values) of the data points. In the case of the high-burnup U-1950 series elements irradiated in subassembly X055B, the failures were distributed evenly throughout the irradiation period between their last interim examination and their final removal from EBR-II.

In the various comparisons that follow, a confidence level was calculated at certain burnup levels (usually 10 at. %) to express the significance of the differences obtained during the comparisons. This confidence level was calculated with the assumption that the lines drawn on the Weibull probability paper were a true representation of the burnup levels at failure.

a. Fuel Density

Postirradiation examination of the fuel microstructures revealed a significant difference between fuel elements containing high-density (> 95% of theoretical) fuel and those containing low-density (77-91% of theoretical) fuel. Because of these observed microstructural differences, the failure ratios of low- and high-density-fueled elements were grouped separately. The results of the Weibull analysis are shown in Figure 463-1.

Comparison of the failure probabilities of all elements containing high- and low-density fuel resulted in a > 99% confidence that more high-density-fueled elements would fail at 10 at. % burnup than those containing low-density fuel. The higher failure frequency of the high-density-fueled elements is believed to be related to their relatively inconsistent microstructural behavioral patterns and a limited ability of this dense fuel to swell into space provided by the original fuel-cladding gap.

b. Cladding Type

Most of the fuel elements were clad in solution-annealed Type 316 stainless steel or in Incoloy 800. Comparison of the performance of fuel elements which were identical except for cladding alloy showed

Fig. 463-1. Percent Failure vs. Peak Burnup for Helium-bonded Carbide Elements; Effect of Fuel density.

no indication that either Incoloy 800 or Type 316 stainless steel was less likely to fail than the other under almost identical conditions.

c. Cladding Thickness

There were no fuel elements tested which had cladding thicknesses between 0.45 mm and 0.50 mm, so the elements were divided into two groups according to cladding thickness: < 0.45 mm and > 0.50 mm. For elements containing low-density fuel, the results of the Weibull analyses comparing the two cladding thicknesses groups are shown in Figure 463-2. At 10 at. % burnup, there was a > 99% confidence that more elements containing low-density fuel with < 0.45-mm-thick solution-annealed Type 316 stainless steel or Incoloy cladding would fail than similar elements clad in > 0.50-mm-thick cladding. Cladding thickness did not appear to have a significant effect on the failure rate of elements containing high-density fuel. Weibull analysis of the performance of these elements showed that elements with either of the two ranges of cladding thickness had a median failure probability of 50% at $7\frac{1}{2}$ at. % burnup.

d. Fuel-Cladding Gap

The fuel-cladding gap, besides influencing the fuel temperature, provides space into which the fuel can swell. The utilization of this space for fuel swelling is complicated by the fracturing of the fuel pellets as a result of thermal stresses at startup. Because of this, the space that was originally in the circumferential fuel-cladding gap can be transferred to space between diametral and radial fuel cracks. Thus, the usefulness of the original fuel-cladding gap as space for fuel swelling is dependent on the shifting of fuel fragments after fracture and the ability of the fuel to swell into cracks between fuel fragments.

The failure probability of elements containing low-density fuel with small (< 0.18-mm), intermediate (0.18 - 0.25 mm), and large (> 0.25-mm) gaps is shown in Figure 463-3. The failure rate of low-density-fueled elements with small gaps was significantly higher than those with intermediate or large gaps. This trend continued as a lower failure rate was found for low-density-fueled elements with large gaps than for

Fig. 463-2. Percent Failure vs. Peak Burnup for Helium-bonded, Low-density-fueled Carbide Elements in Solution-annealed Cladding; Effect of Cladding Thickness.

Fig. 463-3. Percent Failure vs. Peak Burnup for Helium-bonded, Low-density-fueled Carbide Elements; Effect of Diametral Gap Size.

those with intermediate gaps. Confidence that a greater number of failures would occur in elements with an intermediate gap than in those with a large gap was > 90% at 8 at. % burnup and > 80% at 10 at. % burnup.

In order to include the influence of cladding thickness with the fuel-cladding gap, all elements containing low-density fuel were grouped according to cladding thickness (< 0.45 mm thick and > 0.50 mm thick) and gap size. Fuel elements having very small gaps were eliminated from the analysis and two diametral gap sizes were compared: 0.10 - 0.25 mm, and > 0.25 mm. Since there was only one fuel element (unfailed) that had < 0.45-mm-thick cladding and a > 0.25-mm gap, this element was included with the other elements having a > 0.25-mm gap. This comparison is shown in Figure 463-4. The previous conclusion that stronger claddings have a significant effect on failure rates of low-density-fueled elements was confirmed when elements having the same gap size range (0.10 - 0.25 mm) were compared. Confidence that a greater

Fig. 463-4. Percent Failure vs. Peak Burnup for Helium-bonded, Low-density-fueled Carbide Elements; Effect of Cladding Thickness and Diametral Gap Size.

number of failures would occur in elements with this gap size in < 0.45-mm solution-annealed Type 316 stainless steel or Incoloy 800 cladding than in similar cladding > 0.50 mm thick was 99% at 10 at. %. The effect of > 0.25-mm gap sizes on the failure probability of low-density-fueled elements with > 0.50-mm cladding compared to those having 0.10 - 0.25 mm gap sizes was less marked; there was only a 65% confidence that this gap difference would result in fewer failures at 10 at. % burnup.

In elements containing high-density-fuel, the effect of the gap size on failure rate was more pronounced than in those containing low-density fuel. The failure probabilities of high-density-fueled elements according to gap size are shown in Figure 463-5. There was a > 99% confidence that a greater number of failures would occur in high-density-fueled elements with an original diametral gap < 0.25 mm than in those with an original diametral gap > 0.25 mm at burnups up to 10 at. %.

Fig. 463-5. Percent Failure vs. Peak Burnup for Helium-bonded, High-density-fueled Carbide Elements; Effect of Diametral Gap Size.

2. Fuel Restructuring in Helium-Bonded, Low-Density Carbide Fuel Elements

The major restructuring feature in the microstructure of irradiated low-density (< 91% theoretical density) fuel of helium-bonded carbide fuel elements appears in mosaics of fuel-element cross sections as a porous central zone. This zone is characterized by the collection of fission-gas bubbles in the grain boundaries of the fuel. Another feature, sometimes seen in these mosaics, is a dense ring of fuel ~0.25 mm thick between the central porous zone and the unrestructured fuel. The average size of the porous zones and the dense rings were measured from the mosaics and are shown in Table 463-XIV. The central zone has been found to be temperature dependent as evidenced by fuel sections in which heat transfer has been decreased because of fuel chips in the fuel-cladding gap or large cracks in the fuel. When this has occurred, the central zone has been observed to be noticeably oval. The temperature and time necessary to produce the zone have not been determined. The average diameter of the

central porous zone is shown as a function of average linear power in Fig. 463-6. In addition, Fig. 463-6 indicates the presence of the dense fuel ring in the sections studied. For those fuel sections operating at average linear powers between 40 and 60 kW/m (12 - 18 kW/ft), the size of the porous zone can be seen to be sensitive to the size of the original fuel-cladding gap. The sections which had fuel-cladding gaps of 0.10 mm had the smallest porous zones, 0 - 0.37 (ave. = 0.13) of the fuel radius. In contrast, the sections which had fuel-cladding gaps of 0.18 - 0.37 mm had porous zones of 0.34 - 0.88 (ave. = 0.61) of the fuel radius. This difference is a reflection of the lower fuel temperatures produced by the use of a small initial bond gap. All the fuel sections studied operating at 60 - 90 kW/m (18 - 27 kW/ft) had original fuel-cladding gaps of 0.18 - 0.37 mm. For these sections, the size of the porous zone did not appear to be significantly affected by either power level or original gap size.

The existence of the dense fuel ring adjacent to the central porous zone appeared to be related to a specific power range. All but one fuel section in which there was some evidence of a dense ring operated at 60 - 70 kW/m. The sections which operated above 70 kW/m showed no evidence of the dense ring but this absence may be due to the lower burnup that these elements attained. The cause of the fuel densification at this location is as yet unexplained.

Fig. 463-6. Size of Central Porous Zone of Carbide Fuel Elements. Containing 77-91% Dense Fuel vs. Average Linear Power.

TABLE 463-XIV

SIZE OF CENTRAL POROUS ZONES AND INTERMEDIATE DENSE RINGS IN FUEL
SECTIONS OF HELIUM-BONDED, LOW-DENSITY, CARBIDE FUEL ELEMENTS

Element No.	Section No.	Location Above Centerline (mm)	Average Linear Power (kW/m)	Burnup (at. %)	Final Fuel Diam (mm)	Ave Diam of Porous Zone (mm)	Description of Dense Fuel Ring	Thickness of Dense Fuel Ring (mm)
U-92	C	171	49	5.2	6.86	3.84	None	-
"	H	-13	67	7.1	"	5.35	Partial	0.23
U-93	B	159	43	7.4	6.20	0	None	-
"	F	0	56	9.6	"	1.82	"	-
"	H	-121	48	8.4	"	0	"	-
U-94	B	150	53	7.4	7.00	(1/3)2.52 (2/3)2.28	None	-
"	D	61	64	9.0	"	4.54	Full	0.27
"	G	10	67	9.4	"	4.34	"	0.24
U-96	C	175	48	5.1	6.86	5.07	None	-
"	H	0	66	7.0	"	4.87	Full	0.27
U-97	B	68	48	9.1	6.20	0	None	-
"	H	-106	45	8.5	"	2.32	"	-
U-99	C	163	47	5.3	6.86	4.54*	None	-
"	H	-13	62	7.0	"	5.69*	"	-
U-104	C	161	45	5.5	6.86	3.70*	None	-
"	H	-19	60	7.3	"	5.00	Partial	0.20
U-105	B	118	42	8.5	6.20	3.75	None	-
"	E	-4	49	9.9	"	5.44	Partial	0.24
"	H	-95	45	9.1	"	3.50	None	-
U-187	C	121	64	3.9	6.60	4.45*	Partial	0.14
"	E	44	74	4.5	"	4.40	None	-
"	H	0	76	4.6	"	4.58	"	-
U-189	C	146	62	3.8	6.60	4.14	None	-
"	F	-6	79	4.8	"	4.62	"	-
"	J	-95	72	4.4	"	4.50	"	-
U-200	C	101	72	4.2	6.60	4.82	None	-
"	G	-32	80	4.7	"	4.92	"	-
U-206	E	70	83	4.7	6.60	4.76	None	-
"	J	-44	87	4.9	"	5.14	"	-

* Noticeably Oval.

3. Cladding Deformation in Helium-Bonded, Low-Density, Carbide Fuel Elements

A study was made of the diametral changes of all EBR-II helium-bonded carbide fuel elements which contained 77-91% theoretical density fuel pellets. Only those fuel elements which (1) were unfailed or (2) were determined to be failed but had failure regions so small that their locations could not be determined were included in the study. Axial plots of the average diameter of each fuel element were determined from four

profilometry traces taken at 45° intervals. The maximum average diametral increase from these plots was then divided by the average diameter in the upper plenum region of each of the fuel elements (x100) to determine the maximum average % $\Delta D/D$ values. These maximum average % $\Delta D/D$ values, along with other experimental parameters of the fuel elements are shown in Table 463-XV. The maximum average % $\Delta D/D$ values are plotted as a function of peak burnup in Fig. 463-7. This

TABLE 463-XV

**MAXIMUM CLADDING DEFORMATION OF HELIUM-BONDED CARBIDE
ELEMENTS CONTAINING FUEL OF 77-91% OF THEORETICAL**

No.	Fuel Density (%TD)	Cladding Type	Cladding Thickness (mm)	Original Diam. Gap (mm)	Smear Density (%TD)	Peak Linear Power (kW/m)	Peak Burnup (at. %)	Maximum Ave $\Delta D/D$ (%)
U187	86	316SS-SA	0.56	0.18	81	77	4.6	0.36
U200	86	304SS-SA	0.36	0.23	80	83	4.7	0.31
U189	85	INC800-SA	0.53	0.25	79	81	4.8	0.36
U206	90	316SS-SA	0.43	0.25	83	90	5.0	0.48
U99	85	INC800-SA	0.43	0.37	76	62	7.0	0.82
U96	85	INC800-SA	0.43	0.18	81	66	7.0	1.08
U92	85	316SS-SA	0.43	0.20	81	67	7.1	1.54
U104	77	INC800-SA	0.38	0.20	72	60	7.3	0.52
U94	85	316SS-SA	0.43	0.18	81	70	9.4	2.10
U93	86	316SS-SA	0.78	0.10	83	59	9.6	1.69
U97	86	INC800-SA	0.75	0.10	83	55	9.6	1.77
U98	86	INC800-SA	0.38	0.18	82	65	9.6	2.59
U105	77	INC800-SA	0.76	0.20	72	52	9.9	0.97
U106	77	INC800-SA	0.43	0.20	72	59	9.9	1.08

figure shows that the maximum deformation of the claddings correlates well with the fuel element smear density. Elements with smear densities of 72-80% theoretical had approximately half the maximum % $\Delta D/D$ at 10 at. % burnup as those with smear densities of 81-84%. It should be noted that the values of maximum % $\Delta D/D$ include the diameter increases due to irradiation-induced swelling. There was no indication that either solution-annealed Type 316 stainless steel or Incoloy 800 was more effective in decreasing the maximum % $\Delta D/D$ of these helium-bonded carbide fuel elements. However, there was some indication, especially for the fuel elements having smear densities of 81-84%, that increased cladding strength (as indicated by increased cladding thickness) was effective in reducing cladding deformation.

This change in the magnitude of cladding deformation at a smear density of approximately 81% of theoretical corresponds to a previously reported² change in fission-gas release at approximately the same smear density. Figure 463-8 shows the fission gas release as a function of burnup for helium-bonded carbide elements irradiated in EBR-II.

4. Cladding Ovalities in Advanced Fuel Elements

Cladding ovalities, or localized oval regions, have been observed in the cladding of many sodium and

helium bonded advanced LMFBR fuel elements during postirradiation examination. Although observations of this nature have been related to external mechanisms such as wire wrapping in oxide elements,³ no forces external to the cladding were present during the irradiation of these encapsulated elements in EBR-II. Thus, an internal fuel element mechanism, in particular a localized nonaxisymmetric fuel-cladding mechanical interaction, has been postulated as the most likely

Fig. 463-7. Maximum Cladding Deformation vs. Peak Burnup for Helium-Bonded Carbide Fuel Elements Containing 77-91% Dense Fuel.

Fig. 463-8. Fission-Gas Release vs. Peak Burnup for Helium-Bonded Carbide Fuel Elements.

cause of cladding ovalities. Figure 463-9 shows a sample plot of cladding diameter profile measurements. These data were taken at 0° and 90° orientations from the sodium-bonded carbide element W4F. A series of cladding ovalities can be seen along the length of the element. The largest ovality, as measured by the diameter difference, is 0.085 mm located at 270 mm above the bottom reference.

Sodium bonded elements are designed with a large enough gap between the fuel and cladding to eliminate any mechanical interaction if the fuel pellets remained intact during irradiation. However, thermal stresses from initial startup are probably sufficient to crack most pellets, and continued cracking occurs during irradiation because of thermal cycling. Broken fuel pieces can rearrange to contact the cladding or to wedge between broken fuel pieces. Fuel swelling or differential thermal expansion between the fuel and cladding can then deform the cladding, causing an ovality. Thus, the predominant fuel-cladding mechanical interaction in sodium bonded elements is expected to be a nonaxisymmetric one which leads to ovalities.

Postirradiation cladding profile measurements are made at room temperature, but the state of the element under operating conditions is of more

interest. The sizes of ovalities in sodium-bonded elements at operating conditions in EBR-II were calculated from the sizes at room temperature by considering the differential thermal expansion of the fuel and cladding. The calculations assumed no fuel rearrangement during the change in conditions and that all deformations were elastic. Figure 463-10 shows the relationship between the ovality sizes at room temperature and at operating conditions for three linear powers. The range at each linear power results from considering two limiting cases for fuel pellet expansion--as a solid pellet and as a radially cracked pellet. At low linear powers, the ovality size at operating conditions is less than the room-temperature size, but, at high linear powers, the ovality size at operating conditions can be greater than the size measured. Another calculation was performed to relate an ovality size at 400°C (approximate isothermal element temperature during EBR-II shutdown) to the size at operating conditions. Figure 463-11 shows the results. Ovalities are larger at operating conditions than at 400°C for all linear powers of interest. Thus, if fuel cracking or rearrangement and the resulting fuel-cladding contact occurs during reactor shutdown, an ovality will result during the subsequent startup. For helium bonded elements, the higher fuel temperatures, because of poorer heat transfer across the

Fig. 463-9. Sodium-Bonded Carbide Element W4F Cladding Profiles at 0° and 90° .

Fig. 463-10. Relationship Between Ovality Size at Room Temperature and Operating Conditions for Sodium Bonded Elements.

fuel-cladding gap, result in an ovality size at operating conditions that will be larger than shown in Figure 463-10. Specific calculations were not performed because of the uncertainty in the fuel-cladding heat transfer coefficient.

Numerous attempts have been made to observe the fuel-cladding mechanical interaction associated with ovalities in sodium bonded elements during post-irradiation metallography. Fuel chips have been seen in the gap and in fuel cracks, but no clear example of fuel wedged against the cladding has been found.² This is probably due to a rearrangement of the fuel pieces during the sectioning operation.

Ovality deformations and stresses were calculated using a linear elastic model which treats the cladding as an axisymmetric cylindrical thin shell and simulates the mechanical interaction by impressing a localized force on the inside surface of the cladding.⁴ The force was confined to small areas of the cladding as might be expected from contact with pieces of fuel. The calculated ovality shapes compared well with measured shapes in the axial and circumferential directions. (See Fig. 463-12). Cladding membrane stresses

calculated were not large nor were they sensitive to the force distribution. Bending stresses above the yield strength of annealed Type 316 stainless steel cladding were calculated. In some cases, tensile and compressive bending stresses greater than twice the yield strength were calculated. This could lead to plastic cycling or ratcheting with each thermal cycle, and may indicate that a low-cycle fatigue mechanism contributes to sodium-bonded element failure.⁵

An attempt has been made to eliminate fuel-cladding mechanical interaction in sodium bonded elements by the use of a fuel-fragment-restraint mechanism (shroud tubes).² Neutron radiographs taken during an interim examination of elements indicate that they were successful in containing fuel fragments after 3 at. % burnup in EBR-II.

In helium bonded elements, a small initial diametral gap and higher fuel swelling rates lead to an expected fuel-cladding mechanical interaction during irradiation. This interaction will probably have axisymmetric and nonaxisymmetric components. An axisymmetric mechanical interaction can be inferred from the average diametral strains of helium bonded element

Fig. 463-11. Relationship Between Ovality Size at 400°C and Operating Conditions for Sodium Bonded Elements.

Fig. 463-12. Axial Shape of an Isolated Ovality; Measured Data from Element U200.

cladding, and from an examination of postirradiation metallography which has often shown fuel-cladding contact, generally quite uniform around the circumference.² But the presence of ovalities in helium bonded elements indicates that there are nonaxisymmetric contributions to the interaction also. The relative importance of axisymmetric and nonaxisymmetric fuel-cladding mechanical interaction in helium-bonded elements is uncertain at this time.

5. Modeling

The advanced fuel element modeling code UNCLE was placed on disk storage at the LASL computer center and was run on a CDC-7600 machine. The results compared favorably with those generated by ANL.

A data package was prepared which described dimensional parameters, estimated operating conditions, and postirradiation information on 13 helium-bonded, mixed-carbide fuel elements. These data were sent to each of the advanced fuel contractors to aid in their modeling efforts.

The computer code SIGEBR, developed by W-ARD to calculate power, burnup, and fluence values for fuel elements irradiated in EBR-II, was implemented on the LASL computing system. The results of two sample cases run at LASL were essentially identical to those provided by Westinghouse.

The LASL-generated computer code PINTEMP, used for fuel element design calculations, was updated, incorporating more recent gaseous fission product production data. An editing routine was added to select maximum fuel and inner and outer cladding surface temperatures as well as maximum cladding stress.

Calculations have been performed to determine the best choice of ²³⁵U enrichment in the fuel to be used for Phase I and II irradiations. It is desirable to produce a minimum number of isotopic combinations, but to have maximum flexibility and choice of reactor locations. Based on typical LASL fuel parameters and plutonium isotopic distribution, enrichments of 25 and 60 wt% ²³⁵U were found to best meet these qualifications for EBR-II rows 4 and 5.

III. OFF-NORMAL IRRADIATION TESTING AND SAFETY ANALYSES

(N. S. DeMuth, J. F. Kerrisk, and J. H. Smith)

A. Series UL Transient Tests

The UL series tests were a joint effort of Los Alamos Scientific Laboratory (LASL) and Gulf United Nuclear Fuels Corporation (GUNFC) to determine the effect of irradiation on the failure threshold of helium bonded and sodium bonded elements with (U, Pu)C fuel. Table 463-XVI describes the four elements tested. The elements are similar to advanced LMFBR fuel elements currently under steady-state irradiation, except that the peak linear power of the two irradiated elements (tests UL-3 and UL-4) during their steady-state irradiation was only about half that of the present range of interest (80-85 kW/m). Also, the fuel density of the helium bonded elements is higher than is currently considered viable for high burnup performance. The tests were performed in the TREAT reactor. Each test was a single-element test in which the element was contained in a static capsule. No thermal neutron filter was used. The instrumentation for each test consisted of thermocouples located outside the element cladding at the top, axial midplane, and bottom of the fuel column. The thermocouples had ungrounded junctions and thus relatively slow response times.

TABLE 463-XVI
SERIES UL TEST ELEMENTS

	UL-1	UL-2	UL-3	UL-4
Fuel Element No. ^a	263(138A)	264(146A)	265(138)	266(146)
Fuel Material ^b	-----90 vol% (U _{0.85} Pu _{0.15})C + 10 vol% (U _{0.85} Pu _{0.15}) ₂ C ₃ -----			
Fuel o.d. (mm)	6.25	6.09	6.25	6.09
Fuel Density (% Theor.)	-----98-99-----			
Smear Density (% Theor.)	90	77	90	77
Bond Material	He	Na	He	Na
Diametral Bond Thickness (mm)	0.25	0.77	0.25	0.77
Cladding Material ^c	316SS	304SS	316SS	304SS
Cladding Thickness (mm)	0.56	0.38	0.56	0.38
Cladding o.d. (mm)	-----7.62-----			
Fuel Column Length (mm)	-----349 + 3-----			
Burnup (at, %)	0	0	3.56 ^d	3.62 ^d

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

^b Uranium enriched to 60% in ²³⁵U. Fuel was in the form of pressed and sintered pellets.

^c All clad was solution annealed.

^d Irradiated in EBR-II at 36 kW/m in subassembly X055.

Table 463-XVII lists some of the test results for the series UL tests. The power transient shape for each test was nominally the same; a computer-controlled flattop transient with a rapid increase in power, a constant power hold for about 1.4 s, and a rapid drop in power. (See Fig. 463-13). Because of the lack of a thermal neutron filter and the relatively high enrichment of the fuel, there was a severe radial power variation in the fuel. The calculated surface-to-centerline power generation ratio in the fuel was approximately 6. Besides being atypical of a fast reactor, this also complicates temperature calculations in the fuel element during the transient.

A preliminary nondestructive postirradiation examination has been performed on the capsules from tests UL-1 and UL-2. A complete postirradiation examination of the elements from tests UL-3 and UL-4 has been performed. The postirradiation data are still being evaluated so that the following discussions

should be considered as preliminary.

1. Test UL-1

Test UL-1 was designed to produce central melting of the fuel. Temperature calculations indicate

Fig. 463-13. TREAT Reactor Power as a Function of Time for Test UL-1.

TABLE 463-XVII
SERIES UL TEST RESULTS;
TRANSIENT OVERPOWER TESTS

	UL-1	UL-2	UL-3	UL-4
Reactor Energy (MWs)	159	161.5	157	156
Peak Reactor Power (MW)	106	104	101.5	101
Fission Energy (J/g fuel)	912	967	866	891
Peak Fission Power (kW/m)	239	233	221	217
Initial Element Temp. (°C)	260	260	260	260
Peak Fuel Temp. (°C)	2300	1500	1700-2200	1400
Peak Clad Temp. (°C)	850	1000	800-900	950

that the fuel just reached the solidus temperature. Fig. 463-14 shows a plot of the thermocouple temperatures as a function of time during the transient. The only postirradiation examination performed as yet on this element was radiography while still in its inner capsule. Some fuel pellet cracking can be seen as a result of the transient, but no other significant differences were observed. It is assumed that this element is not failed.

2. Test UL-2

Test UL-2 was designed to produce boiling and ejection of the sodium bond, followed by fuel melting. An examination of the thermocouple output from this test (see Fig. 463-15) gave no indication of bond boiling or ejection. Although the response of the ungrounded-junction thermocouples located outside the cladding was relatively slow, the reduction in heat transfer associated with bond ejection should have been observable. The calculated peak bond temperature was 1040 °C, which is about 100 °C above the boiling temperature of the sodium bond in the element. It is currently thought that bond ejection did not occur, and that if any bond boiling occurred it was confined to a very short time during the peak temperatures in the bond region. This was essentially at the end of the transient. Since bond ejection did not occur, the peak fuel temperatures were much lower than anticipated. The peak fuel temperature shown in

Fig. 463-14. Inner Thermocouple Temperatures as a Function of Time During Test UL-1. * : Near top of fuel column; + : Axial midplane of fuel; □ : Near bottom of fuel column.

Table 463-XVII was calculated assuming the sodium bond remained liquid during the transient. The only postirradiation examination performed as yet on this element was radiography while still in its inner capsule.

Fig. 463-15. Inner Thermocouple Temperatures as a Function of Time During Test UL-2. * : Near top of fuel column; + : Axial midplane of fuel; □ : Near bottom of fuel column.

Fig. 463-16. Inner Thermocouple Temperatures as a Function of Time During Test UL-3.
* : Near top of fuel column; + : Axial midplane of fuel; □ : Near bottom of fuel column.

No significant changes were seen as a result of the transient, not even fuel cracking. It is assumed that this element is not failed.

3. Test UL-3

Test UL-3 was designed to duplicate UL-1 with an irradiated element. The total energy deposition and peak linear power were 5-7% below the UL-1 values. The calculated peak fuel temperature shown in Table 463-XVII is somewhat uncertain due to the uncertainty in the fuel-cladding heat transfer coefficient after 3.56 at. % burnup. This element failed as a result of the transient. A perturbation was observed in the thermocouple temperatures starting at about 4.5 s. This can be seen by comparing Fig. 463-16 (from test UL-3) with Fig. 463-14 (from test UL-1). This perturbation probably resulted from gas blanketing of the thermocouples for a short time following failure and release of fission gas from the element. At 4.5 s the transient was essentially complete. During the postirradiation examination, fission gas was found outside the element. Three axial cracks were visible in the cladding, all located in the lower half of the fuel column. The largest crack was 60 mm long and located near the bottom of the fuel

column. Figure 463-17 shows a photograph of the bottom end of the large crack. The material in the crack is sodium or potassium oxide remaining from the cleaning operation after decapsulation. No fuel was found outside the cladding. Profilometry measurements made before and after the transient showed similar cladding ovalities with the post-transient ovalities somewhat larger. The fuel column length increased 0.6% as a result of the transient. Most of the increase came from a breakup and upward expansion of the top pellet. The calculated fission-gas release of the UL-3 fuel was 19%. It is not known what portion of this release occurred during normal steady-state irradiation and what portion was due to the transient.

Most of the postirradiation metallographic and microprobe data from the UL-3 element have been examined. One longitudinal and four transverse metallographic sections were taken from this element. The longitudinal section (section D) was taken to include the top fuel pellet, which had expanded and looked very porous in the radiographs. Figure 463-18 shows a photocopy of this section. The top half of the top fuel pellet contains many large pores. This portion of the

Fig. 463-17. Bottom Portion of the 60-mm-long Cladding Crack from the UL-3 Element. Bottom of the crack located 38 mm (1.5 in.) above the bottom of the fuel column.

Fig. 463-18. Photomosaic of Section D, Element UL-3, Mount Number 5-082.

pellet probably had a higher power generation, and thus higher fuel temperatures, than most of the fuel column because of flux peaking at the ends. Since the top pellet was relatively unrestrained, it may have released a larger fraction of its fission gas than other parts of the fuel. Flux peaking should also have occurred at the bottom pellet, but this portion of the fuel column was highly restrained. The bottom pellet did not look

porous in radiographs. The interface between the top fuel pellet and next lower fuel pellet is also visible in Fig. 463-18. Some axial expansion of the central part of the fuel column at the interface can be seen in this figure. Further analysis will be done in an attempt to determine the relative effects of temperature peaking and fuel restraint on the fuel microstructure.

Three transverse sections were taken at the three axial cracks visible in the cladding. Section M, taken near the large cladding crack and shown in Fig. 463-19 showed a distinctive cracking pattern in the fuel that appeared to be associated with the cladding failure. (See Fig. 463-17). The fuel cracking could have been caused by the propagation of a spherical or cylindrical stress relief wave through the fuel when the cladding failure occurred. The two transverse sections taken above and below the large axial crack (sections Q and H, respectively) showed a chemical interaction at the fuel-cladding interface. Figure 463-20 shows a photomosaic of the fuel-cladding interface and a crack in the fuel (section Q). The porous white phase in the fuel-cladding gap and bridged across the fuel crack contains cladding

Fig. 463-19. Photomosaic of Section M, Element UL-3, Mount Number 5-085.

Fig. 463-20. Photomosaic of the Fuel-Cladding Interface Area in Section Q, Element UL-3, Mount Number 5-086.

components (Fe, Cr and Ni) along with some U and Pu. The U and Pu concentrations in the porous white phase are lower than in the (U,Pu)C fuel. It is thought that this interaction was due to a relatively rapid melting and resolidification of the inside surface of the cladding as might have occurred during the transient. This was

inferred from the low Pu concentration associated with the Fe and Ni, and the presence of Cr in the porous white phase. Where a fuel-cladding interaction has been observed after long exposure, the Pu concentration associated with the Fe and Ni is greater than that in the fuel and Cr is rarely associated with the Fe-Ni-Pu phase. The cause of the postulated inside cladding surface melting was probably the same as the cause of the perturbation of the thermocouple signal that was noted above; viz., a gas blanketing of the fuel element by the release of fission gas from the initial failure. Fission-gas release could have displaced sufficient NaK from the area around the element to temporarily cut off heat transfer at the end of the transient. Thus, the interaction seen at the fuel-cladding interface is believed to be an artifact of a test in a static capsule. If flowing sodium coolant were present outside the element, fission gas released from a cladding failure would be rapidly swept out of the coolant channel.⁶

Small fuel chips were observed in the fuel-cladding gaps in the transverse sections. Many of the fuel chips had indented the cladding, indicating a fuel-cladding mechanical interaction. Fuel chips indenting the cladding have occasionally been seen in the metallography of helium bonded elements which have undergone only steady-state irradiation, but not to the extent observed in the UL-3 element.

Fig. 463-21. Photomosaic of the Cladding Crack at Section Q, Element UL-3, Mount Number 5-086.

Fig. 463-22. Schematic of the UL-4 Capsule, Element, and Sodium Plug, and the Pretransient Temperatures in the Capsule.

Figure 463-21 shows an etched mosaic of the cladding crack at section Q. The intergranular nature of the cladding cracks can be seen in this figure.

4. Test UL-4

Test UL-4 was designed to duplicate UL-2 with an irradiated element. The total energy deposition and peak linear power were 7-8% below the UL-2 values. Thus, the peak fuel and clad temperatures were also slightly lower. This element was found to be unfailed after the transient. The fuel pellets were cracked as a result of the EBR-II irradiation. Some additional cracking and pellet movement was seen in the radio-graphs as a result of the transient, but no other significant changes were observed in the fuel region. The fuel column length increased 3% as a result of the transient. The increase was relatively uniform over the entire fuel column length. No holddown spring was used in the sodium bonded elements.

A long sodium plug (about 75 mm long and containing on the order of 80% of the sodium in the element) was found in the plenum region of the element after the transient. (See Fig. 463-22). The bottom of the sodium plug was about 65 mm above the top of the upper insulator pellet. Individual fission-gas samples were taken above and below the sodium plug. Above the sodium plug the gas was 4.6% fission gas (Kr and Xe) and 95.4% helium. Below the plug the gas was 98.6% fission gas and 1.3% helium. The calculated plenum pressure above the plug was 0.27 MPa and

below the plug was 0.26 MPa. It appears that a release of retained fission gas during the transient displaced the bond sodium up into the plenum. This phenomenon must have occurred either late in the transient or after the transient since no perturbations were seen in the thermocouple temperatures which would indicate a cutoff in heat transfer of the fuel (see Fig. 463-23). The temperature of the plenum around the sodium plug was estimated from the pretransient temperatures in the capsule. Figure 463-22 shows that the estimated cladding temperature was 280-290°C at the location of the sodium plug. Figure 463-24 shows a plot of heat-sink and heater-can temperatures recorded as a function of time after the transient. These temperatures indicate that the sodium plug was probably molten (above 97.8°C) for at least two hours after the transient.

A γ -scan of the section containing the sodium plug (sodium plus cladding) was performed. Aside from cladding components, the only γ -active isotope detected was ^{137}Cs . Peaks in ^{137}Cs activity were found at the menisci of the sodium plug; the peak at the lower meniscus was approximately twice the peak at the upper one. The sodium plug was sampled and

Fig. 463-23. Inner Thermocouple Temperatures as a Function of Time During Test UL-4. *: Near top of fuel column; +: Axial midplane of fuel; □: Near bottom of fuel column.

Fig. 463-24. Heat Sink and Heater Can Temperatures as a Function of Time after the UL-4 Transient.

analyzed for iodine. Samples were scraped from the upper and lower surfaces of the sodium and the bulk of the sodium plug was cut into four additional samples. The amount of iodine in the small surface samples was below the detection limit of the analytical technique. Iodine was detected at 45 ppm, 30 ppm, 24 ppm and 23 ppm in the four bulk samples, listed in order from the lower to the upper locations of the sodium plug. If all the iodine in the lowest bulk sample (45 ppm) were distributed on the lower surface of the sodium plug as NaI, a layer thickness of less than $0.02 \mu\text{m}$ would be obtained. Thus, it seems unlikely that a layer of solid NaI could have formed to hold the sodium plug in place while it was still molten.

The effects of surface tension were examined as a possible mechanism for stabilizing the molten sodium plug. A situation with a high-density fluid (liquid sodium) above a low-density fluid (fission gas) can be stabilized by surface tension in sufficiently small diameter tubes. Although theoretical approaches such as an examination of the Taylor instability⁷ can be applied, the most direct approach found was an examination of experimental work on the velocity of gas bubbles rising in a tube through a liquid.⁸⁻¹⁰ It was found that under certain conditions the gas bubbles would not rise, i.e., a stable condition with a high-density fluid above a low-density fluid could exist.

Bubbles would not rise in a vertical tube if

$$\frac{\Delta\rho g d^2}{4\sigma} < 0.85$$

where $\Delta\rho$ is the density of the liquid minus the density of gas, d is the tube diameter, σ is the surface tension of the liquid, and g is gravitational acceleration. Using $\sigma = 0.16 \text{ N/m}$, $\Delta\rho = 0.85 \text{ Mg/m}^3$, and $g = 9.80 \text{ m/sec}^2$, a liquid sodium plug would be stable if $d < 8 \text{ mm}$. The i.d. of the UL-4 cladding is 6.9 mm, indicating that the effects of surface tension alone could have stabilized the liquid sodium plug.

The fission gas contained in the region above the sodium plug represents a release of 0.4% of the total fission gas generated while the fission gas below the sodium plug represents a release of 10%. Based on the mechanism described above, the 0.4% release corresponds to what occurred during steady-state irradiation while the 10% release corresponds to fission gas released during the transient. About 90% of the fission gas generated during the steady-state irradiation was still contained in the fuel.

Fig. 463-25. Photomosaic of Section L, Element UL-4, Mount Number 5-089.

The postirradiation metallographic data from the UL-4 element were examined. Three transverse metallographic sections were taken from the element at 53 mm above the core midplane (E), 51 mm below the core midplane (J), and 83 mm below the core midplane (L). The sections were similar in many respects to sections from other sodium bonded elements that have been examined after only steady-state irradiation. Figure 463-25 shows the mosaic made from section L. Fuel pellets were cracked. No signs of fuel or cladding melting were seen. The α -autoradiographs and β - γ autoradiographs indicated no redistribution of fissile material or of fission products. Grain-boundary swelling was observed in two of the sections (J and L) near the pellet edges. This swelling could be related to the effects of fission-gas bubbles in the sodium bond,² but it is uncertain whether the swelling occurred during steady-state irradiation or during the transient. A preliminary conclusion of the examination is that all the features observed in the postirradiation metallography could be accounted for by the steady-state irradiation. This conclusion must remain preliminary until other elements have been examined, in particular, similar elements from X055B which have undergone steady-state irradiation but no transient, and the element from transient test UL-2.

B. Transient Temperatures for X055B during Natural Convection Test "F"

Transient thermal analyses were performed to determine the maximum cladding temperatures in subassembly X055B during the EBR-II natural convection test (Test F). At the time of this test, which was conducted near the end of Run 73, 17 of the 19 U1950 series encapsulated experiments were operating at peak linear powers up to 44 kW/m and approaching goal burnups of 12 at. %. Following removal of X055B from EBR-II, gamma scanning indicated that cladding failures had occurred in 8 of 16 high-burnup experiments since their last interim examination at about 8 at. % burnup. Several of the cladding failures were classified as slight because only minute amounts of fission gas or ¹³⁷Cs were detected in the element capsules. The objective

of this analysis was to determine whether the cladding temperatures attained in Test F were sufficiently high to contribute to the observed defects.

In Test F, power-to-flow ratios greater than unity occurred only during the time interval (approximately two minutes) for which the auxiliary pumps were turned off. After the EBR-II control rods were scrammed to initiate Test F, the sequence of operations¹¹ was as follows:

- (a) the primary coolant pumps were tripped five minutes after the scram;
- (b) the auxiliary pump coastdown began twenty minutes after the scram;
- (c) primary pumps turned on to 25% of full flow about two minutes after auxiliary pump shutdown.

At the time that the auxiliary pumps were tripped, the energy generated by fission-product decay was about 1.4% of the fission energy at full power. During coastdown of the auxiliary pumps, a transition from forced- to natural-convection flow was observed in experimental subassembly XX07. The natural-convection flow rate in this subassembly appeared to equilibrate after about 80 seconds at a value about one-fourth of that with the auxiliary pumps operating. Primary pumps were restarted when some subassembly outlet thermocouples indicated temperatures greater than those for normal full-power operation.

Two-dimensional transient heat-transfer calculations were performed with the CINDA¹² code for the time interval that the auxiliary pumps were off. In these calculations, one encapsulated U1950 series fuel element with its associated annular flow channel and shroud was considered. Both radial conduction between elements and axial conduction from the core region were neglected. The steady-state temperature distribution in a fuel element was computed from the decay-heat generation and auxiliary-pump flow rate. Transient calculations were then initiated by allowing the flow rate to coast down in proportion to that measured in XX07.

To determine the effect of natural convection flow on the maximum cladding temperatures for the elements in X055B, two situations were postulated. In the first,

the flow rate in X055B assumed to be proportional (in the ratio of full-power flow rates) to that measured in XX07. With this assumption, the maximum cladding temperatures occurred near the top of the fuel stack and were about 30K below the temperature range for full-power, full-flow operation. The coolant flow and inner cladding surface temperature are shown by the solid lines in Fig. 463-26. Activation of the primary-coolant pumps after ~124 seconds resulted in a very rapid increase in flow with a corresponding decrease in cladding temperature.

In the second situation, calculations were performed to determine the cladding temperature that would occur in the absence of natural-convection-induced coolant flow. In these calculations, the sodium flow was allowed to stagnate following coast-down of the auxiliary pumps. The flow rate and resulting cladding temperatures are shown by the dashed lines in Fig. 463-26. The maximum cladding temperature, which occurred near the midplane of the fuel stack just prior to termination of the test, was calculated to be about 30K above the normal operating range. In this analysis, many factors which would tend to reduce cladding temperatures were neglected, while factors such as flow stagnation, which would tend to increase the cladding temperatures, were included. Thus, the maximum calculated temperature is a limiting value and is not necessarily representative of what occurred during Test F.

The results of this analysis indicated that peak cladding temperatures during Test F could have reached the levels expected for normal operation. Although slightly higher values were obtained for flow stagnation, the general conservatism in these calculations led to the conclusion that these values are higher than those which occurred in Test F.

C. The Consequences of Sodium Bond Loss from an LMFBR Carbide Fuel Element

A potential safety problem with sodium-bonded carbide fuel elements is the behavior of an element following a loss of sodium bond. Sodium bond loss can occur as a result of a cladding failure below the sodium level in the element. Plenum pressures in sodium-

Fig. 463-26. Coolant Flow Rates and Corresponding Cladding Temperatures for Auxiliary Pump Shutdown.

bonded elements are generally higher than the coolant pressure, so that bond sodium will be lost from the element. Following bond loss, the liquid sodium will be replaced by plenum gas--either helium at the beginning of life or a helium/fission-gas mixture later in life. The change from liquid sodium to plenum gas in the bond area reduces the thermal conductance of the bond by about an order of magnitude; thus, high fuel temperatures and, possibly, fuel melting will follow loss of sodium bond. An added complication with loss of sodium bond is the presence of fuel-fragment restraints (shroud tubes) in some elements. Shroud tubes with a high melting point (tantalum) and with a relatively low melting point (Type 316 stainless steel) have been used in fuel elements.

An analysis of the consequences of the loss of sodium bond from a carbide-fueled element during normal steady-state operation has been made. The complexity of the initial state of an element (for example, its burnup, fission-gas release, fuel-cracking pattern, gap size, and linear power) and the further complexity of the phenomena following bond loss preclude a realistic analysis of this accident. Thus, the analysis made was a conservative or limiting assessment of bond loss, and is not representative of what would actually occur. Many simplifying assumptions were made throughout the analysis. These assumptions were conservative in

the sense that they tended to predict worse consequences than would result from a realistic analysis. Even with the conservative assumptions made, the immediate consequences of sodium bond loss are far from catastrophic. Fuel melting will occur, but the fuel will slump and solidify on contact with the cladding.

The cause of sodium bond loss considered is a cladding defect which allows the bond sodium to be lost from the element into the coolant. The sequence of events involved is:

- (a) a cladding defect occurring below the top of the fuel column,
- (b) the loss of the sodium bond through the defect into the coolant,
- (c) gas blanketing of the fuel by plenum gas which reduces the thermal conductance of the bond,
- (d) fuel (and shroud) melting if the linear power is high enough and the gap large enough,
- (e) slumping of the molten fuel (and shroud) against the cladding which increases the thermal conductance of the bond, and
- (f) resolidification of the molten material.

Causes for concern during this sequence are possible cladding melting from contact with the molten fuel, and the possible flow of molten fuel out the cladding defect into contact with the coolant sodium where a molten fuel-coolant interaction (MFCI) can occur.

The likelihood of cladding melting from contact with the molten fuel can be estimated from calculations of the cladding surface temperature following contact with the molten fuel. Experimental evidence is also available from irradiation tests where molten fuel did contact cladding. The likelihood of a severe MFCI is mainly determined by the maximum fuel temperatures attained. If the molten fuel is hot enough to raise the sodium coolant temperature above its homogeneous nucleation temperature, then a vapor explosion is possible.

For the analysis, fuel elements with the design of the "Series U5100 Singly-Clad Carbide Experiments," EBR-II subassembly X-156, were examined (see Table 463-VII). Estimates of the important element parameters were made based on the expected irradiation history in EBR-II. These included the peak linear power

(110 kW/m), the maximum sodium coolant temperature (537°C), the maximum plenum pressure (3.8 MPa at 50% fission-gas release) and the variation of the gap size during irradiation (the gap decreases as irradiation proceeds). Heat transfer calculations were performed for three model elements:

- (a) an unshrouded element with 0.83-mm (33-mil) diametral gap between the fuel and cladding i.d.,
- (b) an element with a Type 304 stainless steel shroud and a 0.53-mm (21-mil) diametral gap between the shroud o.d. and cladding i.d., and
- (c) an element with a tantalum shroud and a 0.53-mm (21-mil) diametral gap between the shroud o.d. and cladding i.d.

These elements cover the range of design parameters in the subassembly. The gaps used were the largest initial gaps in the subassembly. Calculations were performed using initial gap sizes as a conservative assumption since initial gaps are the largest gaps during the life of the element. The element with the stainless steel shroud was also meant to cover the elements with iron and vanadium shrouds. Although the properties of vanadium are somewhat different than stainless steel, the outcome of the calculation is very insensitive to the shroud properties as long as the shroud has melted when the fuel has started to slump.

The calculations were one dimensional (radial) and assumed concentric fuel, shroud, and cladding. Axial conduction and conduction paths through any liquid sodium that might remain around the circumference of the bond were neglected. This assumption is conservative in that it leads to higher fuel temperatures than if these conduction paths were considered. Any pellet eccentricity would also reduce the maximum fuel temperatures.

The CINDA code¹² was used for the numerical heat transfer calculations which determined the maximum fuel temperatures and maximum cladding temperatures. The heat transfer calculations start at an initial steady-state temperature distribution with the sodium bond intact. At zero time, the gap sodium is completely replaced by plenum gas and a transient calculation follows the element temperatures. The plenum gas

was assumed to be 10% helium and 90% fission gas. This amounts to an end-of-life (lowest thermal conductivity) composition with a 40% fission-gas release. Both conduction and radiation were allowed across the gas regions. The fuel, shroud, and cladding radii were corrected for thermal expansion in the steady-state calculation and throughout the transient calculation.

A definition of what occurs whenever the fuel and shroud melt is very difficult. For the purpose of temperature calculations, the following model was used. The fuel was assumed to maintain its structural integrity until the fuel surface temperature was above its liquidus temperature, 2440°C . After this point, the fuel was allowed a specific time to slump out to the cladding, depending on the condition of the shroud. If the shroud had melted, 0.1 s was allowed for the molten fuel and shroud to slump to the cladding. This case covers the stainless steel, iron, and vanadium shrouds. If the shroud had not melted, 0.5 s was allowed for the molten fuel to slump through the shroud slots to the cladding. This case covers the tantalum shroud. The unshrouded elements were also allowed 0.1 s to slump to the cladding. These slumping times are considered conservative estimates of the time required for the molten fuel to move the short distances involved. They are greater than the estimates used in similar calculations of the melting of EBR-II metal fuel.¹³ In addition to the slumping model, temperature calculations were also performed assuming no slumping. These calculations were not meant to represent possible element behavior, but were performed to provide some feeling for the influence of model assumptions on the calculated temperatures.

Once the fuel or shroud has contacted the cladding, a heat-transfer coefficient must be defined between the contact surfaces. The following assumptions were used:

- (a) if two molten surfaces contacted (e.g., fuel and shroud), or if a molten surface contacted a solid surface, a contact coefficient of $28.4 \text{ kW/m}^2 \text{ }^{\circ}\text{C}$ ($5000 \text{ Btu/h ft}^2 \text{ }^{\circ}\text{F}$) was used,
- (b) if two solid surfaces contacted, a contact coefficient of $5.7 \text{ kW/m}^2 \text{ }^{\circ}\text{C}$ ($1000 \text{ Btu/h ft}^2 \text{ }^{\circ}\text{F}$) was used,

- (c) when molten fuel contacted the cladding through the still solid shroud tube, a contact coefficient of $5.7 \text{ kW/m}^2 \text{ }^{\circ}\text{C}$ ($1000 \text{ Btu/h ft}^2 \text{ }^{\circ}\text{F}$) was used to approximate the chance of reduced contact area (the shroud tubes have 25% slot area), and

- (d) once molten fuel, or shroud material, contacted the cladding, the contact was maintained.

The choice of $28.4 \text{ kW/m}^2 \text{ }^{\circ}\text{C}$ for a solid-liquid contact coefficient is conservative for calculating peak fuel temperatures. This value is near the upper limit observed for solid-solid contact¹⁴ and much less than values assumed to model molten fuel-cladding contact with EBR-II metal fuel.¹³ A low value for the conductance will restrict heat transfer and thus predict high fuel temperatures. For the calculation of peak cladding temperatures following slumping of the fuel, the conservative predictions will result from large contact coefficients. To cover this limit, the peak cladding temperatures following slumping in an unshrouded element were also calculated using a contact coefficient of $284 \text{ kW/m}^2 \text{ }^{\circ}\text{C}$ ($50,000 \text{ Btu/h ft}^2 \text{ }^{\circ}\text{F}$). The unshrouded element calculation was done since it predicts the highest peak cladding temperature following slumping. This value for the contact coefficient approaches perfect thermal contact between the molten fuel and cladding and provides upper limit estimates of the peak cladding temperatures. The choice of $5.7 \text{ kW/m}^2 \text{ }^{\circ}\text{C}$ for solid-solid contact is in the range of experimentally observed contact coefficients for solids.¹⁴ None of the calculational results is sensitive to this choice.

Calculations were performed at two core locations: the core midplane where the element power is highest, and the top of the core where the sodium coolant temperature is highest. At the core midplane, the linear power was assumed to be 125 kW/m and the sodium temperature 450°C . At the top of the core, the linear power was assumed to be 110 kW/m and the sodium temperature 540°C . The linear powers are approximately 14% above the linear power of the highest powered element. This conservative assumption was made to account for uncertainties in reactor power and fission rates in the elements. Calculations were also performed using fuel thermal conductivities of +20% and -20% of the normal, in addition to the nominal values, to

TABLE 463-XVIII

MAXIMUM FUEL TEMPERATURES OF CARBIDE FUEL ELEMENTS
DURING LOSS-OF-SODIUM-BOND TRANSIENTS

<u>Shroud</u>	<u>Fuel Slumping</u>	<u>Core Location</u> ^a	<u>Fuel Conductivity</u> ^b	<u>Maximum Fuel Temp., °C</u>
Stainless Steel	Yes	M	N	2775
	Yes	M	+20%	2724
	Yes	M	-20%	2842
	Yes	T	N	2731
	Yes	T	+20%	2689
	Yes	T	-20%	2801
	No	M	N	3163
	No	M	+20%	3095
	No	M	-20%	3264
	No	T	N	3111
	No	T	+20%	3058
	No	T	-20%	3195
None	Yes	M	N	2702
	Yes	T	N	2677
	Yes	T	+20%	2648
	Yes	T	-20%	2720
Tantalum	Yes	M	N	3013
	Yes	T	N	2950
	Yes	T	+20%	2918
	Yes	T	-20%	2996

^a M = Core Midplane, T = Top of Core.

^b N = Nominal.

assess the effect of uncertainty in this property.

Table 463-XVIII lists the maximum fuel (center-line) temperatures calculated for all the cases considered. For the cases where molten fuel slumping was allowed, the tantalum-shrouded element shows the maximum fuel temperatures. This is due to the longer slumping time set for this case and the lower contact coefficient used after molten fuel-cladding contact was established. The results for the unshrouded and stainless steel-shrouded elements where slumping was allowed are quite similar. The results for the stainless steel-shrouded element where slumping was not allowed are approximately 400°C higher than the similar cases where slumping was allowed. The small difference is mainly due to the fact that, by the time slumping starts, the fuel temperatures are beginning to peak out.

Figure 463-27 shows a plot of element temperatures as a function of time for the stainless steel-shrouded element with a nominal fuel conductivity at the top of the core, with slumping allowed. Curve 1 is the fuel center, curve 2 is the fuel surface, curve 3 is the shroud mid-radius, and curve 4 is the cladding inside surface. Shroud melting starts at about 1.1 s, fuel melting starts in the center at about 1.6 s, fuel-shroud contact occurs at 2.5 s, fuel and shroud slumping starts at 3.15 s, and contact with the cladding is established at 3.25 s. The peak fuel temperature occurs in the center at about 3.4 s.

The maximum cladding temperatures calculated were for the unshrouded element, where molten fuel contacts the cladding directly. Figure 463-28 shows a plot of cladding temperatures at the top of the core as a function of time for an unshrouded element. The nominal contact coefficient is 28,4 kW/m² °C while the

Fig. 463-27. Fuel Element Temperatures as a Function of Time following Loss of Sodium Bond with Slumping Allowed. (Curve 1 is fuel center, curve 2 is fuel surface, curve 3 is shroud midradius, and curve 4 is cladding inside surface.)

high contact coefficient is $284 \text{ kW/m}^2 \text{ } ^\circ\text{C}$. The inside surface temperature of the cladding is above the melting range for a few milliseconds after contact, with the high contact coefficient. With the nominal contact coefficient, the entire cladding is always below the melting range. It should be emphasized that the high contact coefficient is a conservative limiting case, i.e., it is essentially perfect contact between the molten fuel and cladding.

The possibility of a severe MFCI requires that molten fuel be released from the cladding defect into the coolant and that the molten fuel temperatures are high enough to raise the coolant temperatures above the homogeneous nucleation temperature of sodium (2027°C). Although there is no evidence that molten fuel will be ejected from a cladding defect following sodium bond loss, calculations of the interface temperature, T_i , following contact between molten fuel and sodium were made, assuming that fuel at the maximum temperatures attained (Table 463-XVIII) was released.¹⁵ This is a conservative assumption since the maximum fuel

temperatures occur at the fuel centerline. Temperatures at the fuel surface adjacent to the cladding failure are approximately 300°C below the fuel centerline temperatures. Figure 463-29 shows a plot of $\Delta T = 2027^\circ\text{C} - T_i$ as a function of fuel thermal conductivity variation around its nominal values. The quantity ΔT can be considered as the safety margin between the calculated sodium interface temperatures and the homogeneous nucleation temperature of sodium. All the interface temperatures calculated are below the homogeneous nucleation temperature. The highest fuel temperatures calculated were at the core midplane (see Table 463-XVIII). But the highest interface temperatures were calculated at the top of the core owing to the higher sodium coolant temperatures at this location. Thus, the smallest margin of safety (smallest ΔT) exists at the top of the core. As the fuel thermal conductivity increases, the maximum calculated fuel temperatures decrease (see Table 463-XVIII). But this decrease in fuel temperatures is more than offset by the effect of increasing thermal conductivity on the interface temperature. Thus, the interface temperatures increase (values of ΔT decrease) as the fuel thermal conductivity increases.

Even though the instantaneous interface temperature resulting from molten fuel-sodium contact is below the homogeneous nucleation temperature of sodium, it is possible that bulk heating of the sodium can raise its temperature above the boiling point (925°C at the EBR-II

Fig. 463-28. Cladding Temperatures as a Function of Time after Molten Fuel-Cladding Contact for Nominal and High Contact Coefficients.

Fig. 463-29. Safety Margin (ΔT) as a Function of Fuel Thermal Conductivity for a Variety of Conditions.

subassembly outlet). Bulk heating of the sodium will depend on the amount of molten fuel that is lost from the element and the rate of loss, as well as the molten fuel temperature. Estimates of the molten fuel loss, in turn, depend on the internal element pressure and the cladding failure size. The final equilibrium temperatures of sodium-fuel mixtures that would result from the loss of molten fuel at 3000°C into sodium coolant at 540°C were calculated based on the following assumptions:

- A moderate cladding failure size of $6 \times 10^{-4} \text{ mm}^2$, or a large cladding failure size of $6 \times 10^{-2} \text{ mm}^2$ exists in the cladding. These sizes are two to four orders of magnitude greater than the failure size estimated from pressurization tests of element U248.
- The molten fuel flow rate through the cladding defect is driven by a 7-MPa pressure differential and can be calculated using a simple orifice flow formula.¹⁶ The use of 7-MPa pressure differential assumes 100% fission-gas release from the fuel at the end of life.
- The total quantity of fuel released to the coolant is controlled by the flow rate and the time required for the cladding defect to be plugged with resolidified fuel. The time required for plugging was calculated from the defect size and the heat transfer between molten fuel and cladding in contact.
- The fuel released is confined to one sub-channel in the subassembly. This assumption ignores radial mixing. Axial mixing was also ignored.

With these assumptions, a moderate size failure would release 0.26 mg of fuel to the coolant before being plugged. The final equilibrium temperature of the sodium coolant-fuel mixture would be 542°C. A large size failure would release 0.26 g of fuel and the mixture would attain a final equilibrium temperature of 700°C. These temperatures are well below the boiling temperature of sodium at the subassembly outlet (925°C).

In summary, this work has assessed the consequences of a loss of sodium bond from a uranium-plutonium-carbide-fueled element during normal steady-state operation. The elements considered were typical of those currently being irradiated in EBR-II. The analysis included elements with and without shroud tubes. Sodium bond loss into the coolant through a clad failure was the mechanism considered.

Maximum fuel temperatures were calculated and found to be in the range of 2700°C to 3000°C when fuel slumping was accounted for. If essentially perfect contact was allowed, inside cladding surface temperatures above the melting range were calculated following contact with molten fuel. If less conservative estimates of the contact coefficient were used in the calculation, no cladding melting was predicted. Sodium coolant temperatures calculated for instantaneous contact with molten fuel and from bulk heating were found to be below the homogeneous nucleation temperature for sodium. Bulk sodium coolant temperatures were also below the boiling temperature of sodium at the exit of the subassembly.

The peak calculated cladding temperatures indicated the possibility of inside surface melting. There are two experiments which relate to this question. Gulf United Nuclear Fuels Corporation (GUNFC) performed a set of GETR irradiations which examined the consequences of sodium bond defects and loss of bond.¹⁷ Evidence of an interaction (probably due to inside cladding surface melting) to about 0.05 mm out of 0.38 mm total cladding thickness could be seen in some instances. No cladding failures occurred in this group of elements. A LASL encapsulated element in EBR-II (K-49) incurred a severe failure and showed

evidence of contact between possibly molten fuel and the capsule wall (Type 304 stainless steel).¹⁸ There was no evidence of capsule melting or interaction between the fuel and the capsule wall. The GUNFC experiments tend to substantiate the calculations performed here, i.e., inside cladding surface melting may occur from contact with molten fuel, but it is rapidly quenched as long as sodium coolant flow is maintained on the outside of the cladding.

The molten fuel-coolant interaction (MFCI) calculations indicate that the sodium temperatures resulting from instantaneous contact with molten fuel will be 300°C to 400°C below the homogeneous nucleation temperature of sodium for unshrouded elements or those shrouded in stainless steel, iron, or vanadium. The tantalum shrouded element may have a smaller margin of safety. This result indicates that small amounts of fuel ejected from a cladding failure will not cause homogeneous vaporization of the sodium coolant. Further calculations also indicate that bulk heating of the sodium coolant by molten fuel will not cause homogeneous vaporization or boiling of the sodium coolant.

Experiments performed by Atomics International have some bearing on MFCI of carbide fuel.¹⁹ These TREAT tests overpowered sodium-bonded UC fuel in stainless steel cladding which was surrounded by sodium and a heat sink. In one test, centerline fuel temperatures of 3150°C, with some indications of temperatures as high as 3800°C, were measured. The maximum pressure measured in the capsule was less than 1.3 MPa. This transient overpower completely destroyed the element. Although these test results cannot be directly translated to the calculations presented here, they showed no significant MFCI under analogous conditions.

As concluding remarks, the generally conservative nature of the analysis presented here should be reiterated. In the calculation of the maximum fuel temperatures attained following the loss of bond, the effect of using initial gap sizes, concentric fuel and cladding, complete loss of bond sodium, end-of-life (lowest thermal conductivity) plenum gas replacing the sodium, no

axial conduction, power that is 14% above nominal, long slumping time for the fuel, and no frothing of the fuel on melting are all conservative. A partial retention of the sodium bond, significant axial conduction (due to a slow loss of bond), eccentric fuel in the cladding, or shorter slumping times would all tend to reduce the maximum fuel temperatures calculated. An allowance for frothing (due to retained fission gas) of the central portion of the fuel at temperatures above its solidus temperature might even prevent melting of the outer region of the fuel. For these reasons, the maximum fuel temperatures calculated should be considered as upper limits, and not necessarily representative of what would be attained following a loss of bond.

The estimates of the amount of fuel that could be lost through a cladding failure and the loss rate are more speculative than the maximum fuel temperature calculations, but still represent a conservative calculation. Current evidence about initial cladding failures indicates that they are much smaller than the hole sizes assumed in this analysis. The estimates presented should be considered as upper limits for initial failures, where the lower limit is no fuel lost.

D. The Consequences of Sodium Bond Loss from an LMFBR Nitride Fuel Element

As part of the safety and hazards analysis for irradiation of unencapsulated ($U_{0.8}Pu_{0.2}$)N fuel elements in EBR-II, transient thermal analyses were performed to determine the temperatures that would result from a hypothetical incident wherein the sodium bond between the fuel and cladding is lost. In these analyses, both fuel melting and decomposition to liquid metal and nitrogen gas were considered as possible high-temperature transformations. The critical fuel temperatures, i.e., the temperatures required to produce a fuel-coolant interface temperature equal to the homogeneous nucleation temperature of sodium (2300 K), were computed and compared with the maximum calculated fuel temperatures to determine the margins of safety for a vapor explosion. In order to evaluate the sensitivity of these margins to the rather large uncertainties in high-temperature fuel (and liquid metal) properties, these

uncertainties were treated parametrically in both the transient heat transfer and the critical fuel temperature calculations. The transient cladding temperatures that resulted from contact with molten fuel were computed to determine whether or not melting of the cladding would occur.

In this study, which was directly related to the fuel elements in subassemblies X163 and X246, one K4 (shrouded) element and two C5 (unshrouded elements, one each with 0.38- and 0.26-mm radial bond gaps), planned for irradiation in E19-B type-subassemblies were considered. The element descriptions and operating conditions used in this assessment are summarized in Table 463-XIX. These elements were selected on the basis of their interior positions in the subassembly, high fuel densities, and high linear powers. For this study, the linear powers were increased $\sim 5\%$ over those for the requested irradiation conditions. Swelling of the fuel and cladding and the fission-gas generation and release to the plenum were estimated as functions of burnup in Ref. 20. At the end of life (12% at, % burnup), about 10% of the fission gases generated (~ 0.017 moles) are expected to be released to the plenum. For the purposes of this study, it was assumed that 20% of the gas that would be generated at the end of life would be released. This released in helium/fission-gas ratios of $\sim 1/5$ for the K4 series elements and $\sim 1/10$ for the C5 series elements.

The following section contains a discussion of the decomposition and melting behavior of the fuel which was postulated to result from loss of the sodium bond through cladding defects of various sizes. The transient thermal analysis following a loss of bond, and the methods used to estimate the potentials for molten fuel-coolant interactions (MFCI) are summarized in succeeding sections. The results of vaporization, decomposition, and melting experiments with nitride fuels together with measured fuel properties are reviewed in Ref. 20. This document also contains details of the methods used to model decomposition and melting during transient heating, to estimate high-temperature properties, and to compute fuel, cladding, and

interface temperatures.

1. Decomposition and Melting of Nitride Fuels

Nitride fuels have been found to vaporize, decompose to liquid metal and nitrogen gas, or melt, depending on the temperature and nitrogen gas overpressure. Based upon the review²¹ of decomposition and melting behavior of nitride fuels, it was concluded that the mixed-nitride fuel will be stable (negligible vaporization and decomposition) at temperatures up to 2770 K for the transient heating conditions that result from a loss-of-bond incident. At higher temperatures, liquid metal and nitrogen gas will be formed if the partial pressure of nitrogen in the element is less than the decomposition pressure at the fuel temperature. The temperatures and nitrogen overpressures for which fuel melting has been observed are as follows:

<u>Fuel</u>	<u>Temperature, K</u>	<u>Nitrogen Pressure, MPa</u>	<u>Reference</u>
UN	3120 $\pm$ 30	0.25	21
PuN	3000 $\pm$ 30	4.0	22
(U _{0.8} Pu _{0.2})N	3040 $\pm$ 30	1.0 to 1.5	22

At temperatures greater than those for which melting occurs, nitrogen overpressures equal to or greater than those listed above will suppress decomposition of liquid fuel.

In order to evaluate the fuel decomposition and melting behavior during transient heating, three types of cladding defects were postulated. The first of these is representative of a very small defect or one which becomes plugged following loss of the sodium bond. In this situation, the nitrogen produced by decomposition of less than 0.5% of the fuel is sufficient to produce a pressure of 1.5 MPa and melting of the remaining fuel can occur.

Next, a very large cladding defect, such as an axial crack running the length of the fuel stack, was postulated. Although such a defect would not be expected to occur under normal operating conditions in EBR-II, it is hypothesized as a limiting case in which nearly complete fuel decomposition is presumed to occur. In such a situation, the rate of decomposition will be

TABLE 463-XIX

COMPOSITE ELEMENT DESCRIPTIONS AND OPERATING CONDITIONS
FOR CLADDING FAILURE ANALYSIS

Element	Fuel Density % TD	Fuel Radius mm	Shroud Radius, mm		Clad Radius, mm		Bond Gap mm	Plenum Volume, cm ³	Linear Power, kW/m ²		Coolant Temp, K	
			Inner	Outer	Inner	Outer			Midplane	Top	Midplane	Top
K4-10	96.0	3.289	3.321	3.398	3.638	3.935	0.265	~14.2	93	75	740	850
C5-14	96.6	3.171	N/A ^b	N/A	3.556	3.937	0.385	~7.2	88	73	730	830
C5-20	95.6	3.292	N/A	N/A	3.556	3.937	0.264	~7.0	91	76	730	840

^aLinear powers increased ~5% over requested irradiation conditions.

^bNot applicable.

limited only by the rate at which nitrogen can be produced, diffuse, or flow to the fuel surfaces, and be released into the coolant. To estimate this rate, the nitrogen pressure at the fuel surfaces was assumed to equal the decomposition pressure at the surface temperature, and the rate of nitrogen loss from the surface was calculated using the Langmuir²³ equation with the assumptions of instant diffusion to the surfaces and an evaporation coefficient of unity.

In the third case, a defect area of one mm² is postulated. This size is four orders of magnitude (10⁴) larger than estimated by Kerrisk²⁴ for element U248, a failed, unencapsulated, sodium-bonded, advanced carbide fuel experiment. For this and smaller defect sizes, the rate of decomposition during transient heating is assumed to be controlled by the rate at which nitrogen can be discharged under pressure into the primary coolant. The nitrogen pressure in the element was assumed to equal the decomposition pressure at the fuel centerline temperature and the discharge rate during transient heating was calculated using the formula for flow of a perfect gas through a sharp-edged orifice.

The effects of the three postulated defect areas on the transient thermal conditions in the elements and the subsequent potentials for a MFCI are discussed in the following sections.

2. Thermal Analysis of Fuel Elements

One-dimensional, transient heat-transfer calculations which assumed concentric fuel, shroud, and cladding, were performed with the CINDA code¹² for the elements and operating conditions listed in Table 463-XIX. The node layouts for shrouded and unshrouded elements and the corrections for transient thermal expansion developed by Kerrisk²⁴ for the U5100 series elements were used in this analysis. The calculations were initiated by replacing the high-conductance sodium bond under normal operating conditions with an end-of-life mixture of helium and fission gas. Fuel swelling which results in a decreasing bond gap and hence, an increasing gap conductance with burnup, was neglected in these calculations.

The properties of solid and liquid (U_{0.8}Pu_{0.2})N and the liquid metals, U and Pu, used in this study are shown in Table 463-XX. For the heat transfer calculations, the melting point of PuN (3000K)²² was used for the solidus temperature. Between 3000 and 3120K, the density was corrected for an assumed volume increase on melting of 20%, and the heat capacity was adjusted to include the heat of fusion, which was estimated as 240 J/g. Reasonable uncertainties in the high-temperature properties were assumed to be ±5% for the densities, ±10% and ±15% for the heat capacities of (U_{0.8}Pu_{0.2})N and the liquid metals, and ±15% and ±25% for the thermal conductivities of (U_{0.8}Pu_{0.2})N and the liquid metals, respectively.

TABLE 463-XX

NITRIDE FUEL AND LIQUID METAL PROPERTIES ^{a, b}
FOR MFCI CALCULATIONS

T, K	(U _{0.8} Pu _{0.2})N			U _L			Pu _L		
	ρ , ^c	C_p , ^c	k_0 , ^d	ρ	C_p , ^c	k_0	ρ	C_p , ^c	k_0
	Mg/m ³	J/g K	W/m K	Mg/m ³	J/g K	W/m K	Mg/m ³	J/g K	W/m K
300	14.17 (25)	0.195 (25)	13.0 (25)						
700	13.94 (25)	0.212 (25)	16.0 (25)						
1100	13.73 (25)	0.230 (25)	18.6 (25)				16.44 (28)	0.175 (27)	18.9 (20)
1500	13.52 (25)	0.247 (25)	20.6 (25)	17.15 (26)	0.204 (27)	54.9 (20)	15.95 (20)	0.175 (27)	22.5 (20)
1900	13.31 (25)	0.264 (25)	22.2 (25)	16.64 (20)	0.204 (27)	61.2 (20)	15.46 (20)	0.175 (27)	25.3 (20)
2300	13.10 (20)	0.282 (25)	23.2 (20)	16.13 (20)	0.204 (27)	66.2 (20)	14.97 (20)	0.175 (27)	27.2 (20)
2700	12.90 (20)	0.299 (25)	23.8 (20)	15.61 (20)	0.204 (27)	69.9 (20)	14.49 (20)	0.175 (27)	28.4 (20)
3000	12.74 (20)	0.312 ^e (20)	23.9 (20)	15.23 (20)	0.204 (27)	71.8 (20)	14.12 (20)	0.175 (27)	28.7 (20)
3060	11.47 ^f (20)	0.315 ^e (20)	23.9 (20)	15.15 (20)	0.204 (27)	72.1 (20)	14.05 (20)	0.175 (27)	28.7 (20)
3120	10.28 ^f (20)	0.318 ^e (20)	23.9 (20)	15.08 (20)	0.204 (27)	72.4 (20)	13.97 (20)	0.175 (27)	28.7 (20)
3400	10.13 ^f (20)	0.330 (20)	23.9 (20)	14.72 (20)	0.204 (27)	73.2 (20)	13.63 (20)	0.175 (27)	28.4 (20)

^a Underlined property values are estimated.

^b Numbers in parentheses are references.

^c Computed from thermal expansion measurements.

^d At theoretical density; to correct for porosity multiply by $(1-P)/(1+P)$.

^e Includes estimated heat of fusion (240 J/g).

^f Includes 20% increase in volume on melting.

Since relatively large uncertainties in high-temperature properties must be considered in estimating the potential for MFCI, the product of density and heat capacity and the thermal conductivity were treated parametrically in this study. The sodium, stainless-steel, helium, and fission-gas properties were the same as used in evaluating the potential for a MFCI with sodium-bonded carbide fuels.²⁴

For a very small postulated cladding defect, where melting of the fuel occurred with less than 0.5% decomposition, the fuel and cladding temperatures calculated for the K4 and C5 series elements are shown in Table 463-XXI. In these calculations, the fuel surface was required to reach a temperature of 3040 K (fuel melting point) before the fuel was allowed to begin to slump, and a slumping time²⁴ of 0.1 sec was allowed

to elapse before the fuel contacted the cladding. For the two elements with the smaller fuel-cladding gaps (K4-10 and C5-20), steady-state temperatures were attained such that the region near the fuel surface remained below the melting point, while the region around the centerline was above the melting point. Because of the similarities in transient thermal responses and peak fuel temperatures for these two elements, only calculations which used nominal property values were performed for element C5-20. The transient temperatures computed with nominal property values under the conditions at the top of the core are shown in Fig. 463-30. The shroud in K4-10 melted after about 2 sec. Fuel centerline temperatures greater than the melting point were reached after ~ 4.6 sec, and the steady-state temperature profile developed after ~ 7 sec.

TABLE 463-XXI

MAXIMUM TEMPERATURE ATTAINED DURING MELTING OF $(U_{0.8}Pu_{0.2})N$ FUEL

Element	Fuel Property Multiplier		Fuel Temperature, K				Cladding Temperature, ^a K	
	k	ρC_p	Core Midplane		Core Top		Maximum	Mid-Radius
			Maximum	Surface	Maximum	Surface		
K4-10	1.15	1.15	3230	2960	3180	2970	N/A ^b	
	1.00	1.00	3250	2950	3210	2970	N/A ^b	
	0.85	0.85	3280	2930	3240	2950	N/A ^b	
	0.85	1.15	-	-	3240	2950	N/A ^b	
C5-20	1.00	1.00	3260	2960	3230	2980	N/A ^b	
C5-14	1.15	1.15	3190	3040	3180	3040	-	-
	1.00	1.00	3240	3040	3230	3040	1610 1820	1340 1460
	0.85	0.85	3310	3040	3270	3040	-	-

^a At top of core.^b Slumping and fuel cladding contact did not occur.^c Lower temperatures calculated with nominal contact coefficient of $1.6 \text{ kW/m}^2 \text{ K}$.

Figure 463-31 shows the temperatures calculated with nominal property values and conditions at the top of the core for element C5-14. At ~ 5.8 sec, the fuel surface reached the melting point and started to slump. After fuel-cladding contact was established, the inner cladding surface attained its maximum temperature very rapidly. Since the maximum cladding temperature is strongly dependent on the value used for the contact coefficient, calculations were performed with two values for the contact coefficient. The nominal value was $1.6 \text{ kW/m}^2 \text{ K}$, and the high value, which approaches that for perfect contact between molten fuel and cladding was an order of magnitude larger. Maximum cladding temperatures greater than the cladding liquidus temperature were found only when the limiting value for the contact coefficient was used, and, in this event, less than 10% of the cladding thickness exceeded the liquidus temperature and for only a few milliseconds.

For the case in which nearly complete decomposition of the fuel was postulated to occur, the maximum fuel temperature reached 2980 K with the nominal fuel property values and 2970 K with ρC_p and k

increased by 20%. In the heat transfer calculations, the heat absorbed in decomposition²⁰ (1.1 kJ/g) was applied over the range from 2800 K to the temperature at which decomposition was complete. This temperature (T_d) and the fuel surface temperature (T_s) necessary for slumping were determined by the following iterative procedure:

- T_d and T_s were estimated,
- fuel properties (density, heat capacity, heat of decomposition, thermal conductivity) were computed for a linear variation in nitride/metal ratio between 2800 K and T_d ,
- fuel temperatures were calculated with CINDA, and transient nitrogen pressures were computed from surface temperatures,
- total nitrogen lost from the fuel was determined by integrating the weight loss rate over the duration of the transients,
- estimates for T_d and T_s were revised, and the procedure repeated.^s

In this way, T_s and T_d were found to be 2930 and 3000 K, respectively, and the decomposed portion of the fuel for elements K4-10 and C5-14 with nominal and increased property values were between 75 and 86%.

Fig. 463-30. Transient Temperatures following Loss of Bond from Element K4-10.

Further iteration was not performed because the changes in T_s and T_d required to achieve more complete decomposition with this model are small compared to the experimental uncertainties (± 30 K) in the decomposition temperature and pressure.

Cladding temperatures were computed for contact with $(U_{0.8}Pu_{0.2})_2$ at 2930 K. For element C5-14, the maximum cladding temperatures calculated with nominal and limiting values for the contact coefficient were 10 K lower and 20 K higher than the corresponding values in Table 463-XXI for liquid nitride.

During transient heating of an element with a cladding defect area of one mm², forced discharge of significant amounts of nitrogen into the coolant would not occur until the nitrogen pressure in the element exceeds the pressure in a coolant channel. For fuel temperatures below ~3000 K, the nitrogen pressure produced by partial decomposition was below the channel pressure, so that in the short time required for heating to this

temperature, a very small amount of nitrogen would effuse through the defect. As the fuel temperature increased further, the nitrogen pressure, which was computed from the decomposition pressure versus temperature measurements,²² rose rapidly to 1.5 MPa. An iterative procedure was required to balance the heat absorbed in partial decomposition and the increase in thermal conductivity for the increasing liquid metal fractions with the fraction of nitrogen released from the fuel. For this procedure, an initial estimate of the fractional decomposition between 2800 and 3100 K was used to determine the heat absorbed and the increase in thermal conductivity. From the fuel temperatures calculated using CINDA, the transient nitrogen pressures in the element were computed. The orifice discharge rates for these pressures were then computed and integrated numerically to obtain the fraction of original nitrogen present in the element that was discharged to the coolant.

Fig. 463-31. Transient Temperatures following Loss of Bond from Element C5-14 with Fuel Melting and Slumping against Cladding.

Revised estimates of the absorbed heat and thermal conductivity were made and the procedure repeated until a balance was achieved.

When the fuel surface was required to reach 3000 K before slumping of the nitride-metal mixture could occur, the amount of fuel decomposition computed with this iterative method was ~ 10%, and the maximum fuel temperature (using nominal property values) was 3120 K for element K4-10. For this element, the use of increased property values (ρC_p and k) resulted in a maximum fuel temperature of 3100 K and a fractional decomposition of ~ 9%. Lower maximum fuel temperatures (3090 and 3080 K) and smaller fractional decompositions (6 and 5%) were computed for element C5-14 with nominal and increased fuel property values. The maximum cladding temperatures that resulted from contact with the molten nitride-metal mixture at 3010 K were lower than those found for melting or gross decomposition of the fuel.

In the next section the fuel-coolant interface temperatures that would result from contact between the molten nitride-metal compositions at their maximum temperatures and the liquid sodium coolant are evaluated.

3. Molten Fuel-Coolant Interactions

The instantaneous interface temperatures produced by contact between molten fuel and coolant were calculated with the following equation,¹⁵

$$T_i = T_c + (T_f - T_c) \frac{\sqrt{(\rho C_p k)_f}}{\sqrt{(\rho C_p k)_f} + \sqrt{(\rho C_p k)_c}},$$

where T is the temperature, ρ is the density, C_p is the heat capacity, and k is the thermal conductivity. In this equation, the subscripts f , c , and i refer to fuel, coolant, and interface, respectively. A necessary condition for the occurrence of vapor explosions results from liquid/liquid interface temperatures greater than the homogeneous nucleation temperature of the colder fluid. The liquid nitride and metal properties listed in Table 463-XX and the liquid sodium properties from Ref. 24 were used to calculate the interface

temperatures.

The critical fuel temperatures, i.e., the molten fuel temperatures required to produce an interface temperature equal to the homogeneous nucleation temperature of sodium (2300 K), are shown in Fig. 463-32 for liquid $(U_{0.8}Pu_{0.2})N$. In this figure, the maximum fuel temperatures listed in Table 463-XXI for increased, nominal, and decreased property values are compared with the critical fuel temperatures calculated with the same properties. For contact between sodium and liquid $(U_{0.8}Pu_{0.2})N$, the smallest margins of safety exist for the coolant temperatures at the top of the core (850 K). The margins computed with increased (decreased) property values are smaller (larger) than those obtained with nominal properties. The maximum fuel temperatures from Table 463-XXI are below the critical temperature by a minimum of ~390 K with nominal property values, and ~ 200 K with increased

Fig. 463-32. Critical Fuel Temperatures for a MFCI between Liquid $(U_{0.8}Pu_{0.2})N$ and Sodium.

property values. Multiplication of the thermal conductivity by 0.85 and the product, ρC_p , by 1.15, resulted in a maximum calculated fuel temperature of 3040 K. The critical fuel temperature, which depends upon the product $(\rho C_p k)_f$, was calculated with nominal property values and compared with the maximum fuel temperature (3040 K) to obtain a margin of ~ 360 K.

In the event of a gross cladding failure for which nearly complete decomposition was postulated, the critical fuel temperatures were computed for a MFCI with $(U_{0.8}Pu_{0.2})_L$. With nominal property values, the critical fuel temperature was ~ 170 K greater than the maximum fuel temperature. When increased property values (+20% on ρC_p and +25% on k) were used to compute the critical temperatures, the maximum fuel temperatures were ~ 30 K below the critical fuel temperatures for an interface with sodium at 850 K. Since the critical temperature depends on the product $\rho C_p k$, the computed margin can be this small only if all these properties are uniformly underestimated by their assumed uncertainties. Also, complete fuel decomposition is an idealized condition produced in laboratory experiments with finely ground powders in a vacuum. That this condition could occur in a fuel element during the transient heating, which follows loss of the sodium bond, is regarded as virtually incredible. Any nitride fuel, dissolved nitrogen, or dissolved fission products in the liquid will increase the margins primarily through a reduction in thermal conductivity. The minimum margin obtained with the increased property values is more than doubled (70 K) if the critical fuel temperature is computed for the maximum calculated liquid metal fraction (0.86). Lastly, use of the fuel surface temperatures rather than the centerline temperatures would result in increased margins of safety.

Critical fuel temperatures computed for partially decomposed fuel, 10% liquid metal and 90% molten $(U_{0.8}Pu_{0.2})N$, were compared with the maximum temperatures found for an element with a defect area of one mm^2 . The smallest margins between the maximum and critical fuel temperatures for sodium at 850 K were ~ 430 K with nominal property values and

~ 240 K with increased property values.

4. Discussion and Summary

The behavior of mixed nitride fuels under the transient heating conditions existing in a fuel element following instantaneous loss of the sodium bond has been investigated for three hypothetical cladding failures. In this investigation, the nitride fuel decomposition and melting behavior during transient heating was evaluated using reasonable assumptions which were based on the behavior observed in isothermal laboratory experiments. Within the framework provided by the experimental data and these assumptions, an endeavor was made to conservatively estimate important parameters, such as the decomposition temperatures and pressures, the fuel melting point, the temperatures for fuel (and metal) slumping, and the rate of decomposition.

In the transient thermal analysis, several important features that would tend to reduce the maximum fuel temperatures, thereby adding conservatism to estimates, were neglected. Fuel swelling, which results in a decreasing bond gap with burnup, was neglected, while the bond gap conductance was computed with an end-of-life mixture of helium and fission gas. The fuel was assumed to be concentric with no areas of fuel-sodium-clad contact over the entire circumference. The linear powers were increased over the requested irradiation conditions, and axial conduction was neglected. Even under these conditions, the elements with small, radial, fuel-cladding gaps (0.26 mm) attained steady-state temperatures such that a molten core was surrounded by fuel at temperatures below 3000 K. The expected fuel-cladding gap at the end of life would have prevented fuel slumping in the element with the large, radial, fuel-cladding gap (0.38 mm) and would have reduced the calculated temperatures in the elements with the small fuel-cladding gap.

A positive margin of safety was found between the temperatures necessary for homogeneous nucleation in liquid sodium and the maximum temperatures calculated for the melted and decomposed $(U_{0.8}Pu_{0.2})N$ for all cases that were evaluated. When the uncertainties assigned to the high-temperature

liquid fuel and metal properties were used as parameters, the smallest margins of safety were computed with the properties (ρ , C_p and k) at their maximum values. For cladding defect sizes up to one mm², the estimated amount of nitride fuel decomposition was ~10%, and the minimum margins of safety were ~390 K with nominal property values and ~200 K with increased property values.

A very large defect, such as an axial crack over the length of the fuel column, was postulated. To evaluate the fuel temperatures and the potential for a MFCI, the fuel decomposition was modeled after that which could occur for temperatures ≥ 2900 K in a vacuum. Under these conditions, fuel decomposition during the transient following loss of the sodium bond was estimated to be ~86%. When the maximum fuel temperatures computed for this case were compared with the critical temperatures for (U_{0.8}Pu_{0.2})₂, margins of safety of ~170 K were found by using nominal property values and ~30 K by using increased property values. When the critical temperatures for a liquid-metal/nitride-fuel ratio of 0.86 were computed, the minimum margin of safety was increased to 70 K. This margin would be increased further by: (1) any property value (ρ , C_p or k) which is lower than the nominal plus its maximum assigned uncertainty, (2) a lower liquid-metal/nitride-fuel ratio, (3) dissolved nitrogen or fission products in the liquid metal, (4) having the fuel-pellet surface rather than the centerline material in contact with sodium, (5) fuel-sodium contact at axial locations other than the top of the fuel stack, and (6) increased high-temperature expansion because of retained fission gases. Further, the assumptions made regarding decomposition kinetics and diffusion rates are very conservative. Any alteration in the analysis that significantly decreases the fraction of metal in the liquid will increase the margin of safety.

IV. ADVANCED LMFBR FUEL PROPERTIES (J. F. Kerrisk, H. D. Lewis, W. M. Olson, and J. F. Andrew)

A. Introduction

The work on the properties of advanced fuels was initiated during the second quarter of FY 1975. The goals of this task over the remainder of the year were: (1) to begin vapor-pressure measurements and gas-phase-species identification studies on the U-Pu-C system using mass spectrometric techniques, (2) to begin thermal-transport measurements on advanced fuel materials using the laser flash method for measuring thermal diffusivity, (3) to initiate measurements of the thermal expansion coefficients of unirradiated carbide fuels and (4) to perform and report the results of critical reviews of the literature on the thermodynamic properties, electrical and thermal transport properties, and the coefficient of thermal expansion and density of the U-Pu carbides.

B. Literature Reviews

1. Thermodynamic Properties

A review of the literature on the thermodynamic properties of the Pu-C system has been completed, and the information summarized in the LASL contribution to an ANL topical report authored by M. Tetenbaum, A. Sheth, and W. Olson.²⁹ This information on the Pu-C system is presented in more detail in a forthcoming LASL report by W. Olson.³⁰

The review was based mainly on the preliminary values for the thermodynamic properties of the Pu-C system reported by Rand at a recent IAEA Consultants Meeting (Grenoble, France, May 1974). Since this represents the best information presently available (including some unpublished data), these values were extracted from this report and, together with comments about the original work and Rand's reasons for his conclusions, assembled into a critical review.

Generally the available thermodynamic data for the Pu-C system are incomplete and inconsistent. Much work remains to be done to resolve these disagreements satisfactorily. Rand attempted to extract the most reasonable values from the conflicting

data, and his values have been used in all cases, although there is some question about his decision.

Much of the difficulty with thermodynamic property measurements on the monocarbide arises from the fact that the range of composition varies with temperature. Apparently, only $\text{PuC}_{0.88}$ exists from room temperature to the melting point. Thus, either metal or Pu_2C_3 could be present in the samples at various temperatures. In cases such as vapor pressure measurements, where a two-phase sample was used, the monocarbide composition would change with temperature. This leads to uncertainties in calculated thermodynamic functions.

The following values were taken directly from Rand's preliminary assessment or were calculated from data presented in it. (Temperature in K.)

Heat Capacity

$$\begin{aligned} \text{PuC} \quad C_p (\text{J/mol K}) &= 77.83 - 7.817 \times 10^{-2} T \\ &+ 6210 \times 10^{-5} T^2 - 1.10 \times 10^6 / T^2 \\ \text{Pu}_2\text{C}_3 \quad C_p (\text{J/mol K}) &= 132.726 - 20.023 \times 10^{-3} T \\ &+ 35.063 \times 10^{-6} T^2 - 16.5163 \times 10^5 / T^2 \end{aligned}$$

Heat of Formation

$$\begin{aligned} \text{PuC} \quad \Delta H_f (298) &= -47.7 \text{ kJ/mol} \\ \text{Pu}_2\text{C}_3 \quad \Delta H_f (298) &= -166 \text{ kJ/mol} \end{aligned}$$

Free Energy of Formation

$$\begin{aligned} \text{PuC} \quad \Delta G_f (298) &= -47.7 \text{ kJ/mol} \\ \text{Pu}_2\text{C}_3 \quad \Delta G_f (298) &= -166 \text{ kJ/mol} \\ \text{PuC}_2 \quad \Delta G_f &= 38150 - 18.9 T \text{ kJ/mol} \\ \text{Pu}_3\text{C}_2 \quad \Delta G_f (300) &= -122 \text{ kJ/mol} \end{aligned}$$

Pu Pressures (Pascals)

$$\begin{aligned} \text{PuC} \quad \ln p &= 105.4 - 0.06 T \\ \text{Pu}_2\text{C}_3 \quad \ln p &= 170.5 - 0.1 T \end{aligned}$$

2. Thermal Expansion and Density

A review report³¹ on the thermal expansion and density of uranium and plutonium carbides has been prepared for publication. This report summarizes the

available data on the thermal expansion and density of UC, PuC, (U, Pu)C, U_2C_3 , Pu_2C_3 , and $(\text{U, Pu})_2\text{C}_3$. A large number of experimental studies on compounds containing only uranium and carbon have been made; however, plutonium-containing compounds have received very little attention. In many cases, the thermal expansion experiments were carried out on specimens of unknown purity. Summaries of selected values for the thermal expansion and density of several carbide compositions in the U-C, Pu-C, and U-Pu-C systems are listed in Tables 463-XXII and 463-XXIII. The quantity α_i is the instantaneous expansion coefficient (value of the derivative of the thermal expansion curve) and α_m is the mean expansion coefficient over the temperature range shown.

3. Electrical and Thermal Transport

A report on the review of the literature on the electrical and thermal transport properties of the U-C, Pu-C, and U-Pu-C systems was written.³² Some of the conclusions from this report which relate to the thermal conductivity of UC, PuC, and (UPu)C are summarized below.

- Quantitative evaluation of the effects of composition, (carbon concentration, U/pu ratio), oxygen and nitrogen impurity concentrations, and porosity on the thermal conductivity of the carbides is not possible with the present state of knowledge.
- The effects of porosity on the thermal conductivity are probably dependent upon temperature, pore-size distribution, and pore morphology.
- Effects of nickel additions, densification phenomena, oxygen and nitrogen impurities, and second-phase, higher carbide concentrations are interdependent. The quantitative evaluation of these effects will require study of conductivity measurements on materials that have been well characterized on the basis of composition, impurity levels, and quantitative microstructural analysis. Base-line studies on the effect of second-phase concentration should include electrical resistivity and metallographic evaluation of the samples used for thermal conductivity measurements.

Suggested values for the thermal conductivity of UC, PuC, and (U, Pu)C from this report are summarized in Table 463-XXIV. The tabulated values are calculated by equations of the form $\lambda = a + bT + cT^2$ where λ is the thermal conductivity in W/m K. The values for

TABLE 463-XXII

RECOMMENDED VALUES FOR THE COEFFICIENTS
OF LINEAR THERMAL EXPANSION ($10^{-6}/^{\circ}\text{C}$)
OF URANIUM AND PLUTONIUM CARBIDES

	$(25^{\circ}\text{C})\alpha_l$	$(1000^{\circ}\text{C})\alpha_l$	$(25-1000^{\circ}\text{C})\alpha_m$
UC	10.1	12.4	11.2
PuC	8.5	13.7 ^a	10.8 ^a
(U, Pu)C	8.8	14.6	11.9
U ₂ C ₃	10.7	12.0	10.6
Pu ₂ C ₃	12.9	16.9	14.9
(U, Pu) ₂ C ₃	9.6	12.8	11.2

^aData for $T = 900^{\circ}\text{C}$.

the conductivity of PuC reflect a significant concentration of Pu₂C₃.

C. Properties Measurement

1. Thermal Transport

Measurements of thermal diffusivity in the U-Pu-C system were to be made at temperatures from ambient to approximately 2000°C. The measurement method is similar to the flash technique first used by Parker.³³ A pulse of energy is deposited uniformly at the front surface of a sample and the diffusivity is calculated as a function of the sample thickness and the time required for the temperature at the back surface of the sample to reach one-half the maximum temperature resulting from the input pulse.

The apparatus consists of a Centorr Series 60 high-temperature furnace (enclosed in an inert-atmosphere glovebox) to control sample temperature, and a Korad K-2 pulsed laser to supply the energy pulse, through a simple optical system, to the sample. The apparatus was set up to measure sample temperatures from ambient to approximately 1000°C using a thermocouple-potentiometer system, and to use the thermocouple output to monitor the back-surface temperature rise. From approximately 700°C to 2000°C, sample temperature was measured by optical pyrometry, and the back-surface temperature was monitored by a photomultiplier system. Back-surface temperature history was

recorded on photographic film from the CRT of an oscilloscope, and the time scale was obtained by superimposing the output of a pulse generator on the back-surface temperature record.

Initial phases of the work in preparation for investigation of the U-Pu-C system consisted of:

- Alignment of the laser and optical system and "proof" of beam uniformity;
- Setup of temperature measurement, timing, and recording systems;
- Completion of QA Experiment Test Plan and calibration procedures for various system components;
- Operational checkout of equipment through diffusivity measurements on "standard" or reference samples;
- Diffusivity measurements on samples of carbide fuel to determine acceptability of sample geometry and temperature-measurement methods.

The basic apparatus used in the diffusivity measurement work proved quite satisfactory. Problems initially encountered in alignment of the pulsed laser beam and optical system were largely the result of imprecision in the positioning adjustment capability of the holder for the right-angle prism required to target the laser beam at the sample surface. This problem was eliminated by replacing the prism with a micromachined copper mirror, supplied by the Product Development Division of ORNL, mounted on an Aerotech laser beam director head. The procedure subsequently developed for alignment of the laser and optical system provided reproducible, uniform energy deposition at the sample front face. Conductive heat loss from the sample, during the measurement time, was minimized. Conduction heat losses were negligible at ten halftimes, where the half-time is the time required for the sample back-face temperature to reach half its maximum value. The thermocouple configuration, which was used for sample back-face temperature measurement up to about 1000°C was modified several times in attempts to maintain good electrical contact at high temperature and minimize noise pickup.

TABLE 463-XXIII

RECOMMENDED VALUES FOR THE LATTICE PARAMETERS AND THEORETICAL DENSITIES OF URANIUM AND PLUTONIUM CARBIDES

Compound	Lattice Parameter ($\text{\AA}$)	Density (Mg/m^3)		
		Isotopic Composition		
		0.7% ^{235}U 99.3% ^{238}U	93% ^{235}U 7% ^{238}U	88-94% ^{239}Pu 6-12% ^{240}Pu
UC	4.9600	13.61	13.46	--
$\text{PuC}_{0.90}$	4.9734	--	--	13.49
$(\text{U}_{0.8}\text{Pu}_{0.2})\text{C}_{0.99}$	4.9639	13.58	13.46	--
U_2C_3	8.088	12.85	12.72	--
Pu_2C_3	8.132	--	--	12.70
$(\text{U}_{0.8}\text{Pu}_{0.2})_2\text{C}_3$	8.096	12.83	12.72	--

The standard experimental procedure required two or three diffusivity determinations at ambient temperature on a molybdenum reference sample. Twelve such determinations resulted in an average thermal diffusivity, $\bar{\alpha}$ of $0.5986 \text{ cm}^2 \text{ s}^{-1}$, and a coefficient of variation (relative standard deviation) $s_{\bar{\alpha}}/\bar{\alpha} = 0.0106$, which compares satisfactorily with values obtained from other experimental runs, reported in Table 463-XXV.

Thermal diffusivities of several molybdenum and U-Pu carbide samples were measured at ambient temperature (16 to 19°C) as part of the equipment check-out. Previous measurements with the equipment used samples which were 10, 16 mm diameter, while current work uses samples prepared from fuel pellets (6.6 mm diameter). Table 463-XXV lists the results of measurements on three molybdenum samples and two U-Pu carbide samples. The measurements indicated good agreement on the molybdenum samples of different diameters. Measurements on a 10, 16-mm-diameter molybdenum sample to approximately 1000°C were also made and compared with previous results (4 years old) on the same sample. The results compared well.

Problems associated with power-supply-generated electrical noise were encountered in measurements of thermal diffusivity of fuel samples when thermocouples were used as the differential temperature detector at the back face of the sample. The noise was essentially eliminated from the thermocouple signal by using a filter tuned to 360 and 720 Hz at the thermocouple input to the oscilloscope amplifier. However, the best filter design developed caused distortion in the zero-time reference pulse.

To circumvent the noise problem, three other back-surface detectors were considered for use with the thermal-diffusivity apparatus. A photomultiplier system which had been previously used has a useful absolute sample temperature range from 800° to 2000°C . A liquid-nitrogen-cooled InSb photodiode having a range from $\sim 350^\circ\text{C}$ to 2000°C could replace both the thermocouple and photomultiplier system. The lack of measurement capability between ambient and 400°C may not be a critical disadvantage. An Ircon photodiode could be used at high sample temperature with the possible advantage of its requirement of ambient temperature operating environment permitting use inside the glovebox.

TABLE 463-XXIV

THERMAL CONDUCTIVITIES OF UC, PuC, AND (U, Pu)C (W/m K)

Equation Coefficients	UC 100% TD		PuC 100% TD	(U _{0.8} Pu _{0.2})C 100% TD	
	50 < T, °C ≤ 700	700 < T, °C ≤ 2300		50 < T, °C ≤ 500	500 < T, °C ≤ 2300
a	21.7	20.2	7.45	17.5	12.8
b	-3.04×10^{-3}	-1.48×10^{-3}	-4.04×10^{-3}	-5.65×10^{-3}	8.71×10^{-3}
c	3.61×10^{-6}	---	1.20×10^{-5}	8.14×10^{-6}	1.88×10^{-6}

Temperature, °C

25	21.6		17.4
100	21.4	7.2	17.0
300	21.1	7.3	16.5
500	21.1	8.4	16.7
700	21.3	10.5	17.9
900	21.5	13.5	19.1
1100	21.8	17.5	20.1
1300	22.1	22.5	20.9
1500	22.4		21.6
1700	22.7		22.1
1900	23.0		22.5
2100	23.3		22.8
2300	23.6		22.9

and a disadvantage being a relatively smaller useful temperature range. It was decided that the InSb photodiode would be used for measurements above room temperature.

Diffusivity measurements were made on the 6.6-mm-diameter specimens of (U_{0.8}Pu_{0.2})C + 13 vol% (U_{0.8}Pu_{0.2})₂C₃ fuel material (98% of theoretical density) up to 1000°C. Table 463-XXVI shows the results, uncorrected for porosity effects, compared with data of Van Craynest³⁴ and Moser and Kruger.³⁵ Van Craynest used two versions of the phase-shift method, while the Moser and Kruger and LASL measurements were by the flash method of diffusivity measurement. Although all of these investigators used well-characterized material, fundamental differences are illustrated in the basic transport property and are typical of differences in the existing literature. For example, the first two sets of data possibly indicate a measureable lattice contribution to the conductivity below 500°C while the third set does not. No basis exists at this time for quantitative comparison of these data on the basis of differences in porosity, composition, or microstructure. Such deficiency

in the present knowledge has been noted in several reviews.^{32,36,37,38,39} At the end of FY 1975, preparations had been made to determine the thermal diffusivities of the following types of samples:

- Single-phase (U, Pu)C, 6.6-mm-diam specimens, 98% dense.
- Single-phase (U, Pu)C, 6.6-mm-diam and 10.16-mm-diam specimens, 87% dense.
- (U, Pu)C + 13 vol% (U, Pu)₂C₃, 10.16-mm-diam, 98% dense.
- (U, Pu)C + 13 vol% (U, Pu)₂C₃, 6.6-mm-diam, 98% dense.

2. Vapor Pressure

Vapor pressures over the U-Pu-C system were to be measured by the Knudsen effusion method. The nature of the vapor species and the pressures relative to known materials were to be determined with a magnetic-sector mass spectrometer (Hitachi RMU-6). Several modifications to the measurement system were planned, including installation of the ion source and Knudsen cell assembly, and a sample preparation box

TABLE 463-XXV

REPRODUCIBILITY OF DIFFUSIVITY MEASUREMENTS AT AMBIENT TEMPERATURE

Sample	Diameter mm	Thickness mm	Thermal Diffusivity ^a cm ² /s	Relative Standard Deviation %
Mo	10.16	2.591	0.593	1.5
Mo	10.16	3.759	0.583	1.6
Mo	6.604	2.553	0.532	1.4
(U _{0.8} Pu _{0.2})C + 13 vol% (U, Pu) ₂ C ₃	6.604	1.410	0.0493	3.8
(U _{0.8} Pu _{0.2})C + 13 vol% (U, Pu) ₂ C ₃	6.604	2.083	0.0521	3.5

^a Average value of 7 to 12 determinations over a 3-week period.

and hood, prior to introducing plutonium samples into the system. During modifications on the Hitachi, another apparatus, incorporating both collection and quadrupole-mass-spectrometric capabilities was to be used to make preliminary measurements in the U-Pu-C system. Although this apparatus is less sensitive than the magnetic-sector instrument in the mass range of interest, it was essentially operational at the start of this program, and could provide preliminary data. The magnetic-sector mass spectrometer was checked out using a gold sample. It was then disassembled, and installation of the hood and preparation box was begun.

The quality assurance document entitled "Experiment Test Plan for Vapor Pressure Measurements in the U-Pu-C System" was written to provide the basic experimental program plan. During preliminary measurements using the quadrupole apparatus, the electron multiplier on the quadrupole failed, leaving only the target-collection system to determine the Pu flux. Initial measurements using only target collection, indicated that sample composition was changing during the run. For this reason, the target-collection method alone is not adequate.

Modifications to the Hitachi RMU-6 and auxiliary equipment required to achieve operation and to obtain calibrated vapor-pressure measurements on plutonium-containing materials are continuing.

3. Thermal Expansion

Preparations were made to measure the coefficient of thermal expansion of carbide fuel materials up to approximately 2400°C. The measurement apparatus consists of a Centorr Series 600 high-temperature furnace designed for horizontal orientation of samples, and two Gaertner filar micrometer microscopes mounted on an Invar bar. The measurement procedure consists of direct determination of the expansion of a sample initially 60 mm long, as a function of temperature. The filar micrometer microscopes, furnace windows, front-surface mirror, thermocouples, and the optical pyrometer used in this work were calibrated according to existing QA calibration procedures.

Measurements were made on a platinum standard at temperatures up to 1500°C in vacuo and in helium. The resulting expansion values were within one percent of the values reported by Hahn and Kirby.⁴⁰

A fixture for holding a line of 10 fuel pellets was made from tantalum and carburized to improve compatibility with the fuel pellets. Poco graphite will be used as a standard in testing the fixture.

TABLE 463-XXVI

THERMAL DIFFUSIVITY OF (U, Pu)C

Temperature, °C	Thermal Diffusivity (cm ² /s)		
	LASL	Van Craynest ³⁴	Moser and Kruger ³⁵
	(U _{0.8} Pu _{0.2})C+13 vol% (U _{0.8} Pu _{0.2}) ₂ C ₃	(U _{0.85} Pu _{0.15})C	(U _{0.8} Pu _{0.2})C
	98%TD	91.5%TD	74.8%TD
100	0.0480	0.0530	---
200	0.0470	0.0515	---
300	0.0465	0.0505	0.0350
400	0.0462	0.0500	0.0357
500	0.0460	0.0497	0.0362
600	0.0460	0.0497	0.0365
700	0.0462	0.0500	0.0370
800	0.0465	0.0510	0.0373
900	0.0467	0.0530	0.0375
1000	0.0470	---	0.0375

TABLE XXVII
ADVANCED FUEL-CLADDING COMPATIBILITY TESTS

D. Fuel-Cladding Compatibility

A study of the compatibility of advanced fuel materials and potential cladding materials will be carried out at LASL. This work was originally started at General Electric (Sunnyvale). The experiments are planned to include (U, Pu)C, (U, Pu)C + (U, Pu)₂C₃, and (U, Pu)N fuel materials and eight potential cladding materials, Types 316 and 330 stainless steels, A-286, M813, PE-16, Inconel 706, Inconel 718, and HT-9. Table 463-XXVII lists an outline of the test conditions in order of test priority. Sodium bonded tests will be run with fuel pellets contained in individual capsules of each alloy. The capsules will be fabricated from alloy tubing. Helium bonded tests will be run using an assembly of alloy wafers and fuel pellets under compression. The alloy wafers will be fabricated from bar stock.

Fuel Composition ^a	Fuel-Cladding Bond	Test Conditions ^b	Alloys
MC+10vol% M ₂ C ₃	Sodium	725°C	c
MC+10vol% M ₂ C ₃	Helium	725°C	d
MN	Sodium	725°C	c
MN	Helium	725°C	d
MC	Sodium	725°C	c
MC	Helium	725°C	d
MC+10vol% M ₂ C ₃	Sodium	600°C	c
MC+10vol% M ₂ C ₃	Helium	600°C	d
MC	Sodium	600°C	c
MC	Helium	600°C	d
MN	Helium	600°C	d
MC+10vol% M ₂ C ₃ ^e	Helium	725°C	d
MN ^c	Helium	725°C	d

^aM = (U, Pu).^bTest times are 1000 h.^cAll alloys.^dAll alloys except HT-9.^eSelected fission products will be added to the fuel-cladding interface.

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