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FAST REACTOR MIXED-CARBIDE FUEL ELEMENT
DEVELOPMENT PROGRAM

Sixth Quarterly Progress Report
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1. INTRODUCTION

The Fast Reactor Mixed-Carbide Fuel Element Development Program is aimed at the establishment of mixed carbides as a competitive LMFBR fuel. The major objective of the program is to develop fast breeder reactor fuel rods of (UPu)C clad with stainless steel or higher strength alloys that will perform satisfactorily to a minimum of 50,000 Mwd/T and a maximum of 100,000 Mwd/T at power levels of 10 to 40 kw/ft.

The program is organized by tasks in the following manner:

Task 1200 – High-Density Pellet Evaluation in EBR-II (10 Rods)

Task 1300 – Annular and Low-Density Pellet Evaluation in EBR-II
(24 Rods)

Task 1500 – Unrestrained Swelling and Fission Gas Release

Task 1800 – Sodium-Bonded Fuel Evaluation

Task 1900 – Fabrication Development

1911 – Quality Assurance and Analytical Methods Development
(Completed)

1930 – High-Power Rods in EBR-II

1941 – Process Improvement

1950 – Low-Power Subassembly in EBR-II (19 Rods)

1960 – High-Power Subassembly in EBR-II (19 Rods)

Task 2400 – Uranium-Plutonium Dioxide Pellet Fabrication (Previously Task 1970).

Progress during the period October 1, 1968 to December 31, 1968 on the currently active tasks of this program is described in this report.

This is the sixth in a series of quarterly progress reports written in partial fulfillment of Contract AT(30-1)-3415 between the United States Atomic Energy Commission and the United Nuclear Corporation.

Prior reports include the following:

Topicals

- UNC-5137 Cihi, J., Hurwitz, S., Rosh, D., and Strasser, A.: The Design and Fabrication of Three (UPu)C Pilot Experiments for EBR-II (Oct. 25, 1965).
- UNC-5170 Andersen, J., Cherry, B., Cihi, J., Powers, R., and Stahl, D.: EBR-II Design and Fabrication Reports of Rods 81 through 90 (Mar. 1967).
- UNC-5198 Stahl, D. and Strasser, A.: Post-Irradiation Examination of High-Density (UPu)C Pellet-Fueled EBR-II Rods Irradiated to 30,000 Mwd/T (Jan. 1968).
- UNC-5229 Powers, R. and Strasser, A.: Compatibility of Several Cladding Materials with Uranium-Plutonium Carbides in Sodium (Nov. 1968).

Progress Reports

Fast Reactor Mixed-Carbide Fuel Element Development Program

- UNC-5193 First Quarterly Progress Report, July through September 1967
(Nov. 15, 1967).
- UNC-5201 Second Quarterly Progress Report, October through December 1967 (Feb. 10, 1968).
- UNC-5217 Third Quarterly Progress Report, January through March 1968
(July 10, 1968).
- UNC-5224 Fourth Quarterly Progress Report, April through June 1968
(Sept. 26, 1968).
- UNC-5233 Fifth Quarterly Progress Report, July through September 1968
(December 12, 1968).

2. HIGHLIGHTS OF THE CURRENT QUARTER

The four sodium-bonded mixed-carbide fuel rods were inserted into the GETR and are operating as designed at 25 kw/ft. These are the first sodium-bonded mixed-carbide rods with a fueled length of 14 in. ever to be irradiated.

The mixed-carbide unrestrained swelling and fission gas release experiments were assembled satisfactorily and shipped to Douglas-United Nuclear for irradiation.

The fabrication of all the mixed-carbide fuel rods for the low-power EBR-II subassembly was completed on schedule.

Post-irradiation examination of nine helium-bonded, high-density mixed-carbide pellet specimens indicated eight intact, including an Incoloy-800 clad specimen with a 0.003-in. initial diametral gap (96.7% smear density). The ninth specimen, having Type 316 clad and a 0.002-in. gap, had three cracks. The rods were irradiated up to 40,000 Mwd/T at 28 kw/ft.

3. PAST RESULTS AND CURRENT PROGRESS

FAST REACTOR IRRADIATION

3.1 TASK 1200 – HIGH-DENSITY PELLET EVALUATION IN EBR-II (10 RODS)

Introduction and Past Results

The objective of the task is to evaluate the performance of high-density (96% of theoretical, minimum) uranium-plutonium carbide pellets with varying size helium annuli between fuel and clad to provide void space for fuel swelling. The experiments are designed to determine the fuel-clad gap and clad strength combination to give satisfactory performance.

Ten doubly encapsulated fuel rods (Rods UNC 81 through 90) were designed, and nine were inserted for irradiation in EBR-II at powers of 20 to 30 kw/ft to burnups of 50,000 to 100,000 Mwd/T. The fuel is high-density, helium-bonded (UPu)C which was prepared by carbothermic reduction of the mixed dioxides followed by cold pressing and sintering of pellets. The cladding is Type 316 stainless steel and Incoloy-800. Eight of the capsules contain full-length prototypic fuel rods, while two capsules contain four segmented experiments each. A detailed description of these experiments is given in Table 1 and UNC-5170.

Rods UNC 87, 89, 90 (28 kw/ft) were discharged at the end of EBR-II Run 29 (August 1968) with a peak exposure of 40,000 Mwd/T. Neutron radiographs of the capsule showed that all three were in good condition.

TABLE 1 — SUMMARY OF CURRENT UNITED NUCLEAR MIXED-CARBIDE IRRADIATION EXPERIMENTS

Task No.	UNC Experiment No.	Description of Experiment	Design Parameters								Operating Conditions					Status
			Pellet Diameter, in.	Fuel Length, in.	Density, % T.D.		a/o Pu	Cladding Material	Maximum Linear Power (Design), kw/ft	Target Burnup, Mwd/T $\times 10^{-3}$	Facility	Operating Maximum Linear Power, kw/ft	Accumulated Burnup, Mwd/T $\times 10^{-3}$			
					Pellet (Type)	Smeared										
1200	81-88	EBR-II rods, helium-bonded, (8 rods)	0.248 to 0.252	14	98 (solid)	81 to 86	20	316 SS and Inco-loy-800	20 to 30	50 to 100	EBR-II	19 to 27	39,600	Rods 81-86 being irradiated, Rod 88 not yet inserted. Rods 87, 89, 90 irradiation complete.		
	89-90	EBR-II multiple specimen rods, helium-bonded (2 rods)	0.249 to 0.252	4	98 (solid)	82 to 90	20	316 SS and Inco-loy-800	24 to 30	40	EBR-II	22 to 28	22,310			
1300	91-114	EBR-II rods, helium-bonded (24 rods)	0.232 to 0.269	14	75 to 84 (solid), 99 (cored)	70 to 85	15	316 SS and Inco-loy-800	20	50 to 100	EBR-II	17 to 23	14,100	19-rod assembly being irradiated.		
1500	115-119	Unrestrained swelling specimens	0.06	0.5	75 to 90 (solid)	—	15	—	Low	20	Hanford	—	—	Fuel and capsule fabrication complete.		
	120-124	Fission gas release specimens	0.375 OD, 0.255 ID	0.500	75 to 98 (annular)	—	15	—	Low	20	Hanford	—	—	Fuel and capsule fabrication complete.		
1800	125-128	Sodium-bonded rods (4 rods)	0.240	14	93 to 98 (solid)	83 to 87	15	304 SS and Inco-loy-800	25	50	GETR	25	—	Rods being irradiated.		
1950	129-147	19-rod low-power subassembly, helium and sodium bonds	0.240 to 0.246	14	75 to 98	70 to 87	15	316 SS, 304 SS, and Inco-loy-800	15	100	EBR-II	—	—	Fuel rod fabrication complete, encapsulation in progress.		
1930	185-190	High-power rods (5 rods)	0.249 to 0.253	14	84 to 96 (solid)	80 to 88	15	316 SS and Inco-loy-800	30	50 to 100	EBR-II	—	—	Capsules completed and shipped to EBR-II.		
1950	191-209	19-rod high-power subassembly, helium and sodium bonds	—	14	84 to 96	—	15	316 SS, 304 SS, and Inco-loy-800	30	30 to 100	EBR-II	—	—	Design completed.		

Current Results

The calculated burnup of the nine rods as of December 31, 1968, in EBR-II is as follows:

1. Rods UNC 81 through 83 (19 kw/ft) – 36,300 Mwd/T
2. Rods UNC 84 through 86 (19 kw/ft) – 39,600 Mwd/T
3. Rod UNC 88 is scheduled for insertion in May 1969.

The post-irradiation examination of UNC 87, 89, and 90 continued at LASL. The rod design and test parameters are given in Table 2. UNC 87 is a standard full-length rod. UNC 89 and 90 are multispecimen rods, which consist of four individually sealed segments or specimens, each with a 2-in. fueled length and its own gas plenum.

The capsules were punctured for fission gas activity. Capsules UNC 87 and 90 did not show any fission product activity, while UNC 89 showed 0.2% of the total gas as xenon and krypton.

The fuel rods were removed from their respective capsules. All the rods were intact with the exception of one specimen in UNC 89, the Type 316 clad multispecimen rod. The specimen with 0.002-in. cold diametral gap and 96.6% smear density had three thin, brittle, longitudinal cracks in the fueled segment. The three cracks were 1-in., 1/2-in., and 1/4-in. long (see Fig. 1).

In comparison, the identical specimen clad with Incoloy-800 was in excellent condition (Fig. 2).

TABLE 2 — DESIGN PARAMETERS OF UNC 87, 89, 90
IRRADIATED IN EBR-II TO 40,000 Mwd/T AND 28 kw/ft (TASK 1200)

(Fuel: MC + 10 v/o M_2C_3 ~97% of Theoretical Density)

<u>Rod No.</u>	<u>Clad Material</u>	<u>Fueled Length, in.</u>	<u>Pellet Type</u>	<u>Diametral Gap, in.</u>	<u>Smear Density, %</u>	<u>Clad Condition after Irradiation</u>
87	Incoloy-800	14	Ground	0.006	94.2	Intact
89	Type 316	2	Unground	0.006	93.8	Intact
		2	Ground	0.002	96.6	Cracked
		2	Ground	0.006	94.2	Intact
		2	Ground	0.009	92.1	Intact
90	Incoloy-800	2	Unground	0.006	93.9	Intact
		2	Ground	0.003	96.7	Intact
		2	Ground	0.006	94.3	Intact
		2	Ground	0.009	92.2	Intact

Fig. 1 — Segment B of UNC 89, Irradiated to 40,000 Mwd/T at 28 kw/ft — Type 316 SS, 0.002-in. Fuel-to-Clad Gap

Fig. 2 — Segment B of UNC 90, Irradiated to 40,000 Mwd/T at 28 kw/ft —
Incoloy -800, 0.002-in. Fuel-to-Clad Gap

3.2 TASK 1300 – ANNULAR AND LOW-DENSITY PELLET EVALUATION IN EBR-II (24 RODS)

Introduction and Past Results

The objective of the task is to evaluate various methods of void deployment in helium-bonded uranium-plutonium carbide pellets to provide void space for fuel swelling. Specifically, high-density annular (97% of theoretical) and low-density pellets (75 and 84% of theoretical) with various smear densities are being evaluated to determine the best fuel rod design.

Twenty-four rods (Rods UNC 91 through 114) were designed for irradiation in EBR-II at powers of 20 kw/ft to burnups of 50,000 to 100,000 Mwd/T. Design variables include power, burnup, clad material and thickness, fuel structure, and fuel smear density. Design details are shown in Table 1.

Nineteen of the 24 rods were fabricated. The fabrication of the other five rods was postponed to FY-1969 for lack of funds.

The 19 rods were accepted by ANL-Idaho, assembled by them into subassembly X033, and then inserted in the reactor. This represents the first mixed-carbide subassembly ever to be irradiated.

Current Results

The 19-rod subassembly (X033) is operating in the EBR-II at about 20 kw/ft, and has accumulated an exposure of 22,300 Mwd/T.

3.3 TASK 1930 – HIGH-POWER RODS IN EBR-II

Introduction and Past Results

The objective of the high-power rod experiments is to extend the investigation of the irradiation behavior of helium-bonded (UPu)C fuel to higher linear power. Fission gas release and restrained swelling data are to be obtained for a total of six rods in an EBR-II subassembly with a maximum linear power of 30 kw/ft. The design variables include Incoloy-800 and Type 316 stainless steel cladding, high- and low-density fuel pellets, and fuel burnups of 50,000 to 100,000 Mwd/T.

The design was completed and the test parameters for the capsules are given in Table 1.

The six rods, UNC 185 through 190, were fabricated, encapsulated in the standard EBR-II capsules, sodium-bonded, and shipped to the EBR-II for irradiation.

Current Results

The insertion of the six rods into the EBR-II has been scheduled for May 1969.

3.4 TASK 1950 – LOW-POWER SUBASSEMBLY IN EBR-II (19 RODS)

Introduction and Past Results

The objective of the experiment is to obtain performance data on uranium-plutonium carbide fuel rods at low powers. Fuel swelling data are of particular interest, because a large portion of an LMFBR will be operating at less than the 20 to 30 kw/ft peak power range at which the carbides are expected to operate.

The design was completed and the test variables are given in Table 1. A program was completed to develop an end-plug design for use with an automatic welder.

The preparation of all fuel rod components has been completed.

Fuel fabrication of the first ($U_{0.85}Pu_{0.15}$)C subassembly to be fabricated in the pilot facility commenced.

Current Results

High-density ($U_{0.85}Pu_{0.15}$)C pellets prepared from a 1.8-kg batch of 60% enriched carbide for Rods UNC 137, 138, 139, 143, 144, 146, and 147 were not acceptable because of a 3% dioxide phase. The dioxide was mainly present as discrete particles having a mean diameter of 25 microns. This is the first time that discernible particles of MO_2 have been detected in carbide at this facility.

A second attempt produced a 1.8-kg batch of acceptable pellets for these rods. Both solid and cored pellets were made. All pellets were surface ground, degreased, and degassed. Low-density pellets of 75 and 84% of theoretical were prepared for Rods UNC 129 and 131 through 136. A summary of the characterization of the fuel for the 19 rods in the subassembly is given in Tables 3, 4, and 5.

TABLE 3 — U-Pu CARBIDE PELLET CHARACTERIZATION FOR THE
LOW-POWER 19-ROD SUBASSEMBLY, UNC 129 THROUGH 147 (TASK 1950)

UNC Rod No.	Clad Material	Pellet Diameter, in.		Sintered Density		
		Intended	Avg, Measured	Intended % T.D. $\pm 2\%$	Avg, g/cm ³	Actual % T.D.
129	316 SS	0.244	0.2440 $\pm$ 0.0007	84	11.54	86
130	316 SS	0.250	0.2496 $\pm$ 0.0004	75	10.34	77
131	316 SS	0.246	0.2458 $\pm$ 0.0004	84	11.49	85
132	316 SS	0.246	0.2458 $\pm$ 0.0004	84	11.49	85
133	316 SS	0.246	0.2458 $\pm$ 0.0004	84	11.49	85
134	316 SS	0.246	0.2458 $\pm$ 0.0004	84	11.49	85
135	Incoloy-800	0.246	0.2458 $\pm$ 0.0004	84	11.49	85
136	Incoloy-800	0.246	0.2458 $\pm$ 0.0004	84	11.49	85
137	316 SS	0.246	0.2459 $\pm$ 0.0003	97	12.94	97
138	316 SS	0.246	0.2459 $\pm$ 0.0003	97	12.94	97
139	Incoloy-800	0.246	0.2459 $\pm$ 0.0003	97	12.94	97
140	Incoloy-800	0.246	0.2460 $\pm$ 0.0005	93	12.16	91
141	316 SS	0.250	0.2501 $\pm$ 0.0004	93	12.31	92
142	316 SS	0.250	0.2501 $\pm$ 0.0004	93	12.31	92
143*	Incoloy-800	0.246	0.2459 $\pm$ 0.0003	97	12.81	96
144*	316 SS	0.246	0.2459 $\pm$ 0.0003	97	12.81	96
145†	304 SS	0.240	0.2399 $\pm$ 0.0005	93	12.18	91
146†	304 SS	0.240	0.2397 $\pm$ 0.0004	97	12.96	97
147†	Incoloy-800	0.240	0.2397 $\pm$ 0.0004	97	12.96	97

*Cored pellets (0.080-in. ID) — all other pellets solid.

†Na bonded, all other rods He bonded.

TABLE 4 — U-Pu CARBIDE PELLET CHARACTERIZATION FOR THE LOW-POWER 19-ROD SUBASSEMBLY, UNC 129 THROUGH 147 (TASK 1950)

<u>UNC Rod No.</u>	<u>Carbon, w o</u>	<u>Plutonium w o</u>	<u>Oxygen, ppm</u>	<u>Nitrogen, ppm</u>	<u>Equivalent Carbon, w o</u>	<u>Lattice Parameter (Å) and Phases</u>	<u>Metallography</u>
129, 131, 132, 133, 134, 135, 136	4.63 ± 0.06	14.54 ± 0.05	2525 175	125 ± 15	4.83	Single-phase (UPu)C $a_0 = 4.9655 \pm 0.0002$	Single-phase (UPu)C
130	4.72 ± 0.04	14.57 ± 0.09	2135 150	95 ± 80	4.88	Single-phase (UPu)C $a_0 = 4.9659 \pm 0.0005$	Single-phase (UPu)C
137, 138, 139, 143, 144, 146, 147	5.11 ± 0.04	14.55 ± 0.02	4355 410	200 ± 10	5.45	77 v o (UPu)C 4.9652 ± 0.003 and 23 v o (UPu) ₂ C ₃ 8.0951 ± 0.0005	80 v o (UPu)C 20 v o (UPu) ₂ C ₃
140, 141	4.53 ± 0.08	14.49 ± 0.06	3365 535	215 ± 2	4.80	Single-phase (UPu)C $a_1 = 4.9621 \pm 0.0003$	Single-phase (UPu)C
142, 145	4.50 ± 0.04	14.57 ± 11	3910 580	215 ± 2	4.82	Single-phase (UPu)C $a = 4.9626 \pm 0.0003$	Single-phase (UPu)C

TABLE 5 — U-Pu CARBIDE PELLET CHARACTERIZATION FOR THE
LOW-POWER 19-ROD SUBASSEMBLY, UNC 129 THROUGH 147 (TASK 1950)

Element	UNC Rods 129, 131, 132, 133, 134, 135, 136	UNC Rod 130	UNC Rods 140, 141, 142, 145	UNC Rods 137, 138, 139, 143, 144, 146, 147
Ag	< 0.5	< 0.5	< 0.1	0.91
Al	< 5	< 5	5	5
As	< 10	< 10	< 10	< 10
Au	< 0.5	< 0.5	< 0.5	< 0.5
B	2.2	0.97	3.4	1.1
Ba	< 10	< 10	< 10	11
Be	< 2	< 2	< 2	< 2
Bi	< 2	< 2	< 2	< 2
Ca	< 10	< 10	< 10	< 10
Cd	< 0.5	< 0.5	< 0.5	< 0.5
Co	< 1	< 1	9.4	6
Cr	16	31	18	42
Cs	< 5	< 5	< 5	< 5
Cu	23	30	39	19
Fe	< 10	88	15	185
Ga	< 10	< 10	< 10	< 10
Ge	< 10	< 10	< 2	< 10
Hg	< 10	< 10	< 10	< 10
In	< 10	< 10	< 10	< 10
K	1	1	5	1
Li	7	7	15	7
Mg	5.9	10.5	10	< 1
Mn	4.4	6.4	3.6	8.6
Mo	< 2	< 2	36	< 2
Na	< 1	< 1	< 1	< 1
Nb	< 5	< 5	< 5	< 5
Ni	< 10	12	< 10	>500
P	< 25	< 25	< 25	< 25
Pb	9	19	7.3	27
Rf	< 5	< 5	< 5	< 5
Ru	< 3	< 3	< 3	< 3
Sb	< 3	< 3	< 3	< 3
Si	30	43	30	23
Sn	< 2	< 2	< 2	< 2
Sr	< 50	< 50	< 50	< 50
Ta	<100	<100	<100	<100
Te	< 10	< 10	< 10	< 10
Ti	1.6	1.7	1.5	< 1
V	< 5	< 5	< 5	< 5
W	< 50	< 50	< 50	< 50
Zn	< 10	< 10	< 10	17
Zr	< 5	< 5	< 5	< 5

A production batch of 280 high-density single-phase $(U_{0.85}Pu_{0.15})C$ pellets was fabricated for assembly of three helium-bonded rods, UNC 140, 141, 142, and one sodium-bonded rod, UNC 145.

All of the fuel rods were loaded. Rods UNC 138 through 144 were inspected and loaded into the standard EBR-II capsules. The 19 rods are scheduled to be shipped by February 1, 1969.

The low-power 19-rod subassembly has been designated X055 and is scheduled for insertion in the EBR-II for Run 33 (~March 1969).

3.5 TASK 1960 – HIGH-POWER SUBASSEMBLY IN EBR-II (19 RODS)

Introduction

The objective of the 19-rod experiment is to extend the current irradiation study to higher powers (30 to 35 kw/ft).

High linear and specific powers at high burnup are necessary in fast reactor fuels to reduce fuel cycle costs significantly. The most critical problem of high-power, high-burnup fuels is the accommodation of fuel swelling without rupturing the clad.

The classical solution is a controlled helium gap between the fuel and clad. As an alternative approach, accommodation for swelling can be provided within the fuel. This can be in the form of a central void (cored pellets) or as uniformly distributed porosity. The third approach is the use of sodium fuel-to-clad bond. The sodium provides expansion space, eliminates clad strain, and reduces fuel temperatures.

The design of the 19-rod subassembly is complete and test variables are given in Table 1.

Current Results

The bids for the cladding material have been received and are being evaluated.

3.6 TASK 1500 – UNRESTRAINED SWELLING AND FISSION GAS RELEASE

Introduction and Past Results

The objective of the experiments is to investigate the unrestrained swelling and fission gas release characteristics of several types of (UPu)C fuel as a function of irradiation exposure and temperature. Since temperature measurement is essential to obtain meaningful data, the experiments have to be carried out in instrumented capsules; this is feasible in thermal reactors only. The problem of flux depression and resulting large ΔT 's, which are not representative of fast reactors, is resolved by the use of small cross-section specimens operating at essentially isothermal conditions.

Design of the two capsule trains was completed and a description of the capsule trains was given in the third quarterly report, UNC-5217.

A capsule to measure the neutron flux in the experimental facility was designed, fabricated, and irradiated. Evaluation of the flux monitor data showed good agreement between the measured and calculated neutron flux in the fuel.

The gas control systems and control consoles have been fabricated by Douglas-United Nuclear at the reactor experiment site.

Fabrication of the various U-Pu carbide fuels for the fission gas release study is complete.

Assembly of the fission gas release capsule train was initiated. Fabrication of 0.060-in. diameter by 0.500-in. long fuel pins for the unrestrained swelling experiment was started. Fuel pins were prepared and assembled into Capsules UNC 117A and 119B.

Current Results

A thermal analysis of the mixed-oxide unrestrained swelling pins has shown that they can be operated in the capsule designed to operate at 1300°C for mixed carbide. The average temperature can be controlled to 1600°C, thus giving a direct comparison between the mixed oxide and mixed carbide at the 1600 C temperature.

Fabrication of the 84 and 93% of theoretical 0.060-in. diameter by 0.500-in. long pins was by cold compaction and sintering. The fuel specimens were pressed and sintered to the diametral tolerance of ± 0.001 in. without grinding. Characterization data for the carbide fuel loaded into Capsules UNC 116B, 117A, and 119B are shown in Tables 6, 7, 8, and 9.

TABLE 6 — CHARACTERIZATION OF 93% T.D. SINGLE-PHASE
(U_{0.85}Pu_{0.15})C PINS LOADED INTO CAPSULE UNC 117A
(TASK 1500)

No.	Weight, g	Length, in.	Average As-Sintered Diameter, * in.	Density, † g/cm ³
1	0.2910	0.4981	0.0596	12.77
2	0.2950	0.5050	0.0596	12.77
3	0.2921	0.4933	0.0600	12.77
4	0.2915	0.4990	0.0598	12.68
5	0.2950	0.4982	0.0603	12.65
6	0.2922	0.4999	0.0598	12.69
7	0.2910	0.4948	0.0598	12.77
8	0.2915	0.4977	0.0601	12.59
Average				12.71 ^{+ 0.06} - 0.12

*The intended "as-sintered" diameter was 0.060 ± 0.001 in. with length of 0.500 ± 0.005 in.

†The theoretical density for (U_{0.85}Pu_{0.15})C is 13.45 g/cm³ as determined from lattice parameter and the composition of the material used. Each of the primary capsules contained eight (UPu)C pins.

TABLE 7 — CHARACTERIZATION OF 93% T.D. SINGLE-PHASE
(U_{0.85}Pu_{0.15})C PINS LOADED INTO CAPSULE UNC 119B (TASK 1500)

No.	Weight g	Length, in.	Average As-Sintered Diameter, * in.	Density, † g/cm ³
1	0.2924	0.4999	0.0599	12.66
2	0.2919	0.5036	0.0596	12.67
3	0.2922	0.5033	0.0596	12.69
4	0.2912	0.5020	0.0596	12.68
5	0.2923	0.5021	0.0598	12.64
6	0.2926	0.5010	0.0598	12.68
7	0.2935	0.5015	0.0599	12.67
8	0.2910	0.5031	0.0600	12.50
Average				12.65 ^{+ 0.02} - 0.15

*The intended "as-sintered" diameter was 0.060 ± 0.001 in. with length of 0.500 ± 0.005 in.

†The theoretical density for (U_{0.85}Pu_{0.15})C is 13.45 g/cm³ as determined from lattice parameter and the composition of the material used. Each of the primary capsules contained eight (UPu)C pins.

TABLE 8 — CHARACTERIZATION OF 84% T.D. (U_{0.85}Pu_{0.15})C
PINS LOADED INTO CAPSULE UNC 116B (TASK 1500)

No.	Weight, g	Length, in.	Average As-Sintered Diameter, * in.	Density, † g/cm ³
1	0.2709	0.5030	0.0603	11.51
2	0.2711	0.5045	0.0604	11.44
3	0.2691	0.2973	0.0605	11.48
4	0.2710	0.5011	0.0605	11.48
5	0.2717	0.4989	0.0605	11.56
6	0.2716	0.5052	0.0602	11.54
7	0.2701	0.5050	0.0604	11.39
8	0.2708	0.4993	0.0605	11.53
Average:				11.49 ^{+0.07} -0.11

*The intended "as-sintered" diameter was 0.060 ± 0.001 in. and length of 0.500 ± 0.005 in.

†The theoretical density for (U_{0.85}Pu_{0.15})C is 13.43 g/cm³ as determined from lattice parameter and the composition of the material used. Each primary capsule contained eight (UPu)C pins.

TABLE 9 — CHARACTERIZATION OF 84% AND 93% T.D. ($U_{0.85}Pu_{0.15}$)C PINS FOR CAPSULES UNC 116B, 117A, AND 119B (TASK 1500)

<u>Element</u>	<u>Capsule 116B</u>	<u>Capsules 117A and 119B</u>
Plutonium, w/o	14.49 ± 0.04	14.19 ± 0.09
Carbon, w/o	4.68 ± 0.05	4.45 ± 0.08
Oxygen, ppm	1105 ± 215	2730 ± 690
Nitrogen, ppm	265 ± 20	415 ± 185
Equivalent carbon*	4.79	4.68
Lattice parameter, a_0	$4.9638 \pm 0.0005A$	$4.9617 \pm 0.0004A$

*The sum of C + 12/16 O + 12/14 N.

Metallography and X-ray diffraction showed that the 84 and 93% of theoretical (UPu)C pins were single-phase with no free metal or dioxide present. A slight increase in porosity was observed at the center section of the pins as a result of the high L/D pressing ratio.

Autoradiography showed a uniform distribution of plutonium.

(UPu)C powder to prepare pins for Capsule UNC 117B was prepared in the previous quarter. The pins were prepared during this quarter. Subsequent to a test sinter, the pins were pressed and sintered at 1750°C for 4 hr. Characterization of the pin loaded into Capsule 117B is given in Tables 10 and 11.

A blend of UO_2 -15 w/o PuO_2 (93% enriched UO_2 and burned metal prepared PuO_2) was prepared for fabrication of the pins for loading of Capsules UNC 118A and 118B by ball milling and sintered at 1700°C – 2 hr – to diametral and density tolerances. Metallographic examination showed a uniform distribution of porosity. Alpha autoradiography showed a uniform plutonium distribution. Characterization data are shown in Tables 12 and 13.

TABLE 10 — CHARACTERIZATION DATA FOR THE
(UPu)(OC) PINS, CAPSULE UNC 117B (TASK 1500)

No.	Weight, g	Length, in.	Average As-Sintered Diameter, in.	Density, g/cm ³
1	0.2881	0.4939	0.0596	12.76
2	0.2888	0.4959	0.0596	12.73
3	0.2860	0.4935	0.0597	12.63
4	0.2892	0.4931	0.0597	12.66
5	0.2873	0.4962	0.0596	12.66
6	0.2879	0.4941	0.0595	12.77
7	0.2860	0.4940	0.0594	12.75
8	0.2835	0.4938	0.0594	12.64

Average 12.70 ± 0.07

TABLE 11 — CHARACTERIZATION DATA FOR
THE (UPu)(OC) PINS, CAPSULE UNC 117B
(TASK 1500)

Plutonium, w/o	13.91 ± 0.33
Carbon, w/o	2.90 ± 0.13
Oxygen, w/o	1.71 ± 0.17
Nitrogen, ppm	280
Lattice Parameters, a_0	$4.9535 \pm 0.0008A$

TABLE 12 — CHARACTERIZATION DATA FOR THE
(U_{0.85}Pu_{0.15})O₂ PINS, CAPSULE UNC 118A (TASK 1500)

<u>No.</u>	<u>Weight</u> <u>g</u>	<u>Length, in.</u>	<u>Average</u> <u>As-Sintered</u> <u>Diameter, in.</u>	<u>Density,</u> <u>g/cm³</u>
1	0.2360	0.4940	0.0599	10.48
2	0.2410	0.5043	0.0596	10.45
3	0.2390	0.5020	0.0597	10.38
4	0.2410	0.5040	0.0597	10.42
5	0.2371	0.5008	0.0598	10.28
6	0.2392	0.5011	0.0598	10.37
7	0.2380	0.4973	0.0598	10.40
8	0.2395	0.5051	0.0598	10.30
			Average 0.0598 ±0.0001	Average 10.38 ±0.07 is 96.4% T.D.

TABLE 13 — CHARACTERIZATION DATA FOR THE
(U_{0.85}Pu_{0.15})O₂ PINS, CAPSULE UNC 118B (TASK 1500)

<u>No.</u>	<u>Weight</u> <u>g</u>	<u>Length,</u> <u>in.</u>	<u>Average</u> <u>As-Sintered</u> <u>Diameter, in.</u>	<u>Density,</u> <u>g/cm³</u>
1	0.2405	0.5052	0.0597	10.38
2	0.2410	0.5054	0.0600	10.29
3	0.2405	0.5016	0.0599	10.38
4	0.2385	0.4990	0.0598	10.38
5	0.2391	0.5004	0.0599	10.35
6	0.2385	0.5000	0.0599	10.33
7	0.2383	0.4995	0.0598	10.36
8	0.2380	0.4945	0.0598	10.35
			Average 0.0598 ±0.0001	Average 10.35 ±0.03 is 96.1% T.D.

Both the unrestrained swelling and fission gas release capsule trains were assembled and shipped to Douglas-United Nuclear for irradiation. A summary of the design parameters and irradiation conditions is given in Table 14.

TABLE 14 — CAPSULE FUEL MATERIALS AND OPERATING DATA (TASK 1500)

Unrestrained Swelling Experiment					
Capsule	Fuel Geometry, *	Fuel	Fuel		Burnup,
No.	in.	Fuel Type†	Density,%	Temp, °C	Mwd/T
116 A	0.060 OD Pin	MC + M ₂ C ₃ ‡	98	1300	20,000
116 B		MC	84	1300	
117 A		MC	93	1300	
117 B		M(OC)	75	1300	
118 A		(UPu)O ₂	93	1300	
118 B		(UPu)O ₂	93	1300	
119 A		MC + M ₂ C ₃ ‡	98	1600	
119 B		MC	84	1600	
Fission Gas Release Experiment					
120 A	0.375 OD × 0.255 ID pellet	MC + M ₂ C ₃ ‡	98	1000	20,000
120 B		MC	84	1000	
121 A		MC + M ₂ C ₃ ‡	98	1300	
121 B		MC	84	1300	
122 A		MC	93	1300	
122 B		MC	75	1300	
123 A		M ₂ C ₃	93	1300	
123 B		M(OC)	93	1300	
124 A		MC + M ₂ C ₃ ‡	98	1600	
124 B		MC	84	1600	

*Length = 0.500 in.

†M = (U_{0.85}Pu_{0.15}), U = 93% U²³⁵‡v/o MC + 10 v/o M₂C₃

3.7 TASK 1800 – SODIUM-BONDED FUEL EVALUATION

Introduction and Past Results

The objective of the irradiations is to evaluate the performance of sodium-bonded, uranium-plutonium carbide rods.

Four rods will be irradiated in instrumented capsules in GETR-RAFT facilities. The test parameters for the capsules are given in Table 1.

The fuel rod is in a sodium-filled inner shell along with a heater designed to melt the sodium prior to startup. Thermocouples are placed in the inner and outer shell. The annulus between the inner and outer shell is filled with NaK.

Fabrication of the control consoles was completed and they were shipped to GETR.

Fabrication of the fuel rods is complete. This represents the first time a long stack (14-in.) of uranium-plutonium carbide fuel pellets has been bonded with sodium.

Assembly of Capsules UNC 125, 126, and 128 has been completed. The capsules were functionally checked and shipped to the GETR.

Current Results

Capsules UNC 125, 126, and 128 were inserted in the GETR in October. The capsules have operated according to design (25 kw/ft) for one and one-half cycles without any problems. Burnup to date is ~10,000 Mwd/T. Capsule UNC 127 was inserted in the GETR in December and has operated for one-half of a cycle according to design (25 kw/ft). Prior to experiment assembly, the unsatisfactory end plug on fuel rod UNC 127 was replaced by welding in a new end plug.

3.8 TASK 1900 – FABRICATION DEVELOPMENT

3.9 TASK 1911 – QUALITY ASSURANCE AND ANALYTICAL METHODS DEVELOPMENT

Introduction and Past Results

Quality control is a continuous program which is directed toward economy and quality through improvement, development, and application of analytical techniques.

Analytical capabilities for plutonium, uranium, carbon, oxygen, nitrogen, oxygen/metal ratio, and lattice parameter for characterization of uranium-plutonium carbide products were established. Each method capability is subject to change, and new methods must be developed to be commensurate with the fuel specification and fabrication process.

X-ray fluorescence analyses have been evaluated to provide rapid analyses for nickel, uranium, and plutonium in a uranium-plutonium carbide matrix. Analyses showed that the method is good for determining Pu and U/Pu within about 1% of the actual value.

A sampling plan was prepared and instituted for the statistical comparison of pertinent physical and chemical variations of uranium-plutonium carbide prepared in the pilot plant equipment.

Current Results

Analytical support was given to the carbide fabrication under Task 1911, as well as work on carbon and plutonium analyses. Subsequently, the task was completed and closed.

Statistical evaluation of long-term performance of the NBS 4J cast iron standard was continued. The performance of the standard was not always a good indication of analytical control on samples. The lower carbon weight percentage of the standard generated smaller volumes of CO_2 than the carbide samples. Absorption difficulties for the amount of CO_2 produced by the standard were minimal. To equate CO_2 collection between samples and standards, the standard sample size was increased from 0.30 to 0.46 grams. This standard sample size generates an equivalent weight of CO_2 yielding a more realistic measurement of carbide sample control.

The feasibility of using ceric sulfate as a control for plutonium accuracy on a day-to-day basis was investigated. Initial work indicates that some difficulty may be experienced with this reagent, as the ceric ion produces a negative diffusion current at the calomel electrode. This cathodic reduction is no problem in itself, but in titrating cerate against ferrous iron, it appeared at first that the ability to establish a residual current or base line at the applied voltage chosen would be questionable.

3.10 TASK 1941 – PROCESS IMPROVEMENT

Introduction and Past Results

The objective of this task is to improve process control, reduce the cost of fabricating carbide fuel, and to develop fabrication methods for oxide fuel.

The carbide fabrication development studies were terminated, because of a shift of emphasis to oxide fuels. The oxide fabrication development, previously Task 1970, was shifted to a new task, Task 2400.

A. Pilot Plant Process

The first step toward cost reduction is a one-step agglomeration of the synthesis feed ($\text{UO}_2 + \text{PuO}_2 + \text{C}$) in a P-K blender, instead of the two-step process of blending the synthesis feed by ball-milling followed by pressing pellets. Kilo-gram batches of uniform UC powder, and subsequently UC pellets, were made by this process. The level of impurity in the starting material was not significantly increased by processing in the pilot plant equipment.

The first (UPu)C batch was processed in the pilot facility utilizing the same process as the UC analog. The synthesized product was homogeneous. Three lots of pellets were pressed and sintered to produce high-density, two-phase, high-density, single-phase, and 84% of theoretical pellets. The results indicate that the pilot process is capable of producing pellet densities to within less than 1% of the intended, with as-sintered diameter and height tolerances of 0.001 and 0.005, respectively.

B. Uranium-Plutonium Carbide Specimens for Thermal Diffusivity Measurement

Fabrication of thermal diffusivity specimens for cooperative property measurements at Argonne National Laboratory is in process. Samples to be prepared

are 75 and 84% T.D. (UPu)C, 90% T.D. single-phase (UPu)C, 97% T.D. (UPu)C + 10 v/o (UPu)₂C₃, 90% T.D. (UPu)(OC), and 90% T.D. (UPu)₂C₃. All samples have been completed and prepared for shipment to ANL.

C. Conversion of Uranium-Plutonium Carbides to Oxides

In order to return U-Pu carbide scrap to the AEC-Richland for reprocessing, it must be received there as oxide, as they have no inert atmosphere to handle carbide. Accordingly, a furnace built for oxidation of Pu metal to PuO₂ was installed for (UPu)C oxidation. A pilot batch of UC was only half converted to oxide at 500°C for 6 hr. An additional oxidation run was made to provide a total of 130 g of converted U-Pu carbide powder. The carbon content was 0.25 w/o. A carbon content of 0.25 w/o is acceptable to the AEC for storage and processing of recoverable scrap.

Since 3½ kg of (UPu)C scrap has to be converted, a larger oxidation furnace is being considered.

Current Results

A. Specimens for Carbide-Sodium-Clad Compatibility Studies

Thirty-five high-density (U_{0.85}Pu_{0.15})C + 10 v/o (UPu)₂C₃ pellets were prepared to specification for compatibility studies at Los Alamos Scientific Laboratory.

The carbide was prepared by carbothermic reduction of compacts prepared from a mechanical blend of depleted UO₂ and PuO₂ at 1550°C under vacuum.

The pellets were sintered at 1550°C-1 hr under vacuum, and surface ground to remove 0.006 in. from the diameter. This treatment was given to remove the single-phase layer of monocarbide that forms during sintering, and thereby to expose the representative (UPu)C + 10 v/o (UPu)₂C₃ structure.

Characterization data for the 35 pellets shipped to LASL is shown in Table 15.

TABLE 15 — CHARACTERIZATION DATA* FOR THE 35 HIGH-DENSITY ($U_{0.85}Pu_{0.15}$)C + 10 v/o (UPu) $_2$ C $_3$ PELLETS SHIPPED TO LASL FOR SODIUM COMPATIBILITY STUDIES (TASK 1941)

Carbon, w/o	4.79 ± 0.05
Oxygen, ppm	3710 ± 50
Nitrogen, ppm	240 ± 20
Equivalent Carbon, w/o	5.09
Plutonium, w/o	14.09 ± 0.06
Lattice Parameter, a_0	$4.9654 \pm 0.0004 \text{ \AA} \text{ (UPu)C}$ $8.1080 \pm 0.002 \text{ \AA} \text{ (UPu)}_2\text{C}_3$
Metallography	90 v/o (UPu)C + 10 v/o (UPu) $_2$ C $_3$
Pellet Density, g/cm 3	13.15 ± 0.04
Pellet Height, in.	0.250 ± 0.005
Pellet Diameter, in.	0.2310 ± 0.0004

*The variability shown was calculated at one standard deviation. The fuel was prepared from depleted uranium.

3.11 TASK 2400 – URANIUM-PLUTONIUM DIOXIDE PELLET FABRICATION

Introduction and Past Results

The purpose of this task is to develop methods for producing well characterized (UPu)O₂ pellets and fuel rods of the LMFBR/FFTF type. The fuel produced will be subjected to a high level of quality control. The quality control methods themselves will be established and improved to the levels required by the specification.

A. Fabrication

Two processes are being evaluated for preparation of press feed in the pilot plant process equipment. The two processes were described in the fourth quarterly report, UNC-5224.

Pressing and sintering studies, with UNC depleted UO₂ and burned PuO₂, to evaluate the effect of variations in cold-formed density showed that a 0.1 g/cm³ change in cold-formed density results in a 0.001-in. change in sintered pellet diameter. The importance of control of press feed characteristics and press performance in obtaining diametral reproducibility is apparent. The density and diametral reproducibility for fabrication of sintered dioxide pellets was ± 0.04 g/cm³ and ± 0.001 in., respectively. The oxygen-to-metal ratio was 1.95, and X-ray diffraction analysis established that complete solid solution was not obtained.

UO₂-25 w/o PuO₂ press feed containing 0.25 w/o PVA and 0.7 w/o zinc stearate was milled in an attempt to reduce particle size and improve homogeneity. Pellets were cold pressed directly from the milled powder and sintered. X-ray diffraction and O/M analysis showed complete solid solution was not obtained and the O/M was 1.94.

B. Characterization

A Perkin-Elmer Shell Sorptometer is being adopted to a glove box to measure surface area of fuel powders.

Development continued of the instrument for measuring the open and closed porosity of fuel pellets by comparison of geometric and helium displacement volumes.

Current Results

A. Fabrication

There was no fabrication work during this quarter.

B. Characterization

A Perkin-Elmer Shell Sorptometer is being set up to measure surface area of plutonium-containing powders. A standard instrument will be adapted to a glove box by placing the sample tube inside the box and keeping the rest of the unit outside. Tests to determine the effect of using filters to prevent contamination outside the box and required connecting lines for glove box setup were completed. A National Bureau of Standards anatase TiO_2 , with a certified surface area of $10.30 \pm 0.2 \text{ m}^2/\text{g}$, was used to evaluate the effect of filters and additional lines on instrument accuracy and precision. Surface area values of 10.43 ± 1.0 , 9.97 , and $10.36 \pm 0.25 \text{ m}^2/\text{g}$ were obtained, respectively. The results indicate that the instrument can be adapted for glove box use, using the necessary filters and additional tubing, without a significant loss of accuracy.

A sample holder assembly has been designed and fabricated which will eliminate the need to bring the liquid nitrogen used for sample cooling into the glove box.

Experimental studies are in process to determine the accuracy and precision of the instrument for measuring of open and closed porosity in 1/4-in. diameter pellets. Porosity is determined by comparison of geometric and helium displacement volumes.

The volume and reproducibility in measurement of the sample chamber, and of a 1/4-in. diameter by 1/4-in. long pellet, were determined. Sample chamber volume was $0.7106 \pm 0.0008 \text{ cm}^3$. Pellet volume was $0.2044 \pm 0.0016 \text{ cm}^3$, compared to 0.2020 cm^3 absolute as determined by geometric measurements. The instrument is capable of meeting the required accuracy of $\pm 0.0020 \text{ cm}^3$ required for displacement volumes.

Three stainless steel samples of about 15, 30, and 45% porosity were tested for primary certification of sample pore spectrum as well as total porosity by the American Instrument Company.

The standard samples now will be tested in the UNC apparatus and their porosities compared with results from the mercury immersion measurements.

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