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DEVELOPMENT REPORT

FAST REACTOR MIXED-CARBIDE FUEL ELEMENT
DEVELOPMENT PROGRAM

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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 1100 – EBR-II Pilot Rods (Three Rods)

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

1920 – Statistical Evaluation of Subassembly in EBR-II (37 Rods)

1930 – High-Power Rods in EBR-II

1941 – Process Improvement

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

Task 2200 – Sodium Compatibility and Bond Testing

2250 – Carbide Stability

Progress during the period January 1, 1968 to March 31, 1968 on the currently active tasks of this program is described in subsequent sections.

This is the third 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 progress 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 Report 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).

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).

2. HIGHLIGHTS OF THE CURRENT QUARTER

The final report on the post-irradiation examination of the first high-density, uranium-plutonium carbide pellet rods irradiated in EBR-II was published and distributed. The report concludes that, if the fuel swelling rate stays the same as it has up to 30,000 Mwd/T at 28 kw/ft, helium-bonded rods will be able to achieve 100,000 Mwd/T at 20 to 30 kw/ft.

The first all uranium-plutonium carbide subassembly ever to be irradiated is operating in the EBR-II at about 20 kw/ft. An exposure of 0.7×10^{20} fiss/cm³ (2000 Mwd/T) has been accumulated. The rods have smear densities in the range 70 to 85% and contain high-density annular pellets (97% of theoretical) and low-density (75 and 84% of theoretical) solid pellets. All are helium bonded.

Bubbles were removed successfully from the sodium fuel to clad bond of the four previously assembled uranium-plutonium carbide rods, UNC 125 through 128. This represents the first long (14-in.) pelletized uranium-plutonium carbide fuel columns that have ever been sodium bonded.

Assembly of the 10 fission gas release specimen assemblies containing uranium-plutonium carbide annular pellets was completed.

Homogeneous 2-kg UO₂ + C blends were made in the pilot plant facility without statistical differences between them in carbon content. High-density (90% T. D.) single-phase UC containing 370 ppm oxygen, and high-density (97% T.D.)

UC + U₂C₃ pellets were prepared from carbide produced using the pilot plant process.

Reaction sintering produced good quality pellets; however a pellet density of only 70% of theoretical was obtained. The minimum acceptable goal was 75% T.D.

Uranium-plutonium carbide specimens were fabricated from various carbide compositions for thermal diffusivity measurement.

Uranium-plutonium dioxide pellets were prepared for analytical development studies. The (U-25 w/o Pu)O₂ pellets were 92% of theoretical density. The sintered pellet diametral variation was less than ±0.001 in.

Measurements of single-phase monocarbides (UPu)C show that the fuel is kinetically and thermodynamically compatible* with stainless steel at 800°C. Measurements at 800°C of (UPu)C + 14 w/o (UPu)₂C₃ and single-phase (UPu)₂C₃ show that they are not thermodynamically compatible. Kinetic compatibility* of (UPu)C + (UPu)₂C₃ was previously established in static capsule tests. Single-phase (UPu)₂C₃ is kinetically incompatible under similar test conditions.

* Kinetic compatibility is an acceptable degree of carbon transfer from fuel to stainless steel within the service period. Thermodynamic compatibility is an acceptable degree of carbon transfer (less than 0.7 w/o carbon surface concentration in stainless steel) in any amount of time.

3. PAST RESULTS AND CURRENT PROGRESS

FAST REACTOR IRRADIATIONS

3.1 TASK 1100 – EBR-II PILOT RODS (3 RODS)

Introduction and Past Results

Three mixed-carbide pilot rods were irradiated to obtain results at a meaningful burnup in the minimum length of time.

The three helium-bonded rods fueled with high-density, hyperstoichiometric structure ($U_{0.8}Pu_{0.2}$)C pellets performed as designed during irradiation at the EBR-II. The rods operated up to a power of 28 kw/ft to a peak burnup of about 10.6×10^{20} fiss/cm³ (30,000 Mwd/T) based on Nd¹⁴⁸ and U-Pu isotopic analyses. This is the highest power at which mixed carbides have been irradiated in a fast reactor. The design conditions are summarized in Table 1.

There were no reactions between the fuel and the type 316 stainless steel or vanadium clad and a slight reaction with the vanadium-20 w/o titanium. Clad sections from all rods were found to be ductile when bent at room temperature.

The fuel rods performed as predicted in these experiments. The good correlation between predicted and actual clad strain was particularly gratifying.

If uranium-plutonium carbide fuel continues to behave in this manner at higher burnups, helium-bonded high-density pellets will be able to achieve 100,000 Mwd/T at 20 to 30 kw/ft.

Current Results

The topical report, UNC-5198, was issued.

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			Cladding Material	Maximum Linear Power (Design), kw/ft	Target Burnup, Mwd/T × 10 ⁻³	Facility	Operating Maximum Linear Power, kw/ft	Accumulated Burnup, Mwd/T × 10 ⁻³		
					Pellet (Type)	Smeared	%PuC								
1100	78-80	EBR-II pilot rods, helium-bonded, (3 rods)	0.251 to 0.252	14	95 (solid)	91	20	V-20 Ti, V, 316 SS	20 to 30	20 to 30	EBR-II	19 to 28	19 to 28	Irradiation and topical report completed.	
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	5800 (81 to 86), 15,000 (87)		
	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 In-coloy-800	24 to 30	40	EBR-II	22 to 28	15,000		
1300	91-114	EBR-II rods, he- lium-bonded (24 rods)	0.232 to 0.269	14	75 to 84 (solid), 99 (cored)	70 to 85	15	316 SS and In- coloy-800	20	50 to 100	EBR-II	—	—	19-rod assembly being irradiated	
1500	115-119	Unrestrained swelling spec- imens	0.06	0.5	79 to 90 (solid)	—	15	—	Low	20	Hanford	—	—	Capsules in fabrication.	
	120-124	Fission gas release spec- imens	0.375 OD, 0.255 ID	0.500	75 to 98 (annular)	—	15	—	Low	20	Hanford	—	—	Capsules in fabrication. Fuel completed.	
1800	125-128	Sodium-bonded rods (4 rods)	0.240	14	93 to 98 (solid)	83 to 87	15	304 SS and In- coloy-800	25	50	GETR	—	—	Fuel rod assembled and capsules in fabrication.	
1950	129-147	19-rod low- power subassem- bly, helium and sodium bonds	0.240 to 0.246	14	75 to 98	70 to 87	15	316 SS, 304 SS and In- coloy-800	15	100	EBR-II	—	—	Fuel rod and capsule hardware in fabrication.	
1920	148-184	37-rod subassem- bly, helium bond	0.241	14	84 to 96 (solid)	78 to 91	15	316 SS, 304 SS, and In- coloy-800	20	100	EBR-II	—	—	Design complete. Work on this task was ter- minated.	
1930	185-190	High-power rods (6 rods)	0.249 to 0.253	14	84 to 96 (solid)	80 to 88	15	316 SS and In- coloy-800	30	50 to 100	EBR-II	—	—	Design and fuel rod hard- ware complete. Fuel in fabrication.	

3.2 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, min.) 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 (UNC rods 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.

Current Results

The calculated burnup of the nine rods presently in EBR-II is as follows:

1. Rods 81 through 86 (19 kw/ft) – 14,100 Mwd/T
2. Rods 87, 98, 90 (28 kw/ft) – 26,400 Mwd/T – scheduled for discharge at the end of run 29 (July 1968) with an exposure of 39,000 Mwd/T
3. Rod 88 – not yet scheduled for insertion; no estimate available.

3.3 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 (UNC rods 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, and assembled by them into subassembly X033. 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 2000 Mwd/T.

3.4 TASK 1920 – STATISTICAL EVALUATION OF SUBASSEMBLY IN EBR-II (37 RODS)

Introduction and Past Results

The objective of the irradiation is to obtain statistical fission gas and restrained swelling data on clad (UPu)C fuel. The variables include high and low density solid fuel pellets, Incoloy-800 and stainless steel clads. Details are given in Table 1.

The design of 37 (UPu)C rods for irradiation in EBR-II was completed. These rods are to be unencapsulated and irradiated at 20 kw/ft to burnups of up to 100,000 Mwd/T. The design was made flexible so that, if unencapsulated rods are not acceptable, the rods can be used in an encapsulated design for 19-rod sub-assemblies. Examination at intermediate burnups was planned.

Current Results

The design was completed and work on this task was stopped. The sub-assembly will not be fabricated since the program emphasis is now directed toward obtaining additional basic data rather than statistical data.

3.5 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 burnup of 50,000 to 100,000 Mwd/T.

The design and predicted performance analyses were nearly completed.

Current Results

The design and predicted performance analyses were completed. They are essentially as reported in the previous quarterly, UNC-5201, and Table 1 of this report.

The hardware for fuel rods UNC 185 through 190 was completed and fuel fabrication started. Assembly of the six rods is scheduled for June 1968.

3.6 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 variables are given in Table 1.

The design of 19 (UPu)C rods for irradiation in EBR-II was completed. Fabrication of fuel rod and capsule hardware continued, and materials for pellet fabrication were ordered. These rods are to be double encapsulated, and irradiated at 10 to 15 kw/ft to burnups of up to 100,000 Mwd/T.

Current Results

Fabrication of fuel rod and capsule hardware is in process.

The PuO_2 and 60% enriched UO_2 for fabrication were received and compaction dies for pelletizing of fuel for all rods ordered. The 60% enriched UO_2 was rejected because the phosphorous impurity content of 500 ppm exceeded the fuel specification of 200 ppm. Fabrication of the 19-rod subassembly will be delayed until an acceptable UO_2 fuel is received. At present, delivery is estimated for the latter part of June 1968.

3.7 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, 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.

The design for unrestrained swelling consists of eight pin-type, 0.060-in. diameter individual specimens in one can, while the design for fission gas release consists of one annular fuel cylinder 0.375-in. OD by 0.255-in. ID per can (Fig. 1). The unrestrained swelling specimen is a pin, to simulate the shape of a fuel rod, and the fission gas release specimen is a thin-walled annular cylinder to provide accurate temperature measurement with a central thermocouple. Two fuel specimen cans are located in a secondary container (capsule) with central thermocouples in both specimen cans (Fig. 2). The temperature in each capsule is individually controlled by a helium-argon gas mixture. Five capsules make up a capsule train (Fig. 3). The design of two pilot capsule trains for irradiation in the Hanford Reactor is complete. The design conditions for the fuel specimens contained in these capsules are shown in Table 2.

The design of the experiment was completed. Fuel capsules, and capsule control system fabrication was in progress. A capsule to be used in measuring flux perturbation due to the capsule materials was designed, fabricated, and shipped to Douglas-United Nuclear for irradiation.

TABLE 2 — FISSION GAS RELEASE EXPERIMENT
DESIGN DATA *

<u>Capsule No.</u>	<u>Fuel Type †</u>	<u>Measured Specimen Density, % T.D.</u>	<u>Fuel Test Temp, °C</u>
120A	(UPu)C + (UPu) ₂ C ₃	97	1000
120B	(UPu)C	84	1000
121A	(UPu)C + (UPu) ₂ C ₃	97	1300
121B	(UPu)C	84	1300
122A	(UPu)C	93	1300
122B	(UPu)C	75	1300
123A	(UPu) ₂ C ₃	93	1300
123B	(UPu)(OC)	90	1300
124A	(UPu)C + (UPu) ₂ C ₃	97	1600
124B	(UPu)C	84	1600

*Burnup (all capsules): 20,000 Mwd/T.

†U/Pu ratio of 85/15.

Fig. 1 — Carbide Fuel Irradiation Experiment Specimen Assembly

TASK 1500
CARBIDE FUEL IRRADIATION EXPERIMENT
CAPSULE ASSEMBLY

Fig. 2 — Carbide Fuel Irradiation Experiment Capsule Assembly

Fig. 3 — Carbide Fuel Irradiation Experiment Capsule Train Assembly

Fabrication of the various U-Pu carbide fuels for the fission gas release study is in process. The carbides are being prepared by carbothermic reduction of the mixed dioxide followed by cold compaction and sintering in the temperature range 1550°C to 1850°C.

The annular, high-density specimens of both (UPu)C plus 10% (UPu)₂C₃, and (UPu)₂C₃ were fabricated. Diametral tolerances were met without surface grinding.

Current Results

Fuel fabrication of the remaining (UPu)C annular fission gas release specimens of 75, 84, and 93% T.D. and (UPu)(OC) of 90% T.D. was complete.

Intended diameter and length specifications of 0.375 ± 0.001 in. OD by 0.255 ± 0.002 in. ID by 0.500 ± 0.005 in. length were obtained without grinding, and intended density and chemical specifications were met. The 10 uranium-plutonium carbide fueled, tantalum specimen cans, 121A and 121B through 124A and 124B, were assembled and the fuel was characterized. The data are shown in Table 3.

The capsule intended to measure the flux in the experimental hole was withdrawn from the reactor and the flux wires counted. The evaluation of the data showed good agreement with the original calculations for neutron flux in the fuel.

The tantalum specimen assemblies (shown in Fig. 1) are in a niobium specimen assembly housing. The niobium secondary containment is contained in a stainless steel outer capsule shell. To prevent possible oxidation of the niobium housing (a Nb-1% Zr alloy) by minor amounts of impurities in the helium-argon control gas, a number of commercial, oxidation-resistant coatings were evaluated. Prototypic niobium alloy housings, coated with Sylcor R512E (an iron-chromium silicide coating), were found satisfactory. The coating will be used in the irradi-

TABLE 3 — FISSION GAS RELEASE EXPERIMENT FUEL SPECIMENS

Fuel Composition	Measured Specimen	Chemical Analysis*				Avg Annular Specimen Dimensions, † in.			Lattice Parameter, Å		Phases by Metallography and Amounts, v/o		
	Density, % T.D.	Carbon, w/o	Plutonium, † w/o	Oxygen, ppm	Nitrogen, ppm	OD	ID	Length	Major Phase	Minor Phase	(UPu)C	(UPu) ₂ C ₃	(UPu)(OC)
(UPu)C	74	4.69 ± 0.05	14.10 ± 0.13	3140 ± 390	210 ± 180	0.3750	0.2540	0.5008	4.9637 ± 0.0005	None detected	100	—	—
(UPu)C	83	4.62 ± 0.08	14.07 ± 0.06	1210 ± 180	180 ± 105	0.3746	0.2533	0.5048	4.9649 ± 0.0005	None detected	100	—	—
(UPu)C	92	4.51 ± 0.05	14.00 ± 0.02	2760 ± 210	245 ± 110	0.3755	0.2540	0.4958	4.9646 ± 0.0003	None detected	100	—	—
(UPu)C+10 w/o (UPu) ₂ C ₃	97	4.78 ± 0.14	14.37 ± 0.08	1505 ± 375	205 ± 160	0.3754	0.2568	0.5000	4.9660 ± 0.0003	8.0948 ± 0.0007	89	11	—
(UPu) ₂ C ₃	93	7.11 ± 0.11	14.11 ± 0.02	1715 ± 230	260 ± 130	0.3759	0.2563	0.4980	8.0931 ± 0.0011	None detected	—	100	—
(UPu)(O _{0.22} C _{0.18})	90	3.72 ± 0.10	13.10 ± 0.18	13,000 ± 1400	370 ± 140	0.3747	0.2535	0.5037	4.9597 ± 0.0002	None detected	—	—	100

*Product variability was calculated about the mean at the 95% confidence level.

†Intended U/Pu ratio 85/15.

‡Intended values were OD = 0.375 ± 0.001 in., ID = 0.255 ± 0.002 in., and length = 0.500 ± 0.005 in.

ation capsule. The machining of the final niobium housings was completed and electrical discharge machining of the thermocouple holes is in progress.

The hysteresis effect previously observed in the W/3 Re was corrected by annealing. Calibration of the W/3 Re-W/25 Re thermocouple wire was completed and sent to the thermocouple vendor for fabrication.

The sample copper-brazed tantalum-stainless steel thermocouple sheath junctions passed their high-temperature thermal cycle tests.

The gas control systems and control consoles are being installed at the reactor experimental site. The gas purification system was completed except for testing and final connections to the control console.

3.8 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 design of the fuel rod and capsule are shown in Figs. 4 and 5. The 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.

The design of the irradiation capsules and control consoles is complete. Fabrication of the capsule components and control consoles is in process. Fabrication of the uranium-plutonium carbide pellet fuels by carbothermic reduction of the dioxides, followed by cold pressing and sintering, and rod assembly are complete. This represented the first time a long stack (14-in.) of uranium-plutonium carbide containing fuel pellets has been bonded with sodium.

Bubbles were removed from the fuel-to-clad sodium bond of UNC 127.

Current Results

Bubbles were removed from the fuel-to-clad sodium bond of UNC 125, 126, and 128 by the process described in the previous Quarterly Report.

Capsule and capsule control system fabrication is in process.

The sample capsule heater completed an operational performance test at 10 kw for 41 hr. The required four heater leads and 10 thermocouples were successfully brazed into a prototypic capsule end plug.

TASK 1800
SODIUM BONDED CARBIDE FUEL IRRADIATION EXPERIMENT
INNER SHELL ASSEMBLY

Fig. 4 — Sodium-Bonded Carbide Fuel Irradiation Experiment Inner Shell Assembly

Fig. 5 — Sodium-Bonded Carbide Fuel Irradiation Experiment Capsule Assembly

3.9 TASK 1900 – FABRICATION DEVELOPMENT

3.9.1 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. The analytical methods and quality control program were found adequate for control of the current process. Each method capability is subject to change and new methods must be developed to be commensurate with the fuel specification, and fabrication process.

Development of a rapid X-ray fluorescence method for analysis of nickel, uranium, and plutonium is in process.

A new technique in preparing and storing the plutonium standard has been initiated. The standard is now made to serve as both a uranium and plutonium standard.

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 Method Improvement

Carbon

Analysis of degassed carbon (degassed to remove chemisorbed oxygen) by

the thermogravimetric method was discontinued. Carbon values obtained by this method were biased low. Carbon content is currently being measured by determining the weight loss of a sample after heating for one hour at 800°C in flowing helium.

Evaluation of NBS standard sample 66A Spiegeleisen with a certified carbon content of 4.39% was discontinued. Analysis indicated a greater variability than observed with the current 2.99% carbon content NBS cast iron standard 4j.

Materials have been ordered to make the thermogravimetric carbon apparatus capable of determining micro carbon levels in oxides. This includes a series of valves and a manometer set up for measurement of CO₂ produced from the ignition of micro amounts of carbon.

Uranium

A large variability has been observed in the determination of uranium from uranium-plutonium solutions. The nitrite oxidation step was found to be time-dependent, and amounts of phosphoric-acid additions critical. It was found theoretically and experimentally that rigid control is necessary in following the established analytical procedure. Modifications in the process should make the method more flexible and reliable.

Oxygen

Pre-outgassing of the inert gas fusion apparatus at 2200°C has reduced the blank rate with an associated improvement in the variability of the method.

The inert gas fusion method was modified by replacing the Hg manometer used for CO₂ detection. The new manometer has a smaller volume which results in greater reliability in the measurements taken.

NBS standard sample No. 1090 is being observed for use as an oxygen standard for the inert gas fusion method. This is an iron sample with a certified oxygen content of 484 ppm.

Nickel

X-ray fluorescence analyses are being evaluated to provide rapid analyses for nickel, uranium, and plutonium in a uranium-plutonium carbide matrix.

To develop a reliable method for the determination of nickel, a series of samples containing explicit ratios of U-Pu carbide, nickel, and ZrO_2 were prepared in a polyester resin base. The following observations were made:

1. Samples imbedded in the resin were stable over a three-month period.
2. The method of analyses for nickel in uranium-plutonium carbide or in UC was not sensitive to the composition of the nickel present. Results for elemental nickel additions or nickel in solid solution were identical.

The sensitivity of this method can be improved if the high background count of the X-ray tube can be reduced. The tube background count is 91,000 counts/50 sec compared to 103,000 counts/50 sec for a uranium-plutonium sample with 0.15 w/o Ni.

Attempts to reduce the background count by filtration of the X-rays from the tube with 0.001-in. iron foil were not successful. Filtration did produce a 12-fold reduction in the background, but reduced the total counts to about the same degree. The cause of the high background count will be determined and corrected.

The precision and reliability of the fluorescence method will be established for Ni in uranium-plutonium carbides after the high background count from the X-ray tube is minimized. Standard samples then will be prepared for reference.

A minor effort was made to determine the ratios of U/Pu intensities on five samples of carbide. The average ratio was 6.952 ± 0.042 . This ratio was obtained in direct comparison of U/Pu (corrected for background) as well as from (U/Zr)/(Pu/Zr) ratios.

Additional U/Pu ratios will be established on the standards to be prepared for nickel analysis. Similar standards also will be prepared for measuring the U/Pu ratio in mixed or solid solution oxides.

3.9.2 Task 1941 – Process Improvement

Introduction and Past Results

The objective of this task is to improve process control and reduce the cost of fabricating carbide fuel.

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. Several 2-kg blends of $\text{UO}_2 + \text{C}$ were made by this process. The granulated products were reacted, comminuted, then cold pressed and sintered.

The characterization of the sintered UC pellets produced by the one-step agglomeration method showed the intended single-phase structure with lattice parameters of $4.9604 \pm 0.0009 \text{ \AA}$ and $4.9616 \pm 0.0004 \text{ \AA}$. The oxygen content was 835 ppm. The impurity level of the final sintered pellets was very low, and indicated that the level of impurity in the starting material is not significantly increased by processing in the pilot plant equipment.

About 130 UC pellets were prepared for experimental sodium-bonding experiments (Task 2200) using the one-step process in the pilot plant facility. The as-sintered diameters of the 130 pellets varied between 0.2460 and 0.2475 in. with an

average density of 12.48 g/cm^3 (92% of theoretical). The pellet densities were generally lower than expected.

The second step toward cost reduction is a one-step synthesis and sintering or reaction-sintering into carbide pellets. Blends of $\text{UO}_2 + \text{C} + \text{phenolformaldehyde}$ or furfuryl-alcohol resin were compacted into pellets and heated to simultaneously cause synthesis and sintering.

The highest density achieved for pellets prepared from both vacuum dried and wet blends which were sintered to 1650°C for 2 hr was 8.11 g/cm^3 (60% T.D.). A minimum density of 10.2 g/cm^3 (75% T.D.) is desired. The pellets prepared from phenolformaldehyde were of reasonable quality and had very little hour-glassing.

Gas chromatographic analysis of decomposed Carbowax 4000 and 6000 temporary binders indicated that an insignificant amount of solid residue remained. Analysis of the residue is in process.

Current Progress

The analysis of Carbowax 4000 and 6000 demonstrated that these binders can be discounted as sources of carbon during normal processing since analyzed residues were less than 0.025%.

Tests were made to determine why UC prepared by the one-step P-K process in the Pilot Plant equipment would not sinter above 92% of theoretical density.

An optical comparison (particles mounted in oil between glass slides) of particle size indicated that the UC granulated product crushed in the pilot plant equipment (vibratory mill for 2 hr) and research equipment (ball mill for 20 hr) gave about the same distribution. Particles ranging from 5 microns to submicron size were obtained with a mean value of 1 to 2 microns.

Pellets with and without 0.15% nickel were cold formed from the ball-milled carbide with 1/2% Carbowax 6000 temporary binder. Pellets with nickel readily sintered to 13.15 g/cm^3 (97% T.D.) after one hour at 1550°C . Those without nickel sintered to 12.26 g/cm^3 (90% T.D.) after four hours at 1800°C . Pellets with nickel addition, and an equivalent carbon content of 5.12 w/o, were 84% UC and 16% U_2C_3 . Those without nickel addition, and an equivalent carbon content of 4.68, were single phase. Characterization data are shown in Table 4.

The discrepancy in sinterability of apparent identical powder size fractions of the same UC product is attributed to an inability to characterize size fraction accurately by optical microscopy. Equipment for photoelectric sedimentation analysis for particle size determination will be used.

Kilogram batches of UO_2 and C containing 1/4 to 1/2 w/o of polyvinyl alcohol temporary binder were prepared by the one-step, P-K process in the pilot plant facility.

The products were sampled according to a statistical plan. Analyses were made for carbon homogeneity on the dry blend before addition of binder, on the agglomerated, and on the dried product. A 5% probability risk was taken in making the following decisions:

1. No area in the P-K blender indicated a significantly low or high carbon value.
2. There was no statistical difference between the mean of P-K dry blend group or of the vacuum dried sampling group.
3. The variability of carbon in the P-K prepared blend is consistent from batch to batch.
4. No detectable oxidation of the UO_2 (in the $\text{UO}_2 + \text{C}$ agglomeration) which would affect carbon yields in the blends occurred during the vacuum drying step for periods up to three hours at 120°C .

TABLE 4 — SINTER TEST ON UC PREPARED BY THE PILOT PLANT PROCESS (TASK 1941)

Nickel Addition, <u>w/o</u>	Forming Pressure, <u>psi</u>	Avg Cold Formed Density, <u>g/cm³</u>	Sintering Temp, °C (Hold Time, hr)	Avg Sintered Density, <u>g/cm³</u>	<u>Chemical Composition *</u>	
					<u>Oxygen, ppm</u>	<u>Carbon, w/o</u>
0.15	45,000	9.16	1550 (1)	13.15	2385 ± 30	4.93 ± 0.08
None	45,000	9.26	1550 (1) plus 1800 (4)	12.26	370 ± 200	4.64 ± 0.02

*The variability shown was calculated about the mean at the 95% confidence level.

The synthesized UC product was statistically characterized for C, O₂, N, and U with respect to position in the reaction crucible. Samples of the reacted product were taken from the inner and outer radius of the reaction container, and also along the length of the container. The results shown in Table 5 indicated that the product was homogeneous with respect to position in the furnace for the variables measured.

Additional kilogram batches of UC are currently being prepared to shake down the equipment to optimize the process, and to gather the statistical data required for production control in the pilot plant facility. UC will continue to serve as the analog for the ultimate mixed uranium-plutonium carbide fuel during this study to conserve plutonium.

A. Reaction Sintering Studies

Reaction sintering (concurrent synthesis and sintering of UO₂ and a carbon source) is being evaluated as a method of preparing 75 and 84% T.D. pellets. Development of this process would eliminate the crushing and comminution steps used in the pilot plant process.

The methods currently being studied for low-density pellet fabrication by reaction sintering are:

1. Blend UO₂ and powdered carbon in the P-K blender. Add water to cause agglomeration and vacuum dry under controlled conditions at 120°C. Screen the agglomerates and compact pellets in the range 30,000 to 80,000 psi. Reaction sinter in vacuum in the range 1650 to 1850°C for one hour, and
2. Substitute a liquid phenolic resin carbon source for the water and part of the powdered carbon described under method 1. Additions of 1/2 w/o Carbowax 6000 or zinc stearate were evaluated as pressing aids, and 0.3 w/o nickel as a sintering aid.

TABLE 5 — CHARACTERIZATION OF UC PRODUCT
PREPARED BY THE PILOT PLANT PROCESS (TASK 1941)

No. of Samples Analyzed	Chemical Composition, * Average Values			
	Carbon, w/o	Oxygen, ppm	Nitrogen, ppm	Uranium w/o
10	5.00 ± 0.06	—	—	—
10	—	895 ± 350	—	—
4	—	—	115 ± 15	—
4	—	—	—	94.71 ± 0.24

*Samples were taken from the top and bottom of the reaction container for spectrochemical analysis. The variability shown was calculated about the mean at one standard deviation.

The significant conclusions which can be drawn from the data obtained for preparation of pellets by method 1 were:

1. Pellets obtained from all tests were very tough in the as-formed condition and could be handled without difficulty.
2. Sintered pellets with and without binders or lubricants and prepared from dry granules were uniform in diameter within each experiment and met the desired diametral tolerance of ± 0.001 in.
3. Sintered pellets prepared with nickel addition or partially dried granules had surface cracks and were hourglassed 0.002 to 0.005 in. An increase in carbon content was observed for pellets prepared and sintered with 0.3 w/o Ni additive.
4. Chemical analysis for oxygen, nitrogen, and carbon content was satisfactory. The maximum reaction sintered pellet was about 60% of theoretical and did not meet the anticipated minimum goal of 75% T.D.

The results of the cursory study using method 1 to prepare low-density pellets by reaction sintering of $\text{UO}_2 + \text{C}$ were encouraging and indicate the feasibility of this process.

Knowledge of the volatile content, carbon yield, and effect of storage on phenolformaldehyde resins was required for the method described in item 2 to assure adequate control of carbon stoichiometry.

The weight loss of the resin after vacuum drying and after pyrolysis is shown in Table 6. The carbon yield (available carbon) was calculated to be 39 w/o, based on the original weight of the liquid resin. The carbon yield did not vary significantly between lots and/or storage.

The significant conclusions which can be shown from the data obtained to date for preparation of the pellets by method 2 are:

TABLE 6 — ANALYSIS OF PHENOLFORMALDEHYDE RESIN
(TASK 1941)

Experiment No. *	Percent Weight Loss		Carbon, w/o (Based on Residue after Vacuum Drying)
	Vacuum Dried at 120°C, 3 hr	Pyrolysis at 700°C, 1 hr, Vacuum	
1	22.8	58.2	93
2	26.0	59.0	93
3	13.0	59.0	93.5
4	Not determined	57.5	95.9
5	Not determined	57.6	Not de- termined

*Resin sample shown in Experiments 1 and 2 was received in January 1968. Resin sample shown in Experiments 3, 4, and 5 was received in April 1967.

1. The optimum drying time for the granulated UC products to achieve maximum cold-formed or sintered pellet density and under the conditions of this experiment was between 10 and 20 minutes. Maximum density was achieved at 1850°C after a one-hour hold. Results are shown in Table 7.
2. The rate of temperature rise during sintering (i.e., about 5°C/minute to 25°C/minute from 1400°C to final temperature) did not significantly influence sintered pellet density.
3. The carbon content of the reaction sintered pellet heated under identical synthesis and/or reaction sintering conditions was difficult to control.
4. Sintered pellets were hourglassed about 0.005 in.
5. The maximum pellet density achieved was 70% of theoretical; the minimum acceptable goal was $75 \pm 2\%$ T.D.

These experiments complete the program for preparation of low-density pellets by reaction sintering (concurrent synthesis and sintering).

B. Uranium-Plutonium Carbide Specimens for Thermal Diffusivity Measurement

Fabrication of thermal diffusivity specimens for cooperative property measurements at Argonne National Laboratory are in process.

The various carbide fuels were prepared by carbothermic reduction of the dioxides with specimen preparation by cold pressing and sintering. Samples will include 97% T.D. (UPu)C + (UPu)₂C₃, 92% T.D. single-phase (UPu)₂C₃, 75%, 84%, 92% T.D. single-phase (UPu)C, and 90% T.D. single-phase (UPu)(OC). The required sample size of 0.250 ± 0.010 in. diameter, 0.030 ± 0.0005 in. thick, with flat sides parallel to ± 0.0005 in. was met without surface grinding.

Characterization of the samples made to date is shown in Table 8.

TABLE 7 — EFFECT OF DRYING TIME, FOR P-K BLENDER PREPARED
GRANULATION, ON REACTION SINTERED UC PELLETS DENSITY
(TASK 1941)

Experiment No.	Drying Time, minutes	Cold Formed Density, * g/cm ³	Sintered Pellet Density, g/cm ³		
			Unground		After Surface Grinding,
			1650°C -1 hr	1850°C -1 hr	1850°C -1 hr
1	5	5.07	6.96	8.34	8.74
2	10	5.31	7.24	8.69	9.32
3	20	5.16	7.10	8.69	9.22
4	40	4.50	6.45	7.94	
5	60	4.43	6.23	7.62	
6	180	4.39	6.22	7.62	

*Pellets were cold formed at 80,000 psi and surface ground (as indicated) after sintering to remove an approximate 0.005-in. hourglass shape to enable accurate geometric density measurement.

TABLE 8 — CHARACTERIZATION OF URANIUM-PLUTONIUM CARBIDE SPECIMENS FOR THERMAL DIFFUSIVITY MEASUREMENTS (TASK 1941)

Composition	Measured Sample Density, g/cm ³	Measured Sample Density, % T.D.	Chemical Analysis *			Phases by X-Ray Diffraction and Lattice Parameter, Å		Phases by Metallography and Amounts, v/o		
			Carbon, w/o	Plutonium, w/o	Oxygen, ppm	MC	M ₂ C ₃	(UPu)C	(UPu) ₂ C ₃	(UPu)(OC)
(UPu)C	10.35	76	4.56 ± 0.00	13.57 ± 0.22	3290 ± 230	4.9647 ± 0.0005	—	100	—	—
(UPu)C	11.22	82	4.56 ± 0.08	13.76 ± 0.30	2000 ± 165	4.9649 ± 0.0005	—	100	—	—
(UPu)C	12.45	91	4.51 ± 0.00	13.44 ± 0.16	3075 ± 30	4.9624 ± 0.0005	—	100	—	—
(UPu)(OC)	12.21	90	3.63 ± 0.14	12.66 ± 0.02	13,900 ± 800	—	—	—	—	100

*Product variability was calculated about the mean at the 95% confidence level.

C. Uranium-Plutonium Dioxide Pellet Fabrication

The fabrication of mixed dioxide UO_2 - PuO_2 pellets (85/15, 80/20, and 75/25 ratios) was initiated to provide samples for analytical method development.

Pellets were prepared by cold pressing blended UO_2 + PuO_2 powders at 40,000 psi containing 1/2 w/o Carbowax 6000, and sintering for three hours at 1650 to 1700°C under vacuum.

The 75/25 uranium-plutonium composition sintered to 92% T.D. These pellets were visually excellent with less than 0.0003 in. hourglassing and diametral variation within the group of pellets of less than 0.001 in.

3.10 TASK 2200 – SODIUM COMPATIBILITY AND BOND TESTING

Introduction and Past Results

A. Compatibility

The objective of the tests is to develop a compatible fuel-sodium-clad system that could be used for sodium-bonded mixed-carbide rods.

The compatibility of solid solution uranium-plutonium carbides, sodium-bonded to potential cladding materials was tested out of pile. Previous results of static capsule tests reported for 1000 and 4000 hr at 800°C have shown no significant carbon transfer between MC + 10 v/o M_2C_3 or single-phase MC ($M = U_{0.80}Pu_{0.20}$) and types 304, 316, or Incoloy-800. Single-phase M_2C_3 was found to carburize type 304 to a depth of 0.010 in. after 1000 hr and 0.020 in. after 4000 hr at 800°C in sodium.

Additional 4000-hr tests in sodium at 800°C were completed with structurally hyperstoichiometric fuel and several types of diffusion barriers. The fuel for these tests was 95% theoretical density MC + 10 v/o M_2C_3 containing 0.11% nickel, 4.83 w/o carbon, 5.01 w/o equivalent carbon. As expected, the diffusion barriers of tungsten, carburized tantalum and niobium, chromized type 316, and type 316 with a zirconium foil liner were compatible with the above fuels.

The carbon activity (a_c) of M_2C_3 was estimated as 0.05 from measured widths of the carbide layer in type 304 in contact with sodium and M_2C_3 , and with sodium and graphite for 1000 hr at 800°C. This value compares to a calculated value at 800°C of 0.016 for types 304 and 316 stainless steel and 0.009 for Incoloy-800.

B. Carbide Stability

The objective of the experiments is to compare the carbon activity of various carbide fuel structures and evaluate their stability with respect to carbon transfer to stainless steel.

Thermodynamic compatibility in fuel-clad systems consisting of various plutonium-bearing carbides sodium bonded to 304 stainless steel is being studied using the United Nuclear Carbon Meter technology.* Thermodynamics provide an alternate approach to the conventional kinetic methods (static capsule tests) of investigating compatibility in such systems. The equilibrium partitioning of carbon between the fuel and the stainless steel is determined, based on measurements of carbon activity in both. Where the carbon activity of a carbide fuel is found to be less than that of a reference stainless steel, thermodynamic compatibility is established. Thermodynamic compatibility is an acceptable degree of carbon transfer in any amount of time. Kinetic compatibility is an acceptable degree of carbon transfer from fuel to stainless steel within the service period.

The method for obtaining data utilizes the gas supply and analysis portion of a United Nuclear Carbon Meter. The fuel to be measured is in a sodium-filled iron capsule. Decarburizing gas – having the composition 5 v/o H_2 , 0.5 v/o H_2O , and balance argon – is passed through a Vycor tube furnace holding each capsule at $800^\circ C$, as shown in Fig. 6. This gas has a negligible carbon activity so that carbon moves from the fuel through the sodium and the iron capsule wall to the exterior where it is converted to CO. The gas is then treated in a catalytic converter to change CO to CH_4 , which is measured in a flame ionization detector. A carbon flux from the specimen is calculated from the rate of carbon removal.

A reference carbon content of 0.7 w/o in type 304 stainless steel was se-

*McKee, J. and Caplinger, W.: Carbon Meter Development, An Interim Report. Presented at the Liquid Metal Instrumentation and Control Symposium, Idaho Falls, Idaho (Mar. 1967).

Fig. 6 — Carbide Stability Test Capsule Configuration

lected to represent the maximum acceptable carbon pickup as the threshold of incompatibility. The carbon level was based on an evaluation of mechanical property data. The corresponding carbon activity was then estimated from carbon flux measurements of Armco iron capsules containing sodium and type 304 stainless steel powder uniformly carburized to 0.7 w/o, single-phase MC, single-phase M_2C_3 , and MC + 14 v/o M_2C_3 .

The assembly of the carbon meter apparatus, intended to supply decarburizing gas to the capsule experiments and analyze the gas from the experiments, was completed. The carbon background of the console was satisfactorily low.

Fabrication of the fuel test pellets was in progress. The various carbides are being prepared by carbothermic reduction of the mixed dioxides followed by cold pressing of pellets and sintering.

Current Results

A. Compatibility

Preparation of the topical report is in process.

B. Carbide Stability

Fabrication of the $(UPu)C + (UPu)_2C_3$ fuel was completed. Carbon flux measurements on 18 capsules are summarized in Table 9.

Typically, fluxes drop to near steady-state values within 15 hr. Actual equilibrium conditions are not obtained because the once-through gas system continually removed the carbon.

Because of carbon depletion, the fluxes tabulated are those obtained by an extrapolation to zero time. For each carbon source, a further extrapolation of flux to zero particle size is made utilizing the reciprocal surface-to-mass ratio. Under the conditions of zero time and zero particle size, the carbon activity of

TABLE 9 — CARBIDE STABILITY CAPSULE DATA SUMMARY

Capsule Number	Specimen Description *	Capsule Wall Thickness, mils	Specimen Oxygen Content, ppm	Specimen Carbon Content, w/o	Flux, † $\mu\text{g}/\text{cm}^2\text{-min}$	1/S/M, ‡ $(\text{cm}^2/\text{g})^{-1}$
1	Control — sodium only	10	—	—	—	—
2	Reference SS powder, 60/80 mesh	10	515	0.74	0.099	0.028
3	Reference SS powder, 100/140 mesh	10	555	0.74	0.099	0.017
4	Reference SS powder, 140/200 mesh	10	610	0.73	0.085	0.012
5	Reference SS powder, 140/200 mesh	20	610	0.73	0.076	0.012
6	MC single-phase, pellets	10	2675	0.73	0.016	—
7	MC single-phase, 100/140 mesh	10	4100	4.78	0.081	0.027
8	MC single-phase, 200/270 mesh	10	4105	4.78	0.123	0.013
9	MC single-phase, 200/270 mesh	20	4140	4.78	0.097	0.013
10	MC + 14 v/o M_2C_3 pellets	10	3650	5.00	0.037	—
11	MC + 14 v/o M_2C_3 , 100/140 mesh	10	3775	5.00	0.417	0.028
12N	MC + 14 v/o M_2C_3 , 200/270 mesh	10	4495	5.00	0.423	0.014
13N	MC + 14 v/o M_2C_3 , 200/270 mesh	20	4507	5.00	0.426	0.014
13§	MC + 14 v/o M_2C_3 , 200/270 mesh	20	4000	5.00	0.446	0.014
14	M_2C_3 , single-phase, pellets	10	595	6.92	0.057	—
15	M_2C_3 , single-phase, 100/140 mesh	10	N.a. ¶	6.92	0.255	0.025
16	M_2C_3 , single-phase, 200/270 mesh	10	N.a.	6.92	0.776	0.012
17	M_2C_3 , single-phase, 200/270 mesh	20	N.a.	6.92	0.965	0.012

*M = $\text{U}_{0.85}\text{Pu}_{0.15}$.

†Flux at zero time from a semilogarithmic extrapolation of flux vs time data.

‡S/M = surface-to-mass ratio.

§Duplicate of 13N, but not encapsulated at the same time.

¶Not available.

the carbon source can be estimated from the calculated activity drop through the capsule wall and that through the gas.

Preliminary estimates of carbon activities determined from the flux data were as follows:

<u>Source</u>	<u>Carbon Activity at 800°C (Relative to Graphite = 1)</u>
MC, single-phase	2.3×10^{-2}
304 SS + 0.7 w/o C	3.5×10^{-2}
MC + 14 v/o M_2C_3	1.1×10^{-1}
M_2C_3 , single-phase	3.3×10^{-1}

The order of estimated activities corresponds well with the order of static capsule compatibility.

These results indicate that MC single-phase is thermodynamically compatible with the reference stainless steel at 800°C, while MC + 14 v/o M_2C_3 and single-phase M_2C_3 are not.

Kinetic compatibility of MC + M_2C_3 was previously established in static capsule tests. Single-phase M_2C_3 was kinetically incompatible under similar test conditions.

The apparatus is being modified to permit direct carbon activity measurements.

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