NDA 2162-1

CARBIDE FUEL DEVELOPMENT

Progress Report

Period of September 15, 1960 to January 31, 1961

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FOREWORD

The Carbide Fuel Development project is part of the AEC Fuel Cycle Development Program. The prime contractor is the Nuclear Development Corporation of America (NDA), and the subcontractor is The Carborundum Company. NDA is performing the conceptual design, fuel evaluation, fuel irradiation, and irradiated fuel reprocessing. The Carborundum Companv is fabricating the fuel. Both companies are building plutonium handling facilities.

This report covers progress from September 15, 1960 to January 31, 1961. Previous progress was reported in:

NDA 2140-2, Carbide Fuel Development – Phase I Report (Oct. 15, 1959)
NDA 2145-1, Carbide Fuel Development – Progress Report (Mar. 11, 1960)
NDA 2145-4, Carbide Fuel Development – Progress Report (June 13, 1960)
NDA 2145-5, Carbide Fuel Development – Progress Report (Aug. 30, 1960)
NDA 2145-6, Carbide Fuel Development – Phase II Report (Nov. 6, 1960)

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1. INTRODUCTION

Fuel made of a combination of UC and PuC has a potential of reducing the fuel cycle cost of existing fast breeder reactors. The fuel cycle cost reduction is anticipated because of increased burnup and increased power generation capability of PuC-UC, compared to presently available metallic fuels. The effect of high burnup and high power generation rate on the stability of PuC-UC is not known. However, based on UC irradiation tests, there is justifiable optimism that carbide fuels will be dimensionally more stable than metallic fuels. As a result of high melting point and good thermal conductivity of UC, there is further expectation that carbide fuels will be capable of high power generation rates.

The Carbide Fuel Development Program is concerned with the technology of the entire PuC-UC fuel cycle. The major goal of the program is to produce PuC-UC and to obtain data on its irradiation behavior for long burnups and at high power generation rates. In addition, other areas of the fuel cycle are being explored to discover potential problems. The program was initiated in May 1959 and as outlined covers a period of about four and one-half years. The program has the objectives as outlined below:

Conceptual Design

1. An analytical study of the effect of substitution of PuC-UC on heat transfer, physics, and cost of existing fast breeder reactors.
2. Conceptual design of rod-type, fuel element configurations which can be substituted directly in existing reactors.

The above was completed and was reported in NDA 2140-2.

Facility Design and Fabrication

1. Design and construction of a facility for carbide fabrication at The Carborundum Company.
2. Design and construction of a facility for carbide evaluation at NDA.

The above was completed and was reported in NDA 2145-6.

Fuel Fabrication and Evaluation

1. Explore various methods of fuel preparation and fabrication into cylindrical pellets, beginning with powders.
2. Evaluate pellets by density measurement, chemical analysis, x-ray diffraction, hardness, metallography, and fuel-cladding compatibility studies. This is essentially complete for UC.

Fuel Irradiation

1. Irradiate clad fuel samples with burnup and maximum fuel temperature as the major variables. A minimum 2% burnup and a minimum 650 °C (1200 °F) fuel temperature were selected to establish the economic advantages of the fuel.
2. Make a post-irradiation examination.

Fuel Reprocessing

Study the reprocessing of irradiated fuel.

Full Scale Fuel Assembly

Design and construct a full scale fuel element assembly for irradiation in an existing fast breeder reactor.

2. SUMMARY

2.1 FUEL FABRICATION AND EVALUATION

Several batches of UC were synthesized in the plutonium boxes. Entirely satisfactory compositions have not been attained as yet, and the difficulties are being investigated and eliminated.

UC with nickel additions was fabricated to improve ultimate density and lower sintering temperatures. Preliminary tests showed that lower sintering temperatures may be possible. Ultimate density has not been improved to date.

Microprobe analysis of the Inconel-X/UC compatibility samples clarified the reaction that had occurred. Analysis of the type 304 sample showed no uranium penetration in 2000 hr at 820°C.

2.2 IRRADIATION TEST

The W1-1 capsule containing two clad UC specimens was inserted in WTR and operated to 5800 MW-d/T by the end of January. The W1-2 capsule is scheduled for insertion in WTR in February.

2.3 PLUTONIUM FACILITY CONSTRUCTION

2.3.1 Facility at The Carborundum Company

The facility has been operating with uranium since November. Modifications of some of the equipment have been required and nearly all of them have been completed.

2.3.2 Facility at NDA

The leaktightness of the glove boxes has been improved. A once-through nitrogen and once-through helium system have been installed parallel to the recirculating helium system.

3. FUEL FABRICATION AND EVALUATION

3.1 INTRODUCTION

The goal of the fabrication studies is to produce a high density combination of stoichiometric PuC and UC by powder fabrication techniques. A high physical density (about 95% of theoretical) is desired to minimize fission gas release and give a high fuel density.

The goal of the evaluation tests is to identify the material by density measurement, chemical analysis, x-ray diffraction, metallography and hardness. Additional out-of-pile tests of fuel-cladding compatibility, thermal stability and coefficient of expansion will survey properties of interest for in-pile tests.

Current studies on UC synthesis and fabrication are being carried out in the plutonium facility to check out equipment and procedures. Studies with plutonium are expected to start during the next quarter.

3.2 CARBIDE POWDER PREPARATION

Several UC batches were synthesized in the graphite resistance furnace and the box helium atmosphere. These were the first batches of UC made in the plutonium glove boxes. The purpose of these runs was to check the operation of the new system and to train operators for future plutonium handling. Initial batches were not reacted completely due to low reaction temperatures or insufficient holding time. Subsequent batches were low in carbon due to non-stoichiometry of the UO_2 or loss of carbon during the mixing operation. Additional mixes with higher carbon contents were synthesized, but a clogged filter in the vacuum line prevented removal of Co from the furnace and produced hyperstoichiometric UC. The filter was cleaned, but the next batch had a high nitrogen content indicating the getters had become saturated. Previous batches did not have detectable amounts of nitrogen. The getter furnaces have been recharged and additional batches will be synthesized.

3.3 CARBIDE PELLET FABRICATION

Compatibility tests between Inconel-X and UC indicated solubility of nickel for UC.* This gave hope that nickel may be a sintering aid for UC, in much the same way as nickel and cobalt are sintering aids to WC. WC-Co compacts are generally sintered at 1400°C, 80°C above the WC-Co eutectic. A WC-Co liquid solution is formed which dissolves some of the WC grain boundaries and produces dense packing of the carbide phase. Upon cooling WC crystals are precipitated from the liquid phase. An attempt was made to simulate this with nickel and UC. The experiments were carried out in the plutonium glove boxes. UC with 5% nickel powder and carbon in the amount of 0.5% of the nickel were milled for 96 hr, cold pressed at 30,000 psi, with

*See NDA 2145-6, pp. 19-20.

1/2% Carbowax-6000 binder and sintered in a tantalum-lined graphite crucible. The results are shown below:

	UC + 5 w/o Ni	UC
Cold Pressed Density, g/cm^3	8.51	8.97
Density after 1450°C – 1/2 hr	11.65	9.10
Density after 1650°C – 1½ hr	11.47	11.77
Density after 1850°C – 1 hr	-	12.56

Nickel reduced the required sintering temperature by 200°C for 87% dense pellets, but did not improve the ultimate density. Longer low temperature sintering of additional UC-Ni compacts is planned.

3.4 FUEL-CLAD COMPATIBILITY TESTS

In order to study possible fuel-clad interactions under conditions similar to those which would prevail in an actual fuel element, a series of diffusion couples were tested. The carbide fuel was held in contact with various cladding materials at 820°C in a helium atmosphere. Following this, the couples were sectioned and the carbide-cladding interface examined by metallographic means, x-ray diffraction and microprobe analysis. Examination of the test specimens is complete except for some of the microprobe analysis.

Microprobe analysis of the Inconel-X sample which had been tested for 1000 hr did not reveal uranium penetration outside the voids formed by the Ni-U eutectic (Fig. 3.11, NDA 2145-6). The white areas adjacent to the voids were identified as Cr_7C_3 by a combination of microprobe and x-ray diffraction analysis. The metallic diffusion layer on the UC half of the interface (Fig. 3.12, NDA 2145-6) was identified as uranium-nickel binary carbides of the M_3C and M_7C_3 type. The nickel concentration increased toward the Inconel-X interface. The nickel penetrated the UC to a 15μ depth. The nickel concentration in the UC was up to 1.5%. The above data indicate that at the test temperature a liquid U-Ni-C phase was formed. Upon cooling, UC with a small amount of nickel was precipitated out of the U-Ni-C solution at the UC interface. At the Inconel-X interface the Ni-U eutectic was formed and on cooling some of the excess carbon formed chromium carbides. The diffusion zone in between formed the binary uranium-nickel carbides.

Microprobe analysis of the type 304 stainless steel specimen tested for 2000 hr showed there was no uranium penetration. An inclusion such as shown on Fig. 3.6, NDA 2145-6 was 75% uranium with no iron, chromium, or nickel. These inclusions are probably adhering UC which have oxidized on storage to U_3O_8 (75 to 76% uranium).

X-ray diffraction analysis of the 2¼ Cr-Mo sample tested for 4000 hr indicated a very slight reaction.

X-ray diffraction analysis of the beryllium sample tested for 4000 hr indicated that it was converted to UBe_{13}. (See Fig. 3.1.)

The Zircaloy-2/UC interface was examined by microhardness measurements. Microhardness in combination with microprobe analyses performed by other investigators* and microprobe analysis performed in this investigation show that ZrC and free uranium metal are formed. (See Fig. 3.2.)

3.5 CHEMISTRY

Work is continuing to check the accuracy of the chemical analysis methods and particularly the carbon analysis method.

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*A. Accary, Third Uranium Carbide Information Meeting, Oak Ridge, Tenn. (Dec. 1960).

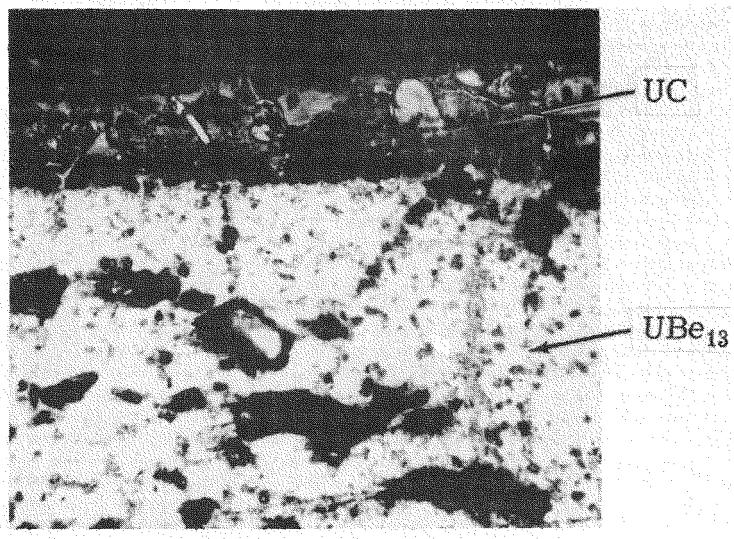

Fig. 3.1 — Beryllium-UC interface tested 4000 hr at 820°C – 500×

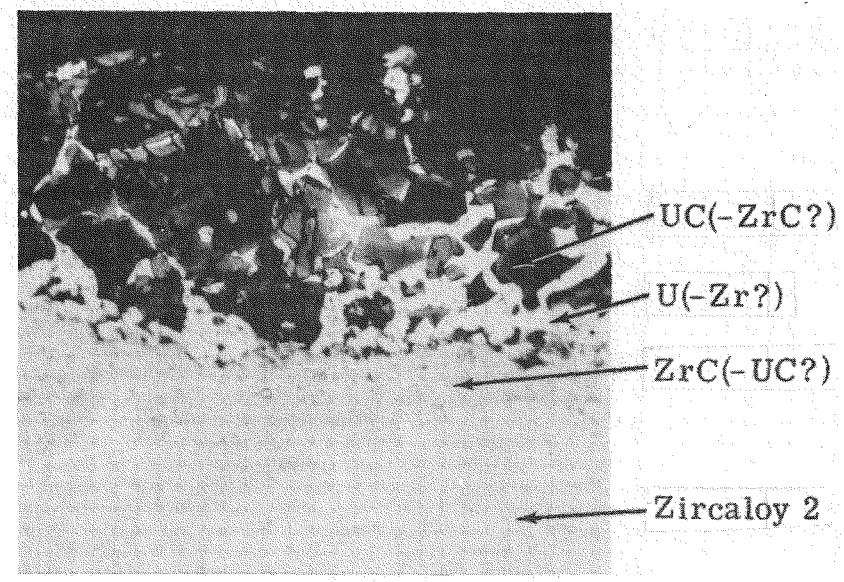

Fig. 3.2 — Zircaloy 2-UC interface tested 4000 hr at 820°C – 500×

4. IRRADIATION TESTS

4.1 INTRODUCTION

In order for PuC-UC fuel to reduce fuel cycle cost, several conditions must be met. The fuel has to be able to achieve high burnup, high operating temperatures and high power. At least 2 a/o burnup is desirable at fuel temperatures above 1200 °F with maximum dimensional stability of the fuel and minimum release of fission products. Power density should be at least equivalent to presently planned fuels. The objective of the irradiation program is to determine whether the high burnup temperature, and power density required can be achieved.

The irradiation test will measure temperatures, burnup, power density, and dimensional stability of the fuel. In addition, measurements of in-pile effective thermal conductivity, fission gas pressure, and fission gas release will be attempted.

4.2 IRRADIATION TEST

The W1-1 and W1-2 capsules and instruments were assembled, their out-of-pile calibration was completed and they were shipped to the test reactor for insertion. A detailed description of the capsules and specimens is given in NDA 2145-6, pp. 32-40.

The W1-1 capsule was started up in WTR on December 21st. Burnup as of the end of January was about 5800 MW-d/T. Negotiations have been concluded to insert W1-2 in the February cycle of WTR.

The performance of all the W1-1 instruments, thermocouples, and heaters has been satisfactory, with the exception of one thermocouple lost after three weeks of operation. The capsule contains two clad UC rods, each having a 0.191 in. diameter × 3 in. long fueled section. One specimen is clad with type 316 SS, the other with niobium. The space between the UC and cladding is filled with helium. The specimen operating conditions are given in Table 1. The temperatures are lower than the planned 1800 °F central fuel and 1000 °F clad surface temperatures. The reasons for this could be: lower reactor flux than estimated, greater flux depression by the experiment than estimated, and better specimen thermal conductivity than estimated. The continuous drop in power generation rate of the specimens is due to a decreasing flux in the experimental hole, which is partly due to the decreasing operating power of WTR.

The drop in UC + He gap conductivity with increased burnup may be due to lower conductivity fission gases in the gap (Table 1). The fuel center and clad surface temperature are measured in this experiment. The fuel surface temperature cannot be measured. For this reason, average UC + He gap conductivities are given, as well as the $\int K d\theta$ values.

Table 2 shows the calculated gap conductances assuming reasonable conductivities for UC. The in-pile thermal conductivity of UC and the He gap conductance are interdependent. The value for one will depend on the assumptions made for the other. Since the UC conductivity will

Table 1 — Operating Data on Wi-1 Specimens

Burnup (MW-d/T)	Central Fuel Temp, °F			Clad Surface Temp, °F			Heat Generation, w/in.			$\int_{ts}^{tc} Kd\theta$,* w/cm			Avg Thermal Cond. of UC+He Gap, Btu/hr-ft²-ft-°F		
	Startup	3000	5000	Startup	3000	5000	Startup	3000	5000	Startup	3000	5000	Startup	3000	5000
Nb Clad UC Sample	1450	1280	1230	840	720	700	860	740	660	27	23	21	4.6	4.6	4.2
Type 316 Clad UC Sample	1490	1405	1290	840	740	720	800	770	750	25	24	24	5.1	4.6	4.3

*A parameter used for the comparison of reactor fuels. See J. A. L. Robertson, $\int Kd\theta$ in Fuel Irradiations CRFD-835 (Apr. 1959).

Table 2 — Fuel and He Gap Conductivities

UC Thermal Conductivity, Btu/hr-ft²-ft-°F (Assumed)	He Gap Conductance, Btu/hr-ft²-°F					
	At Startup		At 3000 MW-d/T		At 5000 MW-d/T	
	Type 316	Nb	Type 316	Nb	Type 316	Nb
10	2500	2200	2200	2200	1900	1800
12	2200	1900	1900	1900	1700	1600
14	1900	1700	1700	1700	1500	1500

be the same in both specimens, the differences in temperature drops should be due to differences in gap conductance. The original estimates of a UC conductivity value similar to the out-of-pile arc cast UC value and a He gap conductance of about 2000 appear justified.

An entirely consistent analysis of the gap conductance data is not possible, probably due to errors in the measurement of heat generation rate and temperatures. In general, however, the conductance across the UC-stainless steel interface is similar to the conductance across the UC-Nb interface. This shows that the fuel-clad gap size is about the same for both specimens in spite of the fact that stainless steel has double the coefficient of expansion of niobium. The differences in conductance values do not account for the greater gap which would be obtained theoretically by stainless steel vs niobium.

5. PLUTONIUM FACILITY CONSTRUCTION

5.1 INTRODUCTION

The Carborundum Company has constructed a facility to be used for fabricating the fuel. NDA has constructed a facility to be used for fuel evaluation, irradiation specimen preparation, and will construct facilities for post-irradiation examination, and "hot" reprocessing.

5.2 FACILITY FOR FUEL CARBIDE FABRICATION AT THE CARBORUNDUM COMPANY

The gas circulation systems have been operating almost continuously since November. Helium gas purity is about 8 ppm oxygen and the purity has been as good as 3 ppm oxygen. Shutdowns and modifications have been required due to equipment failures. During the period when the gas systems were operating, the equipment in the boxes was operated using natural uranium.

Several modifications were required in the helium system. The all-zirconium getters were replaced with more efficient 50-50 a/o zirconium-titanium alloy getters to permit operation of the system 300°F lower. Gas preheaters were installed to permit more efficient use of the getter furnace. The underdesigned wiring of the getter furnaces, as supplied by the vendor, was completely replaced with adequate wiring. A commercial gas analysis unit was installed to replace the ones supplied by the vendor of the gas system. A new helium pump had to be ordered to replace one that had a scored shaft and impellers. The cause of the failure is not known for certain. The new pump is being shipped and will be installed the first week of February. Meantime the facility has been operating on one pump. Modifications on piping and valving have also been made to improve the safety of the system.

The carbon analysis furnace has been converted from an induction heated to a resistance heated one, to enable operation in a helium box, reduce required box space by 50%, and reduce maintenance.

5.3 FACILITY FOR FUEL CARBIDE EVALUATION AT NDA

The leaktightness of the glove boxes was improved considerably by the use of a more flexible, thicker plastic, and a compression seal on the box ports to help seal the plastic to the boxes.

A once-through nitrogen system was installed parallel to the recirculating helium system, and the welding box was adapted to once-through helium. The reason for the additional system was to obtain a lower oxygen content in the boxes at a lower gas cost, and to expedite the startup of the facility which has been delayed by malfunctions in the helium purification system. The nitrogen will be taken from liquid nitrogen bottles of guaranteed -75°F dewpoint. The system has been leak checked and found leaktight. Flushing with helium and nitrogen will start the first week of February.

A Beckman spectrophotometer was checked out and installed in the chemistry box line for colorimetric analysis of iron and nitrogen.

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