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CARBIDE FUEL DEVELOPMENT

Phase IV Report

Period of September 15, 1961 to September 30, 1962

Authors

United Nuclear

A. Strasser

The Carborundum Company

K. Taylor

J. Anderson

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FOREWORD

The Carbide Fuel Development project is sponsored by the USAEC Division of Reactor Development. The prime contractor is the United Nuclear Corporation, Development Division, and the subcontractor is The Carborundum Company. The United Nuclear Corporation is performing the conceptual design, fuel evaluation, and fuel irradiation. The Carborundum Company is fabricating the fuel. Both companies are operating plutonium handling facilities.

The report summarizes progress from September 15, 1961 to September 30, 1962.

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)
- NDA 2162-1, Carbide Fuel Development – Progress Report (Feb. 28, 1961)
- NDA 2162-3, Carbide Fuel Development – Progress Report (June 1, 1961)
- NDA 2162-5, Carbide Fuel Development – Phase III Report (Sept. 30, 1961)
- UNC-5003, Carbide Fuel Development – Progress Report (Feb. 9, 1962)
- UNC-5013, Carbide Fuel Development – Progress Report (May 28, 1962)
- UNC-5030, Carbide Fuel Development – Progress Report (Aug. 10, 1962)

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1. INTRODUCTION

The Carbide Fuel Development program is concerned with the technology of the entire UC-PuC fuel cycle. The major goals of the program are to produce UC-PuC 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.

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 UC-PuC, compared to presently available metallic fuels. The effect of high burnup and high power generation rate on the stability of UC-PuC is not known. However, UC irradiation tests show that carbide fuels are dimensionally more stable than metallic fuels. Because of the high melting point and good thermal conductivity of UC and the formation of solid solutions of UC-PuC with high melting points, there is further expectation that carbide fuels will be capable of high power generation rates.

The program was initiated in May 1959 and as originally outlined covers a period of about four and one-half years. Program objectives are outlined below.

Conceptual Design

1. An analytical study of the effect of substitution of UC-PuC 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 United Nuclear.

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

Fuel Fabrication and Evaluation

1. Explore various methods for preparation of fuel from powders and fabrication of the fuel into cylindrical pellets.

High density (up to 13.2 g/cm^3) UC and UC-PuC pellets have been made. Fabrication work was stopped on this program and continued as work sponsored by the Joint U.S.-Euratom program.

2. Evaluate pellets by density measurement, chemical analysis, x-ray diffraction, hardness, metallography, and fuel-cladding compatibility studies.

This is complete for UC and in process for UC-PuC. The UC-PuC studies also include melting point, thermal stability, and coefficient of expansion measurements.

Fuel Irradiation

1. Irradiate clad fuel samples with burnup and maximum fuel temperature as the major variables. A minimum 2% burnup of all fuel atoms by

fission and a minimum 650°C (1200°F) central fuel temperature were selected to establish the economic advantages of the fuel.

Complete for UC, and experiments for UC-PuC are awaiting insertion in the test reactor.

2. Conduct a post-irradiation examination.

Completed for UC.

2. SUMMARY

2.1 FUEL FABRICATION AND CHARACTERIZATION

Single-phase solid solution UC-PuC powders were prepared by the carbon reduction of a mixture of UO_2 and PuO_2 . A simple method was developed to determine the completion of the reaction by analyzing the exhaust gas from the furnace for CO.

Four groups of solid solution $(\text{U}_{0.8}\text{Pu}_{0.2})\text{C}_{0.95}$ pellets were prepared for irradiation tests. These groups differed in the enrichment of the uranium and in the presence or absence of nickel sintering aid.

Without sintering aid, densities averaged about 92% of theoretical; with sintering aid the average density was about 96% of theoretical.

The second phase, promoted by the use of nickel sintering aid, was identified as sesquicarbide.

2.2 IRRADIATION TESTS

The post-irradiation examination of UC specimens irradiated to 16,400 MW-d/tonne (avg) at an average 1400°F (760°C) fuel temperature and an average of 9.6 kw/ft (315 w/cm) was completed. The effect of radiation on UC was slight: 0.001 in. diametral increase (1.4% volume increase), 0.4% fission gas release, and no microstructural changes.

The $(U_{0.8}Pu_{0.2})C_{0.95}$ irradiation tests were redesigned for insertion in GETR. All of the irradiation specimens were assembled, and assembly of the capsules was started. One fueled and one dummy capsule were shipped to the test reactor site.

3. FUEL FABRICATION AND EVALUATION

3.1 INTRODUCTION

The goal of the fabrication studies is to produce high density solid solution UC-PuC pellets by powder fabrication techniques. High physical density (about 95% of theoretical) minimizes fission gas release and gives a high fuel density.

The evaluation tests are to identify the material by density measurement, chemical analysis, x-ray diffraction, and metallography. Out-of-pile tests of fuel-cladding compatibility, thermal stability, melting point, and coefficient of expansion are also being made to obtain properties of interest for in-pile tests.

3.2 CARBIDE POWDER PREPARATION

3.2.1 Experimental Powder Preparation

Synthesis studies described in the present report were limited to the oxide methods (2 and 3 below). The mixed oxide method (2) was simpler. In previous work essentially single-phase UC-PuC solid solutions had been synthesized by three methods:

1. $2 \text{ PuO}_2 + 7 \text{ C} \rightarrow \text{Pu}_2\text{C}_3 + 4 \text{ CO}$
 $\text{Pu}_2\text{C}_3 + \text{U} + 7 \text{ UC} \rightarrow 10(\text{U}_{0.8}\text{Pu}_{0.2})\text{C}$
2. $4 \text{ UO}_2 + \text{PuO}_2 + 15 \text{ C} \rightarrow 5(\text{U}_{0.8}\text{Pu}_{0.2})\text{C} + 10 \text{ CO}$
3. $5(\text{U}_{0.8}\text{Pu}_{0.2})\text{O}_2 + 15 \text{ C} \rightarrow 5(\text{U}_{0.8}\text{Pu}_{0.2})\text{C} + 10 \text{ CO}$

During Phase III (September 1960 to September 1961) the sesquicarbide method (1) was abandoned since powders prepared by this route required two steps and offered no advantages over the oxide methods (2 and 3).

- | | |
|---|---|
| PuO_2 | Prepared by Dow Chemical Company by the oxidation of the metal, total impurities: <400 ppm; particle size: minus 200 mesh; O/Pu ratio: 2.015. |
| UO_2 | Prepared by United Nuclear Corporation, Chemicals Division, total impurities: <200 ppm; O/U ratio: 2.05. |
| $(\text{U}_{0.8}\text{Pu}_{0.2})\text{O}_2$ | Prepared by the coprecipitation of the peroxides and calcining by General Electric Co., Hanford Atomic Products Operations. Total impurity content was about 5000 ppm with Ca, Cr, Cu, and Fe being present in excess of 500 ppm each; particle size: minus 200 mesh. |
| Carbon | A commercial product prepared by the cracking of methane. Particle size about 0.1 micron; ash content of 0.05%. |

The procedure used in preparing the oxide-carbon mixtures for reaction was as follows. The oxide and calculated weight of carbon were ball milled for 24 hr in a rubber lined mill with stainless steel balls. The mixture was then pressed at 5000 psi, without binder, into pellets 0.6 in. in diameter by about 0.6 in. high, after which each pellet was cut by a thin spatula into four pieces of approximately equal size. Batches of about 50 grams of the pellets were then heated in a graphite crucible in a graphite resistance furnace with a helium atmosphere. During the heating period, helium was aspirated through the furnace at the rate of 30 liters per minute to carry off the carbon monoxide formed in the reaction.

The effect of carbon stoichiometry on the final structure of the carbide powder was studied for the mixed oxide method. Reaction mixes were made containing 100%, 95%, and 90% of the stoichiometric carbon, reacted at 1500°C for 3 hr, and then 1700°C for 1 hr. The results indicated that the material with 95% of theo-

retical carbon was the most desirable, since no sesquicarbide phase was formed (as indicated by x-ray analysis). Table 1, experiment No. 2 shows the results obtained with the preferred carbon content. Lower carbon material may produce an undesirable zeta phase.

Table 2 shows a comparison of the carbide powders produced from the mixed oxide method (2) and from the solid solution oxide method (3) described previously. Several experiments were conducted using these two different starting materials in an effort to synthesize $(U_{0.8}Pu_{0.2})C_{0.95}$ solid solutions. In the first series, compacts of the reactants were heated to 1500°C for 3 hr with an additional hold of 1 hr at 1700°C. The high carbon content for the mixed oxide starting material was attributed to an incomplete reaction. Results are shown in Table 2.

For the second synthesis experiment, mixes were heated to 1550°C with hold times of 5 to 8 hr, followed by heating to 1625°C and 1700°C with hold times of 1 to 2 hr. Reacted pellets were crushed and repelletized before each reheat. The results of these experiments, summarized in Table 3, indicate that an essentially single-phase material can be produced from either method at about 1625°C.

Since the mixed oxide route was simpler it was adopted as the method for synthesis of carbide powders for future work.

An accurate, reliable method was found to determine the completion of the reaction in the synthesis of carbide powders. A Mine Safety Appliance type carbon monoxide tester was connected to the exhaust line from the furnace and the gas sampled periodically. Carbon monoxide, in a concentration of 0.000 to 0.007%, indicated the completion of the reaction. This is approximately the CO concentration in the exhaust gas from the heated empty furnace. The result of several synthesis experiments monitored with the CO indicator are shown in Table 4. Theoretical carbon for these compositions is 4.55%.

Table 1 — The Effect of Carbon Content on The Syntheses of UC-PuC Powders

(Synthesized 1500°C-3 hr; 1700°C-1 hr)

Experiment No.	Intended Composition	Total Measured Carbon, %	X-Ray Analysis
1	$(U_{0.8}Pu_{0.2})C_{1.0}$	5.05	Major (U,Pu)C Weak $(U,Pu)_2C_3$ (There appears to be a double UC-type phase)
2	$(U_{0.8}Pu_{0.2})C_{0.95}$	4.63	Single-phase (U,Pu)C $a_0 = 4.964 \pm 0.001 \text{ \AA}$
3	$(U_{0.8}Pu_{0.2})C_{0.90}$	4.75	Single-phase (U,Pu)C $a_0 = 4.964 \pm 0.001 \text{ \AA}$

Table 2 — Syntheses of UC-PuC Powders from Mixed Oxides and Solid Solution Oxides

(Synthesized 1500°C-3 hr; 1700°C-1 hr)

Experiment No.	Starting Material	Total Carbon in Product, %	X-Ray Analysis
1	UO_2, PuO_2, C	5.21	Major (U,Pu)C Moderate $(U,Pu)_2C_3$ Weak $(U,Pu)O_2$
2	$(U,Pu)O_2, C$	4.75	Major (U,Pu)C Weak/faint $(U,Pu)O_2$

Table 3 — Results of Additional Experiments to Produce $(U_{0.8}Pu_{0.2})C_{0.95}$ by Carbon Reduction of the Oxides

Experiment No.	Starting Material	Reaction Temperature, Time, and Total Carbon				
		1550°C 5-hr Hold	1550°C 8-hr Hold	1625°C +1-hr Hold	1625°C +1-hr Hold	1700°C +1-hr Hold
1	UO_2, PuO_2, C	6.94	-	5.00	4.53*	-
2	$(U, Pu)O_2, C$	-	4.75†	4.75‡	-	4.44§

Results of X-Ray Analysis

*Major $(U, Pu)C$; faint $(U, Pu)O_2$ and $(U, Pu)_2C_3$ $a = 4.968 \pm 0.001 \text{ \AA}$

†Major $(U, Pu)C$; faint $(U, Pu)_2C_3$; weak $(U, Pu)O_2$

‡Major $(U, Pu)C$; faint $(U, Pu)O_2$

§Major $(U, Pu)C$; very faint $(U, Pu)O_2$

Table 4 — Results of Synthesis Experiments at 1625°C to Produce $(U_{0.8}Pu_{0.2})C_{0.95}$ (Monitored by CO Meter)

Experiment No.	Batch Size, g	CO at End of Run, %	Time to Reach Respective % CO,* hr	Total Carbon, %	X-Ray Analysis
1	50	0.005	6½	4.53	Single-phase $(U, Pu)C$ $a_0 = 4.965 \pm 0.001 \text{ \AA}$
2	15	0.007	2½	4.58	Single-phase $(U, Pu)C$ $a_0 = 4.965 \pm 0.001 \text{ \AA}$
3	50	0.005	7½	4.61	Major $(U, Pu)C$ Faint $(U, Pu)O_2$ $a_0 = 4.965 \pm 0.001 \text{ \AA}$
4	50	0.005	5½	4.60	Single-phase $(U, Pu)C$ $a_0 = 4.969 \pm 0.001 \text{ \AA}$

*Note that smaller batches required less time for completion.

3.2.2 Preparation of Powder for Irradiation Test Samples

The carbon reduction of the mixed oxide route reproducibly gave the desired composition, $(U_{0.8}Pu_{0.2})C_{0.95}$, at 1625°C. Therefore, it was used for the synthesis of $(U_{0.8}Pu_{0.2})C_{0.95}$ carbide powders.

Reaction of a 760-g batch of PuO_2-UO_2-C yielded approximately 580 g of essentially single-phase $(U_{0.8}Pu_{0.2})C_{0.95}$ solid solution. In its synthesis, the reaction batch was divided into 16 lots of about 50 g each. Results are shown in Table 5. Chemical analysis for total carbon on the composite of lots 1 through 14 gave 4.65%.

To minimize oxygen content, the composite of lots 1 through 14 was cold pressed without binder and reheated to 1625°C for an additional 2 hr. Analysis of the crushed and milled powders gave total carbon at 4.66%. Lots 15 and 16, which were synthesized subsequently, were milled for 22 hr and blended with lots 1 through 14 by ball milling for an additional 2 hr.

Following the above experiments, $(U_{0.8}Pu_{0.2})C_{0.95}$ powder containing enriched uranium was made by the carbon reduction of the mixed oxides, one part PuO_2 and four parts 24% enriched UO_2 . The procedure was the same as just described for the preparation of analogous carbide powder using normal uranium. About 200 g of the powder were produced in 40-g lots. These experiments are summarized in Table 6.

3.3 CARBIDE PELLET FABRICATION

3.3.1 Preliminary Experiments

The objective was to develop a method for the preparation of high density pellets for irradiation studies.

For all experiments, the reaction clinker was crushed and ball milled for 24 hr. A temporary binder of 1/4 w/o Carbowax-6000 dissolved in trichloroethylene was added to the carbide powder. Pellets of 0.224 in. diameter by about 0.20 in.

Table 5 — Synthesis of $(U_{0.8}Pu_{0.2})C_{0.95}$ Powder Containing Normal Uranium

(1625°C with Hold Time of 5½ hr for Experiments 1 through 14 and 6 hr for Experiments 15 and 16)

Experiment No.	Batch Size, g	Total Carbon, %	X-Ray Analysis
1*	50	4.70	Single-phase $(U,Pu)C$ $a = 4.965 \pm 0.001 \text{ \AA}$
2	10	4.58	Major $(U,Pu)C$ Faint $(U,Pu)O_2$ $a = 4.9645 \pm 0.0002 \text{ \AA}$
3	50	4.63	Single-phase $(U,Pu)C$ $a = 4.964 \pm 0.001 \text{ \AA}$
4	50	4.62	Single-phase $(U,Pu)C$ $a = 4.964 \pm 0.001 \text{ \AA}$
5	50	4.65	Single-phase $(U,Pu)C$ $a = 4.964 \pm 0.001 \text{ \AA}$
6	50	4.57	Major $(U,Pu)C$ Faint $(U,Pu)O_2$ $a = 4.95 \pm 0.01 \text{ \AA}$
7	50	4.66	Single-phase $(U,Pu)C$ $a = 4.965 \pm 0.001 \text{ \AA}$
8	50	4.67	Single-phase $(U,Pu)C$ $a = 4.966 \pm 0.001 \text{ \AA}$
9	50	4.61	Single-phase $(U,Pu)C$ $a = 4.9641 \pm 0.0002 \text{ \AA}$
10	50	4.56	Single-phase $(U,Pu)C$ $a = 4.964 \pm 0.001 \text{ \AA}$
11	50	4.64	Single-phase $(U,Pu)C$ $a = 4.964 \pm 0.001 \text{ \AA}$
12	50	4.64	Single-phase $(U,Pu)C$ $a = 4.964 \pm 0.001 \text{ \AA}$
13	50	4.66	Single-phase $(U,Pu)C$ $a = 4.9642 \pm 0.0002 \text{ \AA}$
14	50	4.59	Single-phase $(U,Pu)C$ $a = 4.964 \pm 0.001 \text{ \AA}$
15	55	4.63	Single-phase $(U,Pu)C$ $a = 4.965 \pm 0.001 \text{ \AA}$
16	55	4.66	Single-phase $(U,Pu)C$ $a = 4.964 \pm 0.001 \text{ \AA}$

*Chemical analyses for total carbon and x-ray were taken prior to making correction for an apparent batching error.

Table 6 — Synthesis of $(U_{0.8}Pu_{0.2})C_{0.95}$, 24% Enriched U
(1625°C-5½-hr Hold)

Experiment No.	Intended Composition	Total Carbon, %	X-Ray Analysis
1	$(U_{0.8}Pu_{0.2})C_{0.95}$	4.62	Single-phase (U,Pu)C $a_0 = 4.965 \pm 0.001 \text{ \AA}$
2	$(U_{0.8}Pu_{0.2})C_{0.95}$	4.59	Major (U,Pu)C Faint (U,Pu)O ₂ $a_0 = 4.964 \pm 0.001 \text{ \AA}$
3	$(U_{0.8}Pu_{0.2})C_{0.95}$	4.63	Single-phase (U,Pu)C $a_0 = 4.964 \pm 0.001 \text{ \AA}$
4	$(U_{0.8}Pu_{0.2})C_{0.95}$	4.65	Single-phase (U,Pu)C $a_0 = 4.964 \pm 0.001 \text{ \AA}$
5	$(U_{0.8}Pu_{0.2})C_{0.95}$	4.67	Single-phase (U,Pu)C $a_0 = 4.963 \pm 0.001 \text{ \AA}$

long were cold pressed at 30,000 psi. Compacts containing nickel were prepared by blending with nickel powder in the ball mill for at least 2 hr. The nickel was prepared by the hydrogen reduction of minus 325 mesh powder at 700°C, followed by ball milling in a rubber lined mill with stainless steel balls.

About 0.10 to 0.15 w/o carbon pickup was usually observed in sintering carbide compacts containing nickel. The normal sintering procedure consisted of heating the compacts in an open tantalum-lined graphite crucible placed in a closed graphite, resistance heated furnace with a helium atmosphere. The temperature was raised gradually to evaporate the temporary binder (1/4 w/o Carbowax-6000) and then more rapidly to 1550°C with a hold period of 1 hr at maximum temperature. It was assumed that the source of the extra carbon was either the temporary binder or the graphite furnace.

To eliminate pickup of extra carbon, which accentuated the formation of the sesquicarbide phase, the sintering procedure was modified. The compacts were first heated to 700°C in an open, tantalum-lined graphite crucible in the graphite furnace while maintaining a flow of helium through the furnace to carry off the vapors from the temporary binder. A tantalum lid was then placed on the tantalum crucible and the sintering completed at 1550°C. This procedure eliminated carbon pickup.

The results of previous work on UC at The Carborundum Company indicated free carbon hindered densification. Therefore, small additions of free carbon were made to study its affect on the sinterability of $(U_{0.8}Pu_{0.2})C_{0.95}$ compacts containing 0.12% nickel. Results appear in Table 7. The nickel sintering aid appeared to be less effective in the pellets containing as little as 0.10 w/o additional free carbon.

At the time of the last summary report (see NDA 2162-5, September 30, 1961, Table 3.8) only a limited amount of work has been done on sintering $(U_{0.8}Pu_{0.2})C_{0.95}$

Table 7 — The Effect of Nickel and Carbon Additions on Sintering of $(U_{0.8}Pu_{0.2})C_{0.95}$

Experiment No.	Composition of Pellet	Temperature and Hold Time	Average Density, g/cm ³	Total Carbon, %	X-Ray Analysis
1	$(U_{0.8}Pu_{0.2})C_{0.95}$	1550°C with a 1/2-hr hold	10.81	4.64	Major (U,Pu)C Faint $(U,Pu)_2C_3$
2	$(U_{0.8}Pu_{0.2})C_{0.95}$ +0.12 w/o Ni		12.90	4.68	Single-phase $(U,Pu)C^*$ $a_0 = 4.9644 \pm 0.0003 \text{ \AA}$
3	$(U_{0.8}Pu_{0.2})C_{0.95}$ +0.12 w/o Ni +0.10 w/o C		12.72	4.76	
4	$(U_{0.8}Pu_{0.2})C_{0.95}$ +0.12 w/o Ni +0.20 w/o C		12.74	4.88	
5	$(U_{0.8}Pu_{0.2})C_{0.95}$	1550°C with a 1-hr hold	11.83	4.60	Major (U,Pu)C Faint $(U,Pu)_2C_3$
6	$(U_{0.8}Pu_{0.2})C_{0.95}$ +0.12 w/o Ni		12.91	4.77	Major (U,Pu)C Faint $(U,Pu)_2C_3$ $a_0 = 4.9644 \pm 0.0002 \text{ \AA}$
7	$(U_{0.8}Pu_{0.2})C_{0.95}$ +0.12 w/o Ni +0.10 w/o C		12.68	4.83	Major (U,Pu)C Weak $(U,Pu)_2C_3$ $a_0 = 4.966 \pm 0.001 \text{ \AA}$
8	$(U_{0.8}Pu_{0.2})C_{0.95}$ +0.12 w/o Ni +0.20 w/o C		12.75	4.94	Major (U,Pu)C Weak $(U,Pu)_2C_3$ $a_0 = 4.949 \pm 0.009 \text{ \AA}$

*Metallographic examination showed a two-phase structure. X-ray analysis was apparently not sufficiently sensitive to pick up the second phase.

without sintering aids. These experiments were carried out in helium at atmospheric pressure at temperatures ranging from 1350 to 1750°C. The highest density was 12.2 g/cm³ (89.5% of theoretical), achieved at the highest sintering temperature, 1750°C.

Additional sintering experiments under Contract AT(30-1)-3118 showed that densities as high as 12.8 g/cm³ could be attained by sintering at 1925°C. Chemical analyses indicated that within experimental error, the uranium to plutonium ratio had not been changed by the high temperature sintering. Therefore, if vaporization took place during sintering, uranium and plutonium vaporized congruently.

3.3.2 Fabrication of (U_{0.8}Pu_{0.2})C_{0.95} Pellets for Irradiation Tests

The following four groups of pellets were prepared.

1. Normal uranium, no sintering aid.
2. Normal uranium, 0.12 w/o nickel.
3. 24% enriched uranium, no sintering aid.
4. 24% enriched uranium, 0.12 w/o nickel.

All pellets were fabricated from slightly hypostoichiometric powders having the intended composition (U_{0.8}Pu_{0.2})C_{0.95}, prepared by carbon reduction of the mixed oxides. A composite of all the powders listed in Table 5 was used for preparation of pellets containing normal uranium. For the enriched uranium pellets, the powders described in Table 6 were used.

Except for sintering temperature, the fabrication procedure was the same for all groups. The carbide powder was ball milled for 24 hr and pressed at 30,000 psi into pellets 0.224 in. in diameter by about 0.2 in. high using 1/4 w/o Carbowax-6000 dissolved in trichloroethylene as the temporary binder. Some cored pellets having the same overall dimensions were also prepared. The sintering was done in helium at atmospheric pressure. The pellets were first heated to 700°C in an open tantalum crucible, in a graphite resistance heated furnace to vaporize the

binder. During this stage, helium was aspirated through the furnace to carry off the binder vapors. Sintering was then completed in a closed tantalum crucible in a stagnant helium atmosphere. For nickel-free carbides, a maximum temperature of 1925°C was held for 1 hr. Compositions containing nickel were held at a maximum temperature of 1550°C for 1/2 hr.

After sintering, the pellets were surface ground to the desired dimensions on a centerless grinder with 1000 mesh SiC suspended in mineral oil.

The pellets prepared for irradiation tests are described in Tables 8 and 9. Representative microstructures are shown in Figs. 1 and 2.

X-ray patterns were obtained on the various batches of pellets represented in the above tables using camera techniques. The pellets of Table 8 were single phase (U,Pu)C with $a_0 = 4.964 + 0.001 \text{ \AA}$ with the exception of batch No. 2 of the solid pellets containing nickel, where a faint sesquicarbide phase was also evident. All batches of pellets in Table 9 were single-phase (U,Pu)C. Here the unit cell was $4.961 \pm 0.001 \text{ \AA}$ in the pellets containing no nickel and $4.964 \pm 0.001 \text{ \AA}$ in the batches containing nickel. These results will be checked by the more sensitive diffractometer method. Oxygen and nickel contents are also being determined.

3.4 METALLOGRAPHY AND MICROPROBE ANALYSES

3.4.1 Introduction

In previous work* it had been found that nickel in amounts as small as 0.1% was an effective sintering aid for both UC and UC-PuC solid solutions, making possible the production of pellets of 95% theoretical density. However, the use of nickel led to considerable quantities of second phase which had not been positively identified at the time of the last summary report.*

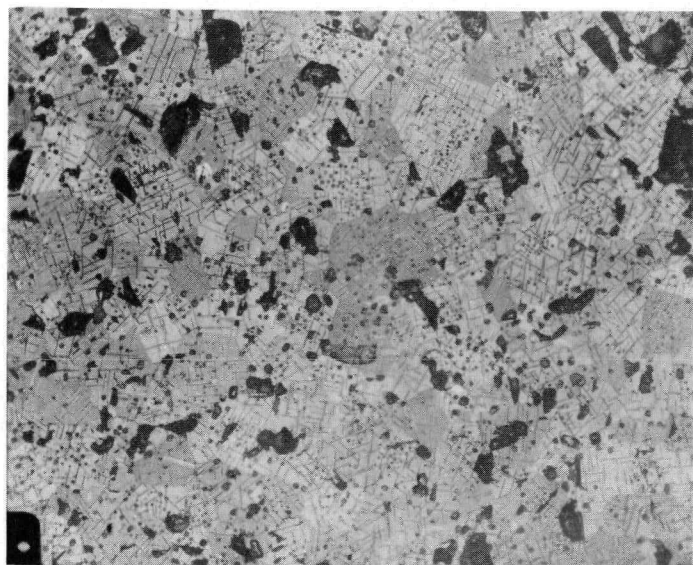
*NDA 2162-5.

Table 8 — Irradiation Pellets of $(U_{0.8}Pu_{0.2})C_{0.95}$ Containing Normal Uranium
(Diameter, 0.191 ± 0.0005 in.)

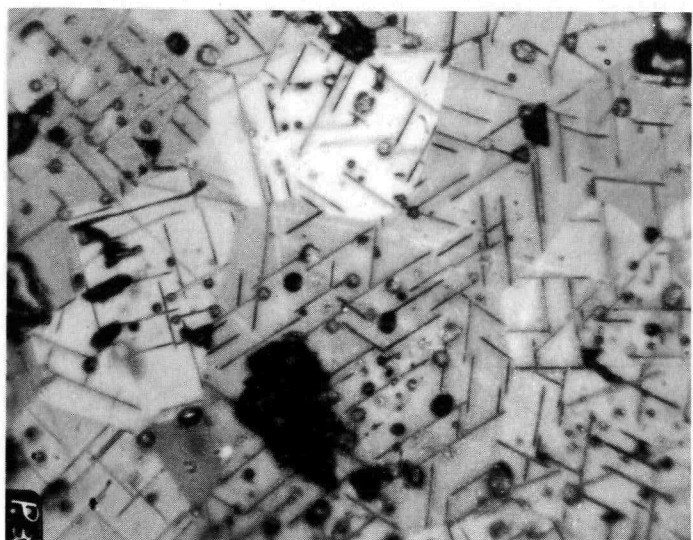
Type	No. of Pellets	Sintering Aid	Density, g/cm ³		Total Carbon, %	Plutonium, %	Nitrogen, ppm
			Range	Average			
Solid	65	None	12.50-12.63	12.54	4.62	19.05	200
Cored	18	None	12.50-12.63	12.54	4.62	19.00	
Solid (batch No. 1)	58	0.12% Ni	12.96-13.14	13.03	4.71	18.80	
(batch No. 2)	78	0.12% Ni	13.04-13.17	13.12	4.63		173
Cored	24	0.12% Ni	13.04-13.20	13.11	4.66		

Table 9 — Irradiation Pellets of $(U_{0.8}Pu_{0.2})C_{0.95}$ Containing 24% Enriched Uranium
(Diameter, 0.175 ± 0.0005 in.)

Type	No. of Pellets	Sintering Aid	Density, g/cm ³		Total Carbon, %	Plutonium %	Nitrogen, ppm
			Range	Average			
Solid	37	None	12.24-12.48	12.38	4.51	18.70	133
Cored	9	None	12.24-12.48	12.38	4.51		
Solid	53	0.12 w/o Ni	12.83-13.11	12.97	4.69	18.30	125
Cored	17	0.12 w/o Ni	12.83-13.11	12.95	4.69		

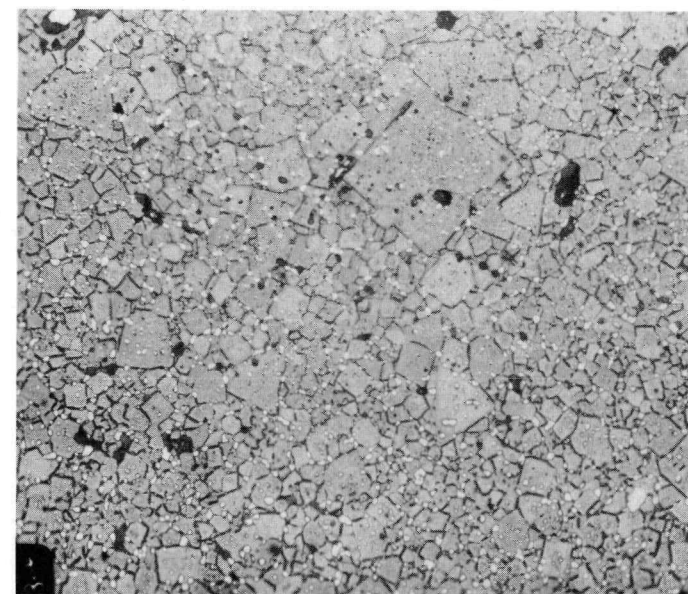


150×

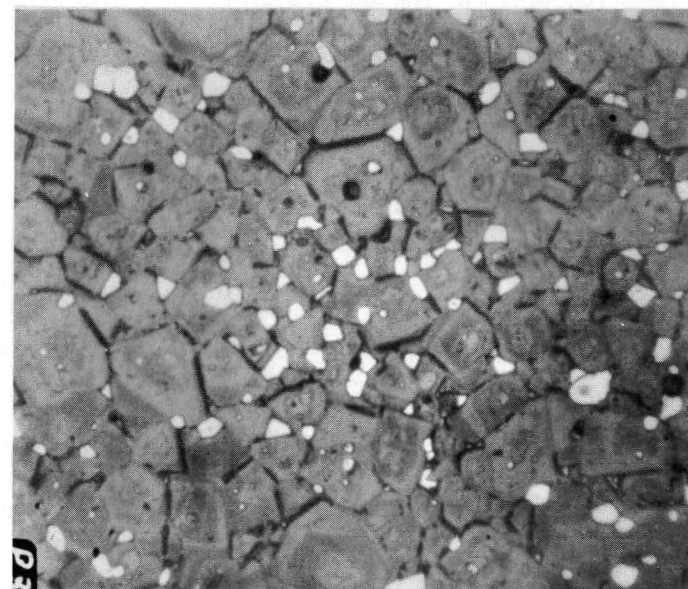


600×

Fig. 1 — $(U_{0.8}Pu_{0.2})C_{0.95}$ — typical structure of irradiation specimen. Etchant: nitric acid-acetic acid-water.



150×



600×

Fig. 2 — $(U_{0.8}Pu_{0.2})C_{0.95} + 0.1 Ni$ — typical structure of irradiation specimen. Etchant: nitric acid-acetic acid-water.

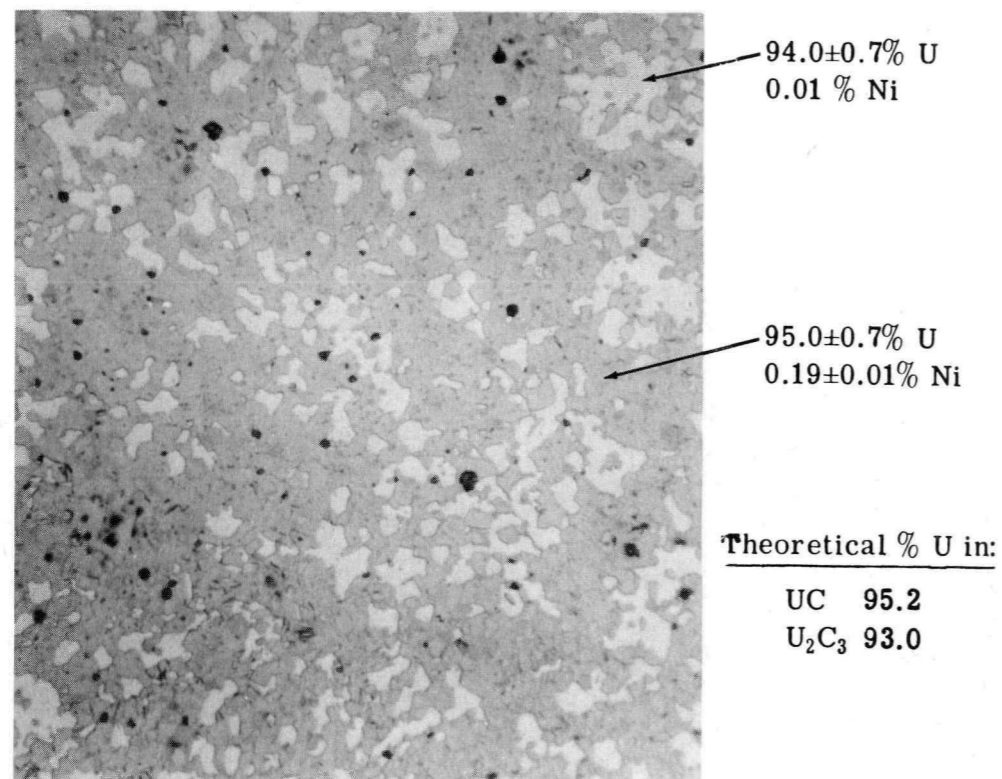
Before producing pellets for irradiation tests, an additional effort was made to determine the structure of the second phase and the location of the nickel in the crystal structure. Microprobe analyses, x-ray diffraction, and metallographic examination of the UC and UC-PuC specimens were made. Microprobe analyses on UC were made by Advanced Metal Research Corporation and on UC-PuC by the Monsanto Research Corporation's Mound Laboratory. As the result of this work and the x-ray and metallographic studies, it was concluded that the second phase was sesquicarbide type.

3.4.2 UC + 0.1% Ni

When nickel additions are made, the sinterings are done at 1500 to 1550°C, a temperature at which U_2C_3 is stable. (See Fig. 3.) If UC samples containing nickel are heated to 1850°C in vacuum, the U_2C_3 type phase disappears and a dicarbide phase forms. (See Fig. 4.)

Microprobe analyses were made on the as sintered, and the sintered reheated specimens. The results are shown on Figs. 3 and 4. The light phase in the as sintered specimen (Fig. 3) is very probably U_2C_3 , with virtually no nickel in solution. The dark phase is conclusively UC with all of the nickel in solution. Nickel was not found as a metal or intermetallic compound.

The light phase in the sintered and annealed specimen (Fig. 4) is very probably UC_2 and the gray phase is thought to be U_3O_8 . Virtually no nickel is in solution in either phase. Although spectral records were obtained on the white and gray phases, no additional detectable elements were noted. The dark phase is conclusively UC with about half the nickel still in solution. The remainder of the nickel has migrated to the grain boundaries, and some probably evaporated. Concentrations in excess of 40 to 50 w/o Ni were detected at the interface of the white, gray, and matrix phases. The nickel enriched regions were much smaller than the electron beam size employed (the beam was 1 to 2 μ) implying even greater concentrations of nickel than noted.

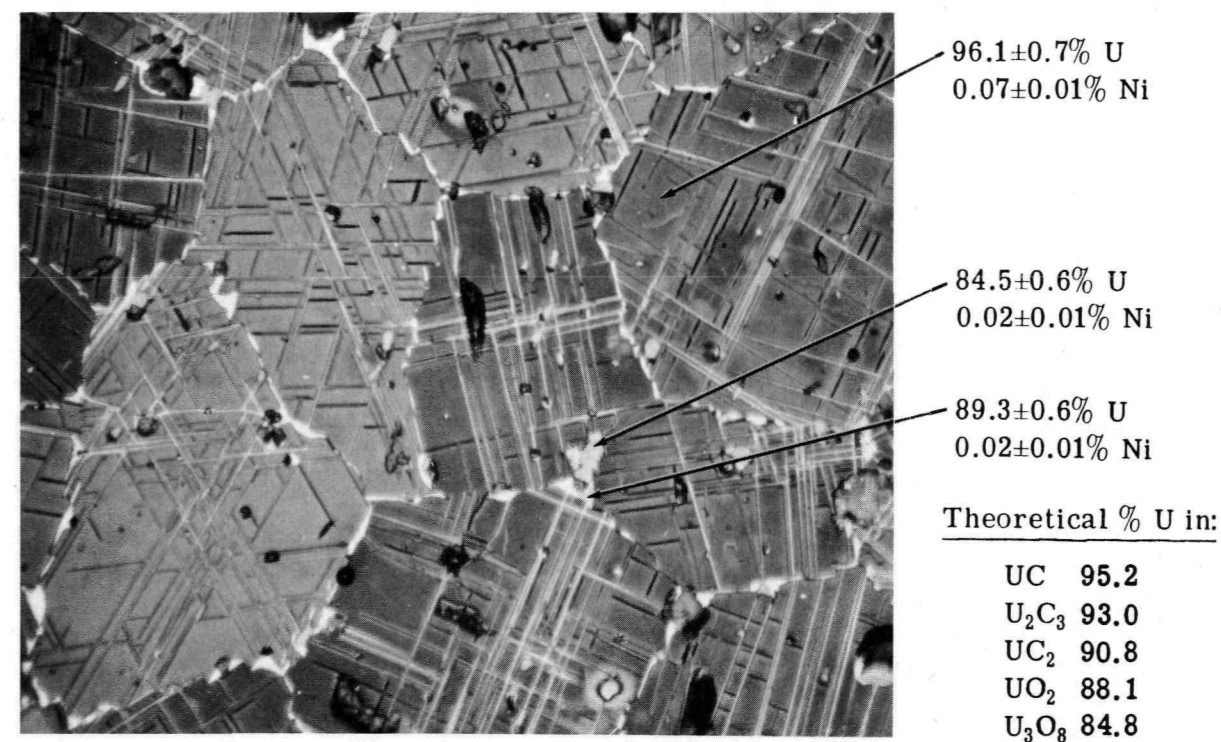


Neg. No. 1299 D

Fig. 3 — UC-0.1% Ni — sintered at 1525°C, 4 hr.
500×. Etchant: nitric acid-acetic acid-water.

X-ray identification

Major UC
 Minor U₂C₃
 Minor UO₂
 One strong Ni type line
 $a_0 = 4.96 \text{ \AA}$ (breadth of lines
 did not allow greater precision)



Neg. No. 1297

Fig. 4 — UC-0.1% Ni — sintered at 1525°C, 4 hr, reheated at
1850°C, 1 hr in vacuum. 500×. Etchant: nitric acid-acetic
acid-water.

X-ray identification

Major UC
 Very faint UC₂
 Very faint UO₂
 $a_0 = 4.960 \text{ \AA}$

The diffusion of the nickel from the UC, together with the peritectoidal decomposition of U_2C_3 at $1780^\circ C$, are the reasons why the U_2C_3 is no longer present in the vacuum annealed sample.

The x-ray diffraction study of the samples confirms the above results. The x-ray results are summarized on Figs. 3 and 4. Diffractometer traces were taken with Cu radiation and Debye-Scherrer photographs were taken with Cu $K\alpha$ radiation. A focusing monochromator camera gave some information, for which the other two methods were not sufficiently sensitive.

3.4.3 $(U_{0.8}Pu_{0.2})C_{0.95} + 0.1\% Ni$

Microprobe analyses were made on the as sintered, and sintered and reheated specimens. The results showed that in both specimens the nickel was concentrated at the grain boundaries. There was no nickel in either the monocarbide or the sesquicarbide phase.* This is different from the UC sample where the nickel was in solution in the monocarbide phase. Absolute uranium and plutonium analyses were not possible, because of the lack of sufficient standards. Standards of PuC and Pu_2C_3 are being obtained. Additional samples will be analyzed when the new standards are available.

Data obtained from the high temperature helium atmosphere annealing of $(U_{0.8}Pu_{0.2})C_{0.95} + 0.1 Ni$ are shown in Table 10 and Figs. 5 and 6. The pellets lost some weight through vaporization and swelled slightly, accompanied by a density decrease. The effect is probably due to the reaction of the residual oxygen. The grain size increased, but no phase changes occurred as with UC. The annealed structure still contains sesquicarbide and no dicarbide. The greater stability of the $(U,Pu)_2C_3$ during the helium atmosphere annealing than

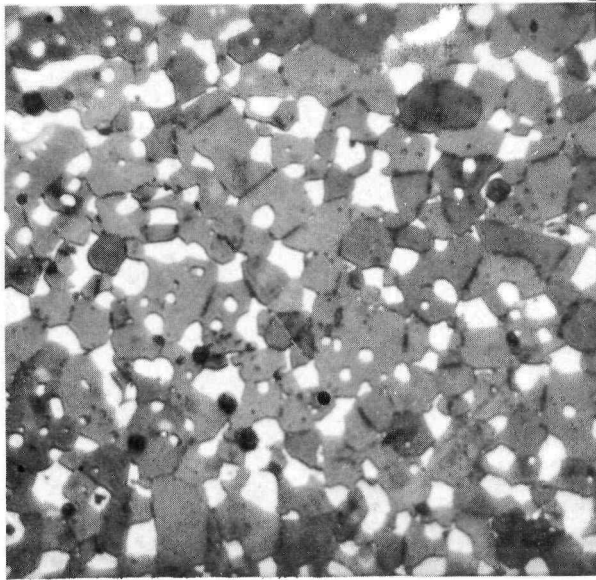
*More recent microprobe analyses show nickel to be in solution in the carbide rather than being concentrated in the grain boundaries.

Table 10 — Effect of High Temperature Anneal on UC-PuC-0.1 Ni Pellets

Experiment No.	Before Anneal				After 3 hr at 1750 °C			
	Intended Composition	Total Carbon, %	Sintering Temp, °C	Density, g/cm ³	Expansion, %		Density, g/cm ³	Weight Loss, %
					Diam	Height		
1*	(U _{0.8} Pu _{0.2})C _{0.95} +0.1% Ni	5.25	1550	12.41	0.16	0.06	12.34	0.252
2†	(U _{0.8} Pu _{0.2})C _{1.0} +0.2% Ni	4.80	1550	12.86	0.21	0.92	12.70	0.304
3†	(U _{0.8} Pu _{0.2})C _{1.0} +0.2% Ni		1450	12.79	0.36	0.61	12.48	1.15

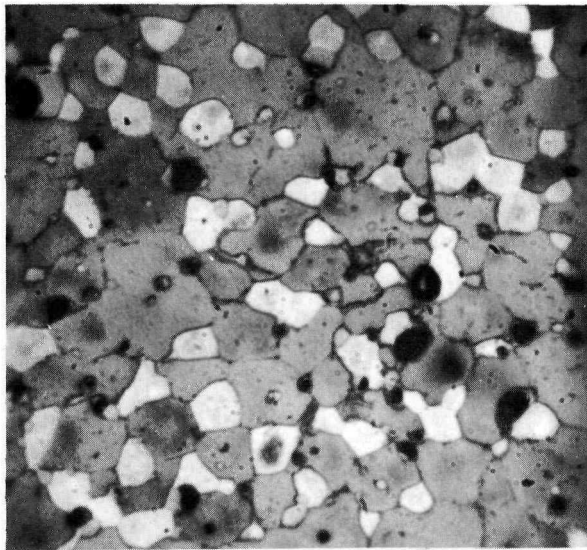
*Powder made from mixed oxides.

† Powder made from sesquicarbide.



Neg. No. P12

Fig. 5 — $(U_{0.8}Pu_{0.2})C_{0.95} + 0.1\% Ni$ — sintered at $1550^{\circ}C$, 1 hr (Exp. 1, Table 10). 600 \times . Etchant: nitric acid-acetic acid-water.



Neg. No. P10

Fig. 6 — $(U_{0.8}Pu_{0.2})C_{0.95} + 0.1\% Ni$ — sintered at $1550^{\circ}C$, 1 hr; reheated at $1750^{\circ}C$, 3 hr in helium (Exp. 1, Table 10). 600 \times . Etchant: nitric acid-acetic acid-water.

the stability of the U_2C_3 during vacuum annealing may be due to the items listed below.

1. The greater thermodynamic stability of $(U,Pu)_2C_3$ compared to the limited stability of U_2C_3 . (PuC_3 melts at $2050^\circ C$ without decomposing.)
2. The lower annealing temperature used for the $(U,Pu)_2C_3$ than for U_2C_3 ($1750^\circ C$ instead of $1850^\circ C$).
3. The extent of oxygen reaction and nickel evaporation may not be as great in helium as in vacuum. Both oxygen and nickel present with monocarbide promote the presence of sesquicarbide.

3.5 CHEMICAL ANALYSIS TECHNIQUES

The chemical analysis techniques used were previously reported in NDA 2145-6 and NDA 2162-5. Improved plutonium and carbon analysis techniques were developed at United Nuclear under Contract AT(30-1)-3118 and are described in the corresponding reports.

4. IRRADIATION TESTS

4.1 INTRODUCTION

In order for UC-PuC 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 of all fuel atoms by fission is desirable at fuel temperatures above 1200°F (650°C) with maximum dimensional stability of the fuel and minimum release of fission products. Power generation rate should be at least equivalent to that of 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 tests measure temperatures, burnup, power generation rate, dimensional stability of the fuel, and fission gas release. Post-irradiation examination of UC specimens was completed during Phase IV. Experiments with UC-PuC specimens are awaiting reactor insertion.

4.2 POST-IRRADIATION EXAMINATION OF UC SPECIMENS

4.2.1 Summary of Test

The post-irradiation examination of the UC irradiation test, capsule W1-1, was completed. A detailed description of the experiment and irradiation conditions was given in NDA 2162-5, pp. 29 to 41.

In summary, 24% enriched UC specimens were irradiated, with stainless steel and niobium clad, immersed in sodium. Each specimen contained 16 fuel pellets,

0.191 in. in diameter, for an approximate 3-in. fueled length. The void space was filled with helium. The UC pellets were made by cold pressing and sintering UC powder, which, in turn, was made by reacting UO_2 with carbon. The carbon content was 4.34 w/o, and the oxygen content was 0.3 w/o. The structure had a small amount of free metal near the pellet OD; the amount was considerably less than one would expect if the oxygen impurity were lower (see Fig. 13). The average density of the pellets was 94% of theoretical.

Irradiation conditions were 1400°F (760°C) average central fuel temperature, 9.6 kw/ft (315 w/cm) average heat generation rate, 1.7 kw/cm^3 average power density, and average $\int_{t_s}^{t_c} k d\theta$ of 25 w/cm.

Based on fuel center and clad surface temperature measurements, the thermal conductivity for the UC + He gap was 4.4 to 5.3 Btu/hr-ft²-°F/ft (0.018-0.022 cgs). Assuming UC conductivities in the range of 10 to 14 Btu/hr-ft²-°F/ft (0.041-0.058 cgs), the gap conductances are 1500 to 2800 Btu/hr-ft²-°F (0.85-1.6 w/cm²/°C). An average gap conductance value of 2000 Btu/hr-ft²-°F agrees with values determined by UO_2 investigators.*

The reverse assumption can be made; for a reasonable gap conductance, the in-pile thermal conductivity of UC is within the range of out-of-pile values.

4.2.2 Visual and Dimensional Examination

Clad Specimens

The appearance and dimensions of the two clad specimens did not change during irradiation. Photographs of the specimens before and after irradiation are shown in Figs. 7 and 8.

*I. Cohen, B. Lustman, and J. Eichenberg, Measurement of Thermal Conductivity of Metal-Clad Uranium Oxide Rods During Irradiation, Fig. 12, p. 17, WAPD-228 (Aug. 1960).

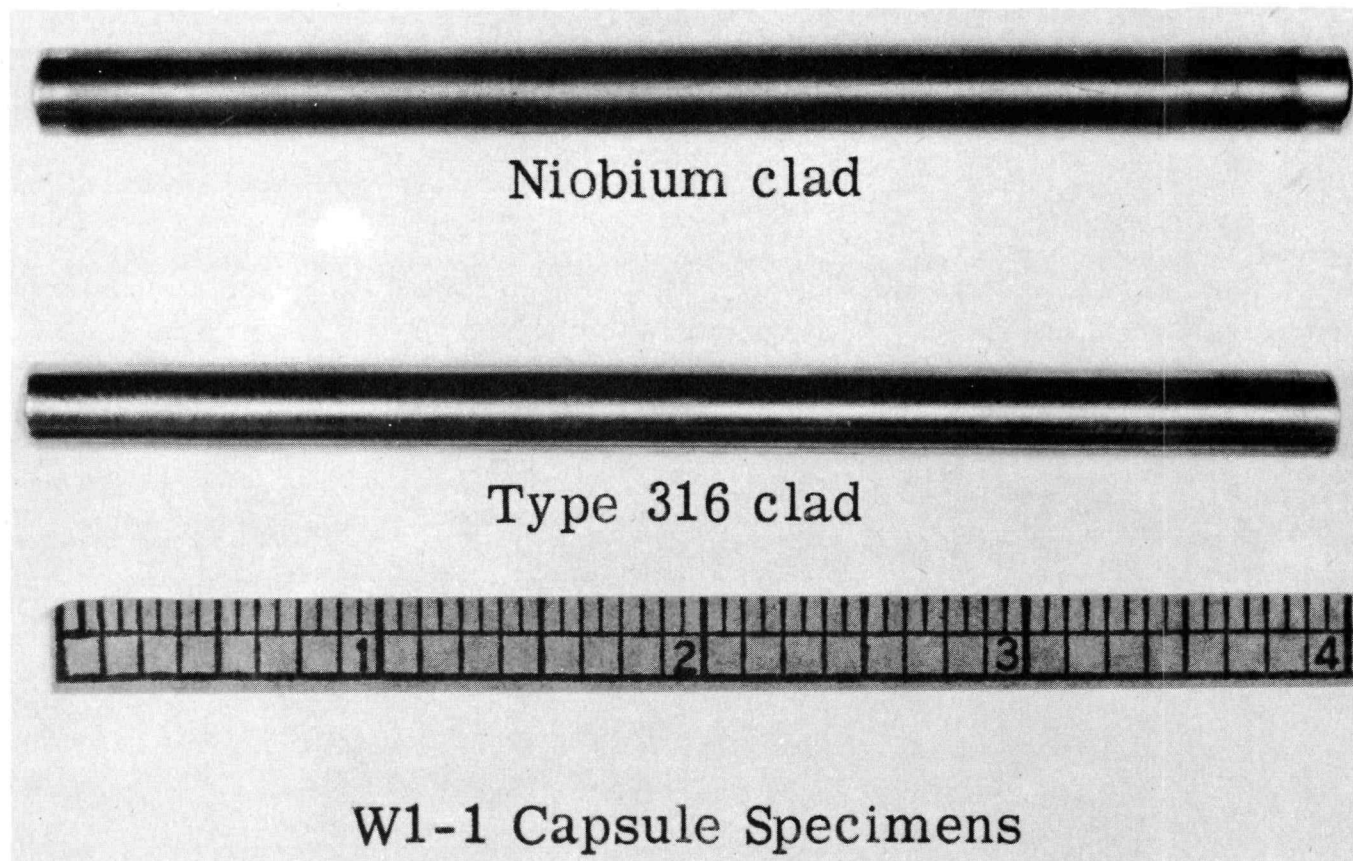


Fig. 7 — Niobium and type 316 clad UC fuel specimens prior to irradiation

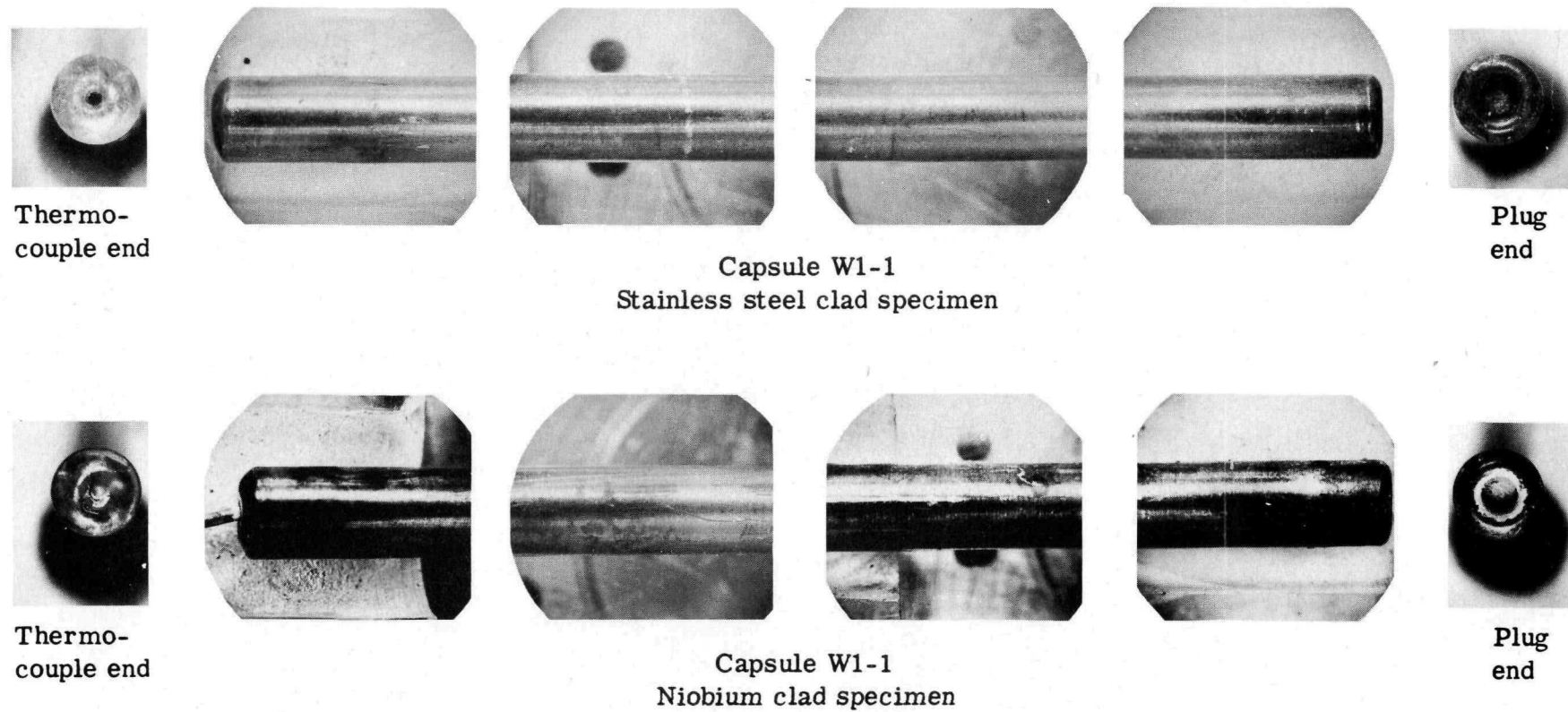


Fig. 8 — Niobium and type 316 clad UC fuel specimens after irradiation

During capsule disassembly the capsule inner shell was punctured and a gas sample taken to determine whether any fission gases had escaped the clad specimen. No activity could be detected in the gas sample bulb after puncturing. Subsequent sectioning of the specimens did not reveal any sodium inside the cladding. The cladding and welds, therefore, remained leaktight throughout the experiment.

The diameter of both specimens was checked every 1/2 in. on two diameters 90° opposed, and found to be identical to the dimensions prior to irradiation. The niobium-clad specimen had a slight bow (~1/64 in.); the stainless steel specimen was straight. The surface appearance of both specimens was the same as prior to irradiation.

Fuel Pellets

The general appearance of the UC pellets after irradiation was the same as prior to irradiation, except that a third of the pellets were cracked into two or more pieces. The pellet diameters increased an average of 0.001 in.

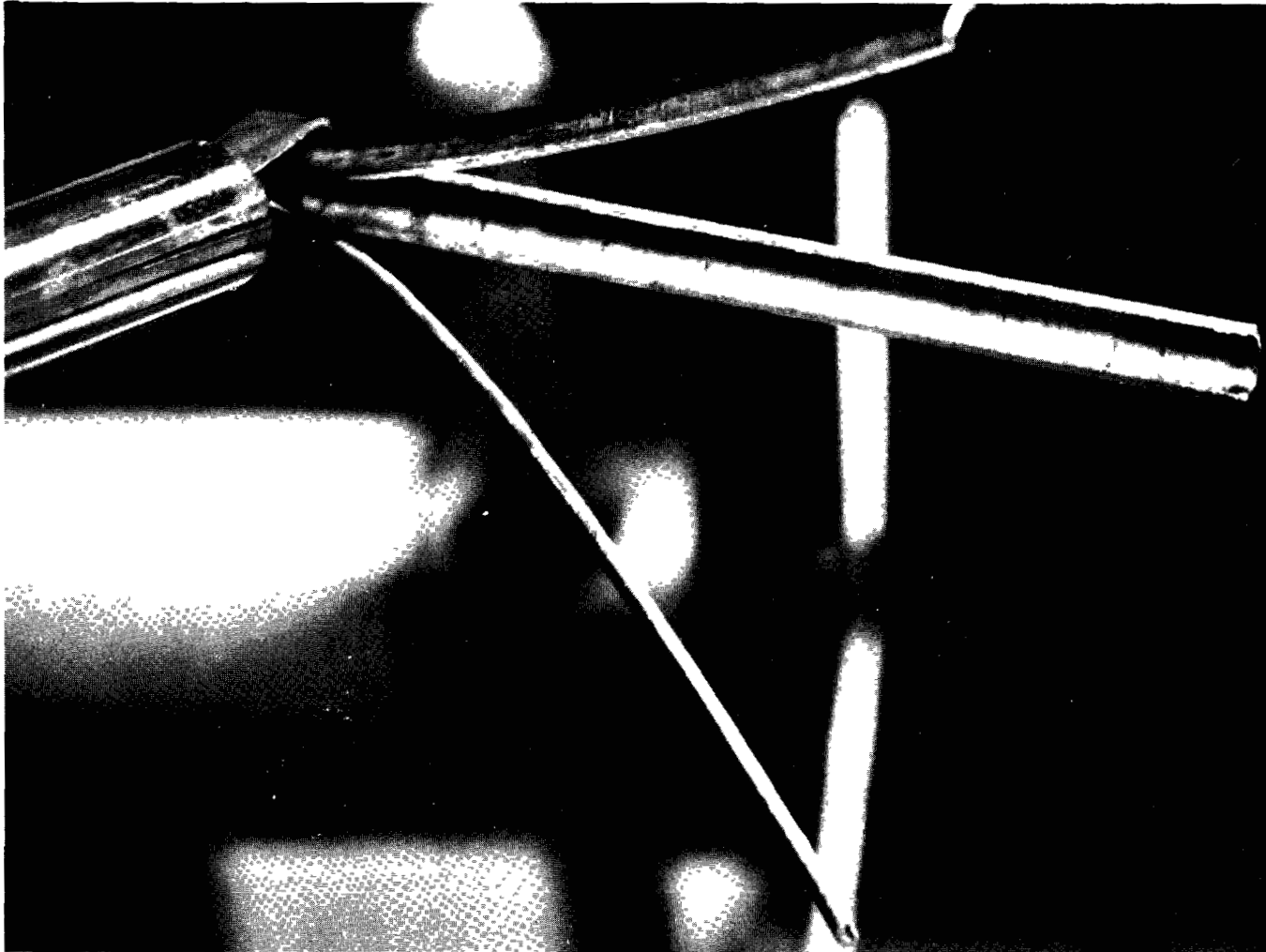
In order to remove the pellets, the cladding of both specimens was slit longitudinally. Fig. 9 shows the opened stainless steel cladding. The UC pellets were removed, examined visually, and measured. The condition of the pellets was as follows:

Stainless Steel-Clad Specimen

15 pellets	One piece (seven of the 15 pellets had a discontinuous small crack along a major diameter)
1 pellet	Cracked in two pieces

Niobium-Clad Specimen

6 pellets	In one piece
2 pellets	Cracked in two pieces
1 pellet	Cracked in four pieces
7 pellets	Cracked in more than four pieces



Neg. No. 3705

Fig. 9 — Type 316 stainless steel cladding, slit open for pellet removal

The fuel was considerably more brittle after irradiation, and the pellets were probably cracked during decladding in the hot lab. The niobium-clad specimen was more difficult to disassemble and required more severe handling, probably accounting for the greater number of cracked pellets. The breaking of the edge around the central hole was probably caused by incipient cracks introduced during the pre-irradiation drilling operation, and aggravated by irradiation. Fig. 10 shows a group of typical, uncracked pellets, Fig. 11 shows pieces of a typical cracked pellet, and Fig. 12 shows a typical uncracked pellet. Discontinuous radial and circumferential cracks were noted in some of the pellets.

A summary of the pellet measurements before and after irradiation is given in Table 11.

The 0.001 in. increase in diameter corresponds to a volume increase of 1.4%. The slightly increased diameter of the pellets corresponds to the ID of the cladding: 0.192 in. + 0.001 - 0.000.

Densities were calculated from dimensional measurements and weights. Post-irradiation measurements were taken on the 19 whole pellets. The density values of the latter are on the low side of the true range, since a slight amount of chipping at the edges reduces the weight without reducing the measured volume. The 2.0 g/cm³ decrease in density corresponds to 2.1%, and is within experimental error of the 1.4% volume increase. The actual density decrease is probably closer to the 1.4% value.

4.2.3 Metallographic Examination

Fuel

The structure and grain size of the UC appeared the same after irradiation as before (Figs. 13 and 14). The irradiated sample had slightly less free uranium metal in the structure; however, the difference is probably a difference between

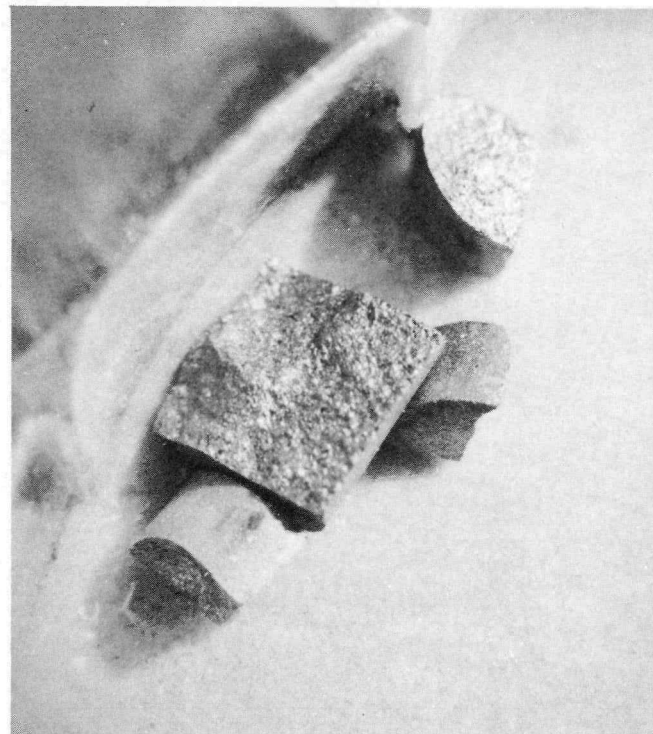
Table 11 — Uranium Carbide Pellets — Comparison of Dimensions
and Densities Before and After Irradiation

	Before Irradiation		After Irradiation	
	Average	Range	Average	Range
Diameter, in.	0.1910	0.1905 to 0.1916	0.1925	0.1917 to 0.1964
Height, in.	0.1833	0.1792 to 0.1874	0.1835	0.1806 to 0.1860
Density, g/cm ³	12.79	12.63 to 13.09	12.52	12.17 to 12.69
Percent of Theoretical Density	93.8	92.7 to 96.0	91.8	89.3 to 93.1



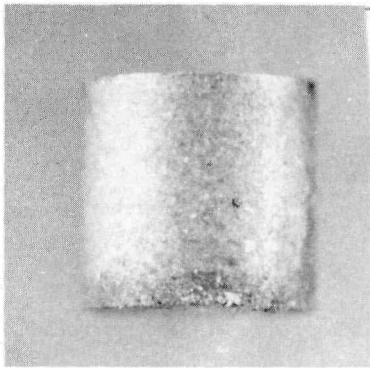
Neg. No. 3713

Fig. 10 — A group of whole irradiated UC pellets from the type 316 clad specimen. (Central hole was drilled prior to irradiation for thermocouple well.)

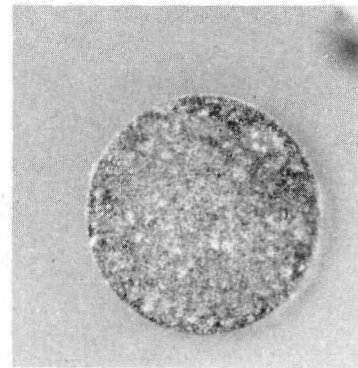


Neg. No. 3716

Fig. 11 — Typical cracked pellet from the niobium clad specimen

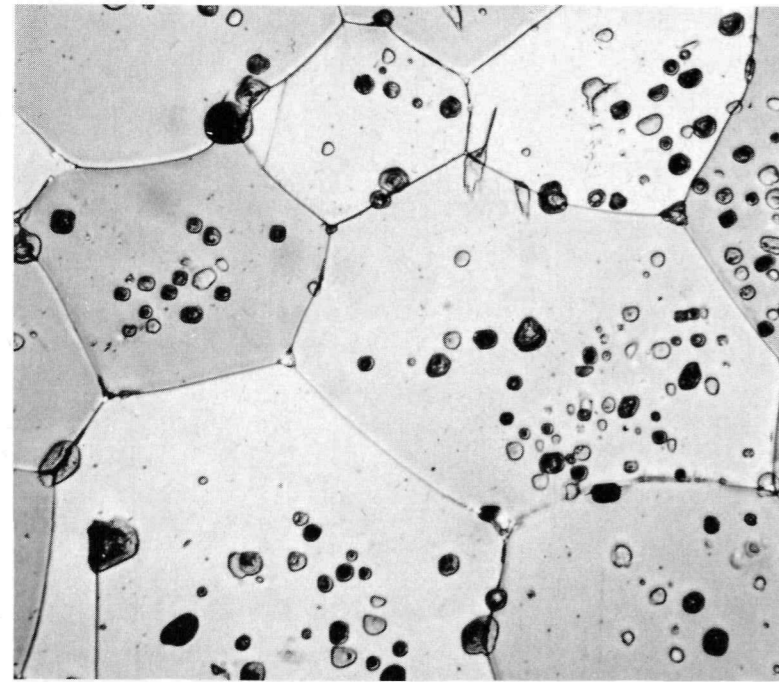


6×

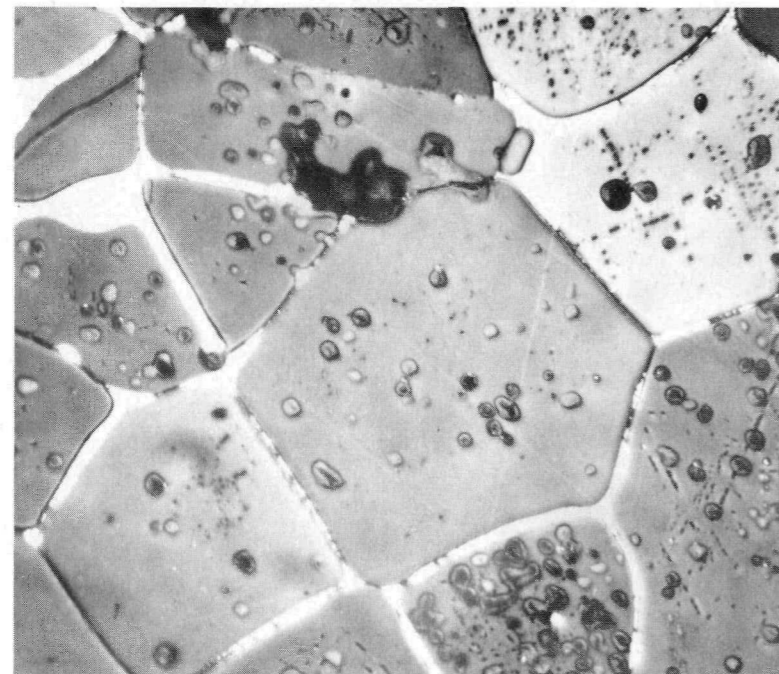


6×

Fig. 12 — Typical irradiated whole UC pellet



Section from center of pellet



Section from near surface of pellet

X-ray Analysis:

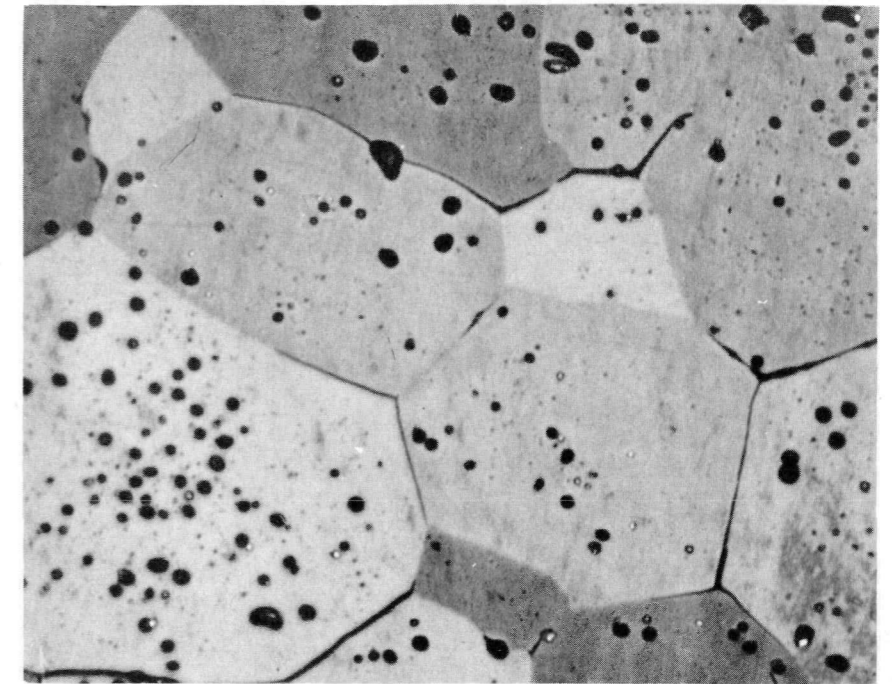
Major UC
 Very faint UO_2
 No UC_2

Chemical Analysis:

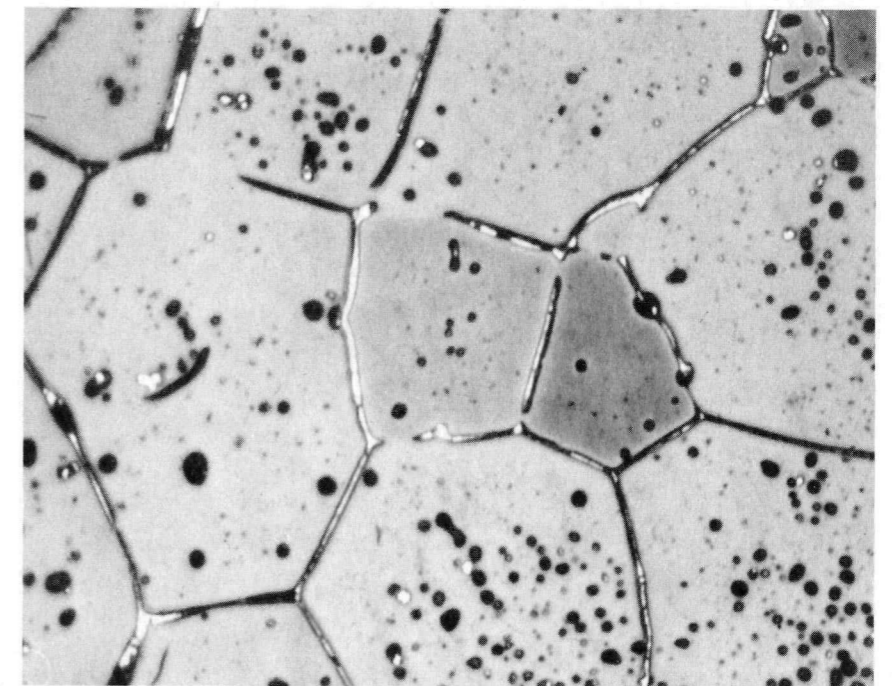
Total carbon 4.34%
 Free carbon < 0.05%
 Uranium 95.37%
 Iron < 0.01%
 Nitrogen 0.04%
 Oxygen 0.32%

Enrichment: 24%

Fig. 13 — UC, unirradiated — 500 \times . The white phase at grain boundaries is metallic uranium. The dots are voids and oxide in the structure. Etchant: nitric acid-acetic acid-water.



Section from center of pellet — irradiated ~10,000 MW-d/tonne — 1400°F



Section from near surface of pellet — irradiated ~24,000 MW-d/tonne — ~1150°F (at 2000 Btu/hr-ft²-°F conductance)

Fig. 14 — UC, irradiated — 500 \times . Some of the metallic uranium oxidized (dark). Etchant: nitric acid-acetic acid-water.

two separate samples. The post-irradiation metallography was performed by Battelle Memorial Institute.

The pellet examined was taken from about the center of the stainless steel clad specimen (Fig. 15). The cross section had discontinuous, circumferential, hair-line cracks at $\sim 1/3$ of the distance from the OD. The highest stress would be expected at the OD: $\sim 40,000$ psi, which is well above the known strength of UC. However, no cracks were found at the OD on any of the pellets. The explanation must lie beyond our limited knowledge of UC mechanical properties.

Metal Specimen Parts

Post-irradiation examination of metal specimen parts, which had been in contact with UC, showed that niobium did not interact and type 316 stainless steel interacted slightly, but without harmful effects on its ductility. The post-irradiation metallography was done by the CANEL Facility of Pratt and Whitney Aircraft Division of the United Aircraft Corporation.

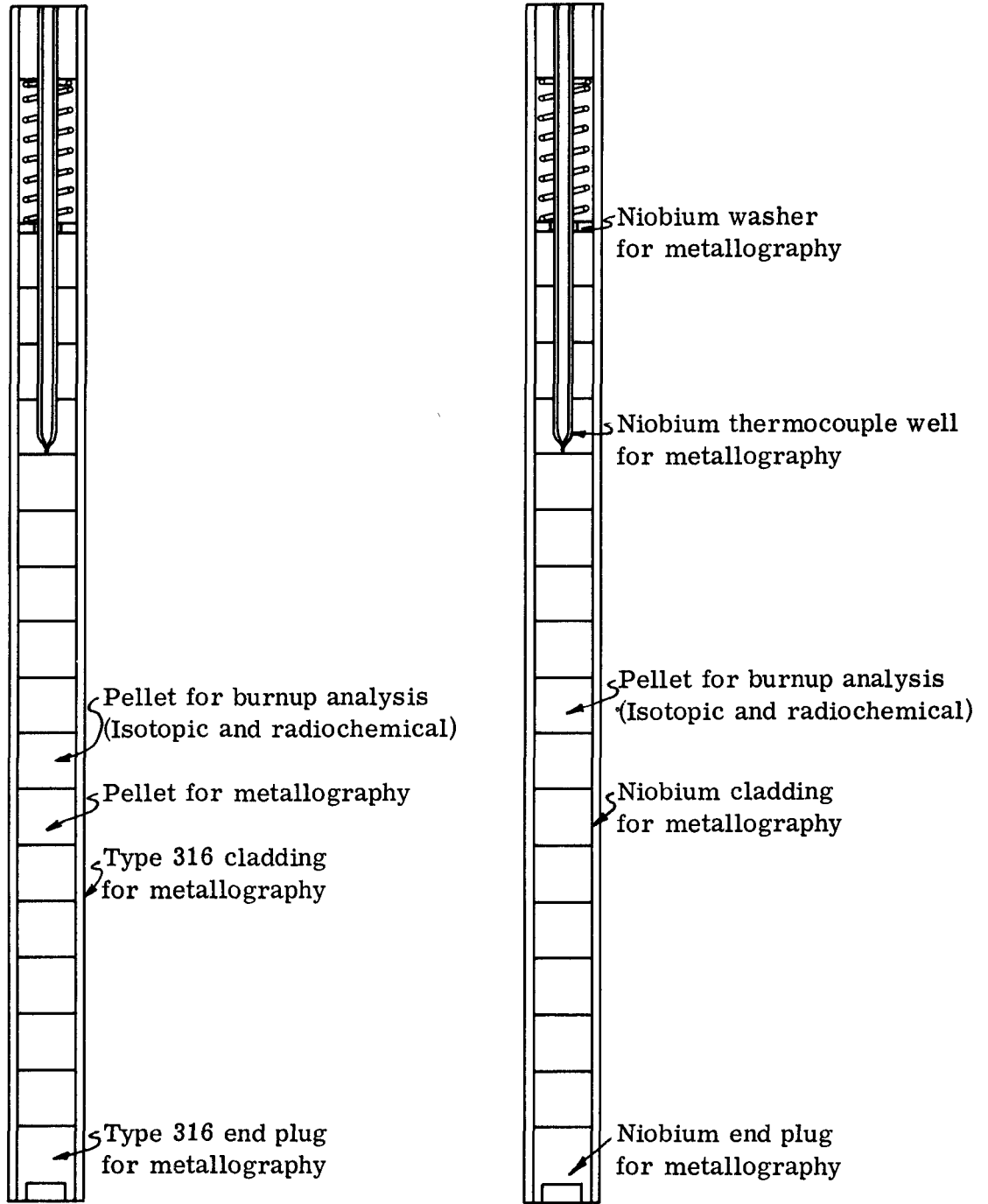
Samples were selected from specimen locations shown schematically in Fig. 15.

The niobium parts operated for approximately 2800 hr as follows.

End of center thermocouple well	1400°F (760°C)	} average temperatures*
Center of specimen end plug	1400°F (760°C)	
ID of cladding	800°F (430°C)	

No metallographic evidence for interaction was found between any of the niobium parts (central thermocouple well, cladding, end plug, washer) and UC. Microhardness measurements gave uniform readings throughout the specimens. The material was ductile in a 90° bend test with the ID in tension. No cracks were seen by macroscopic examination. The end of the thermocouple well is shown in Fig. 16.

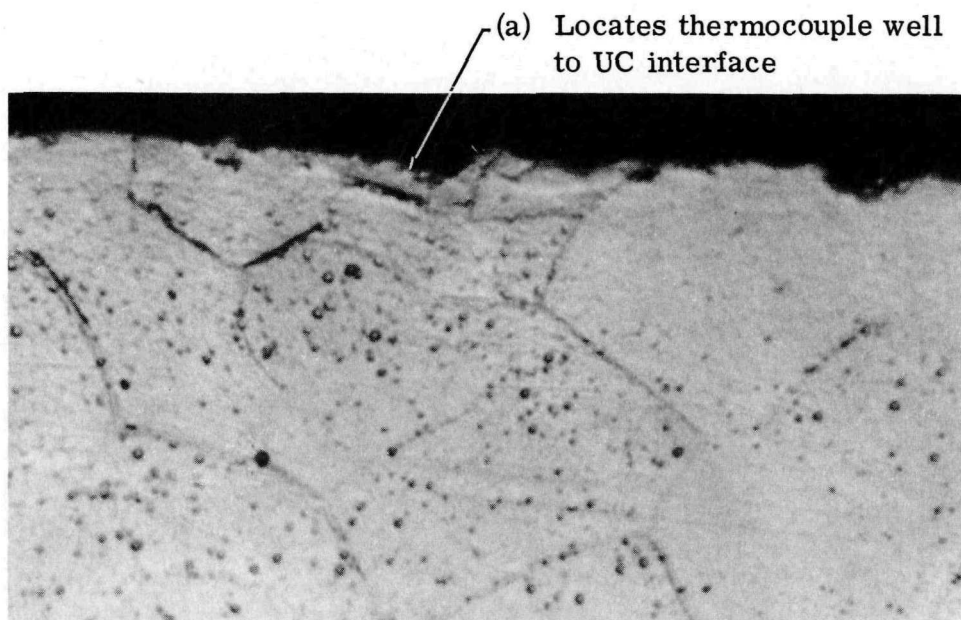
*For exact record, see Fig. 4.5, NDA 2162-5.



Type 316 stainless steel clad specimen

Niobium clad specimen

Fig. 15 — Location of metallographic and burnup samples in irradiation specimens



Etchant: $\text{HNO}_3, \text{HF}, \text{H}_2\text{O}$

880×

Fig. 16 — Nb thermocouple well from Nb clad irradiation specimen

The type 316 stainless steel cladding also operated for about 2800 hr, at an average ID temperature of 920°F (490°C).

A very slight interaction was noted between the steel parts and UC. While some overall carbide precipitation was due to the thermal history of the specimen, a narrow band of the stainless steel clad and end plug adjacent to the UC contained more intergranular carbide precipitate. The microstructures of the steel before and after irradiation are compared in Figs. 17 and 18. A comparison of microhardness measurements before and after irradiation showed hardening of the irradiated specimen due to the general carbide precipitation, and some preferential hardening of the surface near the UC. Results are given in Table 12. The material was ductile in a 90° bend test with the ID in tension. No cracks were seen by macroscopic examination. It can be concluded that the slight interaction is not sufficient to be deleterious to the properties of the cladding.

4.2.4 Fission Gas Release Measurement

The sample taken from the stainless steel specimen showed that 0.4% of the fission gases generated during irradiation was released. This amount corresponds to the volume of fission gas that would leave the surface of the fuel specimen as a direct result of recoil of fission fragments.

The specimen cladding was punctured and gas release measured, using the system shown in Fig. 19. Total gas volume was measured by allowing the gas to escape into an evacuated system of known volume, then measuring the pressure of the system. A fraction of the released gas was expanded into a sampling bulb, sealed, and analyzed on a mass spectrometer.*

The system volume was calibrated by crushing glass vials of known volume in the evacuated system and measuring the resulting pressure. The pressure was measured by a differential pressure transducer. The results are shown in Table 13.

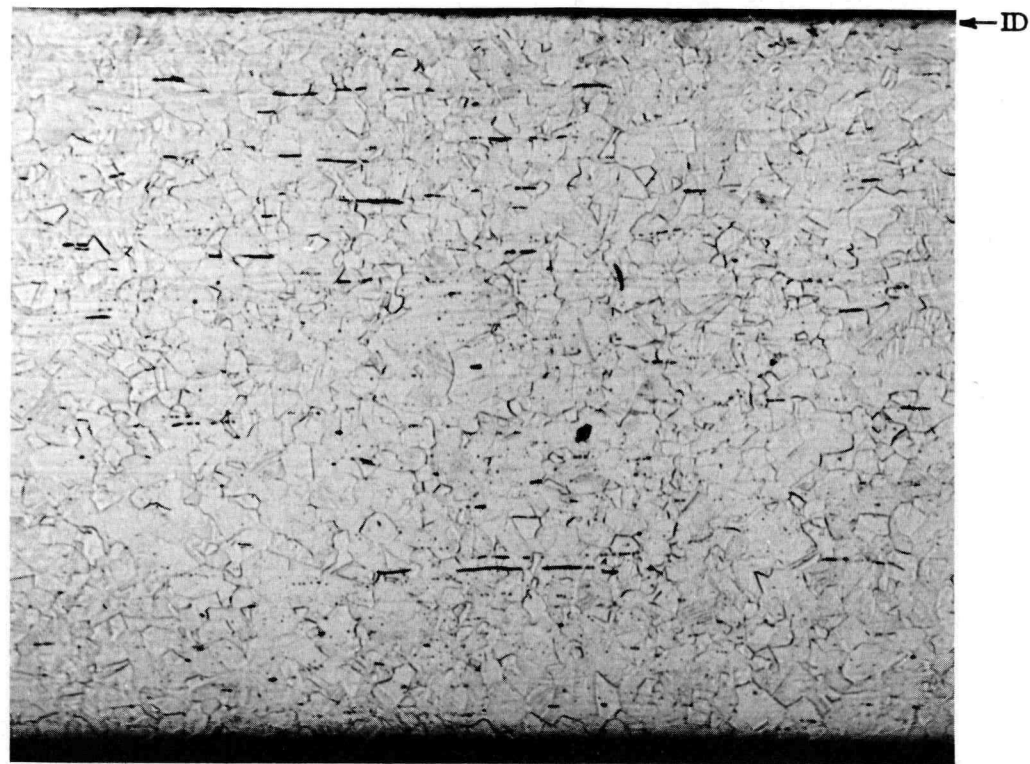
*Mass spectrometer analyses were made by Oak Ridge National Laboratory.

Table 12 — Pre- and Post-Irradiation Hardness
of Type 316 Cladding Tubing

Location	Hardness (DPH), 100-g Load	
	Unirradiated	Irradiated
	(Avg. of 4 Readings)	(Avg. of 2 Readings)
Near OD	263	307
Middle of wall	266	264
Near ID	308	430

Table 13 — Fission Gas Release from UC

Specimen	Total Fission Gas, cc at S.T.P.	Gas Composition	% Release of Theoretical Xe and Kr Produced	% Theoretical Release from Surface Recoil
Type 316 Clad-UC	0.035	81% He 9.1% Xe 5.1% Kr 4.5% N	0.41	0.35

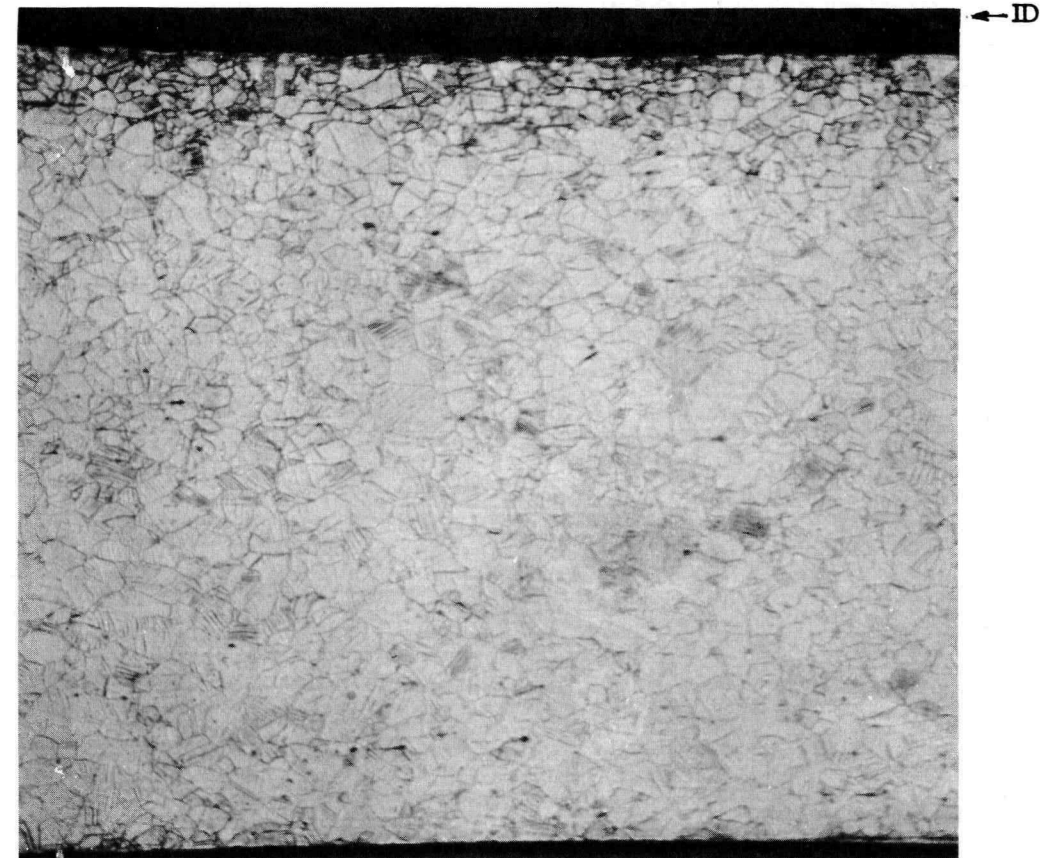


100x

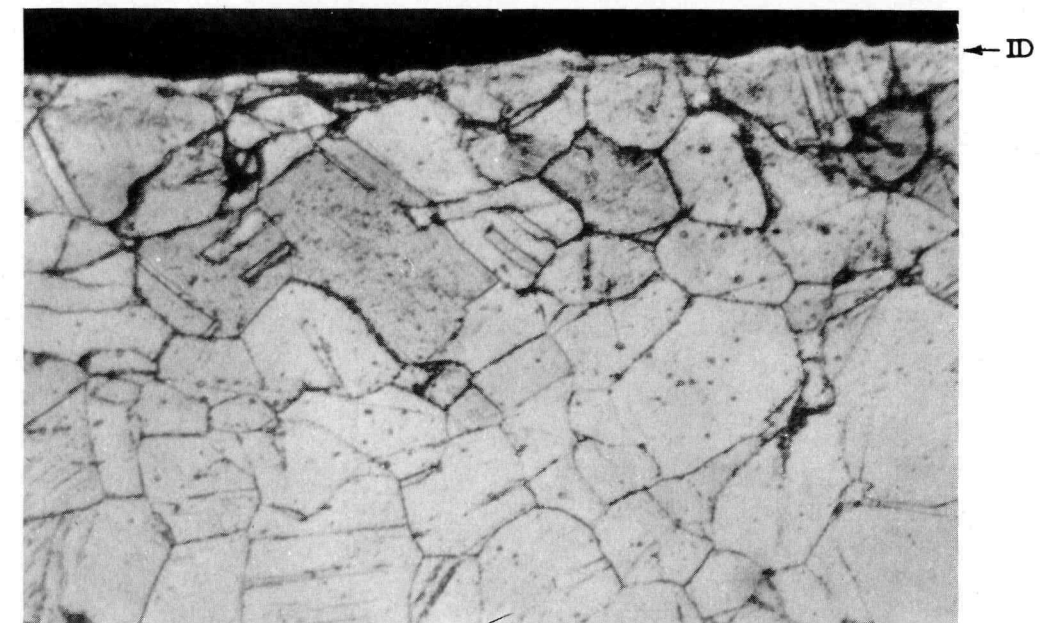


500x

Fig. 17 — Type 316 stainless steel cladding, unirradiated — electrolytically etched



100x



500x

Fig. 18 — Type 316 stainless steel cladding, irradiated — Kallings etch

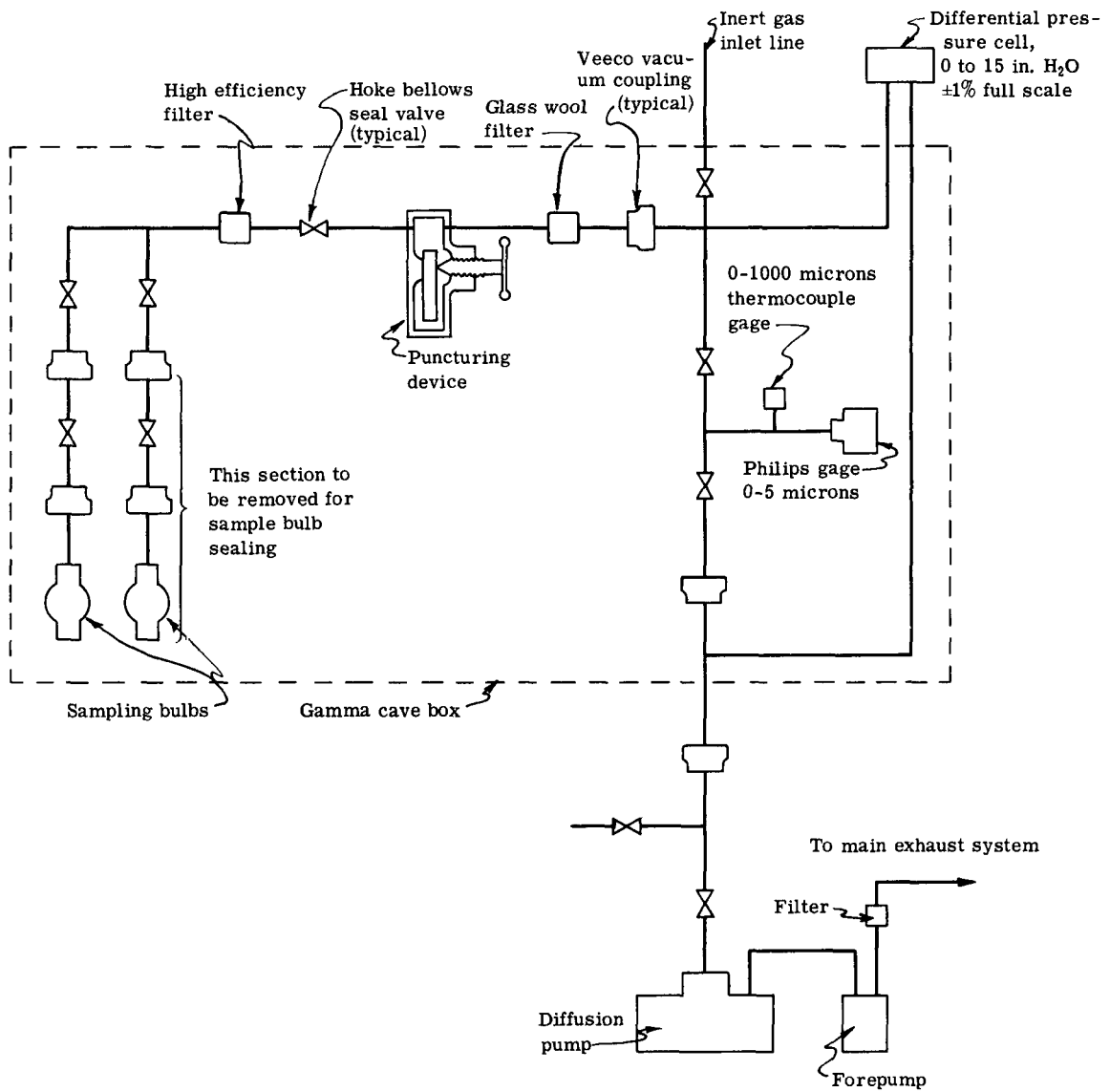


Fig. 19 — Schematic of fission gas collection system

4.2.5 Burnup Determinations

The average burnup of the UC was 16,100 to 16,400 MW-d/tonne. Burnup varied radially between 9000 and 25,000 MW-d/tonne.

The UC burnup was estimated by three different methods.

1. In-pile Calorimetry – calculation from total power generated by the specimen during the test.

The power generation data were obtained from in-pile heat flow measurements taken three times daily.

2. Radiochemical Analysis – mass spectrometer analyses for Cs¹³⁷.

The analyses were made on an aliquot from a solution of one complete pellet taken from each specimen (Fig. 15), by the Phillips Petroleum Co., Idaho Falls, Idaho.

3. Isotopic Analysis – mass spectrometer analyses for U²³⁴, U²³⁵, U²³⁶, and U²³⁸.

The analyses were made on the same sample as the Cs¹³⁷ analyses.

The results are compared in Table 14. The isotopic analyses are considered the most reliable. The variation among the analyses is within the range expected from past experience.

The flux depression produced a radial burnup variation in the 24% enriched fuel. The flux depression was calculated by the use of the Taraba-Paine data* and the average fuel isotopic burnup. Table 15 shows the results.

*F. R. Taraba and S. H. Paine, The Radial Distribution of Thermal Neutron Flux in Cylindrical Fuel Specimens During Neutron Irradiation, ANL-5872 (Aug. 1959).

Table 14 — Comparison of Average Burnup (MW-d/tonne) Determinations of the UC Specimens

	Calorimetric, in-pile	Radiochemical, Cs ¹³⁷	Isotopic, U
Stainless Steel Clad Specimen	18,600	15,800	16,100
Niobium Clad Specimen	18,400	14,800	16,400

Table 15 — Radial Variation in Burnup

	Maximum Burnup, MW-d/tonne	Minimum Burnup, MW-d/tonne
Stainless Steel Clad Specimen	24,600	9000
Niobium Clad Specimen	24,900	9200

4.2.6 Conclusions

The UC produced by the oxide-carbon reaction, and formed by cold pressing and sintering (4.34 w/o C, 0.32 w/o O, with a slight amount of free uranium metal) has excellent stability and fission gas retention ability at an average maximum fuel temperature of 760°C (1400°F) up to 16,400 MW-d/tonne burnup.

Niobium does not react with the UC tested at 800°F (430°C) for 2800 hr in-pile. Stainless steel reacts slightly, at 490°C (920°F) for 2800 hr in-pile, but the reaction is not harmful to its properties.

4.3 DESIGN FOR $(U_{0.8}Pu_{0.2})C_{0.95}$ IRRADIATION TESTS

The basic design conditions for the $(U_{0.8}Pu_{0.2})C_{0.95}$ irradiation tests are given in Table 16.

Table 16 — Design Irradiation Conditions for $(U_{0.8}Pu_{0.2})C_{0.95}$ Specimens

Capsule No.	Clad Surface Temp, °F (°C)	Fuel Center Temp, °F (°C)	Power,		Burnup, MW-d/tonne
			kw/ft	kw/cc	
61	1060 (570)	1870 (1020)	14.4	2.56	16,600
62	1060 (570)	1870 (1020)	14.4	2.56	33,200
63	1065 (575)	1875 (1025)	14.0	3.04	66,400
64	1370 (740)	2660 (1460)	20.0	3.56	16,600
65	1275 (690)	2565 (1405)	20.0	3.56	33,200
66	1300 (705)	2575 (1410)	21.0	3.75	66,400

Each capsule contains two specimens identical or similar to the UC irradiation specimens shown in Fig. 15. One specimen contains $(U_{0.8}Pu_{0.2})C_{0.95}$ with 0.12 w/o nickel sintering aid and the other specimen contains $(U_{0.8}Pu_{0.2})C_{0.95}$ without nickel. The highest burnup capsules (Nos. 63 and 66) contain 24% enriched uranium to shorten the required irradiation time. (The other capsules contain normal urani-

um.) The enriched fuel pellets are slightly smaller in diameter in order to operate at the desired power level and temperature. The ratio of void volume to fuel volume is also greater, to accommodate a greater volume of fission gas.

The specimens for the lower power capsules (Nos. 61 to 63) have niobium central thermocouple wells. The cladding and end plugs are also niobium, so that reliable welds can be made. The specimens for the higher power capsules (Nos. 64 to 66) do not have provision for center fuel temperature measurement. The cladding and end plugs are stainless steel, because this is potentially one of the most economical fuel clads for a fast central station power plant.

The capsule design is an improved version of the one used for the UC irradiation. The specimens are immersed in sodium contained in the finned inner stainless steel shell. A radiograph of a typical assembly in an inner shell is shown in Fig. 20. The inner shell fits tightly into the outer stainless steel shell. The instrument conduit is welded directly to the outer shell. The conduit and the annulus between the two shells are filled with helium.

A total of up to 10 thermocouples are located at the specimen center, specimen cladding surface, and in the annulus between the inner and outer shells, to measure radial heat flow. Sheathed electric heaters in the inner shell are used to calibrate capsule thermal resistance; that is, to obtain the relationship between radial temperature drop and a known amount of power.

A Ni-Co flux monitor wire is wrapped around the specimens, for post-irradiation burnup determination by Co activation analysis.

Each capsule has an instrument junction box at the end of the conduit to provide a gas-tight seal for the conduit. An instrument console, connected to the junction box, serves to record temperatures, and to control and measure power to the capsule heaters.

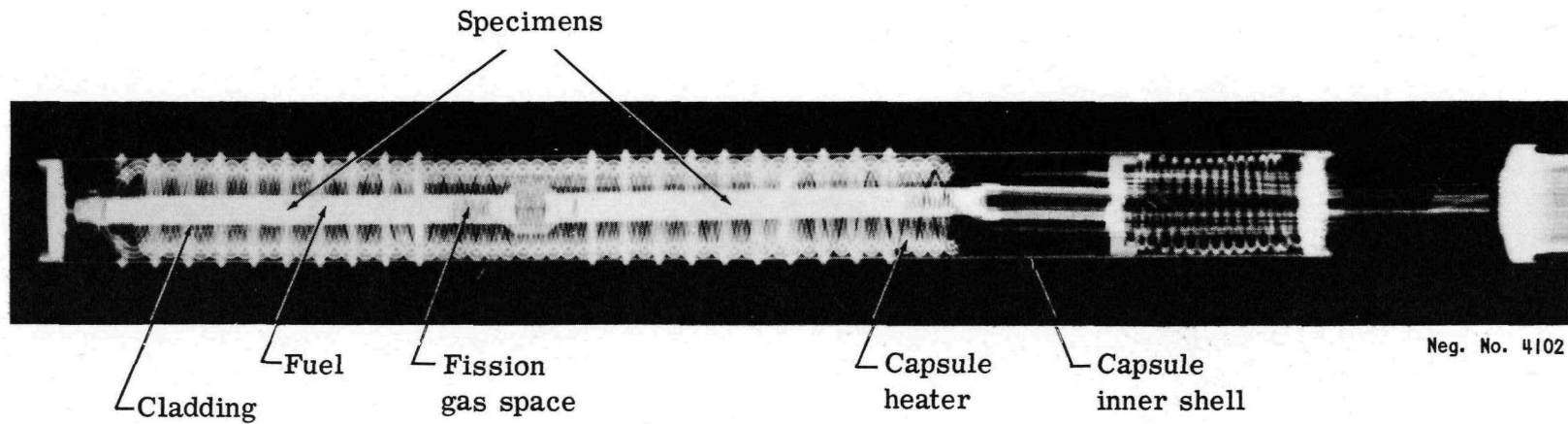


Fig. 20 — Radiograph of a typical irradiation capsule

The MTR and WTR were originally chosen as the most desirable test reactors based on available fluxes, cost, and schedule. The capsules were designed and their components fabricated to suit these reactors. Subsequently, WTR closed operations, and MTR informed United Nuclear that the required number of experimental locations with the required fluxes were no longer available. A re-evaluation of the remaining available test reactors, the ETR and GETR, recommended GETR.

Four of the capsules were recommended for GETR reflector positions and two for core positions. The desired core positions had not been used for capsule irradiation before, and there were two potential mechanical difficulties: (1) insertion of the capsule past a number of obstructions, and (2) vibration of the capsule and conduit due to water turbulence. To assure successful operation of the capsules, a dummy capsule will be operated in one of the proposed positions for one cycle. The capsule will contain flux monitors, to make an improved flux estimate possible. The dummy capsule design was completed by GETR.

4.4 CONSTRUCTION OF $(U_{0.8}Pu_{0.2})C_{0.95}$ IRRADIATION EXPERIMENTS

4.4.1 Fuel Specimen Assembly

The 12 specimens were assembled, leak-tested, and decontaminated.

A typical set of pellets for one irradiation specimen with a central thermocouple is shown in Fig. 21.

The method of loading is shown pictorially in Fig. 22. The loading device fits tightly into a counter-bore in the cladding. Aluminum foil, which serves to prevent cladding contamination, is wrapped around the cladding and is taped to the loading device. This assembly is put into the plutonium glove box, the pellets are loaded, the tape and loading device are removed, and the end plug is placed in the cladding and welded. The specimen is removed to the decontamination box, where

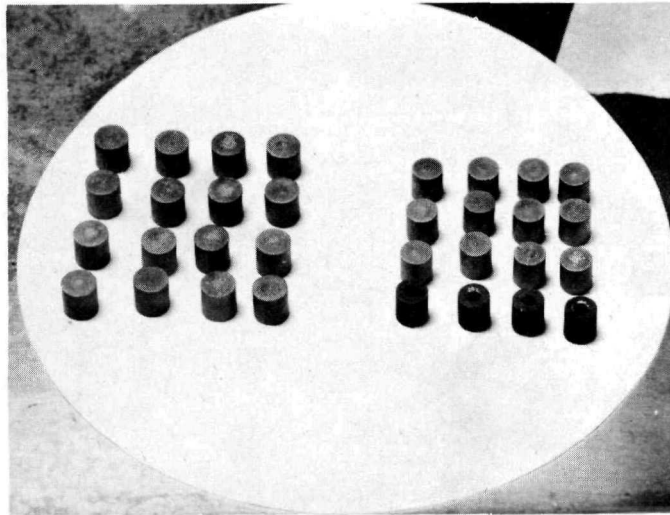
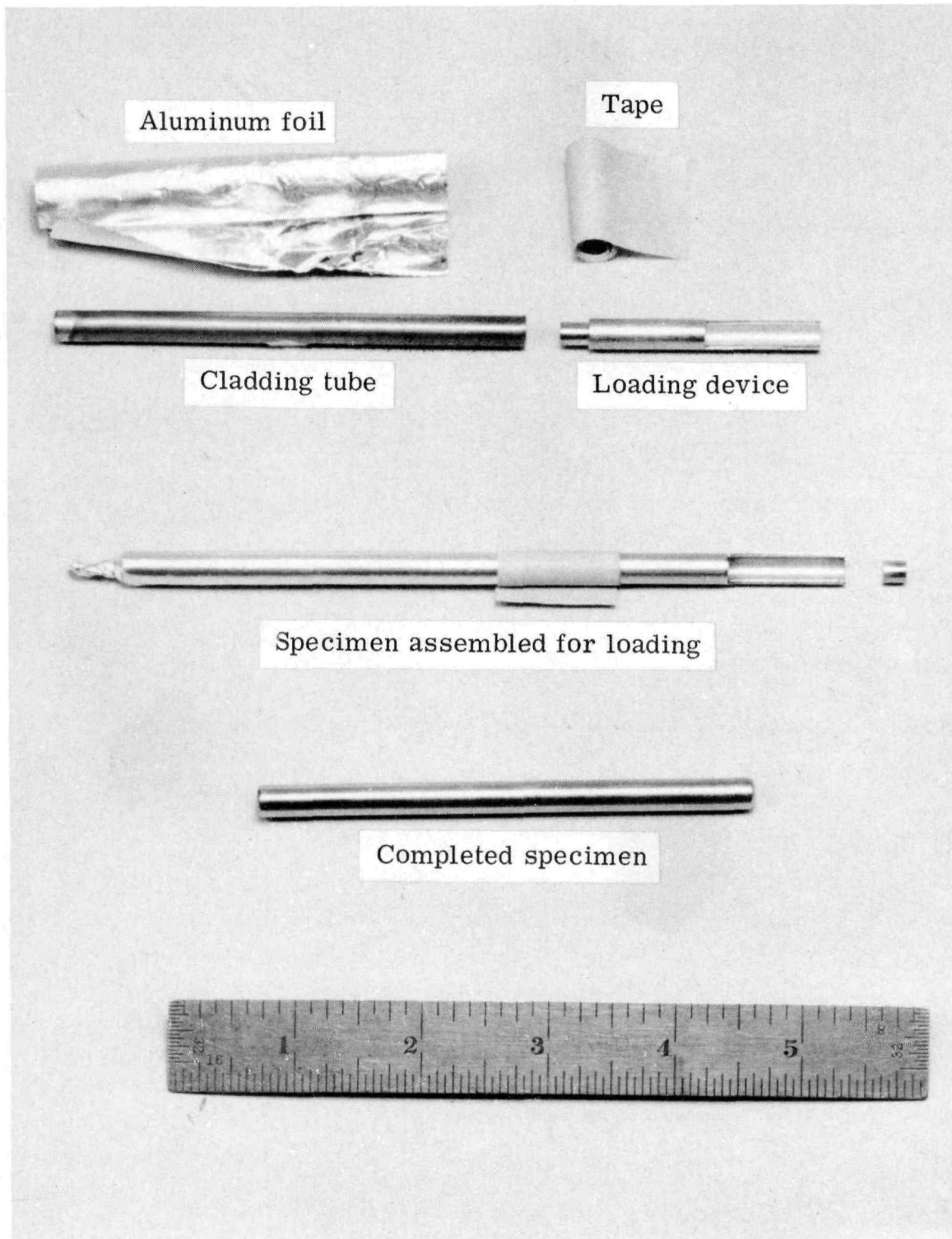


Fig. 21 — Typical group of $(U_{0.8}Pu_{0.2})C_{0.95}$ pellets for irradiation testing



Neg. No. 4092

Fig. 22 — Irradiation specimen loading method

the aluminum foil is removed and the specimen is monitored, decontaminated, and helium mass-spectrometer leak-tested.

The only chance for contamination of the specimen is at one end plug during and after welding. The contamination has been very slight (200 counts/min) and could be removed easily and completely by detergent or dilute nitric acid solutions.

Central holes for thermocouples were drilled in several $(U_{0.8}Pu_{0.2})C_{0.95} + 0.1 Ni$ pellets, using a tungsten carbide drill. A large percentage of the pellets had a laminar-type fracture. Cored pellets were then fabricated, as described in Section 3.3.2, to ease the drilling problem. Since the pressing and sintering process does not produce the hole size and concentricity of the required tolerances, the cored pellets were made with about a 0.060 in. hole and reamed to size with a tungsten carbide drill. Pellets with 0.0705 ± 0.0005 in. holes centered within the required 0.003 in. were produced satisfactorily by this process.

4.4.2 Experiment Assembly

The assembly of capsules 61, 64, and the dummy capsule was completed. The assembly of capsules 62 and 65 was initiated. Associated instrument consoles, cables, and junction boxes were completed.

Capsule 61 and the dummy capsule were shipped to and received by GETR.

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