MASTER

UNC-5015

FUEL CYCLE DEVELOPMENT PROGRAM

QUARTERLY PROGRESS REPORT
JANUARY 1 TO MARCH 31, 1962

June 8, 1962

Work Performed under Project 2801
Contract AT(30-1)-2374 with the
United States Atomic Energy Commission

UNITED NUCLEAR
CORPORATION

DEVELOPMENT DIVISION
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White Plains, N.Y.

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New Haven Research Laboratories
E. Gordon, Manager

Quarterly and Annual Reports Published under Fuel Cycle Development Program

NYO-2684	First Quarterly Progress Report Period Ending September 30, 1959
NYO-2685	Quarterly Progress Report October 1 to December 31, 1959
NYO-2687	Quarterly Progress Report January 1 to March 31, 1960
NYO-2688	The Development of Uranium Carbide as a Nuclear Fuel First Annual Report May 1, 1959 to April 30, 1960
NYO-2689	Quarterly Progress Report April 1 to June 30, 1960
NYO-2690	Quarterly Progress Report July 1 to September 30, 1960
NYO-2691	Quarterly Progress Report October 1 to December 31, 1960
NYO-2692	Quarterly Progress Report January 1 to March 31, 1961
NYO-2693	Quarterly Progress Report April 1 to June 30, 1961
NYO-2694	The Development of Uranium Carbide as a Nuclear Fuel Second Annual Report May 1, 1960 through August 31, 1961
NYO-2695	Quarterly Progress Report July 1 to September 30, 1962
UNC-3001	Quarterly Progress Report October 1 to December 31, 1961
UNC-3006	Low Temperature Sintering of Uranium Dioxide
NYO-2686	The Carbides of Uranium; An Annotated Bibliography

SUMMARY

This report presents the progress made by United Nuclear Corporation under Contract AT(30-1)-2374 during the period January 1 to March 31, 1962.

Under Task I, the permanent shutdown of the Westinghouse Testing Reactor at the end of the quarter forced a revision in plans for programming the remainder of the irradiation of the two uninstrumented capsules whose testing was in progress. The minimum estimated burnup at this point, based on hot cell data obtained from the pilot capsule, was 13,100 MW-d/ton U. It was decided to continue the testing of only one capsule in another reactor until the original goal of 20,000 MW-d/ton U is reached. The irradiation of the second capsule is to be terminated so that it can serve as a control.

Fabrication was initiated on enriched UO_2 pellets for incorporation in full scale fuel rods to be irradiated in the Vallecitos Boiling Water Reactor. A wet nitrogen pyrohydrolysis step in conjunction with oxidation-reduction cycling is being used to attain a satisfactory density exceeding 95% of theoretical at 1150°C.

Under Task II, apparatus and procedures being used for measurement of thermal conductivity and thermal expansion of sintered and cast uranium carbide are described. The coefficient of linear thermal expansion for a single specimen of 4.37 w/o carbon sintered uranium carbide was determined to be 11.8×10^{-6} mm/mm-°C, while that for a cast uranium carbide specimen containing 4.73 w/o carbon was 10.6×10^{-6} mm/mm-°C over the range from room temperature to 1000°C. Hardness data for 4.4 and 4.8 ± 0.1 w/o carbon sintered and cast uranium carbide and for 5.2 w/o carbon cast material are also presented.

Irradiation test specimens prepared by United Nuclear Corporation during the last report period were given a pre-irradiation examination and were encapsulated by Battelle Memorial Institute. Three capsules, two using a sodium bond between fuel and container and one using an interference fit between fuel and container, were loaded in the MTR and appear to be operating under the required conditions.

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1. DEVELOPMENT OF A LOW COST FABRICATION PROCESS FOR URANIUM OXIDE FUEL (Task I)

R. B. Holden, N. Fuhrman, L. D. Hower, Jr.

1.1 INTRODUCTION

Practically all of the work conducted on Task I during the reporting period was concerned with irradiation testing of UO_2 pellets produced by the low temperature sintering process via the nonstoichiometric route. Of the three capsules originally inserted in the Westinghouse Testing Reactor, two remained under irradiation until March 23, when the reactor was permanently shut down. Since the burnup in each capsule had not reached the desired 20,000 MW-d/ton U level, a plan for the completion of the tests had to be devised. Hot cell examination of the pilot capsule, which had been removed from the WTR after four cycles, was completed at the General Electric Vallecitos Atomic Laboratory, and the data developed were useful in establishing new test conditions for continuation of the irradiation of one of the capsules in another reactor.

During this period, the fabrication of several fuel rods containing UO_2 pellets for a high burnup irradiation experiment in the Vallecitos Boiling Water Reactor was also initiated.

1.2 IRRADIATION TESTING PROGRAM

1.2.1 Status of WTR Irradiations

Until near the end of this quarter, two capsules, each holding two specimen tubes containing enriched UO_2 pellets, as described in Table 1.1, were under irradiation in the WTR. Hot cell data (see UNC-3001) from examination of the pilot capsule removed earlier had indicated that the actual burnup rate was less than that planned. Arrangements for extending the irradiation period were being made, therefore, to assure that the burnup goal of 20,000 MW-d/ton U would be reached before the two capsules were removed. After 10 cycles had been completed, however, the WTR was permanently shut down, necessitating a revision of plans for continuing the test program. A summary of the irradiation units supplied by WTR up to this point is presented in Table 1.1.

Since all three capsules were of similar design and were exposed to essentially the same flux (see Table 1.1), the data developed from the pilot capsule were used to estimate the burnup level in the two capsules at shutdown. Extrapolation of flux monitor data leads to a maximum burnup level of 16,200 MW-d/ton U after 10 cycles. More credence is placed in an estimate of 13,100 MW-d/ton U based on the Ce^{144}/U burnup determination on the UO_2 in Specimen Tube 1 (shown in Table 1.2).

Since the burnup in either case is apparently still significantly less than 20,000 MW-d/ton U, it was decided to continue the irradiation of one of the capsules until this level is reached. To minimize uncertainties in the interpretation of final results arising from the transfer of an uninstrumented experiment to another reactor, the irradiation of the second capsule is to be terminated so that it can serve as a control. Capsule OLM-1-2 was selected as the latter in view of the fact that it contains hydrogen sintered pellets introduced originally for experimental control.

Further irradiation of OLM-1-3 has been assigned to MTR-ETR and the flux requested for test resumption will be based upon reverting to the initial heat generation conditions experienced in the WTR.

1.2.2 Hot Cell Examination of Pilot Capsule

Early in the quarter, a report on the post-irradiation examination of the pilot capsule (OLM-1-1) was received from the General Electric Vallecitos

Table 1.1 — Summary of Irradiation Units Supplied by WTR

Capsule No.	No. of WTR Cycles	Total I.U.*	I.U./Cycle
OLM-1-1	4	28.71	7.17
OLM-1-2	10	75.08	7.51
OLM-1-3	10	70.62	7.06

*One irradiation unit (I.U.) = 10^{20} nvt in.^{3}

Table 1.2 — Irradiation Capsule Data

Capsule No.	Specimen Tube No.	Geometric Pellet Density, % of Theoretical	Initial Diametral Clearance, in.	Sintering Temp, °C	Burnup,* MW-d/ton U
OLM-1-1 (Pilot)	1	95.1	0.002	1300	5950
	7	98.1	0.006	1200	7640
OLM-1-2	2	95.3	0.002	1300	—
	3	96.3	0.004	1550†	—
OLM-1-3	6	97.3	0.004	1000	—
	8	97.9	0.006	1200	—

*From Ce^{144}/U determinations.
†Hydrogen-sintered.

Atomic Laboratory. All of the preliminary results presented in the previous quarterly report (UNC-3001) were confirmed, except for the exposure data calculated from flux monitor analysis. It was learned that an incorrect Co^{60} absorption cross section of 41 barns was used as a basis for the exposure data originally reported. Since the appropriate cross section consistent with the WTR neutron energy spectrum is 29 barns, the values have been corrected and are reported in Table 1.3.

The burnup in each specimen tube calculated from the average exposure is presented in Table 1.4, where it may be compared with those determined by radiochemical measurement.

It should be noted that the radiochemical burnup data listed for Tube No. 7 were obtained on an end pellet rather than on a more representative specimen from the center of the stack, as in the case of Tube No. 1. The effect of flux suppression on going from the externally located monitors to the centrally positioned fuel can be clearly seen, therefore, only in the case of the latter.

It was hypothesized previously that the fuel in Tube No. 7 was operated at a higher temperature than that in Tube No. 1. This arose from a consideration of the difference in initial diametral clearance (0.006 vs 0.002) and the finding of a 100-fold greater Kr^{85} release in Tube No. 7. Further support for this view is presented in Figs. 1.1 and 1.2 in the form of composite photomicrographs of a representative pellet from each tube showing the variation of grain size from the outside to the center of the pellet after irradiation. The UO_2 in Tube No. 1 (Fig. 1.1) shows no difference, whereas that in Tube No. 7 (Fig. 1.2) exhibits definite evidence of a grain growth pattern in the central region.

In earlier work, to promote detectable grain growth in stoichiometric pre-irradiation specimens in out-of-pile annealing experiments, it was found necessary to soak the specimens at 1300°C for one week. The central fuel temperature during the irradiation was probably of the same order. P. Murray[1] has summarized percent gas release vs fuel center temperature data for several irradiation programs conducted with hydrogen-sintered UO_2. It is noteworthy that the

Table 1.3 — Pilot Capsule Exposure Data

Specimen Tube	Location of Monitor	Exposure, 10^{20} nvt At Location	Average
1	Top	4.83	
	Full length	4.37	4.51
	Bottom	4.32	
7	Top	5.07	
	Full length	4.41	4.77
	Bottom	4.83	

Table 1.4 — Comparison of Burnup Measurements
(in MW-d/ton U of uranium)

Method	Tube No. 1	Tube No. 7
From flux monitor analysis	7460	7840
Ce^{144}/U	5950	7640
Cs^{137}/U	5670	8210

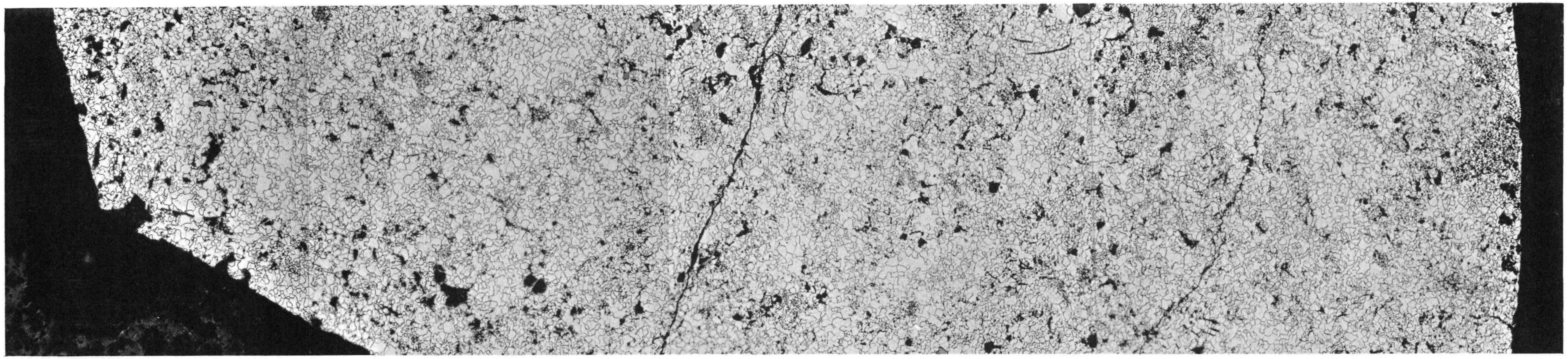

Fig. 1.1 — Microscopic appearance of representative pellet from Tube No. 1 after irradiation, composite, as etched. (100×)

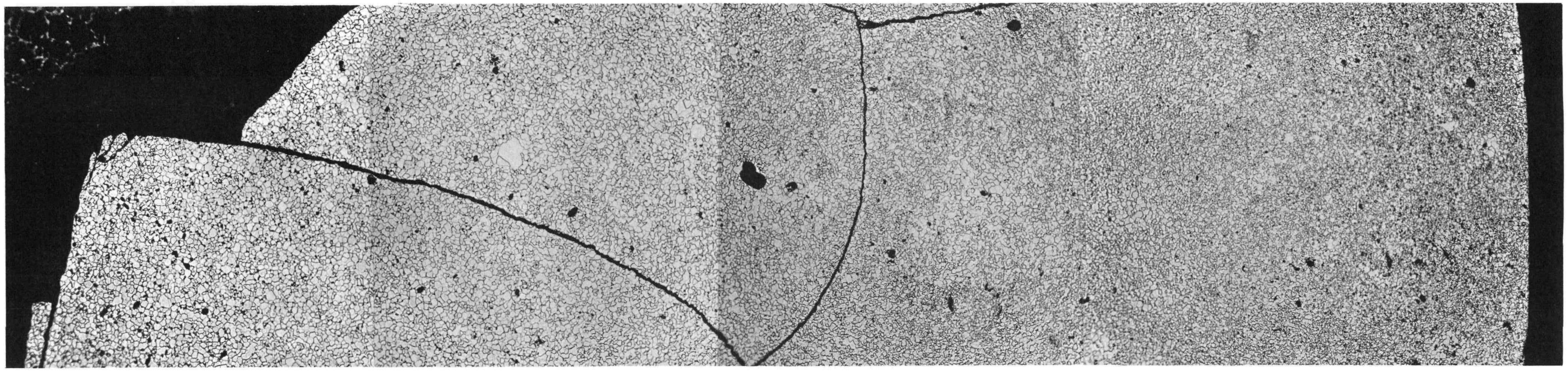

Fig. 1.2 — Microscopic appearance of representative pellet from Tube No. 7 after irradiation, composite, as etched. (100×)

0.2% release observed in Tube No. 7 falls within the range experienced with conventionally fabricated pellets at a similar fuel temperature.

1.2.3 Irradiation of Fuel Rods in VBWR

Work was initiated on the preparation of UO_2 pellets for the fuel rods to be fabricated for the high burnup irradiation program in the Vallecitos Boiling Water Reactor. In what was expected to be a straightforward set of sintering trials to establish shrinkage behavior, low temperature sintering experiments were conducted with the Spencer enriched (5%) oxide received from GE/APED to obtain data for the design of a die to produce the desired pellet diameter.

The maximum pellet density attained with the as-received material was 90% of theoretical at a sintering temperature of 1200°C. In view of the goal of 95% T.D. set for this program, the oxide was subjected to a standard oxidation-reduction cycling treatment to improve its low temperature sinterability. A density increase to only 93% of theoretical was achieved with this activation treatment after sintering at 1300°C for 1 hr in nitrogen, followed by 1 hr in hydrogen.

The effect of activation by multiple oxidation-reduction cycling was investigated next. Two oxide batches, subjected to double and triple cycles, respectively, were formed into compacts at 20, 30, and 40 tsi. These were sintered at 1300°C for 2 hr in nitrogen, followed by 1 hr in hydrogen. The highest density obtained from both batches was only 90% of theoretical with the compacts pressed at 20 tsi. All sintered pellets exhibited unusual surface irregularities in the form of small raised areas resembling blister formation, which were more prominent with the triple-cycled batch.

These results were reminiscent of the "oversintering" phenomenon observed with a depleted Spencer oxide lot during the process development phase of the project. In that case, an optimum sintering temperature (1100 to 1200°C) and soaking time (1 hr) were found to produce satisfactory high density pellets.

In view of this past experience, subsequent sintering trials were conducted under milder furnace conditions. Enriched oxide subjected to single and double oxidation-reduction cycles was sintered next at 1100°C for 1 hr in nitrogen followed by 1 hr in hydrogen. Compacts of each activated batch were formed at several pressures and a highly sinterable Davison oxide was included for experimental control. The results of this run are reported in Table 1.5 under the appropriate sintering conditions.

All pellets exhibited good physical integrity, and the batch activated by one oxidation-reduction cycle, when pressed at 30 tsi, yielded the highest sintered pellet density of 94.1% of theoretical. It is noteworthy that this was the highest density attained with the enriched oxide up to that point. The Davison control pellets, on the other hand, did not shrink to their normal high density of at least 95% of theoretical.

A sintering experiment was then performed at a higher temperature of 1165°C and with a longer soaking time to promote greater densification. Included in the run were activated Spencer oxide batches which had been air-roasted to produce intermediate O/U ratios of 2.31 and 2.33 for the single and double-cycled materials, respectively. The experimental data presented in Table 1.5 indicate that the batches with higher excess oxygen levels yielded somewhat higher densities; however, the use of a higher sintering temperature and longer soaking time did not raise the maximum pellet density above that obtained at the lower temperature in the previous experiment. Moreover, the more severe furnace conditions were apparently responsible for the slight irregularities, in the form of small blisters, observed on the surface of the pellets.

Finally, the enriched oxide was subjected to a modified oxidation-reduction cycling treatment. Deviating from the standard $1\frac{1}{2}$-hr oxidation in air at 500°C, followed by a 1-hr reduction in hydrogen at 510 to 520°C, the as-received oxide was processed as follows:

	Treatment	Time, hr	Temp, °C
Step 1	Oxidation in air	$1\frac{1}{3}$	500
Step 2	Soak in wet nitrogen – 10 cfh (saturated with H_2O at room temp)	1/2	500-520
Step 3	Soak in dry nitrogen – 10 cfh	1	500-520
Step 4	Reduction in dry hydrogen – 10 cfh	1	500-520

The use of wet nitrogen to provide a pyrohydrolytic atmosphere was prompted by earlier work which had demonstrated that it was effective in removing the low temperature sintering inhibitor, fluorine, from ceramic grade UO_2.

The resulting material was air-roasted, formed into compacts at several pressures, and sintered at 1150°C for only 1 hr in nitrogen, followed by 2 hr in hydrogen, to remove all residual excess oxygen.

The results in Table 1.5 show that a combination of oxidation-reduction cycling with an intermediate pyrohydrolysis step in nitrogen markedly improved the sinterability of the oxide. Pellets of at least 95.7% T.D. were produced from compacts formed at all three pressures investigated. Maximum density, 96.3% T.D., was produced from compacts pressed at 40 tsi. All pellets exhibited good physical integrity with an hourglass pattern averaging 2 mils on the diameter. The final O/U ratios determined on two representative specimens were 2.011 and 2.012, respectively.

In view of the satisfactory pellet densities obtained with the modified activation treatment, the remainder of the enriched oxide lot received from GE/APED will be similarly processed and fabricated into pellets.

1.3 CONCLUSIONS

The major conclusion to be drawn from the irradiation testing results to date is that the behavior of stoichiometric oxide pellets prepared by low temperature sintering is not unlike that of conventional hydrogen sintered pellets when irradiated to moderate burnup levels at fuel center temperatures sufficiently high to promote some grain growth.

The finding that a pyrohydrolysis treatment in wet nitrogen improves the sintering characteristics of a ceramic grade oxide beyond the level achieved with multiple oxidation-reduction cycling, suggests that the effect of impurities on the low temperature sintering mechanism may be a fruitful area for further investigation.

1.4 WORK PLAN FOR THE NEXT QUARTER

The irradiation of Capsule OLM-1-3 will be resumed in the MTR or ETR and the fabrication of fuel rods for irradiation in the VBWR will be completed.

Table 1.5 — Summary of Enriched (5%) Spencer Oxide Sintering Data

Sintering Conditions			Oxide Treatment	O/U Ratio	Forming Pressure, tsi	Geometric Density,* g/cm^3	
Temp, °C	Cycle, hr in N_2	Cycle, hr in H_2				Green	Sintered
1100	1	1	Oxidation-reduction, single cycle, air roasted 16 hr at 180°C	2.39	15	5.17	10.11
					20	5.64	10.23
					30	5.94	10.32
			Oxidation-reduction, double cycle, air roasted 16 hr at 160°C	2.39	15	5.06	10.15
					20	5.49	10.22
					30	5.90	10.10
			Davison Lot 3, oxidation-reduction, single cycle, air roasted 16 hr at 160°C	2.38	20	5.90	10.36
1165	2	2	Oxidation-reduction, single cycle, air roasted 16 hr at 140°C	2.31	20	5.63	10.13
					30	5.97	10.01
					40	6.21	10.09
			Oxidation-reduction, double cycle, air roasted 16 hr at 140°C	2.33	20	5.40	10.19
					30	5.93	10.05
					40†	6.16	9.80
			Oxidation-reduction, single cycle, air roasted 16 hr at 180°C	2.39	20	5.83	10.24
					30	5.82	10.30
					40	6.23	9.93
			Oxidation-reduction, double cycle, air roasted 16 hr at 180°C	2.39	20†	5.61	10.06
					30†	5.93	9.91
					40†	6.22	9.81
			Davison Lot 3, oxidation-reduction, single cycle, air roasted 16 hr at 160°C	2.38	20	5.89	10.46
1150	1	2	Modified oxidation-reduction, single cycle with intermediate wet N_2, air roasted 16 hr at 180°C	2.41	20	5.76	10.53
					30	6.14	10.50
					40	6.38	10.56
			Davison Lot 3, oxidation-reduction, single cycle, air roasted 16 hr at 160°C	2.38	20	5.95	10.54

*All values represent the average of two samples; theoretical density = 10.97 g/cm^3.
†Samples contained cracks after sintering.

2. THE DEVELOPMENT OF URANIUM CARBIDES AS A NUCLEAR FUEL FOR LOWERING THE COST OF NUCLEAR POWER (Task II)

H. S. Kalish, F. B. Litton, J. Crane

2.1 INTRODUCTION

Effort during this period was directed toward the preparation of specimens for property studies and the determination of physical properties as a function of composition and means of fabrication. As in the case of irradiation test specimens, this work includes sintered uranium carbide at 4.4 and 4.8 ± 0.1 w/o carbon levels and cast uranium carbide at 4.4, 4.8, and 5.2 ± 0.1 w/o carbon levels.

Irradiation test specimens sent to Battelle Memorial Institute were encapsulated and loaded in the MTR. The pre-irradiation examination of the specimens carried out at Battelle is described in this Quarterly Report.

2.2 PHYSICAL PROPERTY DETERMINATIONS OF URANIUM CARBIDE

The objective of Phase 3 of this program is the determination of physical properties of uranium carbide as a function of compositional and fabricational variables. For all the properties to be determined – thermal conductivity, thermal expansion, and hardness at elevated temperature – sintered carbide specimens at 4.4 and 4.8 ± 0.1 w/o carbon and cast carbide specimens at 4.4, 4.8, and 5.2 ± 0.1 w/o carbon will be tested.

2.2.1 Thermal Expansion

The expansion characteristics of uranium carbide are being determined in the temperature range from room temperature to 1000°C in vacuo, using a Leitz dilatometer. The data are obtained with the aid of a mirror galvanometer which permits tracing a curve by a light source on light sensitive film which is the plot of linear expansion vs temperature. From this plot the coefficient of linear thermal expansion can be determined for any given temperature interval.

The specimens used for this work were obtained by grinding a casting or sintered bar to required dimensions – nominally, 25.4 mm long by 8 mm diameter. Three separate specimens are being run for each of the five combined fabrication and composition variables.

The dilation curve for a 4.37 w/o carbon sintered uranium carbide specimen is shown in Fig. 2.1. A distinct jog in the curve was evident at 660°C, and a lesser but perceptible irregularity in the curve was seen at 770°C. Since the specimen did contain a substantial amount of free uranium, it is believed that these two deviations from the smooth curve correspond to the alpha-to-beta and beta-to-gamma transitions for uranium. The coefficient of linear thermal expansion over the temperature range 25 to 1000°C was 11.8×10^{-6} mm/mm-°C.

The dilation curve for a 4.73 w/o carbon cast uranium carbide specimen is shown in Fig. 2.2. There were no evident breaks in the curve. The coefficient of linear thermal expansion over the temperature range 25 to 1000°C was 10.6×10^{-6} mm/mm-°C.

Duplicate and triplicate samples of 4.4 ± 0.1 w/o carbon sintered and 4.8 ± 0.1 w/o carbon cast material are currently being tested for thermal expansion. Specimens representing the remaining conditions of fabrication and carbon content are being prepared. The final data, upon completion of triplicate runs and evaluation of scatter, will be plotted in terms of coefficient of linear thermal expansion as a function of temperature up to 1000°C.

2.2.2 Thermal Conductivity

Determination of thermal conductivity of uranium carbide as a part of this program is being conducted by Lexington Laboratories, Inc., of Cambridge, Massachusetts, on specimens prepared by United Nuclear Corporation. Determinations

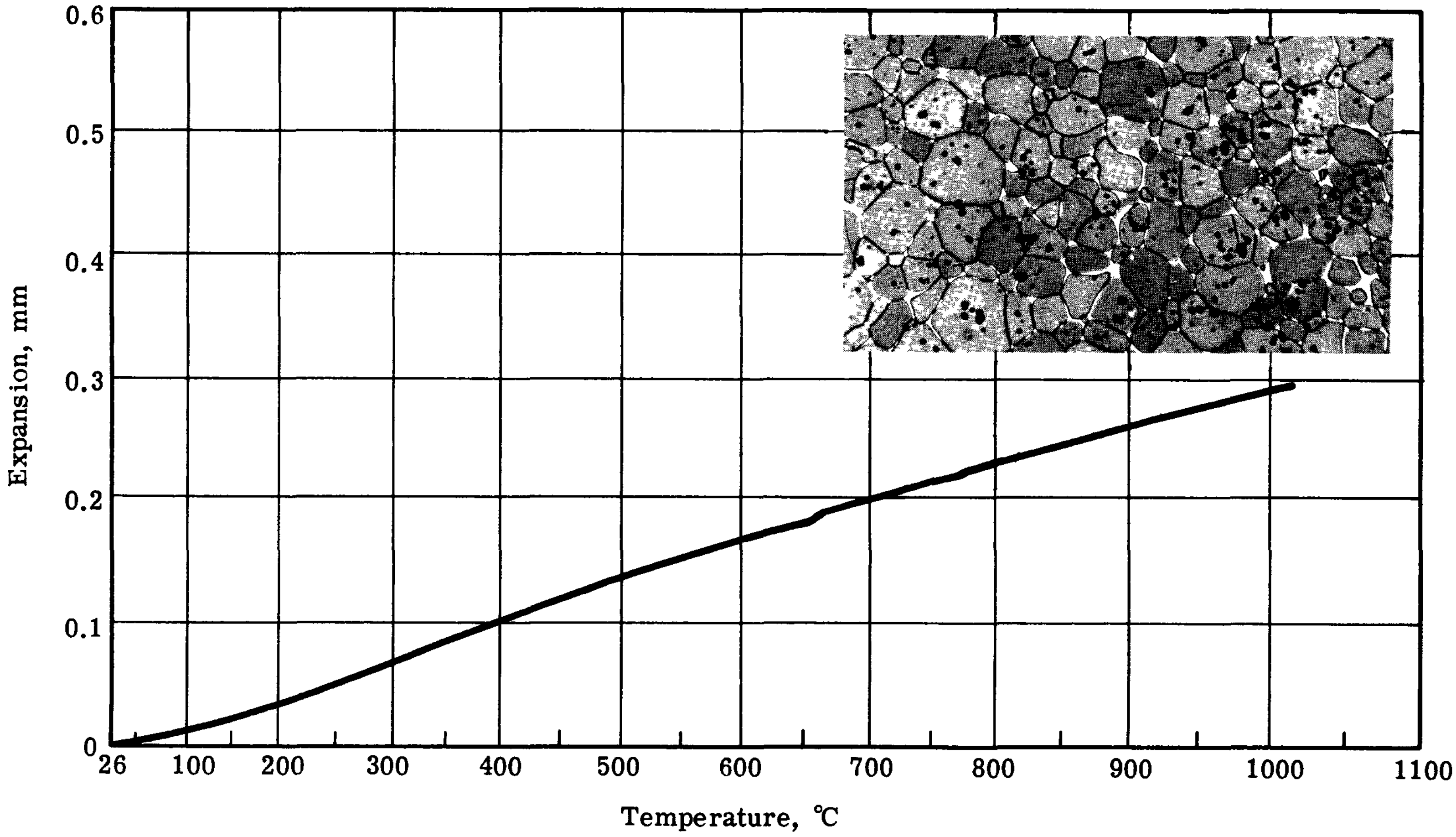

Fig. 2.1 — Plot of expansion vs temperature for a sintered uranium carbide specimen containing 4.37 w/o carbon; density 13.83 g/cm^3 (99% of theoretical density). Inset shows representative microstructure for this specimen at 100×.

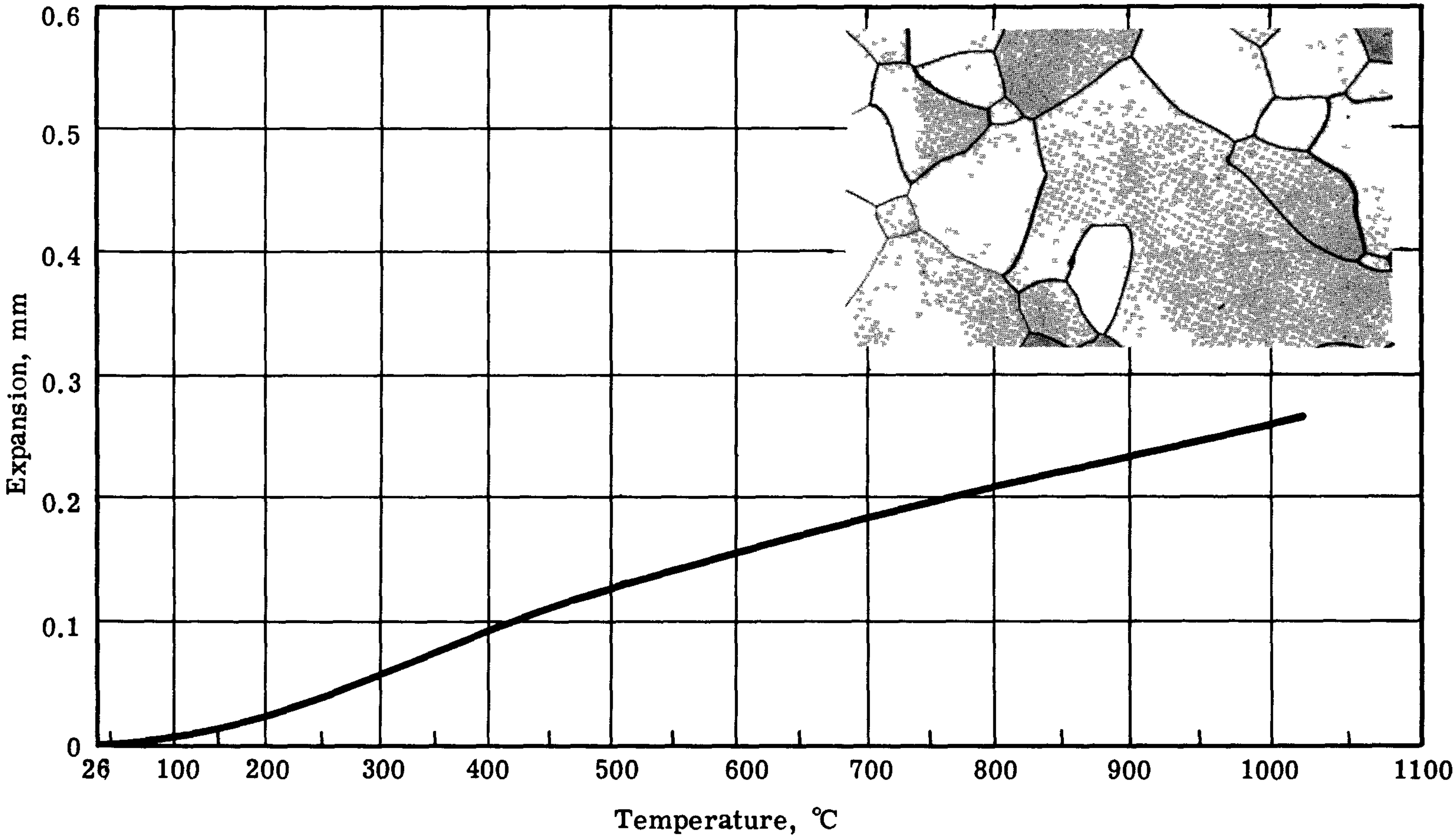

Fig. 2.2 — Plot of expansion vs temperature for a cast uranium carbide specimen containing 4.73 w/o carbon; density 13.42 g/cm^3 (98% of theoretical density). Inset shows representative microstructure for this specimen at 100×.

of the coefficient of thermal conductivity will be made at six temperatures in the range of 200 to 1000°C for each of five uranium carbide specimens.

The apparatus and method being employed by Lexington Laboratories is that described by Francl and Kingery.[2] The method depends on achieving steady state heat flow in one direction through the unknown and through two standards in thermal contact with each end of the unknown. Although the principle of the comparative method is straightforward, lateral heat flow and interfacial thermal resistance constitute two sources of error in the determination. Lateral heat flow is minimized by adjusting the power into seven individually controlled heaters surrounding the standards and unknown until temperatures indicated by thermocouples attached to the specimens are in good agreement with those measured by thermocouples in adjacent locations in the guard heaters. Interfacial resistance to heat flow is reduced by providing good mating surfaces between the unknown and the standards, and through the use of platinum foil at the mating interfaces.

After uniform heat flow through the specimens is established for the required temperature, the temperature drop along each of the specimens is measured. The coefficient of thermal conductivity can then be determined using the equation for uni-directional heat flow:

$$q = \frac{k_1 A_1 \Delta t_1}{\Delta 1_1} = \frac{k_2 A_2 \Delta t_2}{\Delta 1_2} = \frac{k_3 A_3 \Delta t_3}{\Delta 1_3}$$

where q = heat flow

k = coefficient of thermal conductivity

A = mean specimen cross-sectional area

Δt = temperature drop

$\Delta 1$ = thermocouple separation.

The coefficient for the unknown can thereby be determined, using the known coefficient for either of the standards. The use of two standards provides an additional check of the data.

The standards being used for these tests are sintered high-purity slip-cast alumina containing 10% porosity. Tests are being run in vacuo. The uranium carbide specimens are nominally 7/8 in. long by 1/2 in. in diameter. Thus far, three uranium carbide specimens are undergoing thermal conductivity measurements at Lexington Laboratories: sintered uranium carbide containing 4.43 w/o carbon, sintered carbide containing 4.82 w/o carbon, and cast uranium carbide containing 4.71 w/o carbon. Determinations from these specimens are not yet complete. Remaining thermal conductivity samples of cast 4.4 and 5.2 ± 0.1 w/o carbon uranium carbide are being prepared.

2.2.3 Hardness

Room temperature hardness measurements were made using a Knoop indenter and an applied load of 500 grams. Averages of five readings taken on each specimen are shown in Table 2.1. Micro-examination of the specimens in the area of the indentations showed no evidence of cracks associated with the hardness impressions. It was evident, however, that porosity in the sintered material made evaluation of hardness as a function of carbon content difficult since porosity tended to increase with increasing carbon content. The cast material increased slightly in hardness from substoichiometric uranium carbide to UC, and then decreased substantially going from UC to UC + UC_2.

2.3 IRRADIATION PROGRAM

(Work performed at Battelle Memorial Institute, under the supervision of
J. E. Gates and R. F. Dickerson)

The objective of the irradiation program is to investigate the effect of variables in the composition and fabrication on the stability of uranium carbide at a fuel surface temperature of 750°C, fission heat rate of 1.5×10^6 Btu/hr-ft^2, and a fuel burnup of 15,000 and 30,000 MW-d/ton U.

The program consists of the irradiation of five uranium carbide specimens in each of three instrumented capsules in the MTR. The uranium carbide specimens were fabricated by arc melting and powder metallurgical techniques from 12% enriched uranium. The carbon content was varied from about 4.4 to 5.2 w/o.

Table 2.1 — Hardness of Sintered and Cast Uranium Carbide

Specimen No.	Fabrication Method	Carbon Content, w/o	Knoop Hardness Number, kg/mm^2
E 6-1	Sintered	4.31	680
E 5-1	Sintered	4.44	585
E 1-1, 2, 3	Sintered	4.79	471
E 1-4, 5, 6	Sintered	4.86	598
19D-728-4SM38-2	Cast	4.43	705
19D-709-4SM18-1	Cast	4.71	743
19D-715-4SM24-1	Cast	5.19	573

A description of the irradiation test specimens was included in the Quarterly Report[3] issued by United Nuclear Corporation on February 1, 1962. However, in order that a detailed post-irradiation study of microstructural and physical property changes might be made, a complete pre-irradiation examination of both the fuel and assembled specimens was performed by Battelle Memorial Institute.

Specimen encapsulation, capsule fabrication, irradiation surveillance, and post-irradiation examination will be carried out by Battelle.

The weight, density, and physical dimensions of each fuel specimen were measured. After separately encapsulating the fuel specimen in niobium-1% zirconium alloy cans, the weight and physical dimensions of each fuel container were recorded. The data are shown in Table 2.2.

Since little was known of the swelling characteristics of uranium carbide at the irradiation test temperatures, the cladding was designed to permit about 10% swelling without creating excessive stresses in the niobium-1% zirconium alloy encapsulating material. In order to satisfy these conditions and maintain the desired thermal conditions during irradiation, a sodium bond was incorporated between the fuel and cladding. The capsule design is shown in the previous Quarterly Report.[3] A sketch of the encapsulated specimen for capsules UNC-1-1 and UNC-1-2 is shown in Fig. 2.3. Each specimen can was machined to 0.415 in. ID

Table 2.2 — Pre-Irradiation Physical Properties of Specimens

Specimen		Physical Measurements of Fuel				Physical Measurements of Fuel Container		
UNC No.	BMI No.	Density, g/cm^3	Diameter, in.	Length, in.	Weight, g	Diameter, in.	Length, in.	Weight, g
		Cast Specimens – 19D-728-4SM38 – 4.45 w/o Carbon						
1A	A-1	13.79	0.3758	0.7389	18.4774	0.475	1.50	33.61
1B	A-2	13.80	0.3758	0.7481	18.6945	0.475	1.50	33.82
3A	A-3	13.79	0.3747	0.7403	18.3897	0.435	1.50	30.98
		Cast Specimens 19D-709-4SM18 – 4.72 w/o Carbon						
1C	B-1	13.62	0.3751	0.7151	17.5422	0.475	1.50	32.83
2A	B-2	13.60	0.3748	0.7341	17.9507	0.475	1.50	33.08
2B	B-3	13.53	0.3757	0.7406	18.1368	0.435	1.50	31.17
		Sintered Specimens – 4.86 w/o Carbon						
E1-6	C-1	12.73	0.3755	0.7305	16.7971	0.475	1.50	31.93
E1-4B	C-2	12.74	0.3750	0.7195	16.4656	0.475	1.50	31.60
E1-4A	C-3	12.63	0.3750	0.7214	16.4003	0.435	1.50	28.93
		Cast Specimens 19D-715-4SM24 – 5.19 w/o Carbon						
2B	D-1	13.33	0.3758	0.7246	17.4906	0.475	1.50	32.62
3A	D-2	13.34	0.3745	0.7051	16.9182	0.475	1.50	32.05
3B	D-3	13.34	0.3742	0.7327	17.5684	0.435	1.50	30.14
		Sintered Specimens – 4.31 w/o Carbon						
E6-2B	E-1	13.70	0.3759	0.7389	18.3488	0.475	1.50	33.48
E6-3	E-2	13.70	0.3756	0.7371	18.2352	0.475	1.50	33.37
E6-4A	E-3	13.71	0.3757	0.7475	18.5190	0.435	1.50	31.03

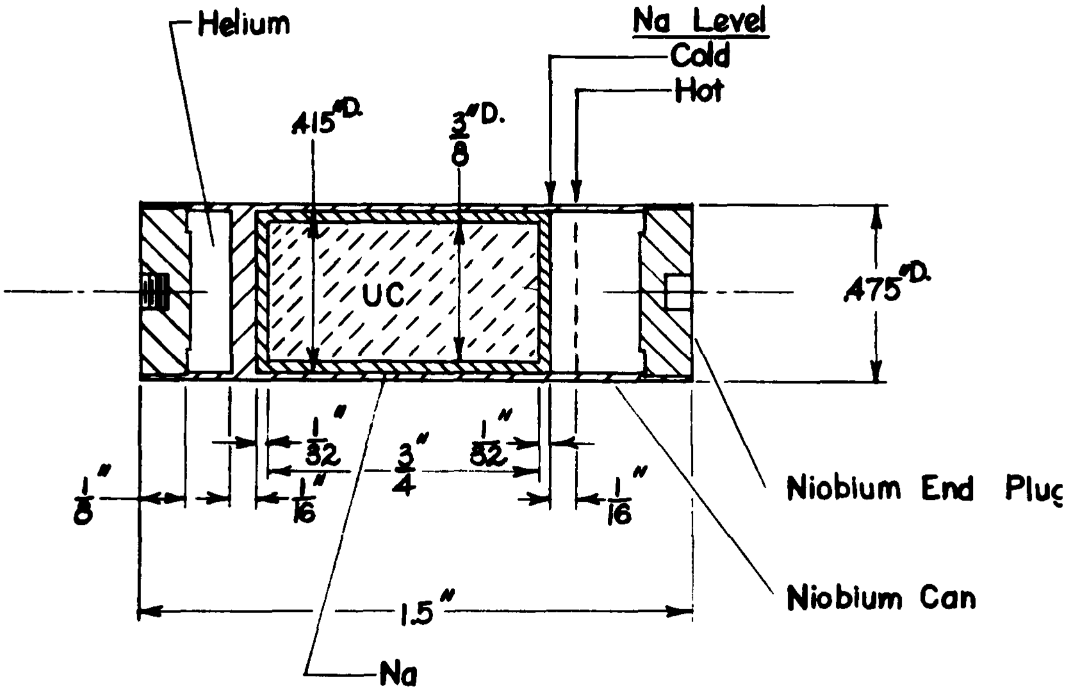

Fig. 2.3 — Design of fuel container using a sodium bond between fuel and cladding material

and 0.475 in. OD. A void space filled with helium was provided at one end of the specimen to reduce heat losses through the ends of the can. Expansion space was provided in the fuel container above the sodium to accommodate fission gases which might escape from the fuel during irradiation. The fuel in each container was centered in the sodium by a 0.015 in. diameter molybdenum wire spiraled around the fuel.

During the period of capsule design and fabrication, the compatibility of uranium carbide and sodium at the irradiation test temperatures was investigated. Although earlier studies at Battelle had suggested that these two materials were compatible at about 800°C, it was decided that further studies should be performed. Prototype specimens of uranium-4.4 w/o carbon alloy and uranium-5.2 w/o carbon alloy were immersed in sodium in prototype niobium-1% zirconium alloy fuel containers. This study was carried out at 1500°F (816°C) for 260 hr in a vacuum furnace. The physical appearance of the specimens after testing did not show evidence of reaction. However, the microstructure of the uranium-4.4 w/o carbon alloy revealed a slight reaction of the metallic uranium in grain boundaries extending to the surface. This reaction was observed to a depth of about 0.001 in. The use of polarized light during the metallographic examination indicated that the reaction product was UO_2, suggesting an oxidation of the metallic uranium. The microstructure of the uranium-5.2 w/o carbon alloy specimen showed surface chipping and microcracks, indicative of slight oxidation of the uranium carbide. Since the sodium used in these studies had been gettered with zirconium chips prior to the test to reduce its oxygen content, it is suspected that the oxygen may have emanated from surface contamination of the specimens. The cladding used for the uranium-5.2 w/o carbon alloy showed a thin, 0.0001 in.,niobium carbide layer on the inner surface, suggesting that the sodium acted as a carbon transfer medium. The degree and type of attack on the fuel, however, was interpreted to be so slight that it would be of no consequence during the period of the irradiation study.

Although the results of the compatibility study using sodium were favorable, the design of the fuel container was altered in the third capsule (UNC-1-3) to eliminate the use of the sodium bond between fuel and cladding, so that a direct comparison could be made in the irradiation study between a sodium bond and a helium filled capsule. The containers were redesigned, as shown in Fig. 2.4, to provide a 0.001 in. annulus between the fuel and cladding which would result in an approximate 0.001 in. fit at 1500°F (816°C).

The encapsulation of all fuel specimens was performed in a helium atmosphere dry box. The cans containing sodium as a bonding material, which were inserted in capsules UNC-1-1 and UNC-1-2, were prepared in the following manner. A predetermined quantity of freshly gettered sodium, necessary to completely immerse a fuel specimen, was placed in the can for each specimen. Each fuel specimen with the spiraled molybdenum wire intact was then placed on top of the sodium and the end caps heliarc welded in place. The specimens were then heated to a temperature just above the melting point of sodium to permit the fuel to submerge in the molten sodium. The specimens were then taken from the dry box and immediately checked with a helium leak detector to determine the integrity of the welds. Radiographs of each specimen were taken to establish the position of the fuel and determine whether the fuel was totally covered with sodium. The specimens for capsule UNC-1-3, which did not contain sodium, were assembled by merely sliding the spacers and fuel into the can and heliarc welding the end caps in place. These specimens were also taken from the dry box and checked for helium leakage.

Photographs were taken of the fuel and welded fuel container. The series of typical views are shown in Figs. 2.5 through 2.19.

The microstructural studies of both the arc-cast and powder metallurgically produced uranium-4.4 ± 0.1 w/o carbon alloy specimens showed metallic uranium in the grain boundaries. The cast specimens were composed basically of larger grains and contained less porosity than the sintered specimens. The photomicrographs of these specimens are shown in Figs. 2.20 and 2.21. The arc-cast uranium-4.8 ±0.1 w/o carbon specimens exhibited much larger grains and were considerably more dense than those produced by powder metallurgical techniques. The microstructure of these specimens is shown in Figs. 2.22 and 2.23. A second phase, UC_2, was detected near the surface of the sintered uranium-4.8 ± 0.1 w/o carbon alloy fuel specimens indicating some inhomogeneity. The specimens of uranium-5.2 ± 0.1 w/o carbon alloy produced by arc casting showed, in general, a structure of uranium dicarbide platelets uniformly distributed throughout uranium monocarbide grains. This microstructure is shown in Fig. 2.24.

The three capsules were fabricated and shipped to the MTR site. Capsule UNC-1-2 was inserted in the MTR for Cycle 170 on February 19, 1962. However, the reactor loading was changed prior to Cycle 170, resulting in an unanticipated increase in the thermal neutron flux from 1.8×10^{14} to 2.4×10^{14} nv. Consequently, the capsule was raised during the cycle so that only the bottom specimen in the capsule operated at the desired temperature. In raising capsule UNC-1-2, the lead tube was jammed against a clamp and fractured. Helium pressure was maintained throughout the cycle in order to prevent water leakage into the capsule. At the end of the cycle approximately 6 ft of the lead tube was removed and the fractured section replaced by another tube which was mechanically sealed to the capsule.

The reactor flux pattern was studied during Cycle 170 and an area having the specified flux selected for relocating capsule UNC-1-2 and inserting capsule UNC-1-1. Capsules UNC-1-1 and UNC-1-2 were inserted on March 12, 1962, for Cycle 171. These capsules operated in the new location at the desired temperature (about 1500°F surface temperature) as indicated by temperature readings of 1190 ± 100°F at the control thermocouples. The third capsule, capsule UNC-1-3, was inserted in the MTR for Cycle 172 on April 2, 1962. This capsule also operated at the desired temperature at the time of this report.

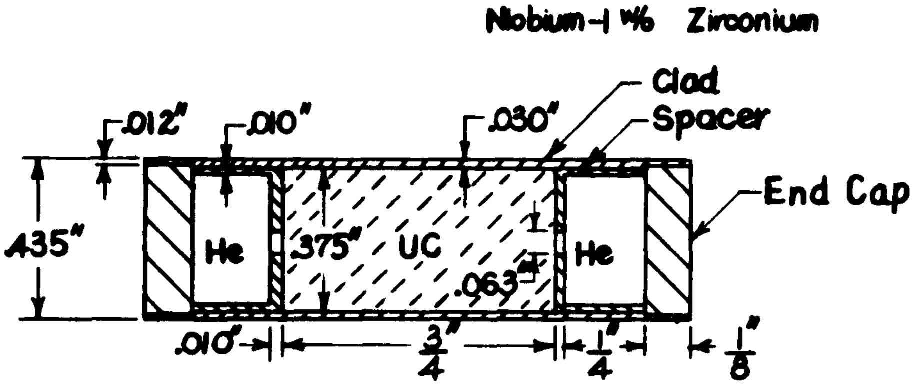

Fig. 2.4 — Design of fuel container using an interference fit between fuel and cladding material

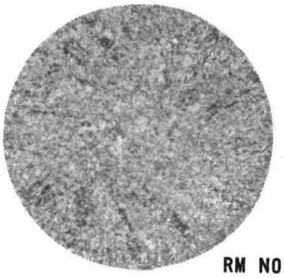

Fig. 2.5 — Transverse view of unclad cast 4.45 w/o carbon UC fuel specimen. (4×)

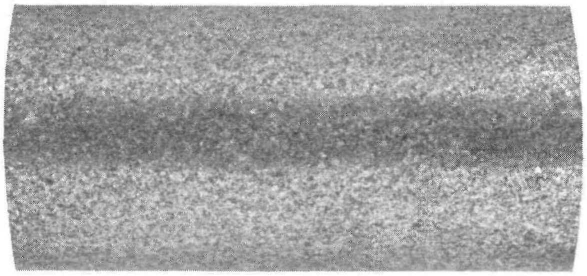

Fig. 2.6 — Longitudinal view of unclad cast 4.45 w/o carbon UC fuel specimen. (4×)

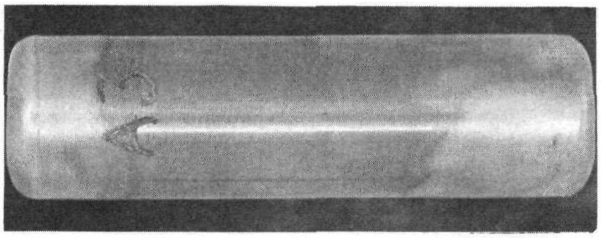

Fig. 2.7 — Longitudinal view of welded fuel container. (2×)

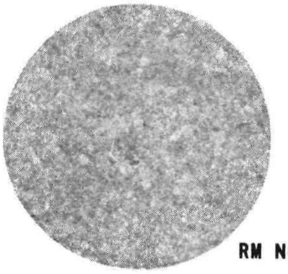

Fig. 2.8 — Transverse view of unclad cast 4.72 w/o carbon UC fuel specimen. (4×)

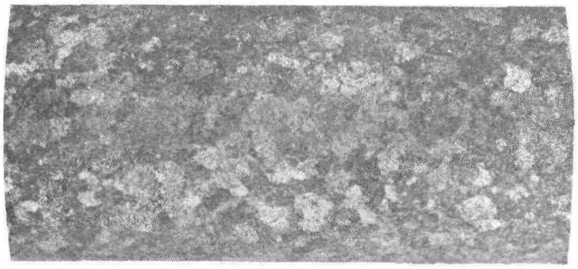

Fig. 2.9 — Longitudinal view of unclad cast 4.72 w/o carbon UC fuel specimen. (4×)

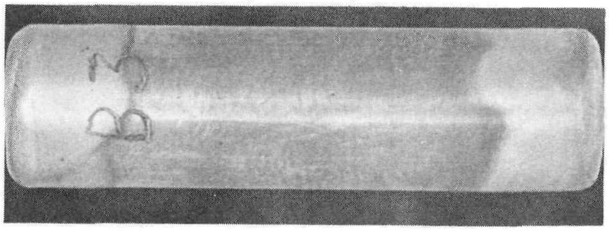

Fig. 2.10 — Longitudinal view of welded fuel container. (2×)

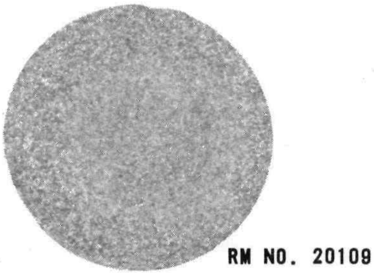

Fig. 2.11 — Transverse view of unclad sintered 4.86 w/o carbon fuel specimen. (4×)

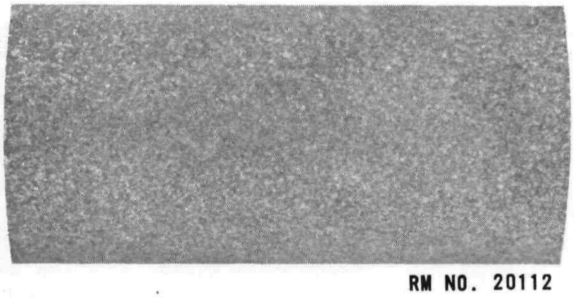

Fig. 2.12 — Longitudinal view of unclad sintered 4.86 w/o carbon fuel specimen. (4×)

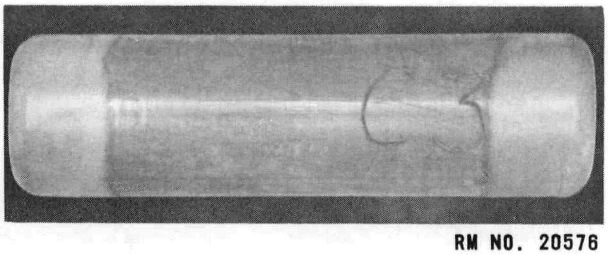

Fig. 2.13 — Longitudinal view of welded fuel container. (2×)

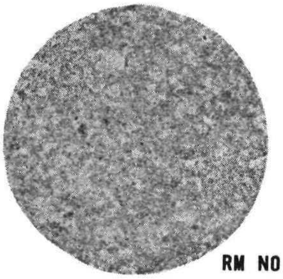

Fig. 2.14 — Transverse view of unclad cast 5.19 w/o carbon fuel specimen. (4×)

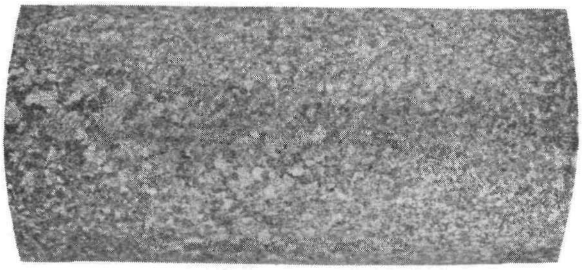

Fig. 2.15 — Longitudinal view of unclad cast 5.19 w/o carbon fuel specimen. (4×)

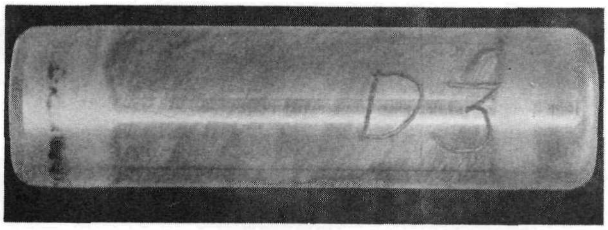

Fig. 2.16 — Longitudinal view of welded fuel container. (2×)

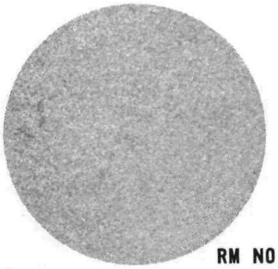
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Fig. 2.17 — Transverse view of unclad sintered 4.31 w/o carbon fuel specimen. (4×)

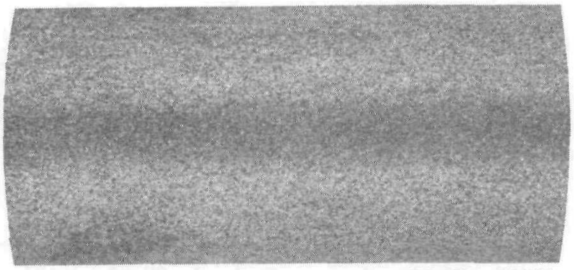
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Fig. 2.18 — Longitudinal view of unclad sintered 4.31 w/o carbon fuel specimen. (4×)

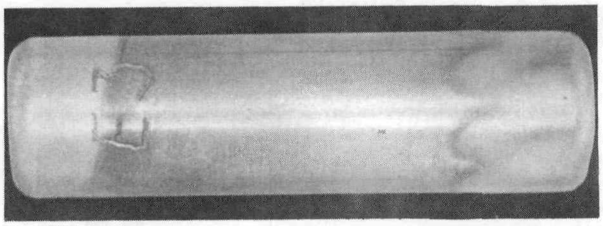
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Fig. 2.19 — Longitudinal view of welded fuel container. (2×)

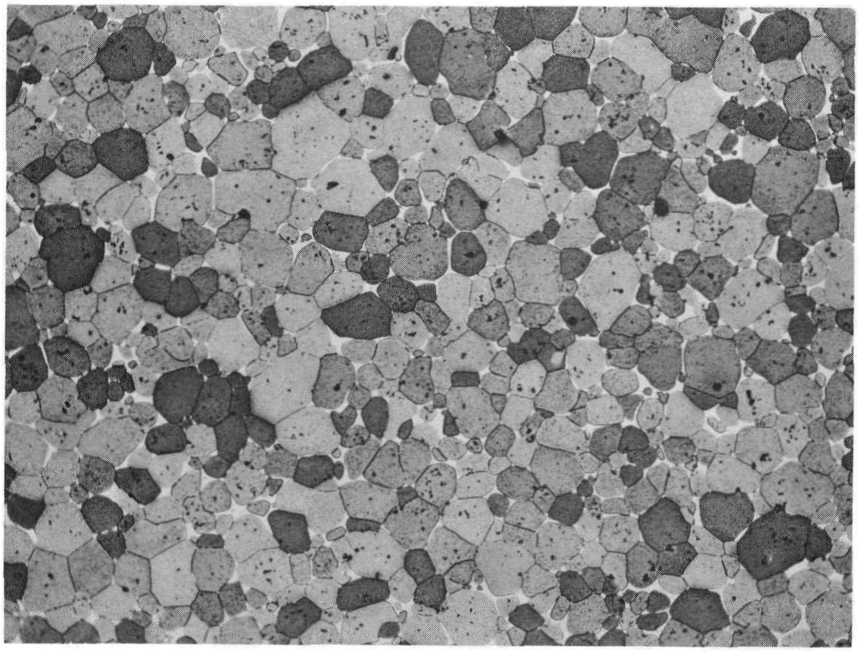

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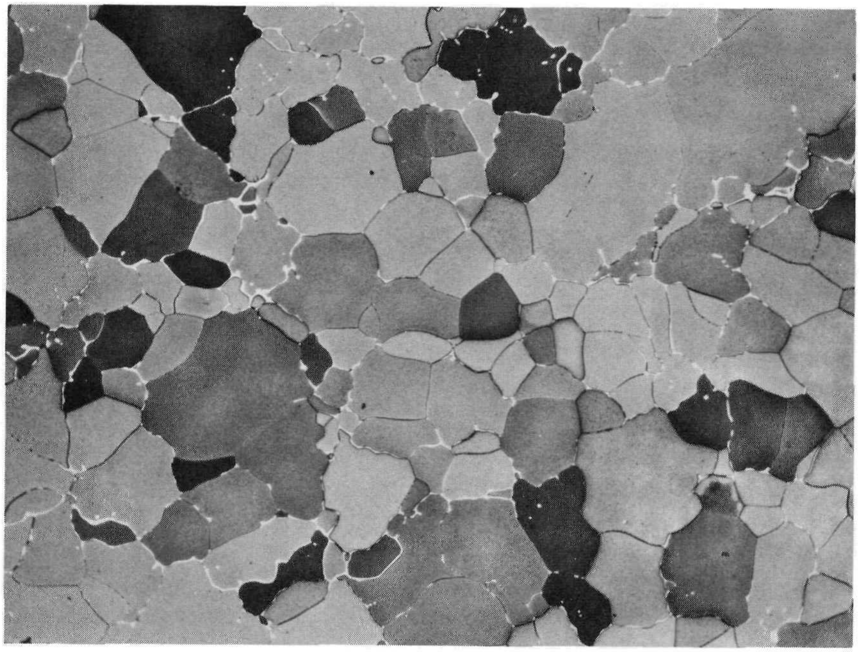
Fig. 2.20 — Typical microstructure of sintered uranium-4.31 w/o carbon fuel alloy. (100×)



RM NO. 20268

Fig. 2.21 — Typical microstructure of arc-cast uranium-4.45 w/o carbon fuel alloy. (100×)

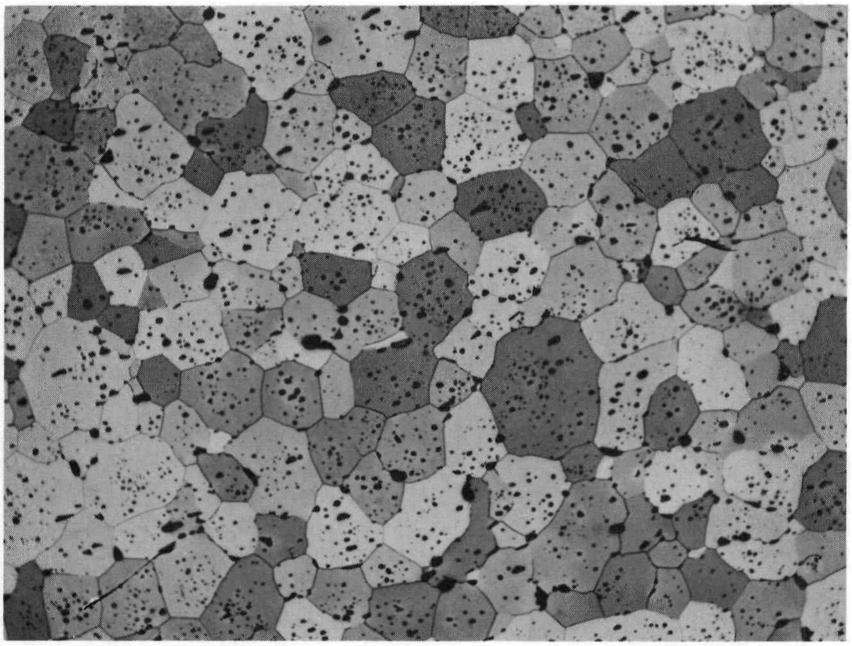


Fig. 2.22 — Typical microstructure of sintered uranium-4.86 w/o carbon fuel alloy. (100×)

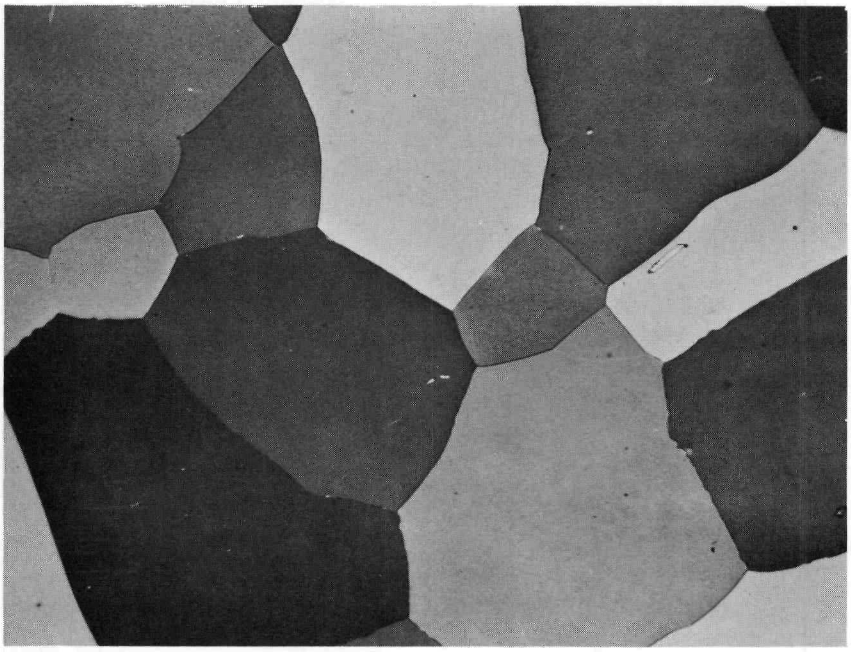


Fig. 2.23 — Typical microstructure of arc-cast uranium-4.72 w/o carbon fuel alloy. (100×)

Fig. 2.24 — Typical structure of arc-cast uranium-5.19 w/o carbon fuel alloy. (100×)

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3. United Nuclear Corporation Fuel Cycle Development Program, Quarterly Progress Report, October 1 to December 31, 1961, UNC-3001 (Feb. 1, 1962).

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