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CARBIDE FUEL DEVELOPMENT

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FOREWORD

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

The report covers progress from April 1, 1962 to June 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).

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1. INTRODUCTION

Fuel made of a combination of UC and PuC has a potential of reducing the fuel cycle cost of existing fast breeder reactors. The fuel cycle cost reduction is anticipated because of increased burnup and increased power generation capability of 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 Carbide Fuel Development Program is concerned with the technology of the entire UC-PuC fuel cycle. The major goal of the program is 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. 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 13.1 g/cm^3) UC and UC-PuC pellets have been made. Fabrication work has been stopped on this program and is being 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 initiated for UC-PuC. The UC-PuC studies will 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. This is complete for UC, and experiments for UC-PuC are under construction.
2. Make a post-irradiation examination. This is completed for UC.

2. SUMMARY

All of the UC-PuC powder required for the natural uranium irradiation specimens, and some of the powder required for the enriched uranium irradiation specimens was synthesized.

All of the natural uranium UC-PuC pellets required were fabricated by pressing and sintering, and are being ground to size. The average density of the $(U_{0.8}Pu_{0.2})C_{0.95}$ pellets was 12.54 g/cm^3 (92.4% of theoretical) and of the $(U_{0.8}Pu_{0.2})C_{0.95}$ plus 0.1 w/o Ni pellets was 13.15 g/cm^3 (96.8% of theoretical).

Cored pellets were fabricated by pressing and sintering, and the holes are being reamed to size to provide a place for an accurately located central thermocouple.

The initial clad irradiation specimens were assembled, and the assembly of the first irradiation capsule was started. The test reactor choice has been narrowed to ETR and GETR. The GETR is recommended for at least four of the six capsules.

3. FABRICATION OF THE UC-PuC IRRADIATION TESTS

3.1 FUEL SYNTHESIS

All of the powder required for the natural uranium irradiation specimens was synthesized. Reaction of the 760-gram batch of PuO_2-UO_2-C yielded approximately 580 g of an essentially single-phase $(U_{0.8}Pu_{0.2})C_{0.95}$ solid solution. The synthesis was carried out by heating 50-g lots of the reaction mixture at 1625°C with a hold time of about $5\frac{1}{2}$ hr. The reaction end-point was determined by monitoring the exhaust gas, drawn from the heated furnace, for CO.

Chemical and x-ray analyses for the individual lots are shown in Table 1. Theoretical carbon for the $(U_{0.8}Pu_{0.2})C_{0.95}$ solid solution is 4.55%. The value by chemical analysis was 4.65% (shown in Table 2).

The addition of nickel, used as a sintering aid, to the UC-PuC materials appears to inhibit complete reaction of residual oxides and carbon. Therefore, the composites of lots 1 through 14 were divided into two equal parts, cold-compacted, and reheated to 1625°C for an additional 2 hr to react further any residual oxide. The product was pulverized, and ball-milled for 24 hr in a rubber-lined mill using stainless steel balls. Chemical analysis of the products for carbon and after additional heating are shown in Table 2.

Lots 15 and 16 were synthesized subsequently, crushed in the Spex mixer, ball-milled for 22 hr and blended with lots 1 through 14 by ball milling an additional 2 hr.

Table 1 — Results of Synthesis Experiments to Produce $(U_{0.8}Pu_{0.2})C_{0.95}$ Powder (1625°C with Hold Time of $5\frac{1}{2}$ hr for Expts. 1 through 14 and 6 hr for Expts. 15 and 16)

Experiment No.	Batch Size, g	Total Carbon, %	Results of X-ray Analysis
1*	50	4.70	Single phase (U,Pu)C a = 4.965 ± 0.001 Å
2	10	4.58	Major (U,Pu)C. Faint $(U,Pu)_{02}$ a = 4.9645 ± 0.0002 Å
3	50	4.63	Single phase (U,Pu)C a = 4.964 ± 0.001 Å
4	50	4.62	Single phase (U,Pu)C a = 4.964 ± 0.001 Å
5	50	4.65	Single phase (U,Pu)C a = 4.964 ± 0.001 Å
6	50	4.57	Major (U,Pu)C. Faint $(U,Pu)O_2$ a = 4.95 ± 0.01 Å
7	50	4.66	Single phase (U,Pu)C a = 4965 ± 0.001 Å
8	50	4.67	Single phase (U,Pu)C a = 4.966 ± 0.001 Å
9	50	4.61	Single phase (U,Pu)C a = 4.9641 ± 0.0002 Å
10	50	4.56	Single phase (U,Pu)C a = 4.964 ± 0.001 Å
11	50	4.64	Single phase (U,Pu)C a = 4.964 ± 0.001 Å
12	50	4.64	Single phase (U,Pu)C a = 4.964 ± 0.001 Å
13	50	4.66	Single phase (U,Pu)C a = 4.9642 ± 0.0002 Å
14	50	4.59	Single phase (U,Pu)C a = 4.964 ± 0.001 Å
15	55	4.63	Single phase (U,Pu)C a = 4.965 ± 0.001 Å
16	55	4.66	Single phase (U,Pu)C a = 4.964 ± 0.001 Å

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

Table 2 — Results of Compositing and Reheating the Lots of (U,Pu)C Powder Shown in Table 1 (Reheat to 1625°C with Hold Time of 2 hr)

Batch No.	Material	Total Carbon, %	Results of X-ray Analysis
1	A composite of Batches 1 through 14 (Table 1) prior to additional heating at 1625°C. Material crushed to about −325 mesh	4.65	Not obtained
2	A composite of Batches 1 through 14 (Table 1) after reheating to 1625°C for 2 hr and ball-milling	4.66	Single-phase (U,Pu)C
3	Batches 15 and 16 (Table 1) were blended in a ball mill for 2 hr with the above, subsequent to their reheating		

Work began on the synthesis of the powder required for the enriched uranium irradiation specimens. The batch materials of 24% enriched UO_2, PuO_2 and carbon were blended by ball-milling for 24 hr in a rubber-lined mill, using stainless steel balls. Chemical analysis for total carbon gave 11.62%. A 50-g batch of the reaction mix was pelletized and heated to 1625°C with a hold time of $5^1/_2$ hr. The reaction end-point was monitored by determining the CO present in the exhaust gas drawn from the heated furnace. Chemical analysis for total carbon on this product gave 4.62%. X-ray analysis showed single-phase (U,Pu)C with a unit cell size, $a_0 = 4.965 \pm 0.001$Å. The synthesis of the remainder of the 200 g of enriched powder required, will be completed next month.

3.2 PELLET FABRICATION

The cold pressing, sintering, and some of the grinding of the natural uranium UC-PuC pellets were completed. The pellets were pressed in the usual manner (30,000 psi, 1/4 w/o Carbowax). The cold-pressed pellets were preheated to about 700°C in an open tantalum crucible to volatilize the binder; the final sintering was completed in a closed tantalum crucible.

About 185 solid pellets were fabricated having the composition $(U_{0.8}Pu_{0.2})C_{0.95}$ plus 0.12 w/o nickel. The pellets were divided into groups of 65, 60, and 60. These were sintered, in three separate runs, at 1550°C with a hold time of 1 hr. Results are shown in Table 3. The first group of 65 solid sintered pellets was surface ground to 0.191 ± 0.0005 in. in diameter using 400 mesh silicon carbide suspended in Convoil-20 (diffusion pump oil). A change to a higher vapor pressure, more easily-removable mineral oil has been made since. A portion of the sintered pellets from the other groups is presently being ground to size.

A sintering experiment was conducted to produce thirty $(U_{0.8}Pu_{0.2})C_{0.95}$ plus 0.12 w/o nickel pellets with a 0.060 in. core. The sintering temperature was 1550°C and the hold time was 1/2 hr. The results are shown in Table 4.

Some of the 30 cored pellets, shown in Table 4, were ground to 0.191 ± 0.0005 in. in diameter.

Table 3 — Results of Sintering $(U_{0.8}Pu_{0.2})C_{0.95}$ + 0.12 w/o Ni at 1550°C for 1 hr

Experiment No.	No. of Pellets Sintered	Density Range, g/cm^3	Avg. Density g/cm^3	Total Carbon,* %		X-Ray Analysis
				Carborundum	UNC	
1	65	12.96-13.14	13.03	4.71	4.69 4.66	Single phase (Pu, U)C $a_0 = 4.95 \pm 0.01$ Å
2	60	13.04-13.17	13.10	4.63		Major (Pu, U)C; Faint $(Pu,U)_2C_3$
3	60	13.12-13.16	13.14	4.62		Major (Pu, U)C; Faint $(Pu,U)_2C_3$ $a_0 = 4.964 \pm 0.002$ Å

*Additional chemical analyses of Exp. No. 1:
19.04% plutonium (by counting)
170 and 160 ppm nitrogen
in process $\left\{\begin{matrix}\text{uranium}\\ \text{plutonium}\end{matrix}\right\}$ (by titration)

Table 4 — Result of Sintering
$(U_{0.8}Pu_{0.2})C_{0.95}$ + 0.12 w/o Nickel
Cored Pellets at 1550°C for 1/2 hr

Pellet Density, g/cm^3	Total Carbon, %	X-Ray Analysis
13.11-13.20 (Avg.-13.15)	4.66	Single Phase (U,Pu)C a_0 = 4.964 ± 0.001 Å

About 130 pellets were fabricated having the composition $(U_{0.8}Pu_{0.2})C_{0.95}$. The pellets were divided into two groups of 65. These were sintered, in separate runs, at 1950°C with a hold time of 1 hr. Pellet densities for the groups, determined prior to surface grinding, were about 12.60 g/cm^3. Results of chemical analyses for total carbon gave 4.50%. X-ray analyses showed single-phase (U,Pu)C with a unit cell size, a_0 = 4.963 ± 0.001Å. An attempt to surface grind a few of the sintered pellets was unsuccessful. Removal of about 0.005 in. from the pellet diameter revealed a high porosity area which covered approximately one-third of the periphery of the specimen. Metallographic examination confirmed this finding. No further attempts to surface grind were made.

Two sintering experiments were conducted with powder, from the combined batches of Table 2, to determine whether excessive temperature may have caused vaporization and porosity in the above pellets. To re-establish the optimum sintering temperature, straight $(U_{0.8}Pu_{0.2})C_{0.95}$ pellets were heated to (1) 1925°C, and (2) 1950°C with hold times of 1 hr, the sintered pellets were surface ground, and densities determined. Examination of the ground pellets from the 1950°C run revealed several small areas on the periphery which appeared to be somewhat porous. However, the pellets appeared to be satisfactory. The pellets sintered at 1925°C ground to a smooth continuous surface with no apparent imperfections. Results are shown in Table 5. The results of these experiments indicate that the optimum sintering temperature for straight $(U_{0.8}Pu_{0.2})C_{0.95}$ pellets is probably 1925°C.

Table 5 — Results of Sintering $(U_{0.8}Pu_{0.2})C_{0.95}$ Pellets With No Sintering Aid

Experiment No.	Sintering Temp and Hold Time	Pellet Density and Range	Total Carbon, %
1	1925°C-1 hr	(12.68-12.78) Avg. -12.73	4.56
2	1950°C-1hr	(12.71-12.83) Avg.-12.77	4.62

Subsequently, 110 pellets having the composition $(U_{0.8}Pu_{0.2})C_{0.95}$ were fabricated from the same powder. Of these pellets, 85 were pressed solid and 25 were pressed with a core. The pellets were sintered at 1925°C with a hold time of 1 hr. Results are shown in Table 6.

Table 6 — Result of Sintering $(U_{0.8}Pu_{0.2})C_{0.95}$ Pellets at 1925°C for 1 hr

Pellet Density, g/cm^3	Total Carbon, %	Plutonium, %	X-Ray Analysis
12.50-12.63* (Avg.-12.54)	4.62	19.05	Single-phase (U,Pu)C $a_0 = 4.964 \pm 0.001$ Å

*Solid and cored pellets.

Some of the solid and cored pellets were surface ground to 0.191 – 0.005 in. in diameter. The core size was about 0.060 in. in diameter.

An effort was made to determine the true temperature used in the synthesis and sintering studies performed in the graphite resistance heated furnace. For this study, several attempts were made to melt chemically pure platinum in the resistance furnace. For the lowest temperature experiment the metal was placed

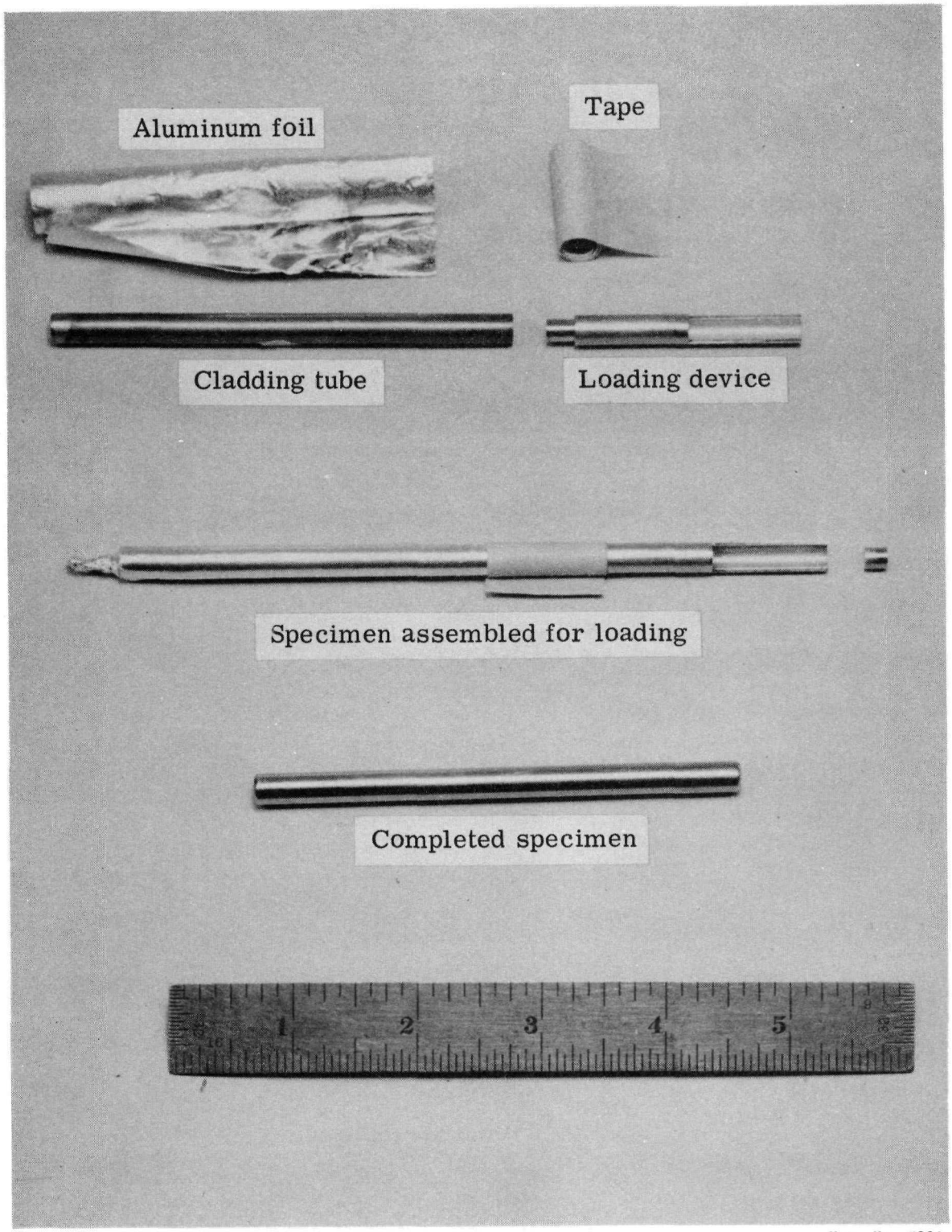
on an alumina plate, inside a closed tantalum crucible. The furnace was heated to 1650°C with a 1/2-hr hold time. This temperature was measured on the heating element with an optical pyrometer, uncorrected for glass absorption or emissivity. The melting point of pure platinum is reported[1] to be 1769°C; however, in this experiment the platimun melted at an optical reading of 1650°C. According to Hansen,[2] the freezing point of platinum is reduced to 1734°C when melted in a graphite crucible and the solid solubility of carbon is about 0.25 w/o. Assuming that the platinum has dissolved carbon, and using the measured correction for the absorption of the laminated glass window, an additional correction factor of at least 40°C is required at this temperature. Experiments on calibrating the temperature in this furnace will continue.

3.3 FUEL SPECIMEN ASSEMBLY

Three irradiation specimens without central thermocouples were assembled. The method of loading is shown pictorially in Fig. 1. The loading device fits tightly into a counterbore 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 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. To date, the contamination has been very slight (200 counts/min.). The contamination is removed easily and completely by detergent. A radiograph of a typical specimen is shown in Fig. 2.

Central holes for thermocouples were drilled in several UC-PuC-0.1 Ni pellets, using a new centering collet. The holes were centered within the required 0.003 in. However, the pellets had a laminar-type fracture. Cored pellets were then fabricated, as described in Section 3.2, to ease the drilling problem.



Neg. No. 4092

Fig. 1 — Irradiation specimen loading method

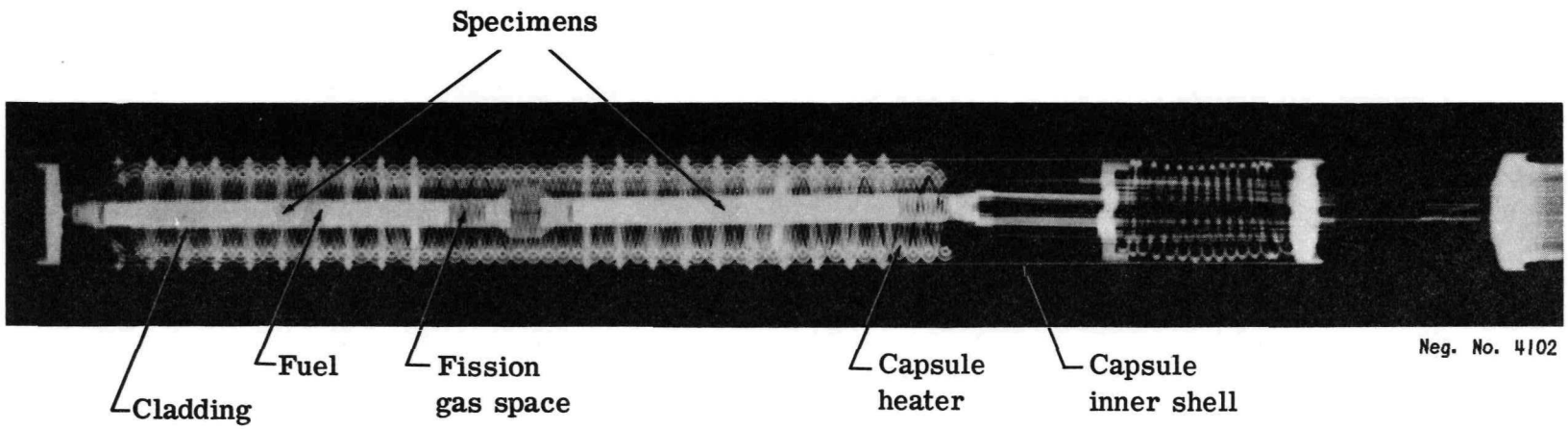


Fig. 2 — Radiograph of a typical irradiation capsule

Since the pressing and sintering process does not produce the required hole size and concentricity, the cored pellets were made with about a 0.060 in. hole and reamed to size. Initial pellets with 0.070 in. to 0.071 in. holes centered within the required 0.003 in. were produced satisfactorily by this process.

3.4 EXPERIMENT ASSEMBLY

The assembly of Capsule No. 64 was initiated.

Inspection showed that four used instrument consoles, in addition to the two new ones recently assembled, are in operating condition. Replacement instruments with improved accuracy and more suitable ranges were ordered. One instrument console will be used for each of the six capsules.

3.5 TEST REACTOR LIAISON

Since WTR and MTR have become unavailable for the irradiation program, negotiations have been in progress to determine the technical feasibility, schedule and cost of irradiating in GETR and ETR.

Four of the six capsules and possibly all six capsules are recommended for GETR irradiation. The technical feasibility of irradiating two of the high power capsules in GETR is currently being investigated.

4. REFERENCES

1. Engelhard Industries Inc., Catalog of Baker Platinum Div., p. 4 (1958).
2. M. Hansen, "Constitution of Binary Alloys," p. 377, McGraw-Hill Book Co., 1958.

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