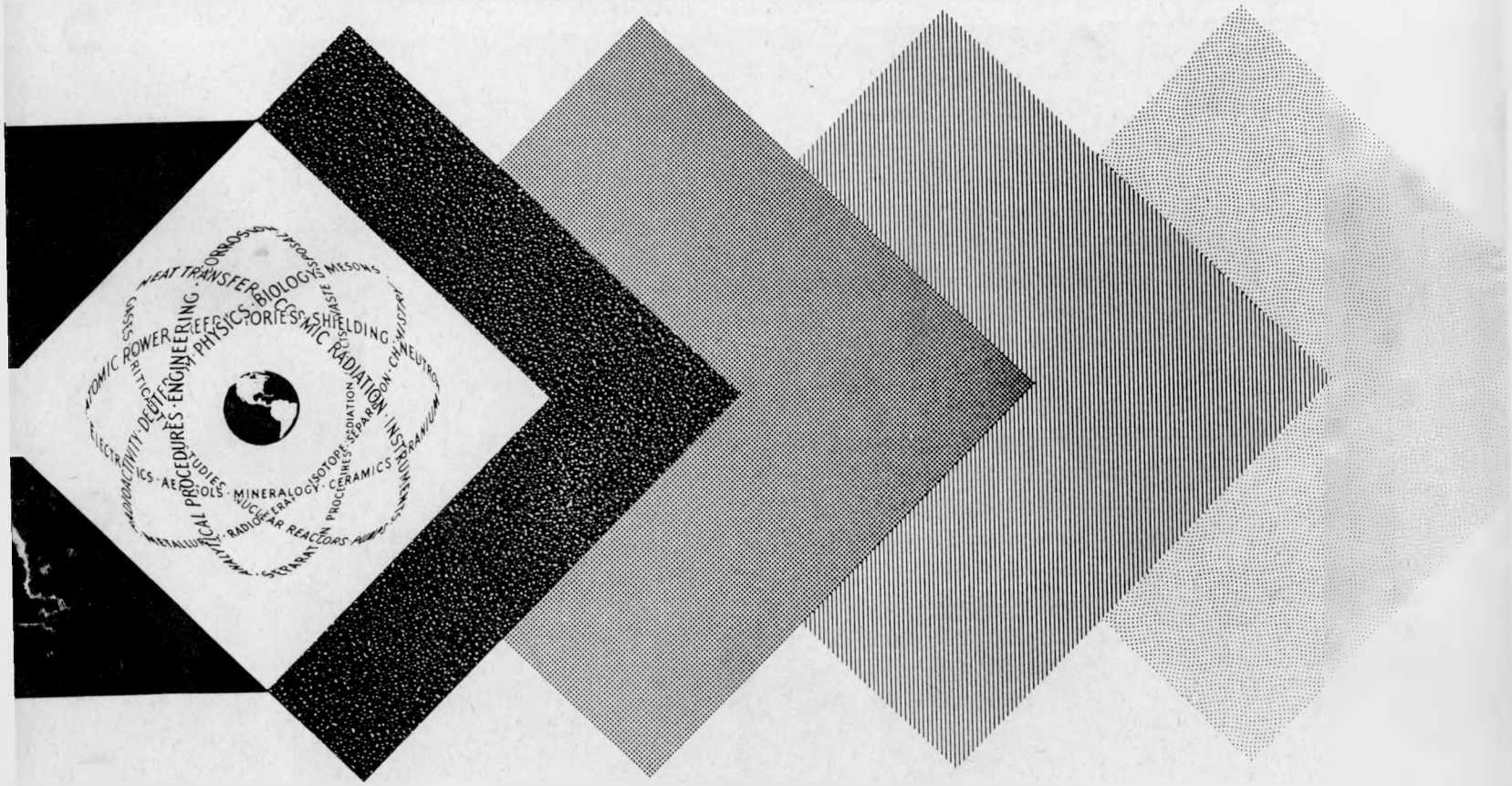
SCNC-309

METALLURGY AND CERAMICS

QUARTERLY TECHNICAL PROGRESS REPORT FOR PERIOD ENDING MARCH 31, 1960

May 1960

Sylvania-Corning Nuclear Corporation
Bayside, New York



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SCNC-309

QUARTERLY TECHNICAL PROGRESS REPORT FOR PERIOD ENDING

March 31, 1960

J. L. Zambrow, Director of Engineering

Contract: AT-30-1 GEN-366
Division of
Reactor Development

Issued: May, 1960

SYLVANIA-CORNING NUCLEAR CORPORATION
Bayside, Long Island, New York

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I. LOW COST CERAMIC FUEL ELEMENTS BY ISOSTATIC PRESSING

(J. Fugardi)

Objective: The objectives of this program are the fabrication of low cost, high quality ceramic, rod-type fuel elements through the application of hot isostatic pressing techniques.

Hot isostatic pressing techniques have been developed by Sylcor[1] for cladding UO_2 sintered pellets, "green" compacted pellets, and for both cladding and densification of UO_2 powder tamped into the cladding. The latter technique, namely the use of hot isostatic pressing to clad and densify the consolidated powder column, appears to be the most promising method.

Work has been performed on various types of powders, such as ceramic grade, high-fired and fused types. A reactor requirement based on irradiation of experimental fuels and fuel elements is that UO_2 fuels must have nearly stoichiometric U/O ratios. Fused UO_2 powder meets this requirement best. In working with the fused UO_2 powder, various methods have been tried to improve its density, such as: ball milling, hammer milling, special blending of 100% UO_2, and blending 75% coarse fused UO_2 with 25% fine ceramic. The best densities, 92-99% of theoretical, have been obtained with a mixture of 75% coarse fused powder blended with 25% ceramic grade UO_2. The U/O ratio of the powder was 2.01. Using blends of 100% fused UO_2, densities of 90-92% of theoretical have been obtained.

In all samples made to date, the powder was consolidated in the cladding by tamping prior to hot isostatic pressing. This method was satisfactory for producing short samples but as the length or number of samples increase the process becomes inefficient. A method which lends itself to consolidating the UO_2 powder for either quantity production or for parts of long lengths is the vibratory method. Both sonic and ultrasonic methods are being considered.

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[1] J. Fugardi and J. L. Zambrow, "Low Cost Ceramic Fuel Elements by Isostatic Pressing", SCNC-289 (July, 1959).

A. Ultrasonic Vibratory Packing

Aeroprojects Corporation has been conducting some ultrasonic vibratory compaction studies on UO_2 powder mixtures in stainless steel tubes supplied to them by Sylcor. These vibratory packed assemblies will be subsequently hot isostatically pressed. Aeroprojects has reported that they have obtained a density of 9.3 g/cc or 85% of theoretical density for a column of this fused UO_2 powder which had been ultrasonically vibrated into a tube 0.439" O.D. x 0.015" wall thickness. During the process a slight load was placed on the top of the powder column, using a ram. The density obtained by the ultrasonic process is equal to that obtained by a tamping process where the powder is loaded in small increments and compacted at 40 tsi. The tamping process requires a restraining die to prevent distortion of the stainless steel tubing whereas the ultrasonic process does not.

An attempt at consolidating to further density will be made by applying ultrasonic energy at both ends of the powder column. The second source of ultrasonic energy will be operated both in-phase and out-of-phase with the lower transducer to induce a maximum densification of the UO_2 powder column.

The powder used in these experiments was Spencer fused UO_2. The blend used was obtained by performing a series of experiments designed to obtain a high tap density. Table I tabulates the blend obtained, and another blend reported by General Electric as having the best vibratory packed density.

TABLE I

Powder Blend	Particle Size Distribution				Density	
	-20 +60	-20 +100	-140 +200	-325	Tap, g/cc	% Theo.
Spencer No. 12	-	64	9	27	7.90	72.0
G.E.	60	-	15	25	7.92	72.2

B. Sonic Vibratory Packing

A sonic transducer coupling has been designed and fabricated for holding the 0.439" O.D. tubing while the UO_2 powder is consolidated. Arrangements are being made for carrying out the experiments at Dayton T. Brown, Inc., Copiague, Long Island. The variables of frequency, force, time, and column length will be investigated. Various particle size distribution will be tried to obtain the highest consolidation by this method.

C. End Cap Design

End caps for the clad UO_2 powder have been designed to give a continuous cladding of stainless steel around the UO_2 core. The samples for autoclave test described in the next section used this design. The end caps are formed by compacting stainless steel powder between the electron beam-welded end cap and the tamped UO_2 powder column. An X-ray of several samples, taken after the samples were sealed by electron beam-welding the second end cap in place, shows the cold compacted end plug and UO_2 core (Figure 1). Note the line of demarcation between the fully dense stainless end cap and pressed stainless steel powder compacts. The second X-ray was taken of a group of samples after hot isostatic pressing and is shown in Figure 2. The stainless steel end plug is shown densified through the application of heat and pressure. The stainless steel powder used was type 304L prealloyed metal powder. The chemical analysis of this powder is as follows:

C	0.02%	P	0.013%
Si	2.38%	Cr	18.28%
Mn	0.93%	Ni	8.64%
S	0.013%	Fe	Balance

The screen analysis of the powder is:

+100 mesh	4.7%
-100 +140	13.1%
-140 +200	20.2%
-200 +325	26.0%
-325	36.0%

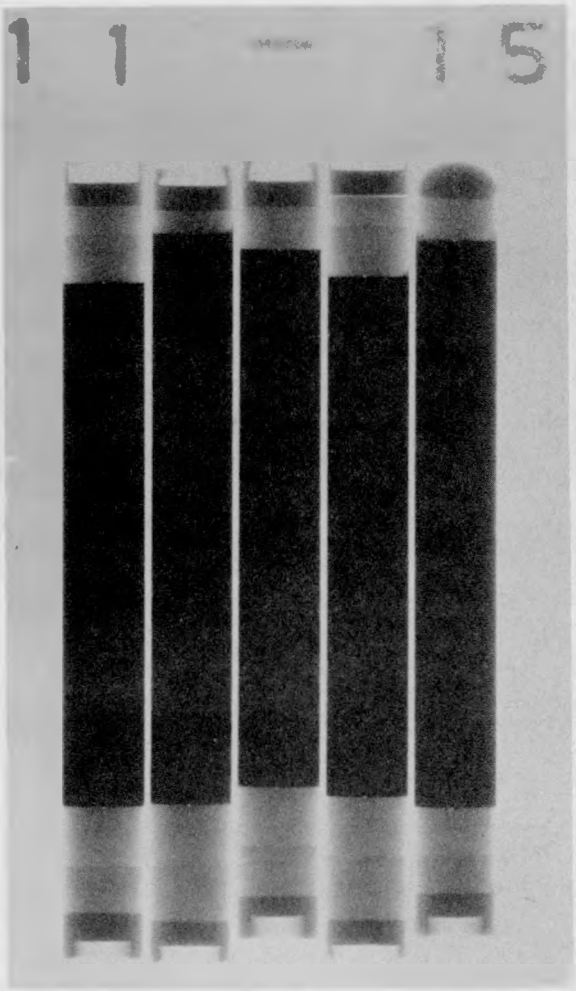

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Figure 1 - X-ray print of samples before hot isostatic pressing.

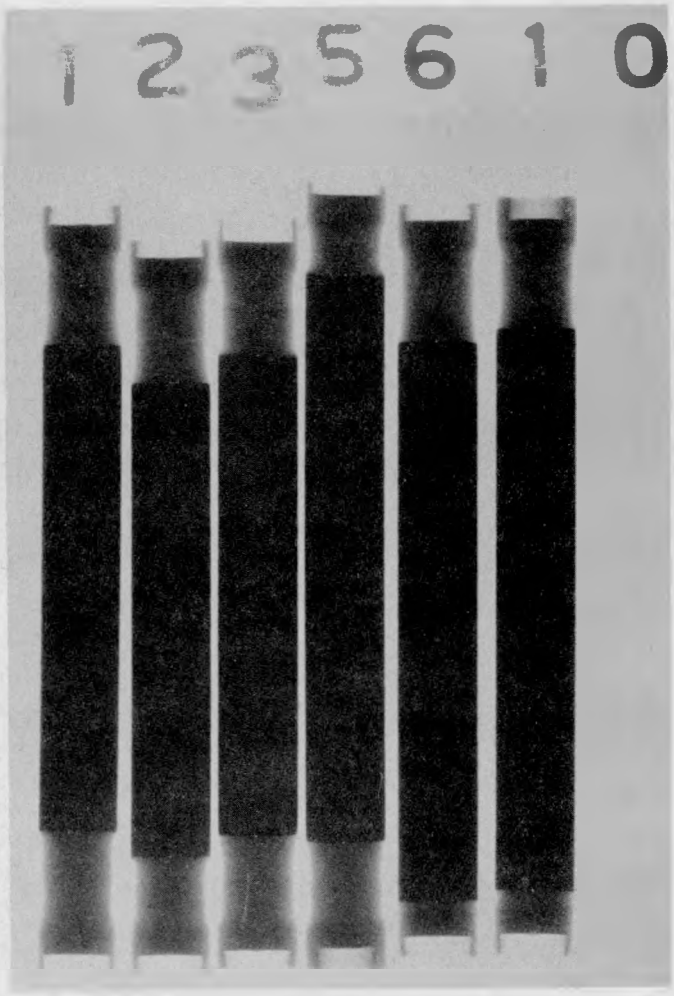

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Figure 2 - X-ray print of samples after hot isostatic pressing.

The unusually high silicon content of the powder, over the normal 0.58% silicon obtained in 304L grades of stainless steel, is caused by addition of silicon required in making the prealloyed metal powder.

In the photomicrograph of the hot isostatically pressed end caps (see Figure 3), precipitates can be seen at the grain boundaries. These precipitates have been caused by the high silicon content of the prealloyed powder.

D. Testing of the Cladding

Both autoclave and corrosion type tests have been performed on hot isostatically pressed samples.

1. Autoclave Testing

Four samples have been submitted for autoclave testing, as follows:

a. A sample of as-received 304L tubing.

b. A sample of as-received 304L tubing with electron beam-welded end caps.

c. Samples of hot isostatically pressed UO_2 & core and stainless steel powder end d. plugs.

A photograph showing these samples prior to autoclave testing is shown in Figure 4. The steam autoclave test was performed at 750°F, 1,500 psi for 36 hours.

Initial data on the autoclave test indicated that all samples remained intact upon completion of the test. Upon the return of these samples, they will be sectioned and examined metallographically for any changes in the structure of the cladding or the end cap. The weight loss of material will also be reported.

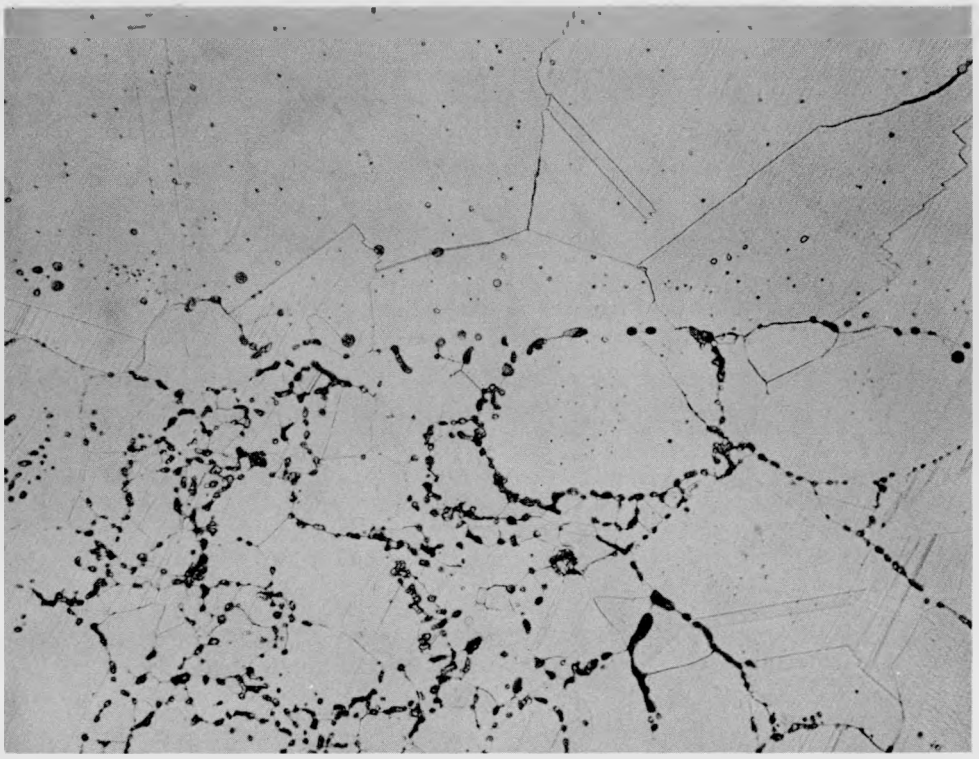

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Figure 3 - Photomicrograph of bond between stainless steel tubing and compacted stainless steel powder after hot isostatic pressing.

2. Corrosion Testing

The corrosion test performed is an ASTM test, designated as A240. Another name for this test is the "Strauss Test". It is a standard specification for testing the corrosion-resistance of chromium and chrome-nickel steel plate, sheet and strip for fusion welded unfired pressure vessels.

This test was applied to a bar of 304 stainless steel measuring 1/2" x 3/4" x 4" which had been hot isostatically pressed at the standard pressing conditions, namely, 1200°C, 12,000 psi of argon for 60 minutes. The purpose of this test in this case was to determine whether or not carbides had formed at the grain boundaries during the cooling portion of the isostatic cycle. The test consisted of immersing the sample in a solution of boiling cupric sulfate for 72 hours. The sample was then bent 180° around a 1/2" diameter pin, and examined under a magnification of no more than 30X. Cracking indicated disintegration caused by acid attack at the grain boundaries. A photograph of the sample after this test is shown in Figure 5, and indicates no evidence of cracking.

E. Sample Preparation Data

In preparing a series of fifteen samples from an as-received blend of fused UO_2, the following data has been obtained:

1. Screen Analysis of Powder Used:

Mesh fraction	% by weight
-20 +100	50.4
-100 +140	9.8
-140 +200	8.7
-200 +270	4.2
-270 +325	4.8
-325	22.1

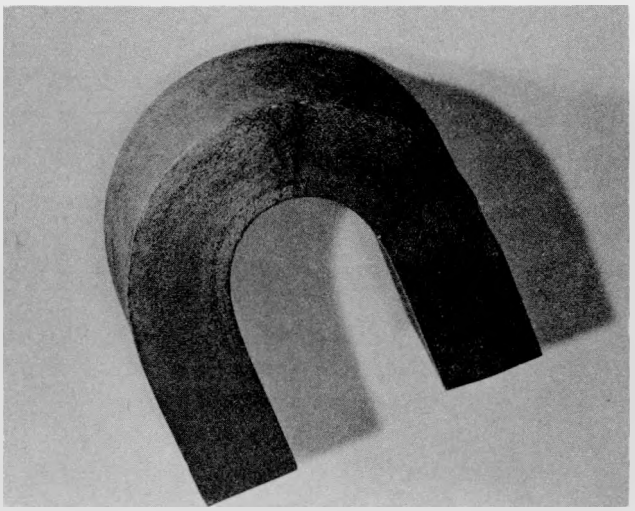

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Figure 4 - Photograph of Strauss test sample.

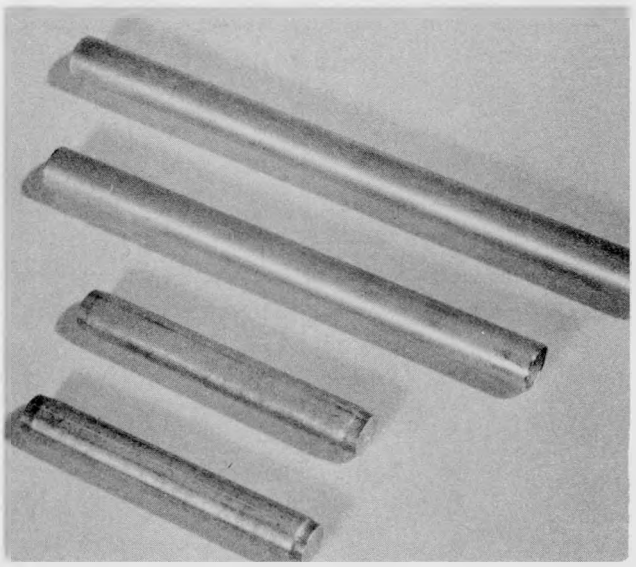

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Figure 5 - Photograph of autoclave samples before testing.

2. Powder Density:

	g/cc
a. Apparent	5.45
b. Tap	7.37
c. Pycnometer	10.78
d. Tamped	9.26
e. Hot isostatic	9.82

3. Compaction Ratios:

a. Length-to-diameter ratio 6:1.

b. Diametrical reduction from cold tamped column to hot isostatically pressed samples, %, $\frac{\Delta D}{D} = 2.33$.

c. Length reduction from cold tamped column to hot isostatically pressed sample, %, $\frac{\Delta L}{L} = 0.99$.

d. Deviation of sample from round after isostatic pressing $\pm$ 0.003".

NOTE: This distortion is principally due to the use of a two-piece restraining die during the tamping process. The use of vibratory methods to consolidate the powder column should greatly reduce the sample roundness.

e. Cladding wall thickness does not change.

II. URANIUM DIOXIDE FUEL ELEMENTS (Y. Cavallaro, J. P. Mathern and R. M. Powers)

Objective: Improvement in the thermal conductivity of UO_2 pellets by the addition of small amounts of other oxides.

A. Sample Preparation

During this period, work on the preparation of UO_2 doped with more than 4 m/o Y_2O_3 was continued, and crack-free samples containing 5 m/o and 5.6 m/o Y_2O_3 have been prepared. It was found that at additive levels greater than 4 m/o Y_2O_3 the samples had a tendency to crack, particularly during the reduction stage of the sintering operation. This occurred even though the hydrogen partial pressure was maintained at a low level during the reduction step (i.e., 10 v/o hydrogen in argon), and the cooling rate from 1300°C to room temperature was maintained at about 60°C per hour. However, if the samples were completely imbedded in alundum bubbles during sintering and reduction, crack-free samples were obtained. Using the latter technique, the following densities were obtained for the large thermal conductivity samples:

Composition, m/o	Density, g/cc	Theo. density* for composition, g/cc	% of Theoretical
5.0 Y_2O_3 - UO_2	9.36	10.45	89.6
5.6 Y_2O_3 - UO_2	9.12	10.39	87.8

* By mixture of components.

These densities are somewhat below those generally obtained with undoped UO_2, and attempts are being made to improve the density of the 5.6 m/o Y_2O_3 - UO_2 composition by a higher temperature (1500°C)

treatment in argon followed by reduction in hydrogen. These results will be reported in the following period. A photomicrograph of a 5 m/o Y_2O_3 - UO_2 sample is shown in Figure 6. The sample shows an average grain size of about 12μ and a porosity of .01.

During this period, UO_2 samples doped with 6 m/o CaO and with a combination of 1 m/o CaO - 1 m/o ZrO_2 were prepared. No difficulty was experienced in the preparation of these samples and both compositions showed good densities. Both samples were sintered by the standard technique, i.e: 3 hours in argon and 1 hour in a hydrogen-argon mixture at 1300°C, the samples being brought up to temperature and cooled back to room temperature at a rate of about 60°C/hour. Characteristic data for these samples are tabulated below.

Composition, m/o	Density, g/cc	% of Theo. Density	O/U Ratio	Grain Size, μ	Lattice Constant a_o
6 m/o $CaO-UO_2$	10.16	95%	1.99	9-18	5.454Å
1 m/o CaO-1 m/o ZrO_2-UO_2	10.15	93%	2.00	7	5.466Å

Photomicrographs of these two compositions are shown in Figures 7 and 8.

In order to obtain an independent check on the thermal conductivity measurements made at this laboratory preliminary attempts have been made to prepare the larger diameter samples which would be required for thermal conductivity measurements by the radial heat flow method employed at Armour by Hedge[2]. This method requires a 5-inch high (5 Kg UO_2) sample or stack of samples having a minimum diameter of 2-1/2 to 2-5/8 inches. In preliminary experiments, granulated UO_2 powder was hydrostatically pressed (cold) in

[2] Personal communication to R. M. Powers by J. C. Hedge.

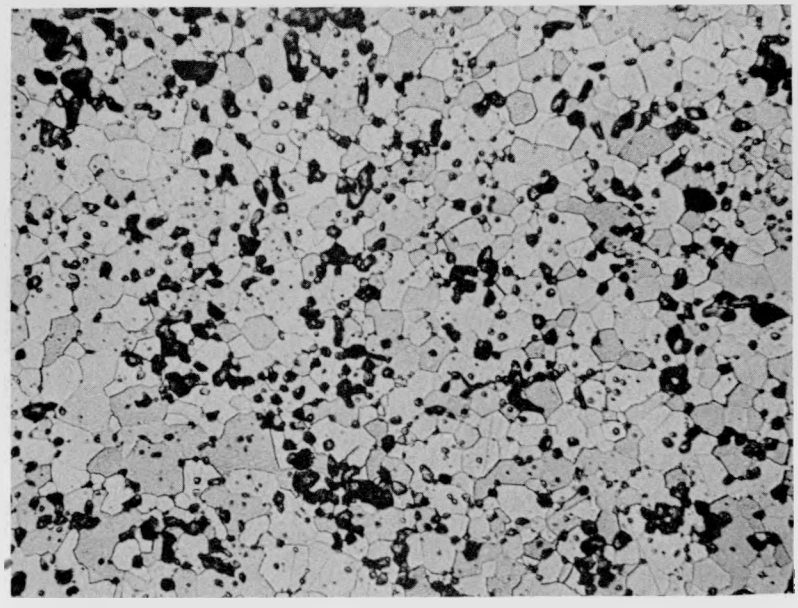

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Figure 6 - Photomicrograph of the 5 m/o Y_2O_3-UO_2 sample. This sample had been sintered and reduced while completely imbedded in alundum grain and was found to be crack-free. Its density was 9.36 g/cc (89.6% of theoretical) and the measured grain size was found to average about 12μ.

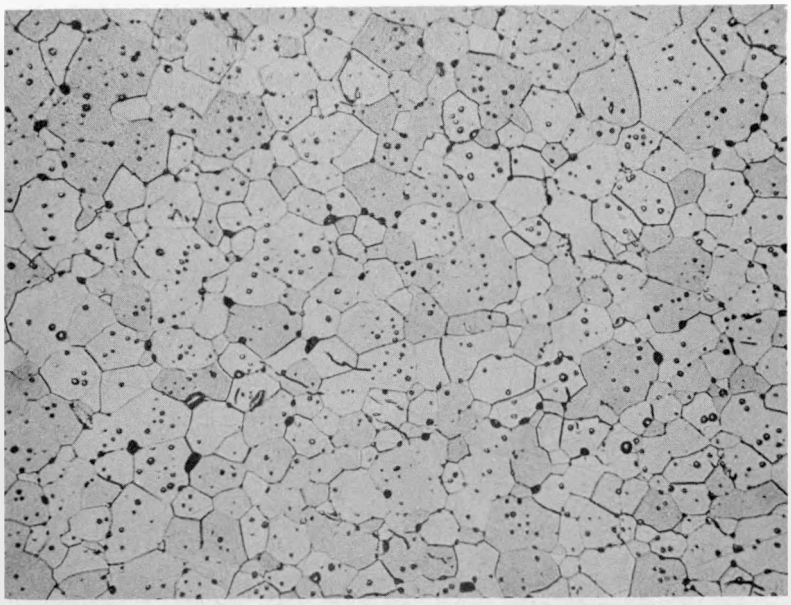

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Figure 7 - Photomicrograph of a 6 m/o $CaO-UO_2$ sample having an O/U ratio of 1.99. Density of this sample was 10.16 g/cc or 95% of theoretical.

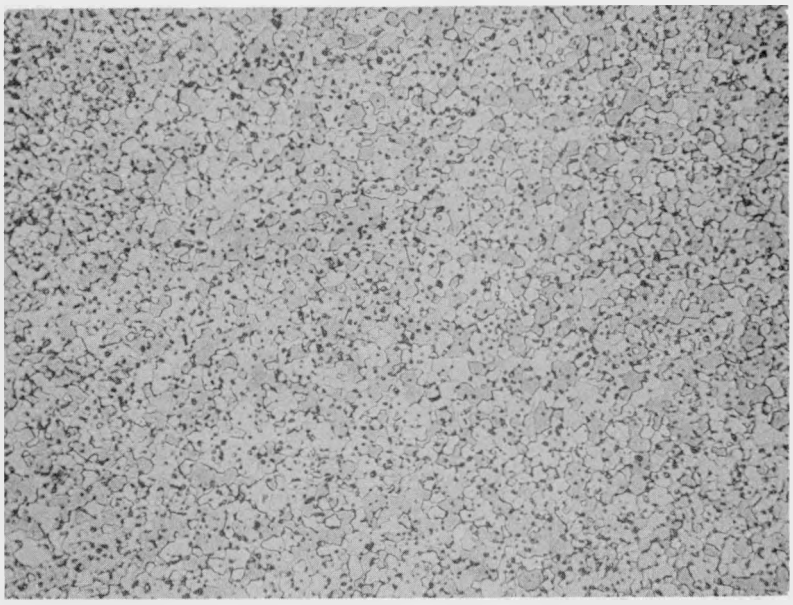

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Figure 8 - Photomicrograph of a 1 m/o CaO-1 m/o ZrO_2-UO_2 sample having an O/U ratio of 2.00. Density of this sample was 10.15 g/cc or 93% of theoretical.

a plastisol mold, forming a green compact about 2.4 inches in diameter and 0.5 inch high. Upon sintering the compact imbedded in alumina bubbles, shrinkage reduced the diameter to approximately 1.9 inches. The compact showed a small hairline crack, but otherwise was of satisfactory density, 10.69 g/cc or 97.5% of theoretical. A similar UO_2 disc was pressed hydrostatically and sintered to a dense (10.7 g/cc) stoichiometric body with sintered dimensions approximately 2-1/2" diameter x 1/4" thick. The cracking observed in this case was attributed to the hydrostatic compaction step and to the fact that the sample stuck to the mold. This situation is to be remedied by proper lubrication and modification of the mold design.

It is very important to supply crack-free samples for the radial heat flow thermal conductivity test because of the danger of extending these cracks during measurements and thereby invalidating the results.

UO_2 samples are also being fabricated containing 1 and 4 m/o BaO additions.

B. Thermal Conductivity Measurements UO_2 Standard Samples

Standardization by means of three UO_2 samples was continued in the low temperature thermal conductivity furnace during this quarter after furnace modifications had been made. Thus, the upper sample pedestal was changed from stainless steel to firebrick to reduce the longitudinal heat leakage above the sample, and a higher resistance Kanthal-wound heater was employed to provide a higher upper temperature for the system. At the same time, the conductivity of the lower pedestal was increased to provide a better heat sink, the leakage from which would be decreased by the application of heat to the lower sample heater. Hollow alumina bubbles of low conductivity were also substituted for the crushed firebrick used previously as a radiation shielding.

These modifications of themselves did not seem to improve the results obtained with the three stoichiometric UO_2 standard samples before improvements had been made. As can be seen from the results shown in Table II which were obtained under conditions of typical balance at two temperature levels (run No. 2), the middle sample showed a

TABLE II

SAMPLE AND GUARD TEMPERATURES WITH THREE UO_2 SAMPLES UNDER APPARENT BALANCE. SMALLER TEMPERATURE DROP IN MIDDLE SAMPLE INDICATES APPARENT HEAT LOSS. (RUN NO. 2)

	Interfacial Temperature Drops, °C	Guard Temperature °C	Sample Temperature °C	Sample Temperature Drops, °C	Temp. Difference Between Guard and Sample, °C
Upper		704	706		-2
Sample			650	56	
	18	642			+1
Middle			632		
Sample			592	40	
	27	578			-1
Lower			565		
Sample		524	522	43	+2
Upper		486	486		0
Sample			456	30	
	24	444			0
Middle			432		
Sample			412	20	
	34	394			-1
Lower			378		
Sample		354	354	24	0

smaller temperature drop than should be expected for axial heat flow alone. This run was therefore discontinued and additional guard thermocouples were added to the system at positions opposite the middle of each sample as well as opposite the top and bottom sample heaters. The automatic heat balancing obtained by the use of Micromax controllers connected between the upper sample and upper guard couples and between the lower sample and lower guard couples was also discontinued and manual control substituted for all of the heaters. Also, silver foil was substituted for platinum at the interfaces to reduce the magnitude of the interfacial drops. The results after using additional guard thermocouples showed that a balance obtained by means of the four guard couples previously used did not necessarily insure a linear drop along the guard heaters corresponding to that along the sample stack. A marked tendency was found for the middle guard couple of each heater, particularly at elevated temperatures, to show a temperature lower than that to be expected from a linear drop. This effect is thought to be caused by the tendency of the mullite guard to produce a constant temperature zone opposite each heater because of the uniformity of the heater windings. The poor thermal conductivity of the mullite at elevated temperatures tends to accentuate this effect. However, it has been possible, by proper adjustment of the guard heaters to approach linear temperature drops along the mullite guard at least opposite the middle and bottom samples and thus to approximate a true balance. When balance was established in this manner a decreased temperature drop across the middle standard sample was no longer observed. Results obtained during this run (No. 3) using three standard UO_2 samples at various temperatures are shown in Table III, and plots of sample and guard temperatures for typical conditions are shown in Figures 9 and 10.

Under conditions of true thermal balance where there is no transfer of heat between the sample and its surroundings (guard heaters), the product of the temperature drop (Δt) along a unit length for a sample of constant cross-section and its thermal conductivity (k) is constant so that

$$(k_1)\ (\Delta t_1) = (k_2)\ (\Delta t_2) = (k_3)\ (\Delta t_3)$$

where the subscripts denote the upper, middle and lower samples, respectively. When this equation is satisfied

TABLE III

RESULTS OF SEVERAL THERMAL CONDUCTIVITY STANDARDIZATION RUNS USING THREE UO_2 PELLETS AS SAMPLE STACK

Date & Time of Balance	$k_1\Delta t_1$ [a] $\times 10^3$	$k_2\Delta t_2$ [a] $\times 10^3$	$k_3\Delta t_3$ [a] $\times 10^3$	Δt_2 °C	Avg. t_2 °C	$k_2 \times 10^3$ Calc. [b]	$k_2 \times 10^3$ Kingery [c]
	cal/sec/cm					cal/sec/°C/cm	
3/24 (1:15)	748.0	693.0	653.4	77	690	9.1	9.0
3/24 (3:15)	747.6	712.0	695.8	80	705	9.0	8.9
3/23 (12 N)	572.0	571.2	602.7	51	493	11.5	11.2
3/23 (8:10)	618.0	577.2	585.6	52	502	11.6	11.1
3/18 (10:15)	470.8	501.6	508.4	44	480	11.1	11.4
3/14 (11:15)	528.0	468.0	466.2	39	460	12.7	12.0
2/25 [d] (10:30)	520.8	392.0	460.1	40	612	12.3	9.8
2/15 [d]	354.0	254.0	331.2	20	422	17.1	12.7

(a) k values obtained by Kingery[3] and corrected to 95% density used.

(b) Calculated $k_2 = \dfrac{k_1 \Delta t_1 + k_3 \Delta t_3}{2(\Delta t_2)}$

(c) Values of thermal conductivity for UO_2 corrected to 95% density.

(d) Data taken during run No. 2. Middle UO_2 sample in stack showed smaller temperature drop than expected under conditions of axial heat flow alone although guard-sample couples appeared to be in balance.

[3] W. D. Kingery, et al, "Thermal Conductivity Data for Several Pure Oxide Materials Corrected to Zero Porosity", J. Am. Cer. Soc. 37 (2) pp.107-110, (1954).

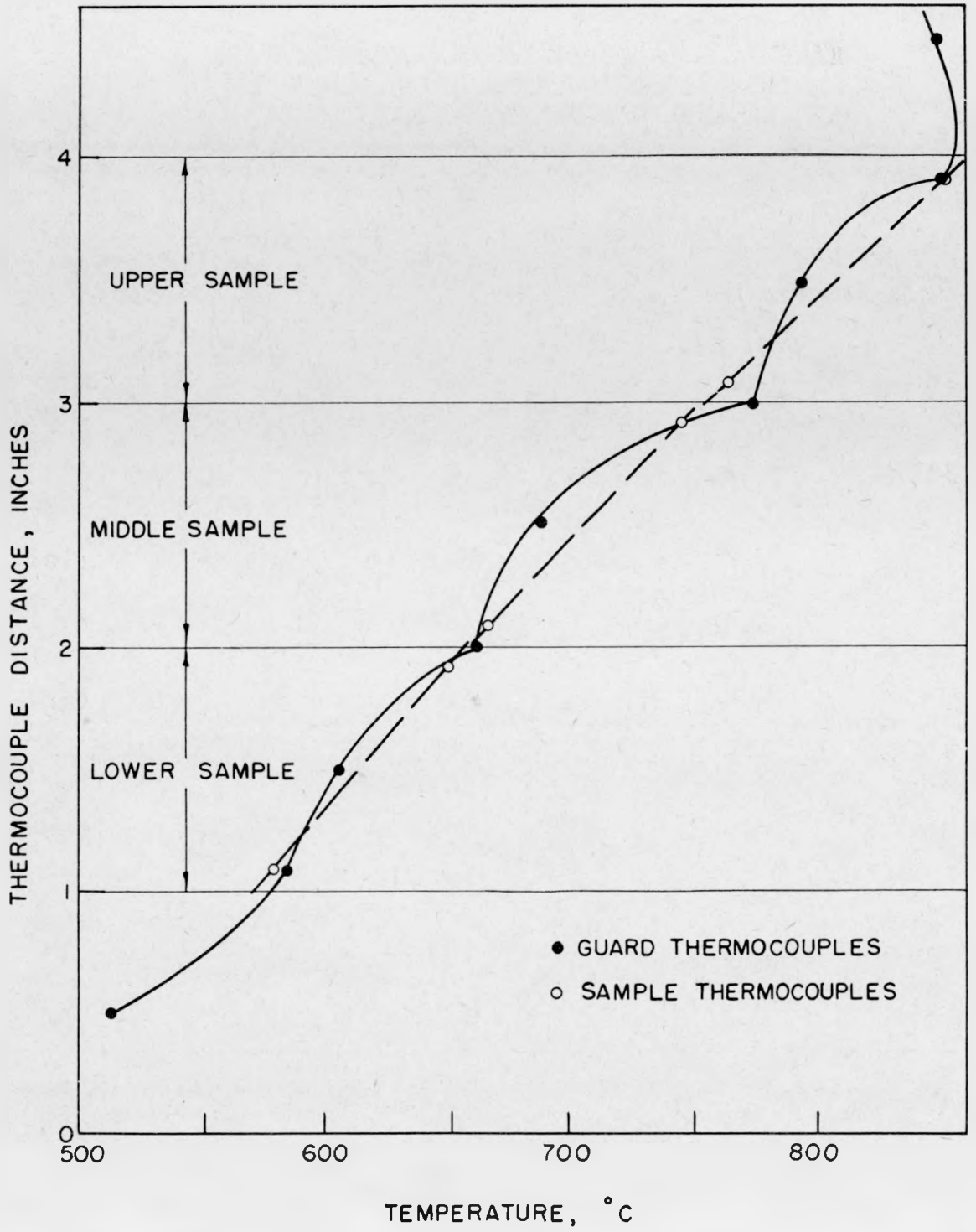

Figure 9 - Temperature profile of sample stack (3 UO_2 pellets separated by thin silver foil) and of furnace guard for balance taken at 1:15, 3/24. Although a linear temperature drop in the guard corresponding to that of the sample stack has not been attained, the calculated thermal conductivity of the middle sample compared well with that given by Kingery (see Table III) since the lateral heat flux from each sample as determined by the areas between the two curves is relatively comparable.

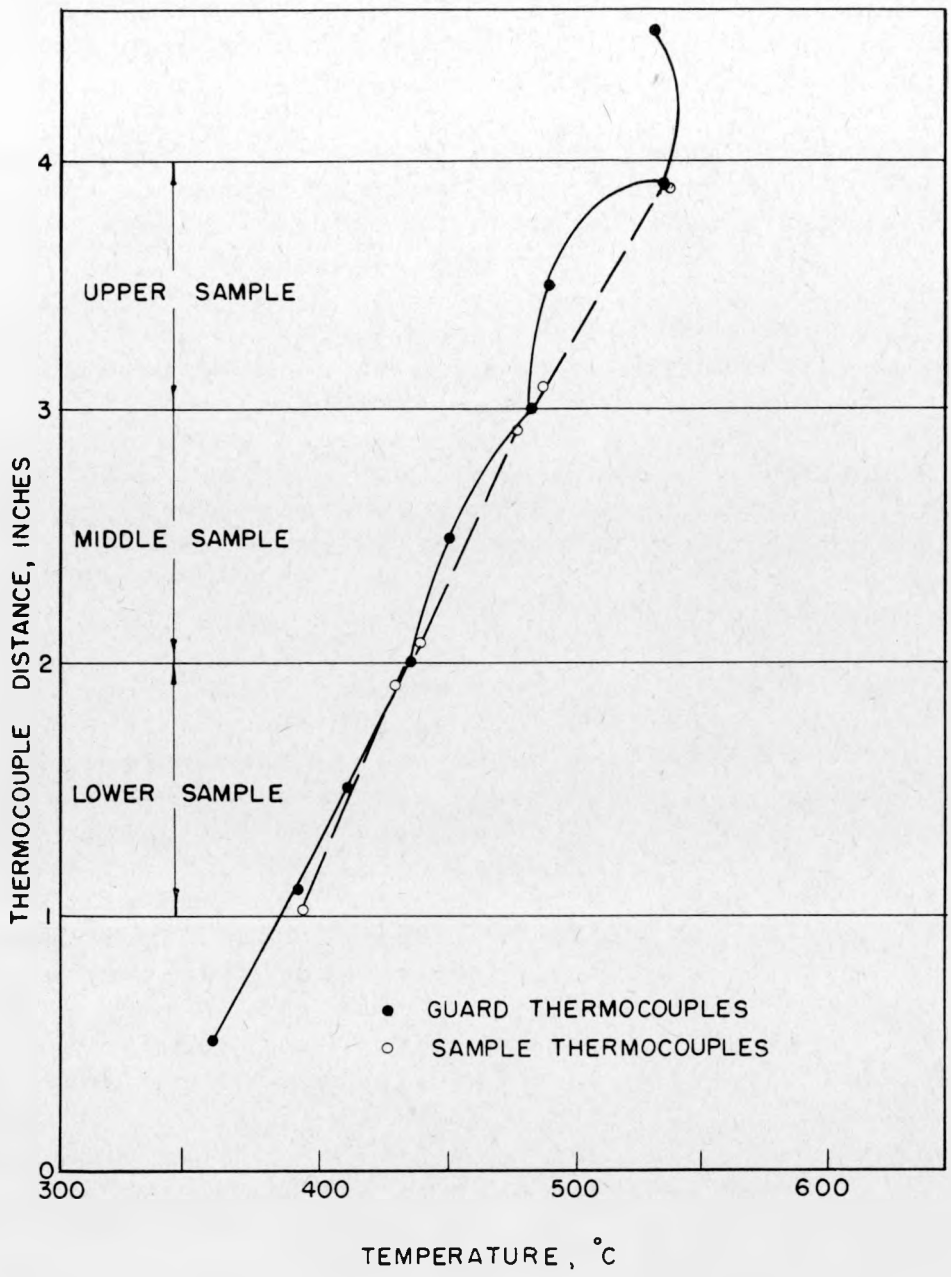

Figure 10 - Temperature profile of sample stack (3 UO_2 pellets separated by thin silver foil) and of furnace guard for balance taken at 11:15, 3/14. An almost linear temperature drop in the guard for the lower and the middle samples has been attained, but not for the upper sample. This is reflected in the k_{at} values for each sample (from top to bottom: 528.0, 468.0 and 466.2). The middle and lower samples show a nearly constant heat flux with negligible lateral heat losses whereas the upper sample shows considerable loss of heat, laterally, to its environment.

one can deduce that there is no lateral loss or gain of heat, from sample to sample in the stack.

As can be seen from Table III, the ($k\Delta t$) values for the three samples in the stack are not constant since it has not been possible to achieve linear temperature drops along both the guard and samples simultaneously in the present furnace. Under certain conditions, however, and with the aid of the additional thermocouples, a pseudo-balance can be achieved which results in good correlation between the Kingery values for UO_2 and those calculated from the sample temperature drops. Figure 9 shows a graph of such a pseudo-balance where the amount of heat loss from each sample to its surroundings is about equal, as can be estimated by the area between the guard and the sample temperature curve. The data for this graph are those shown under 3/24-1:15 in Table III.

The graph shown in Figure 10 shows almost linear drops in the sample and the guard temperatures for the middle and lower samples but not for the upper one. (These are given in Table III under 3/14-11:15.) As can be seen from the ($k\Delta t$) values for each of the three samples, the values for the middle and lower samples are nearly equal but the value for the upper sample which is not in balance with the guard is considerably higher. Thermal conductivities calculated from an average of the upper and lower sample data, therefore, show some deviation from the Kingery data which is being used as a standard. However, good agreement is obtained using the data from the middle and lower samples only.

Since it has not been possible to obtain true thermal balances between the samples and the guard along the entire stack of samples, accurate thermal conductivity measurements cannot be made in this low temperature thermal conductivity furnace. With the aid of the additional thermocouples, measurements giving qualitative indications of improvement in thermal conductivities at temperatures up to 700°-800°C are possible. This low temperature furnace is therefore to be used for obtaining qualitative measurements on UO_2 samples doped with yttria, calcium and possibly barium.

A higher temperature furnace (Kanthal-wound) modified to overcome the disadvantages of the low temperature furnace had been nearly completed. The primary modifications are:

1. Longer muffle to minimize end-effects.

2. Non-uniform furnace windings opposite the sample stack designed so as to produce a temperature gradient rather than a constant temperature zone opposite each sample.

3. Use of an alundum sleeve between the mullite furnace tube and the samples which, because of its higher thermal conductivity at elevated temperatures, is expected to induce a smoother guard temperature gradient than has been obtained with the mullite guard.

4. Increase in the top temperature of the system to 1200°C.

During the coming quarter, it is expected first to calibrate this high temperature furnace with three UO_2 samples and then to measure the thermal conductivity of yttria and calcium doped samples up to 1200°C.

C. Additional Studies

1. Furnace Checkout

The high temperature tungsten resistance furnace shown in Figure 11 to be used to measure the electrical resistivity of samples of UO_2 and UO_2 containing additives as a function of temperature as well as to determine the solidus temperature of such samples, has been completed and tested during this period. Vacua of 10^{-5} mm Hg have been maintained up to 1200°C. By substituting an atmosphere of purified argon for vacuum, temperatures slightly in excess of 3000°C have been reached. Minor changes, such as redesign of the baffles into a single rigid unit, redesign of the sample holder and increasing the water flow to electrodes, have been made since the first trial runs.

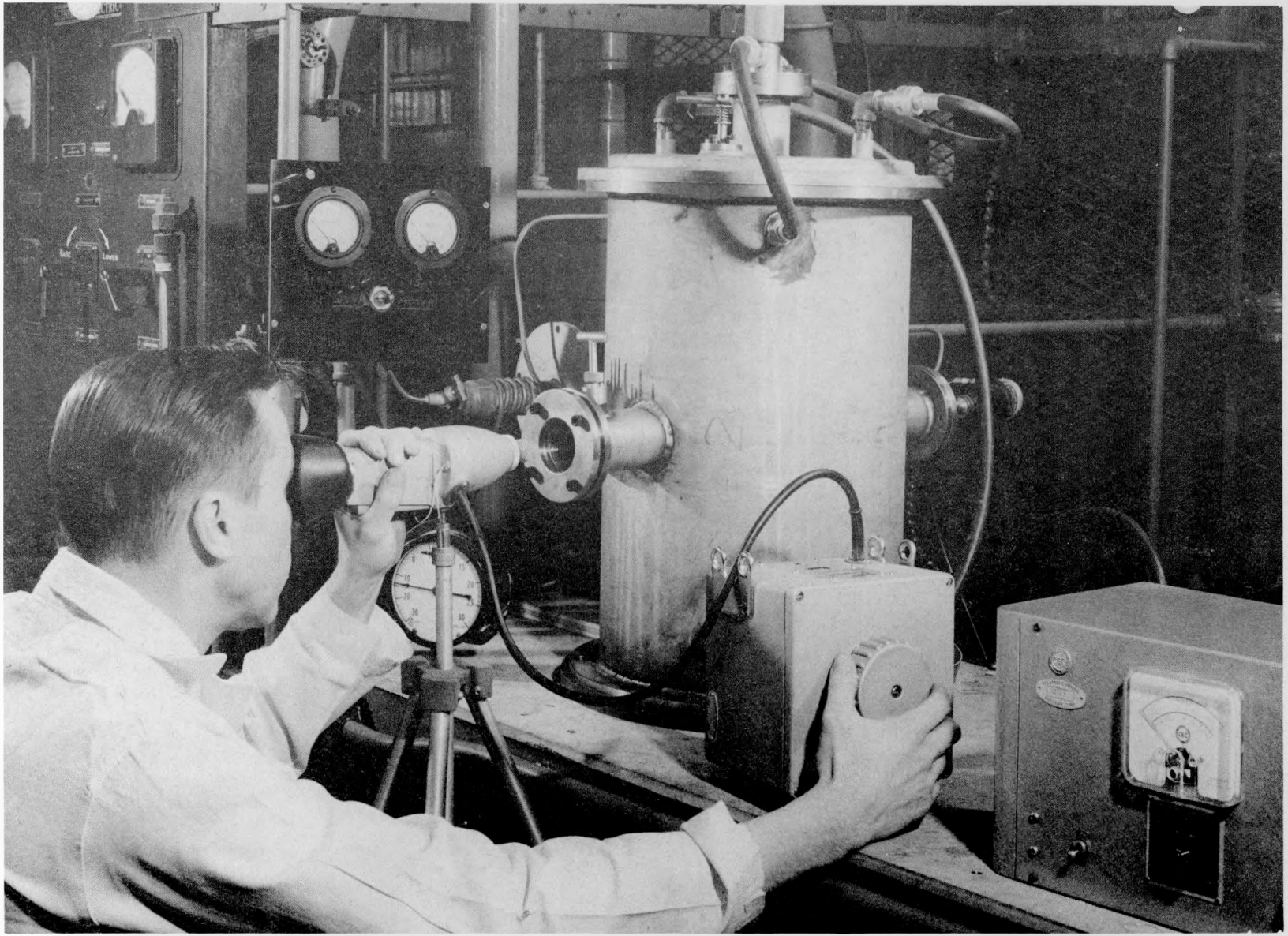

Plate No. 10720. Fig. 11-High Temperature Furnace for Determining Melting Points and Resistivity.

2. Solidus Temperature Determination

A sample of UO_2 containing 4 m/o of Y_2O_3 similar to that supplied for the Chalk River tests was heated without melting to a corrected temperature of about 2650°C. From these data, it appears that an addition of 4 m/o Y_2O_3 to UO_2 does not depress the melting point of UO_2 by more than 100°C. This test is to be repeated using a redesigned sample holder. The new holder should prevent the black body holes in the sample (0.040" diameter x 0.2" deep) from shifting out of the line of sight and thus permit the solidus temperature to be observed.

3. Electrical Resistivity vs Temperature

Electrical resistivity measurements have been started on UO_2 and its solid solutions at room temperature. To minimize the large contact resistance that arises in such measurements, a probe method is being employed. Both DC and AC probe methods are to be tested at room temperature before measurements in the furnace are begun. It was found that UO_2 samples after fabrication are covered with a surface layer of higher conductivity than that of the bulk of the sample. This layer must be ground off before meaningful resistance measurements can be made. This surface skin on UO_2 appears to give rise to capacitive effects so that the AC resistance at room temperature as measured by a high impedence AC vacuum tube voltmeter appeared to drift with frequency. The effect of humidity on the resistivity of these high resistance UO_2 samples is also appreciable so that final measurements must be carried out in the vacuum furnace. A sample holder has been designed and constructed for measurement of resistivity up to 2200°C, in the vacuum furnace.

4. Corrosion Testing

Corrosion testing of UO_2 samples containing various percentages of the solid solution additions CaO, Y_2O_3, $CaO-ZrO_2$ has begun. These samples are supported on shelves of stainless steel screen within a conventional autoclave bomb. The corrosion rates are being measured as a function of time in 680°F steam at 2,000 psi.

D. In-Pile Thermal Conductivity Tests at Chalk River

During this period two capsules containing stoichiometric UO_2 and two capsules containing stoichiometric UO_2 + 4 m/o Y_2O_3 were shipped to Chalk River for in-pile thermal conductivity testing. Such a capsule is shown in Figure 12.

This test is expected to indicate whether or not an overall improvement in the average thermal conductivity of UO_2 is obtained over the operating range of the fuel (i.e., from 300°C to 2750°C) by minor additions of Y_2O_3 to UO_2. The principle of the test[4,5] is to generate identical amounts of fission heat in a doped UO_2 sample as there would be in a UO_2 control sample. This is accomplished by adjusting the enrichment of U-235. The capsule wall thickness and diameter, the pellet-to-clad clearance, the pellet diameter and density, are carefully reproduced from test to test. A capsule is irradiated in a standard neutron flux in the hydraulic rabbit test hole of the NRX reactor. Coolant flows and temperature are also standardized.

In a uniform neutron flux it is assumed that equal rates of heat generation are obtained by employing equal numbers of U-235 atoms per unit volume of oxide. It is also assumed that negligible changes in flux occur as a result of substituting small quantities of yttrium (σ = 1.38 barns) for U-238 (σ = 2.88 barns) because of the preponderant effect of the U-235 (σ = 5.80 barns) on this flux. Simple calculations indicate a 0.2% increase in flux would be expected in a UO_2 pellet 4.6% enriched in U-235 by substituting 8 atom percent of yttrium for 8 atom percent of U-238.

For the first test at Chalk River a total heat flux ($\int k d\Theta$) of 80 watts/cm is to be employed. This is sufficient to cause central melting of the standard stoichiometric UO_2 pellets. If an improvement in the

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[4] A. S. Bain and J. A. L. Robertson, "UO_2 Irradiations of Short Duration", CRFD-825, AECL No. 806, April 1959.

[5] J. G. Melvin, "The NRX Hydraulic Rabbit Facility", AECL No. NE1 107, Feb. 1959.

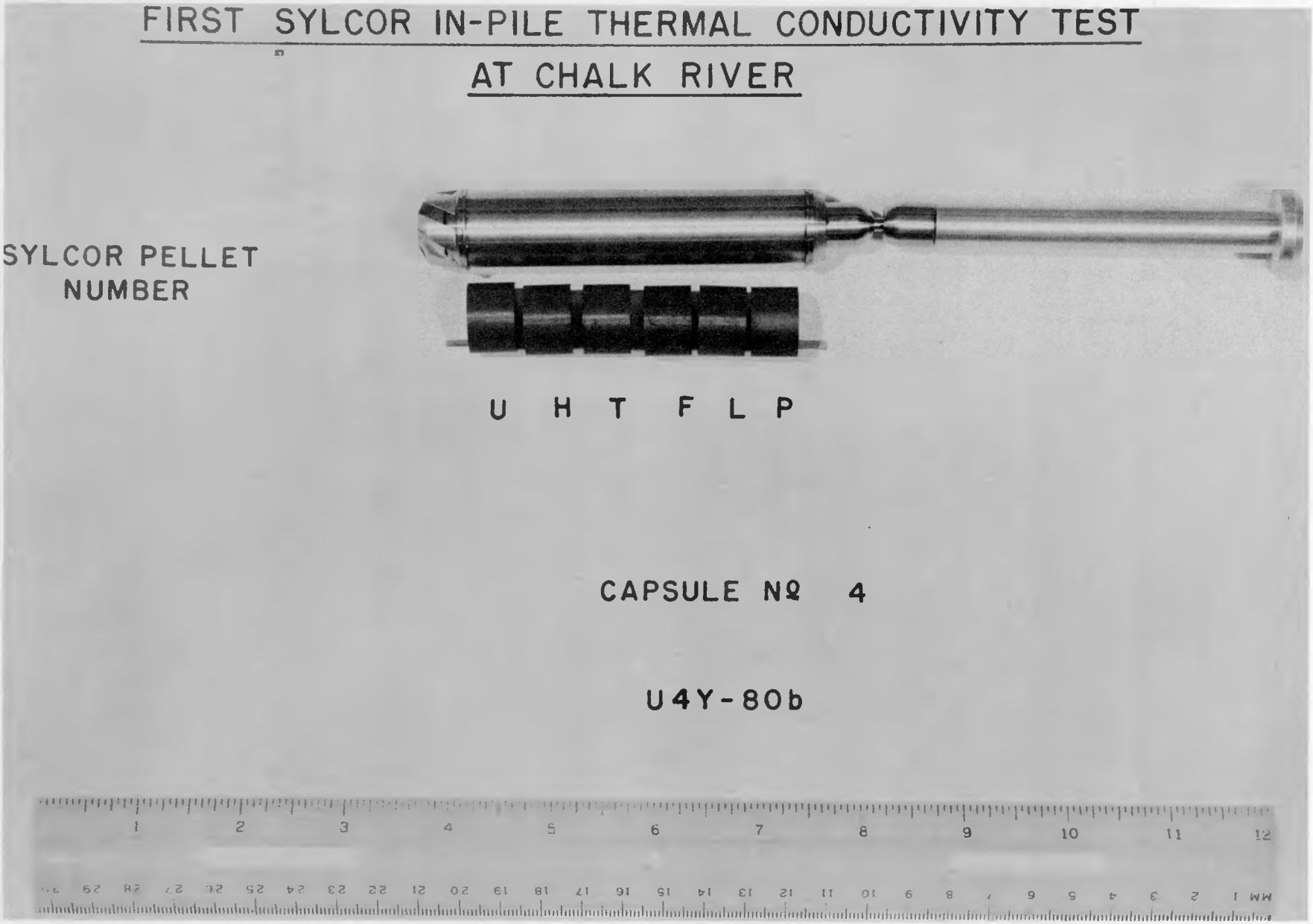

Plate No. 10701. Fig. 12 - In-Pile Thermal Conductivity Test Capsule.

thermal conductivity of UO_2 over the operating range of the fuel is obtained by addition of 4 m/o Y_2O_3 in solid solution no central melting should be observed for the doped samples.

Irradiation times of 3 minutes are to be employed to allow thermal equilibrium to be obtained under the standard flux conditions of 5×10^{13} n/cm^2/sec at NRX position N-9 and one sample irradiation is to follow on the heels of the other. Cobalt monitors are included in an appendage to the capsule to check this flux.

The following compositions are to be irradiated in the first test, all at a $\int k d\theta$ of 80 watts/cm.

Order of Irradiation	Sample Composition	Sample No.	w/o* U-235	v/o $U_{235}O_2$
1	UO_2	U80a	4.60	4.656
2	UO_2+4 m/o Y_2O_3	U4Y-80a	4.95	4.656
3	UO_2	U80b	4.60	4.656
4	UO_2+4 m/o Y_2O_3	U4Y-80b	4.95	4.656

* These enrichments are being checked by the New Brunswick Laboratories of the U.S. Atomic Energy Commission.

The enrichment required to achieve an $\int k d\theta$ of 80 watts/cm was taken from a recent graph kindly supplied by J. A. L. Robertson[6] for stoichiometric $UO_{2.00}$. Pellets were prepared by established techniques in which Mallinckrodt ceramic grade UO_2 was blended with yttrium nitrate solution, prefired to 1000°C, hydrogen reduced, then reoxidized to a O/U ratio of 2.5-2.6. After granulation with 0.5% PVA this powder was pelletized in a Stokes R-4 press at 20 tsi then fired 3 hours in argon at 1300°C followed by 1 hour in a 1/10 H_2-argon mixture at 1300°C and cooled to room temperature in the same atmosphere. The samples for the Chalk River test have been characterized by density, metallography, O/U ratio, and X-ray diffraction.

6 Personal communication to R. M. Powers from J. A. L. Robertson.

E. Post Irradiation Examination

Samples stored by Chalk River personnel in the receiving flask are to be cooled for at least one day then transferred to a hot cell for examination. There they will be cut in two and photographed. The radius of the melted zone and/or the grain growth zones in the UO_2 and the 4 m/o Y_2O_3 doped UO_2 are to be measured from the photographs, and the average thermal conductivity determined from these results.

III. IRRADIATION OF ISOSTATICALLY PRESSED OXIDE SAMPLES

(S. D. Strauss)

Objective: Determination of the behavior of stainless steel-clad, isostatically pressed UO_2 samples relative to that of standard, pelletized UO_2, under prolonged neutron irradiation.

A. Preparation of Irradiation Samples

1. Process Test Run

Finalization and testing of the hot isostatic pressing procedure, to be used in preparing the irradiation test samples, was successfully accomplished during this quarterly period. The following procedure was used to prepare a single dummy test sample of normal UO_2 in a stainless steel jacket.

a. The UO_2 powders are sieved and blended. Fused UO_2 has been used in the dummy sample, whereas the irradiation samples will be prepared from two types of powder:

(1) a mixture of 75 w/o fused and 25 w/o ceramic grade UO_2,

(2) 100 w/o fused UO_2.

The mixed powders yield 93-94% of theoretical density as compared to 90-92% for 100% fused UO_2 powder. Use of ceramic grade powder, however, results in a lower green density and, therefore, in a higher degree of distortion during the hot pressing operation. Consequently, duplicate samples of both the fused and the mixed powders will be prepared for irradiation.

b. The powders are tampled into type 304 stainless steel tubing. The tube dimensions are 0.40 inch I.D. by 0.015 inch wall thickness. The powder is added in increments of approxi-

mately 1/4 inch, and tamping pressures of approximately 40 tsi are applied. The steel tube has been contained within a split-type restraining dye, in this step, in order to minimize its distortion during tamping, but since a deviation of ± 0.003 inches from roundness occurred as a result of using the split die, the irradiation samples will be pressed in an expendable solid die.

c. Stainless steel end plugs are inserted in both ends of the tube, and the tube is sealed by electron beam welding.

d. The assembly is isostatically pressed at 14,000 psi for one hour, at a temperature of 1200°C.

In step c, stainless steel powder is compacted between each stainless steel end plug and the tamped UO_2 column. Hot isostatic pressing densifies the compacted steel powder and establishes a bond both between the powder particles and between the cladding and the compacted powder plug. Both chemical and screen analysis of the prealloyed steel powder is given in Section I of this report.

Because of the high silicon content, which is necessary for the manufacture of the powder, precipitation occurs at the grain boundaries as a result of isostatic pressing, as described in Section I. An autoclave test now being carried out on an assembly made with this powder will determine whether the precipitates are objectionable.

X-ray inspection of the dummy sample showed the oxide to be homogeneous and the surface and interfaces to be free of irregularities or defects (see Figures 13 and 14). The oxide density was found to be 90% of theoretical. A helium leak test and dye-penetrant test indicated there were no surface cracks or leaks in the completed sample. The irradiation samples are expected to show equally good characteristics, with slightly higher oxide densities anticipated for those incorporating the mixture of 75 w/o fused and 25 w/o ceramic grade oxide.

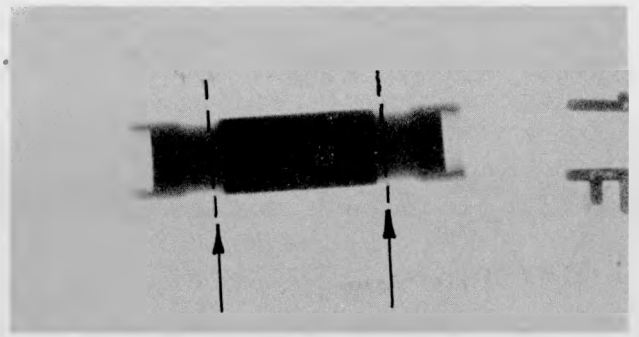

Plate No. 23108 1X

Figure 13 - X-ray of isostatically pressed, fused normal UO_2 in stainless steel jacket. Arrows indicate portion to constitute dummy irradiation sample (see below).

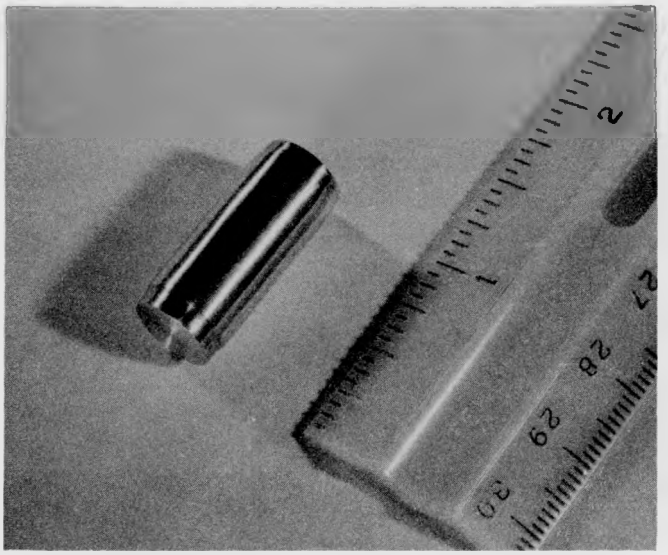

Plate No. 10711

Figure 14 - Photograph of dummy irradiation sample of fused UO_2 isostatically pressed in stainless steel jacket.

2. Procurement of 19% Enriched UO_2

Approximately 425 grams of uranium metal, enriched to 90%, was sent to the Spencer Chemical Corporation for conversion to UO_2, purification, and downgrading to a final enrichment of 19%. This was accomplished, and a portion of the product was delivered to the Mallinckrodt Chemical Corporation for pelletization.

Spencer reported, however, that the product contained 332 ppm of aluminum impurity, and as such, did not meet the requirements for reactor grade powder. Spencer was therefore requested to reprocess the material in order to reduce the aluminum content to 50 ppm or less. Since the purity specifications were set by Spencer, no difficulty is anticipated in their being able to obtain the desired purity level. The Mallinckrodt shipment has been returned, and reprocessing has begun. Receipt of the oxide in both powder and pellet form is expected in May, 1960.

B. Capsule Design and Fabrication

All capsule components were fabricated during this quarter. Encapsulation of the irradiation samples and checkout of the capsules will proceed when the samples become available.

C. Work Plan for Next Quarter

Fabrication and encapsulation of the 19% enriched irradiation samples is scheduled during the next quarter. As a result of the prolonged delay in procuring reactor grade UO_2, insertion of the capsules in the reactor has been rescheduled from May to the end of July, 1960. A Request for Irradiation, Form AEC-320, has been filed with the AEC requesting irradiation space in either the Materials Testing Reactor or the Engineering Test Reactor. Temperature, material and weight characteristics of the capsules have been supplied to the Reactor Operator[7].

[7] NDA Form ET-822, March 2, 1960.

IV. IRRADIATION OF URANIUM-YTTRIUM ALLOYS

(S. D. Strauss)

Objective: Determination of the characteristics and dimensional stability of two uranium-yttrium alloys under neutron irradiation at high temperatures.

A. Preparation of Irradiation Samples

The irradiation samples of uranium-yttrium alloys were successfully prepared during this quarter. The hydriding equipment used for hydriding both the uranium and the yttrium had been checked out using normal uranium. The following procedure was used for preparing 12 enriched samples (10 for irradiation and 2 for metallographic inspection):

1. Uranium of 90% enrichment was hydrided at 450°F, decomposed at 1000°F, and ground to -270 mesh powder.

2. Normal uranium was hydrided, decomposed, and ground to -270 mesh powder (same temperatures).

3. Yttrium was hydrided at 1472°F and ground to 5.6 micron average particle size.

4. The enriched uranium was separated into two equal charges of 25 grams, and sufficient normal uranium was added to yield one blend of 55% enriched uranium powder and one blend of 22% enrichment. The two blends were then mixed separately for six hours in a V-blender.

5. The 55% enriched uranium powder was added to YH_2 powder to form a mixture of 25 w/o U-75 w/o Y; the 22% enriched uranium powder was added to YH_2 powder to form a powder mixture of 50 w/o U-50 w/o Y. Each mixture was blended for six hours.

6. Benzene saturated with camphor was added to the mixtures; the benzene was then evaporated in an argon atmosphere, and the blends mixed for six hours.

7. Six green compacts of each alloy composition were pressed in the 1/2 inch diameter, double-acting die at 50 tsi.

8. The samples were wrapped in tantalum foil, placed in an alundum boat with zirconium getter, and sintered in vacuo for twelve hours at 1100°C.

The ten samples to be irradiated were machined to final dimensions. Density determinations were then made on all 12 pieces, including the two which were prepared for the purpose of metallographic inspection. The results are presented below:

TABLE IV

Densities of 25 w/o U-75 w/o Y Alloy
Irradiation Samples

Sample	Length inches	Diameter inches	Density gm/cc	U-235 Density gm/cc	% Theoretical Density
1	0.755	0.147	5.25	0.751	95.5
2	0.753	0.184	5.28	0.755	96.0
3	0.753	0.213	5.32	0.761	96.7
4	0.755	0.230	5.30	0.758	96.4
5	0.754	0.254	5.28	0.755	96.0
A	-	-	5.32	0.761	96.7

TABLE V

Densities of 50 w/o U-50 w/o Y Alloy
Irradiation Samples

Sample	Length inches	Diameter inches	Density gm/cc	U-235 Density gm/cc	% Theoretical Density
6	0.756	0.157	7.08	0.803	97.1
7	0.752	0.193	7.13	0.809	97.8
8	0.755	0.217	7.13	0.809	97.8
9	0.752	0.244	7.16	0.812	98.2
10	0.755	0.271	7.14	0.810	98.0
B	-	-	7.16	0.812	98.2

The metallographic samples A and B were sectioned and examined. In both alloys, the uranium was found to be homogeneously dispersed in an yttrium matrix. Photographs and photomicrographs of the irradiation samples are presented in Figures 15, 16, 17 and 18, respectively.

B. Capsule Design and Fabrication

Design of the irradiation capsule was completed during the quarter. In its final form, the capsule consists of inner and outer stainless steel cylinders separated by a gas barrier. The test specimens are located at the center of the inner cylinder immersed in sodium. Their temperature is controlled by changing the heat transfer area between the two cylinders. This is accomplished in the following manner:

1. The inner cylinder contains a gas bulb within the sodium, which experiences a change in internal pressure when the sodium temperature changes.

2. This pressure change is transmitted to a bellows which is affixed to the outer cylinder at one end and attached to the inner cylinder at the other.

3. An expansion or contraction of the bellows moves the inner cylinder in an axial direction with respect to the outer one. The two cylinders have opposing lands and grooves, and an axial displacement changes the overlap of the lands. This changes the heat transfer area and rate in such a way as to maintain a constant temperature within the inner cylinder.

A calibration capsule is being built to simulate in-pile operation. This will be used to determine the initial bellows pressure necessary to provide the desired sensitivity and range of control. Fission and gamma heating within the capsule are simulated by means of two heaters. The original heaters failed, and the capsule is now being reassembled with new heaters.

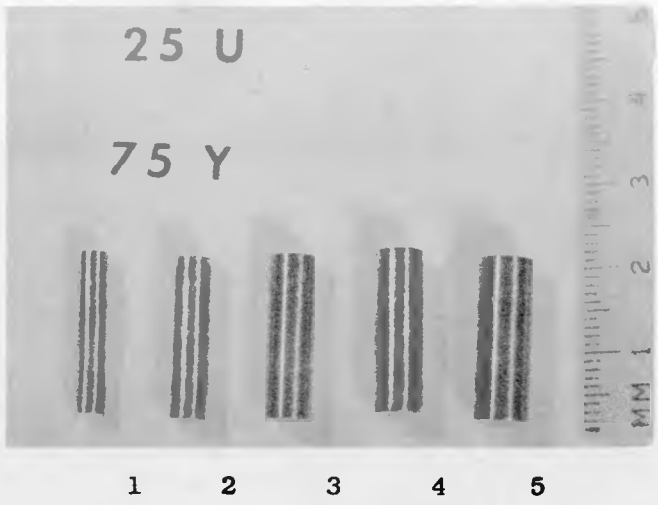

Plate No. 10713

Figure 15 - Samples of 25 w/o uranium - 75 w/o yttrium alloy ready for encapsulation. Diameters were selected to produce central temperatures of 500-900°C.

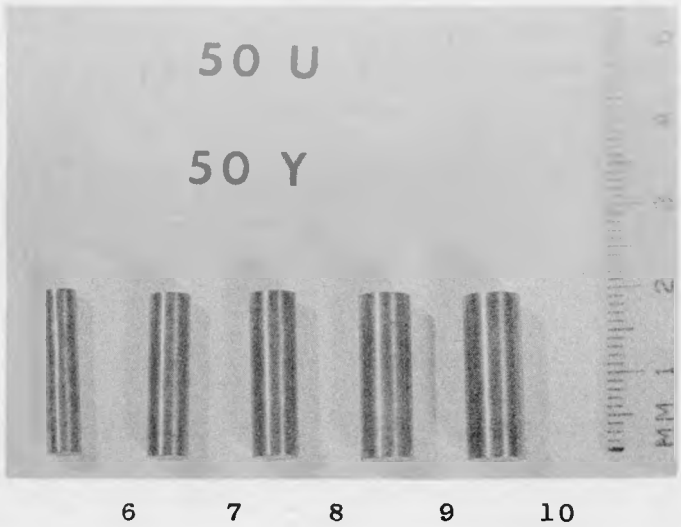

Plate No. 10712

Figure 16 - Samples of 50 w/o uranium - 50 w/o yttrium alloy ready for encapsulation. Diameters were selected to produce central temperatures of 500-900°C.

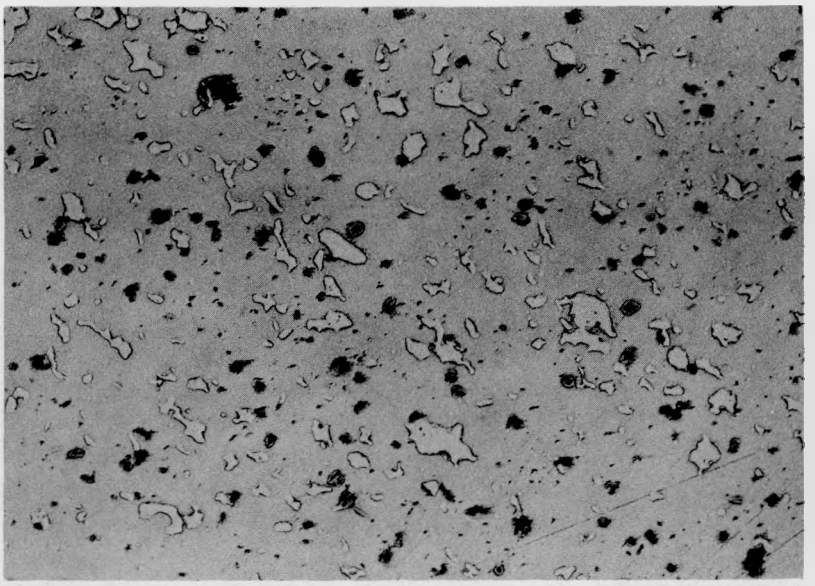

Plate No. 24004 100X

Figure 17 - Photomicrograph of 25 w/o uranium 75 w/o yttrium alloy (sample A) showing uranium islands (light) in yttrium matrix.

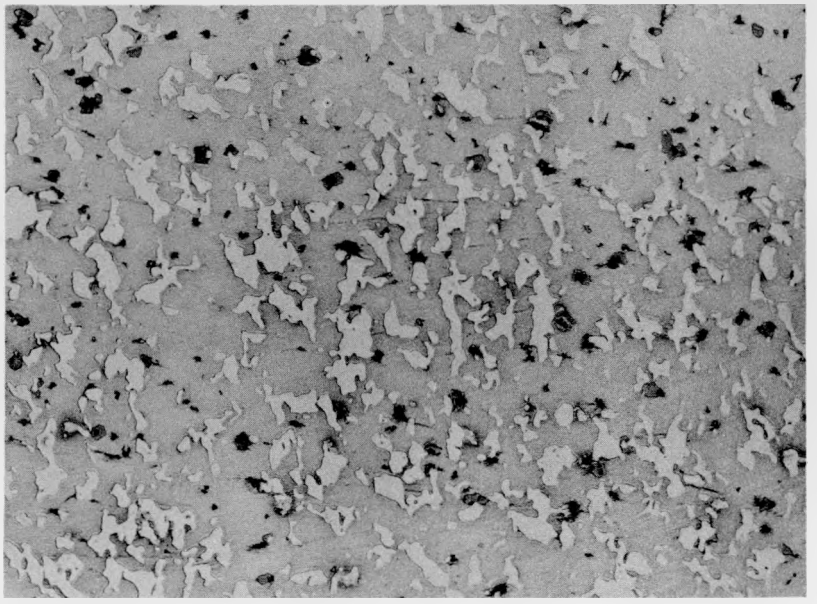

Plate No. 24003 100X

Figure 18 - Photomicrograph of 50 w/o uranium - 50 w/o yttrium alloy (sample B) showing uranium islands (light) in yttrium matrix.

Fabrication of the components for the in-pile capsules will proceed after completion of the calibration test.

C. Irradiation

The required irradiation time has been increased from 3 months to 5 months, in order to obtain the desired burnup and specimen temperatures with an acceptable heat generation rate. Analysis of the heat to be generated in the larger diameter specimens, in an effective neturon flux of 8×10^{13} n/cm^2-sec, indicated that an excessive amount of heat would be released into the reactor coolant. The effective flux has therefore been reduced to 4×10^{13} n/cm^2-sec. As a result, the irradiation time must be increased to 5 months in order to obtain a cumulative fission density of 4×10^{20} fissions per cubic centimeter of alloy. Table VI summarizes the conditions for the irradiation test.

D. Work Plan for the Next Quarter

Completion of the calibration test, construction of the capsule components, encapsulation of the samples and insertion into the reactor are scheduled during the next quarter. Actual start of the irradiation has been postponed from May to June because of the delay in assembling the calibration capsule.

A Request for Irradiation, Form AEC-320, has been filed with the AEC requesting irradiation space in either the Materials Testing Reactor or the Engineering Test Reactor. Temperature, material and weight characteristics of the capsules have been supplied to the Reactor Operator[8].

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[8] NDA Document ET-821, March 2, 1960.

TABLE VI

Uranium-Yttrium Alloy

Specimen	Burnup Fiss/cc	Material	Enrichment	ϕth, eff.	Irr.Time	Specimen Diameter	Center Temp. °C	Surface Temp. °C	Capsule No.
1	4×10^{20}	25 wt%U	55%	4×10^{13}	5 months	.147	500	-450	SR-1
2	4×10^{20}	25 wt%U	55%	4×10^{13}	5 months	.184	600	-450	SR-1
3	4×10^{20}	25 wt%U	55%	4×10^{13}	5 months	.213	700	-450	SR-1
4	4×10^{20}	25 wt%U	55%	4×10^{13}	5 months	.230	800	-600	SR-2
5	4×10^{20}	25 wt%U	55%	4×10^{13}	5 months	.254	900	-600	SR-2
6	4×10^{20}	50 wt%U	22%	4×10^{13}	5 months	.157	500	-450	SR-1
7	4×10^{20}	50 wt%U	22%	4×10^{13}	5 months	.193	600	-450	SR-1
8	4×10^{20}	50 wt%U	22%	4×10^{13}	5 months	.217	700	-600	SR-2
9	4×10^{20}	50 wt%U	22%	4×10^{13}	5 months	.244	800	-600	SR-2
10	4×10^{20}	50 wt%U	22%	4×10^{13}	5 months	.271	900	-600	SR-2