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METALLURGY AND CERAMICS (M-3679, 23rd Ed.)

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PHYSICAL AND MECHANICAL PROPERTIES OF YTTRIUM-URANIUM ALLOYS

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August 1959

Sylvania-Corning Nuclear Corporation
Bayside, New York

AEC Research and Development Report

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

Issued: August, 1959

Classification cancelled (or changed to UNCLASSIFIED)
by authority of Letter from Declassification Branch, Dated Jan. 11, 1960
by JB TIE, date 2-4-60

SYLVANIA-CORNING NUCLEAR CORPORATION
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I. SUMMARY

Since yttrium and uranium are immiscible with each other in both the solid and liquid states, an alloy of these two metals would consist of a mixture of yttrium and uranium. By means of powder metallurgy techniques, mixtures of yttrium hydride powders and uranium powders were fabricated into solid yttrium-uranium alloys which were homogeneous dispersions of metallic uranium in a matrix of yttrium. The most satisfactory fabrication procedure was to blend yttrium hydride and uranium powders, compact them at 50 tsi, and sinter at 1100°C in vacuum. Full-density alloys with uranium contents ranging from 5 w/o to 85 w/o, which contained several particle size fractions of uranium, were fabricated by this process. Metallographic examination of the various alloys indicated that a structure consisting of discrete particles of uranium in a matrix of yttrium was obtained in alloys containing up to 65 w/o of uranium. Alloys containing 85 w/o of uranium, on the other hand, consisted of discrete yttrium particles in a uranium matrix.

These alloys exhibited no dimensional changes when they were held for extended periods of time at 1000°C; they showed no reaction with sodium at 650°C. The results of elevated temperature hardness and tensile tests for various alloys indicated that they had reasonable mechanical strength up to at least 600°C. The response of the alloys to thermal cycling tests was shown to be largely dependent on composition and uranium particle size.

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II. INTRODUCTION

The work discussed in this report was performed during the period between July 1, 1958, and June 30, 1959, and was sponsored by the Reactor Development Division of the AEC.

Much attention has been given to various types of dispersions, usually of uranium compounds in a metal matrix, as potential fuel materials. Although these dispersions have generally been very stable to radiation, they have usually been deficient in several other respects: (1) low uranium content in the fuel, (2) reaction between the dispersant and the matrix during fabrication, and (3) limitation to low-temperature operation. It was demonstrated at HAPO[1] that a dispersion of uranium in magnesium could be successfully irradiated to a very high burnup, which was attributed to the immiscibility of uranium and magnesium in both the solid and liquid states. The low melting point and poor corrosion properties of magnesium were limiting factors in this system.

The selection of the yttrium-uranium alloy system was based on previous work at Sylcor which demonstrated that yttrium and uranium were also immiscible with each other in both the liquid and solid states[2] (see Figs. 1 and 2, for an arc-melted button). This immiscibility was confirmed by Ames[3] who reported a maximum solubility of 0.14% yttrium in uranium at 1225°C. Since yttrium and

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[1] M. D. Freshley and G. A. Last, "Irradiation of Mg Matrix Fuel Material to High Exposures," HW-43973.

[2] Sylvania-Corning Nuclear Corp. reports SCNC-254, 261, 265, and 279.

[3] J. F. Haefling and A. H. Doane, "The Immiscibility Limits of Uranium with Rare Earth Metals," Ames Laboratory, Contribution No. 611.

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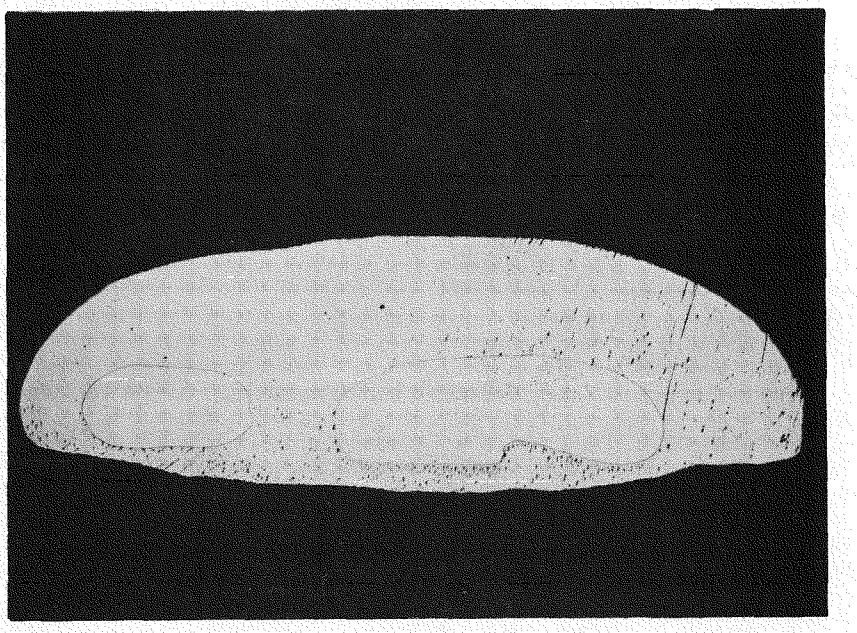

Plate No. 18784 5X

Fig. 1. Arc-melted yttrium-uranium buttons showing two islands of uranium surrounded by yttrium.

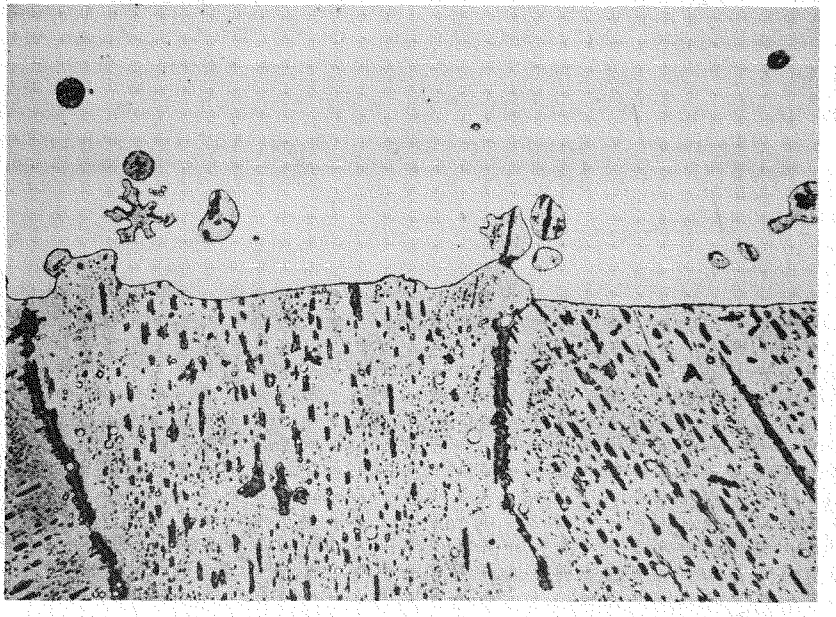

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Fig. 2. The uranium-yttrium interface in the arc-melted button shown in Figure 1; the darker areas are yttrium.

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uranium are immiscible, an alloy of these materials would actually consist of a mixture of the two metals. By the use of powder metallurgy techniques, mixtures of yttrium hydride and uranium powders were fabricated into solid yttrium-uranium alloys which were homogeneous dispersions of metallic uranium particles in a matrix of yttrium. An yttrium-uranium fuel material would have the following significant advantages:

1. A dispersion of discrete uranium particles in a continuous matrix of yttrium would have the improved dimensional stability characteristics which are associated with dispersion-type fuels, and should be capable of withstanding high burnups.

2. Since the fuel material would be metallic uranium, the dispersant would have a maximum uranium content, as compared to dispersions containing uranium compounds or uranium alloys as the dispersant.

3. Due to the immiscibility of uranium and yttrium in each other, they would not undergo any changes as a result of interaction, nor would any brittle intermetallic compounds be formed.

4. Because the components are compatible and stable at elevated temperatures, the alloy could presumably be used as a high-temperature fuel material.

5. Since the components are immiscible in the liquid state, reprocessing the fuel element after radiation would involve a comparatively simple melting operation to separate the uranium from the yttrium.

This program, therefore, had the following objectives: to conduct a study of the yttrium-uranium alloy system as a potential fuel element material, to investigate and choose the best methods of fabricating the alloys, and

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to determine some of the mechanical and physical properties of these alloys. Some pertinent properties of yttrium are listed here for reference purposes:

Melting point	- 2826°F
Boiling point	- 5840°F
Density	- 4.47 gm/cc
Crystal structure	- hexagonal, close packed
Thermal neutron cross section	- 1.38 barns/atom

III. PREPARATION OF ALLOYS

A. Preparation of Yttrium Hydride Powders

Arc-melted yttrium metal, made at the Ames Laboratory,[4] was used as a starting material for yttrium hydride powder. Analysis of this material by vacuum fusion showed the following: oxygen = 0.168%, nitrogen = 0.013%, and hydrogen = 0.0077%. An analysis for zirconium was not carried out. This material, however, usually contains 0.5-1% of zirconium which is picked up from the zirconium crucible during reduction of yttrium trifluoride. A photomicrograph of the as-received yttrium is shown in Figure 3. It has a coarse grain structure usually found in cast material.

Prior to hydriding, the yttrium ingot was cut into 1-1/2" cubes and the surfaces were cleaned with abrasive paper. Attempts to clean the surfaces with either nitric or sulfuric acid in concentrations

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[4] O. N. Carlson, F. A. Schmidt, and F. H. Spedding, "Preparation of Yttrium Trifluoride with Calcium," ISC-744.

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Fig. 3. Arc-melted yttrium as-received from Ames Laboratory; the coarse-grained cast structure should be noted.

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varying from 1% to 50% were unsatisfactory. In all cases the yttrium reacted quite violently and the surfaces became discolored.

Yttrium hydride, YH_3, was prepared by hydriding the yttrium cubes at 800°C in purified hydrogen, cooling to 300°C in hydrogen, and holding the material at this temperature until there was no further hydrogen pickup. This procedure caused the massive yttrium to disintegrate into a coarse yttrium hydride powder. In order to determine the effect of the grinding operation on the chemical analysis of the powder, one batch of this coarse hydride was ground to a -200 mesh powder in a mortar and pestle, and another batch of similar material was ground to a -200 mesh powder in a hammer mill. The data in Table I indicate that, except for slightly higher iron pickup in the hammer mill, the two batches were similar with respect to chemical analysis. All subsequent yttrium hydride was ground in a hammer mill to a -200 mesh powder, unless otherwise specified. All grinding operations were done inside a dry box in an argon atmosphere. Vacuum decomposition of the yttrium hydride powder for two hours at 800°C resulted in a material which analyzed as shown in Table I. This analysis indicates that the YH_3 was reduced to approximately YH_2.

A similar yttrium hydride powder was prepared by hydriding massive yttrium metal at 800°C and cooling it under vacuum. This also yielded yttrium hydride corresponding to approximately YH_2 without going through the procedure of decomposing the YH_3. The YH_2 which was prepared in this manner was very friable and could be readily ground to a -200 mesh powder.

B. <u>Preparation of Uranium Powders</u>

Uranium powders were also prepared by the hydride method. Uranium bars (1 inch in diameter) were pickled in 50/50 nitric acid, hydrided at 250°C with purified hydrogen, and decomposed under vacuum at 450°C. The decomposed uranium in the form of a friable sinter cake was ground

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TABLE I

ANALYSIS OF YTTRIUM HYDRIDE POWDER[a]

	Ground in mortar and pestle		Ground in hammer mill		Partially decomposed
	+200 Mesh	-200 Mesh	+200 Mesh	-200 Mesh	yttrium hydride (800°C)
Hydrogen, %	2.99	3.07	3.01	3.03	1.94
Iron, %	0.006	0.008	0.008	0.013	0.014
Nitrogen, %	0.012	0.013	0.012	0.012	0.014

[a] The theoretical hydrogen content of YH_3 is 3.29%; that of YH_2, 2.22%.

in a micropulverizer and screened to the desired mesh fraction. Since the uranium powder was pyrophoric, it was always handled in an argon atmosphere. A 3% molybdenum-uranium alloy powder was prepared by the same process, using the uranium alloy as a starting material.

C. Mixing of Powders

Due to the differences in particle size and density between the yttrium and uranium powders, some difficulty was encountered in obtaining homogeneous mixtures of the two powders. Since the heavy uranium particles tended to segregate in the powder mixtures, the alloys contained stringers of uranium particles rather than discrete particles surrounded by yttrium. The following procedure was used to make a homogeneous powder mixture:

1. Weighed quantities of YH_2 and uranium powders were mixed for fifteen minutes in a twin-cone blender.

2. A solution of camphor in benzine was added to the mixed powders; the mixture was stirred, and the benzine was removed by evaporation in a vacuum chamber.

3. The mixture was tumbled for thirty minutes in a twin-cone blender, passed through a 100-mesh screen to break up any lumps, and then mixed for another thirty minutes.

D. Hot Pressing

Powder mixtures containing 65 w/o of uranium and 35 w/o of YH_3 were hot pressed under vacuum, in graphite dies, at temperatures ranging from 1100° to 1300°C, for periods up to thirty minutes, and at a pressure of 4 tsi. Hot pressing at temperatures above the melting point of uranium (1133°C) resulted in considerable loss of material during the operation, apparently due to extrusion of the molten uranium between the punch and the die, and into pores in the graphite die. Even when a dense grade of

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graphite was used, and a snug fit between the punch and the die was insured, some material loss and a wide density variation in the resulting material occurred. Hot pressing at 1100°C for fifteen minutes (below the melting point of uranium) eliminated this loss of material and resulted in densities of 7.5-7.6 gm/cc (86% of theoretical). All hot-pressed compacts reacted with the die to some extent; there was considerable scoring of the die on ejection. The reaction of the compact with the graphite die was more severe at higher hot pressing temperatures. The chemical analysis of hot-pressed material is given in Table II. The data indicate that hot pressing under vacuum for fifteen minutes at temperatures up to 1200°C does not completely decompose the yttrium hydride.

TABLE II

ANALYSIS OF 35 w/o YH_3-65 w/o URANIUM AFTER HOT PRESSING UNDER VACUUM

	Hot-pressing conditions		Analysis, %		
Sample	Time, min	Temp., °C	H_2	O_2	N_2
HP-3	15	1200	0.5	0.15	0.019
HP-4	15	1125	0.28	0.12	0.022

Since the results obtained with hot pressing were not as favorable as those obtained with cold pressing and sintering under vacuum, the use of the hot-pressing method was discontinued. The higher hydrogen values obtained at 1200°C were probably due to differences in the heating cycle.

E. Cold Pressing and Vacuum Sintering

The most satisfactory procedure for making yttrium-uranium alloys consisted in: (1) cold compacting the mixed YH_2-uranium powders, and (2) sintering the green compacts

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under vacuum. The sintered material was fully dense and had minimum contamination. Figure 4 shows the relationship among compacting pressure, green density, and green strength for a 65 w/o uranium-35 w/o yttrium powder mixture. A -200 mesh yttrium powder and -270 +325 mesh uranium were used. The graph indicates that the green density and green strength increased quite rapidly with pressures up to 40 tsi. Above 40 tsi, the density increased at a much slower rate. Since compacting at pressures above 50 tsi did not result in any improvement in the sintered material, pressures of 25 and 50 tsi were, therefore, used to prepare material for this program.

A series of compacts was pressed and sintered in order to compare material made with YH_2 and with YH_3. Powder mixtures consisting of 35 w/o of yttrium hydride (200 mesh), and 65 w/o of uranium (-35 +140 mesh) were compacted at 50 tsi into 7/16" diam. x 1/2" long pellets. The pellets were sintered under vacuum for two hours at 1200°C: the pellets made with YH_3 were severely cracked after sintering, whereas those made with YH_2 did not show any cracking. The formation of cracks in the YH_3 material was probably due to the rapid evolution of hydrogen at low temperatures, and also to the volume change. Metallic yttrium has a density of 4.47 gm/cc, YH_3 3.96 gm/cc, and YH_2 4.29 gm/cc. The volume change from YH_3 to metallic yttrium is quite large as compared to the change from YH_2, and is more likely to result in damage to the compact. Since YH_3 starts to decompose at a lower temperature than YH_2, hydrogen is released before sintering has started and while the compact is comparatively weak. All subsequent yttrium hydride powder for use in sintered material was used as YH_2.

At sintering temperatures close to or above the melting point of uranium (1133°C), uranium bleeds to the surface and forms small pools on the surface of the compact. This is apparently due to shrinkage of the compact during sintering, which forces out the molten uranium. The results obtained on sintering a 65 w/o uranium-yttrium alloy, prepared from -40 +140 mesh uranium powder and -200 mesh yttrium hydride powder, are shown in Table III.

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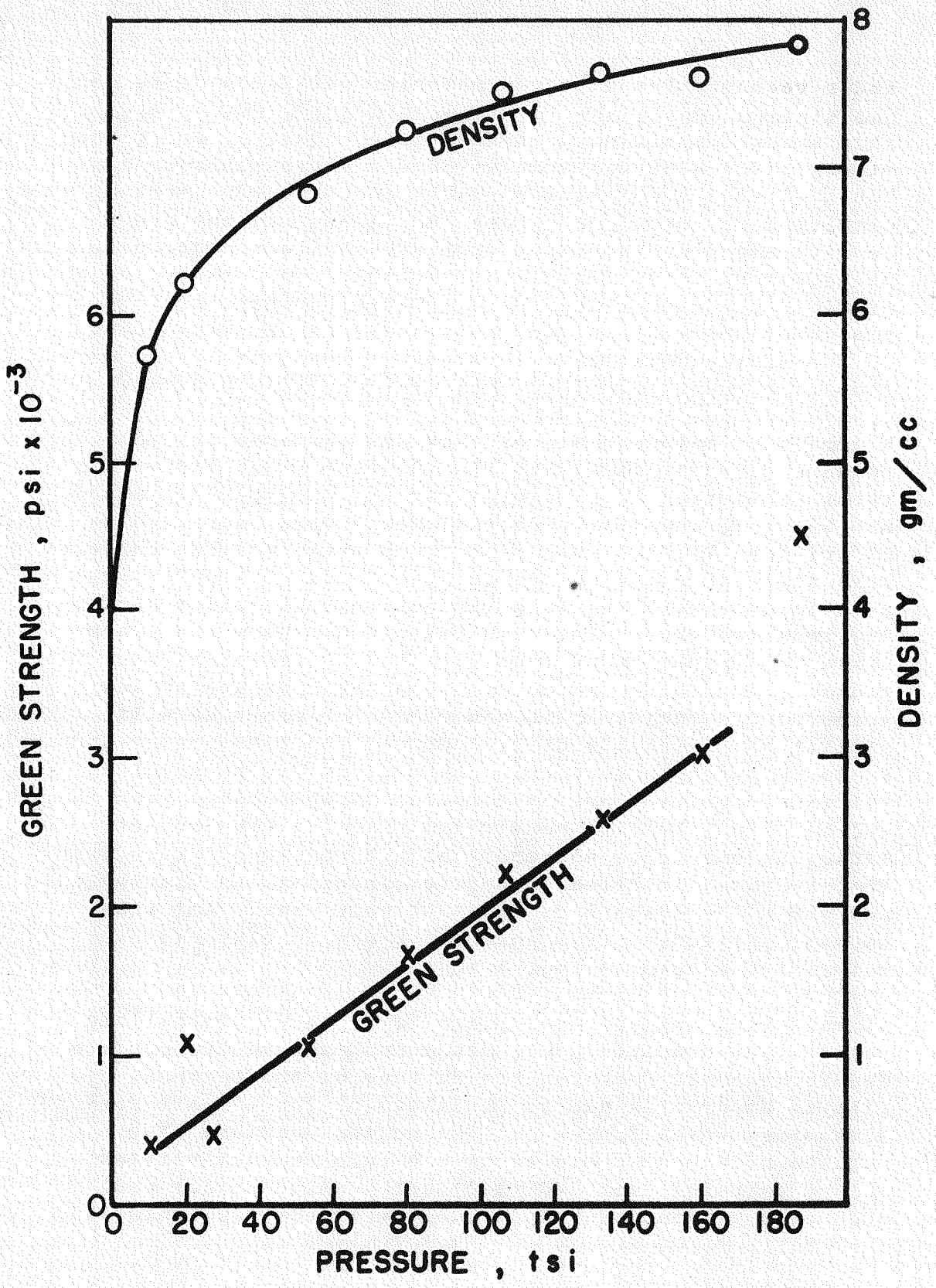

Fig. 4. Compacting pressure versus green strength and density for powder mixture of 65 U-35 Y.

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TABLE III

EFFECT OF COMPACTING PRESSURE AND SINTERING CONDITIONS ON DENSITY AND SURFACE OF A 65 w/o URANIUM-YTTRIUM ALLOY

Sample	Compacting pressure, tsi	Sintering conditions Time, hr	Sintering conditions Temp., °C	Density,[a] gm/cc	Surface appearance
5	25	2	1125	8.3	Clean
5	25	Resintered 1 hr	1200	8.8	U on surface
9	25	4	1125	8.6	Clean
10	25	1/2	1200	8.2	Clean
13	25	2	1200	8.5	Clean
14	25	2	1250	8.7	Clean
18	25	2-1/2	1250	8.8	U on surface
1	50	1/2	1200	8.6	U on surface
2	50	2	1200	8.7	U on surface
7	50	2	1125	8.6	Clean
15	50	6	1125	8.7	Clean

[a] Calculated density, 8.8 gm/cc.

The data in Table III indicate that the 65 w/o uranium-yttrium composition with the specified particle sizes sinters readily at temperature of 1125°C and higher, and that the optimum sintering conditions vary with the density of the green compact. The uranium apparently bleeds to the surface only at temperatures above the melting point of uranium, and then only as the compact approaches full density. Compacts formed at 50 tsi show uranium on the surface after a shorter sintering time, for a given sintering temperature, than compacts pressed at 25 tsi. This confirms the observation that molten uranium is forced to the surface by shrinkage of the compact.

Another difficulty encountered in sintering at temperatures above the melting point of uranium is due to the tendency of the particles of uranium to flow together and agglomerate into larger particles, as illustrated in Figures 5 and 6. This alloy consisted of 65 w/o uranium-35 w/o yttrium, -140 +270 mesh uranium and -200 mesh yttrium. The material shown in Figure 5 was sintered for two hours at 1250°C, under which conditions the molten uranium particles flowed together and formed large pools. The uranium particles in the material shown in Figure 6, which had been sintered for ten hours at 1100°C, retained their original size.

Since sintering at temperatures above the melting point of uranium resulted in an unsatisfactory material, the sintering cycle used for the balance of this program was standardized at twelve hours at 1100°C unless specified otherwise.

IV. EVALUATION OF ALLOYS

A. Effect of Composition and Particle Size on Density and Chemistry of Alloys

A series of alloys with uranium contents ranging from 5 w/o to 85 w/o was prepared to determine the effects of composition and particle size on the density and

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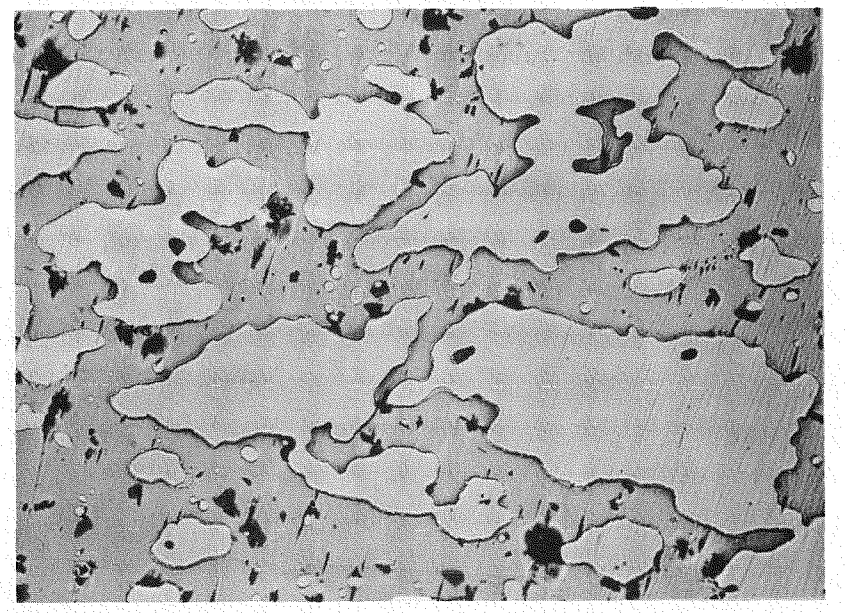

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Fig. 5. Alloy containing 35Y-65U (-140 +270 mesh uranium particles) sintered above the melting point of uranium; the uranium particles have flowed together and agglomerated into large particles.

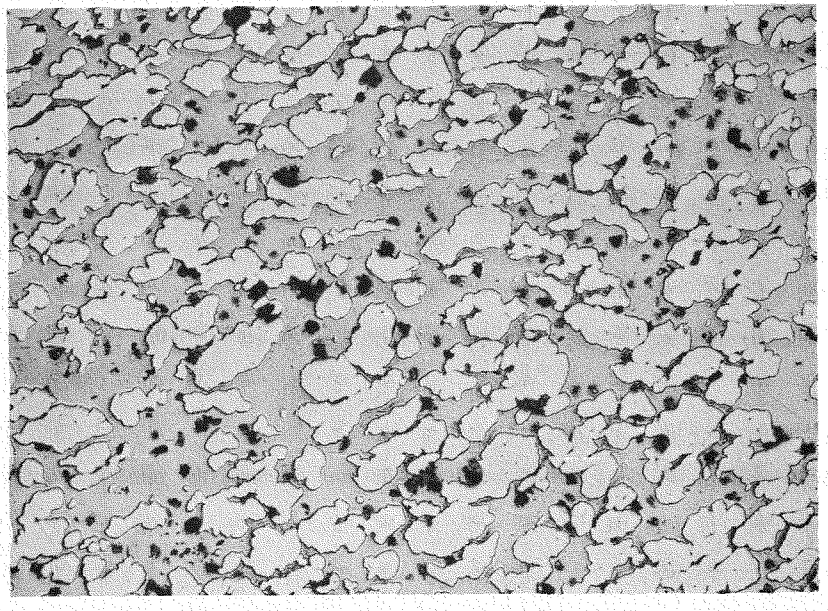

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Fig. 6. Same alloy shown in Figure 5 except that the material was sintered below the melting point of uranium; the uranium particles have retained their original size. The lighter areas are uranium.

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chemistry of the resulting alloys. Four particle size fractions of uranium powder were used: -40 +140, -140 +270, -270 +325, and -325 mesh. Three YH_2 powders with different sizes (as measured on the Fisher sub-sieve sizer were used: 30, 11.5, and 5.6 microns. The mixed powders were pressed at 50 tsi and sintered under vacuum for 12 hours at 1100°C. The results are summarized in Table IV.

The following conclusions can be drawn from the data in Table IV:

1. Sintering under vacuum for twelve hours at 1100°C reduced the hydrogen content to less than 50 ppm.

2. The uranium particle size did not have any significant effect on the density of the sintered compact. In all cases, essentially full density was obtained.

3. Although some oxygen in the sintered alloy was due to the presence of oxygen in the starting yttrium powder (0.17%), some oxygen was also picked up during preparation.

4. The particle sizes of the uranium and yttrium powders and the composition of the alloys had no significant effect on the nitrogen and hydrogen contents of the sintered alloys. The high yttrium alloys and the finer yttrium powders had higher oxygen contents.

B. Metallography

The effects of uranium content and uranium particle size on the microstructures of the alloys are shown in Figures 6-9. Alloys containing 25 w/o of uranium (Figs. 8 and 9) consist of discrete uranium particles dispersed in an yttrium matrix. Increasing the uranium content to 65 w/o results in an alloy in which some of the uranium particles have sintered together; the structure, however, still consists of uranium particles in an yttrium matrix (Fig. 6). When the uranium content is increased to 85 w/o, the

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TABLE IV

EFFECT OF COMPOSITION AND PARTICLE SIZE ON DENSITY AND CHEMICAL ANALYSIS OF YTTRIUM-URANIUM ALLOYS

Sample	Composition, w/o	Uranium particle size, mesh	Yttrium particle size, μ	Density, gm/cc	Chemical analysis, % O_2	N_2	H_2
58	5U/95Y	-40 +140	30	4.45	0.248	0.006	0.0026
57	5U/95Y	-140 +270	30	4.44	0.242	0.012	0.0034
59	5U/95Y	-270 +325	30	4.43	0.273	0.012	0.0029
30A	25U/75Y	-40 +140	30	5.25	0.294	0.027	-
31A	25U/75Y	-140 +270	30	5.24	0.282	0.021	-
32A	25U/75Y	-270 +325	30	5.33	0.196	0.022	-
46	50U/50Y	-140 +270	30	6.9	0.073	0.010	0.0022
47	50U/50Y	-325	30	6.1	0.085	0.011	0.0024
19	65U/35Y	-40 +140	30	8.9	0.165	0.023	0.0047
20	65U/35Y	-140 +270	30	8.9	-	-	-
24	65U/35Y	-270 +325	30	8.3	-	-	-
33	85U/15Y	-40 +140	30	11.3	0.175	0.018	-
34	85U/15Y	-140 +270	30	12.1	0.067	0.018	-
35	85U/15Y	-270 +325	30	12.1	0.235	0.018	-
67	65U/35Y	-325	11.5	8.6	0.275	0.011	0.0018
68	65U/35Y	-140 +270	11.5	8.77	0.204	0.010	0.0014
84	65U/35Y	-140 +270	5.6	-	0.173	0.013	0.0031
85	65U/35Y	-325	5.6	-	0.210	0.017	0.0027

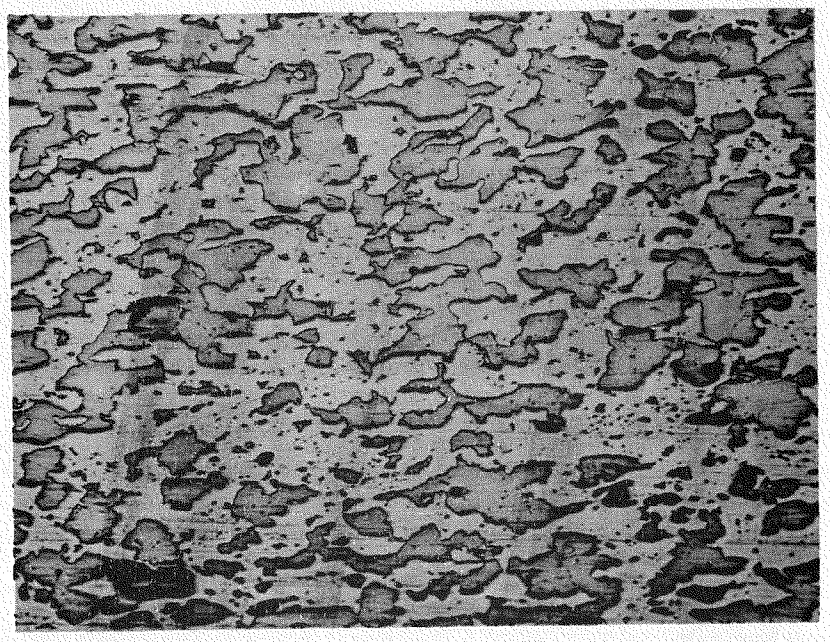

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Fig. 7. Alloy containing 85U-15Y. The darker yttrium particles are dispersed in a matrix of uranium.

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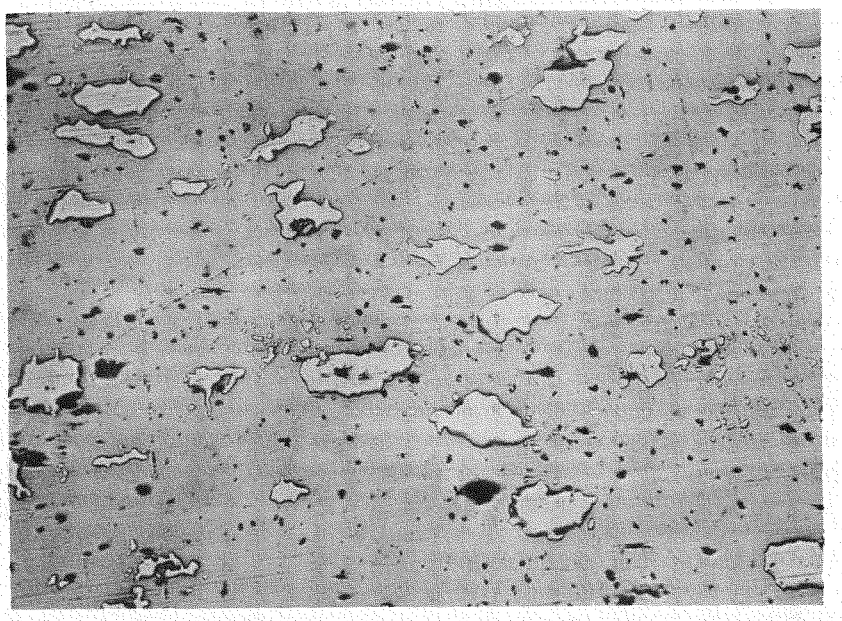

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Fig. 8. Alloy containing 75Y-25U, with -140 +270 mesh uranium particles. The coarse particles of uranium are spread comparatively far apart.

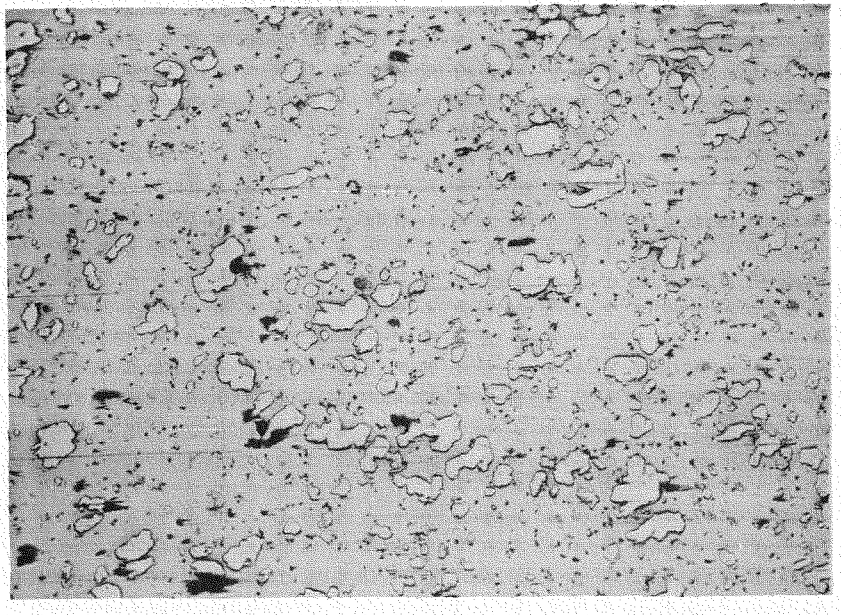

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Fig. 9. Alloy with same composition as that shown in Figure 8 except for finer uranium particles, -270 +325 mesh. Finer particles are closer together than coarser particles.

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structure is reversed, resulting in a dispersion of yttrium particles in a uranium matrix (Fig. 7). Comparison of the structures shown in Figures 8 and 9 indicates that the use of a coarser uranium particle size results in fewer discrete particles which are, on the average, farther apart.

Metallographic examination of alloys made from fine yttrium powders did not indicate any differences in structure in comparison with those made from coarse yttrium powders. After 30% reduction by cold rolling, no significant amount of stringering or other structural change was evident.

C. Thermal Stability

Table V shows the results of thermal stability tests on the 65 w/o U-35 w/o Y alloy. These tests were carried out to determine whether any dimensional changes occurred in the alloy after it was held at elevated temperatures for extended periods of time. The test samples were approximately 3/8" in diameter x 3/8" long, and were originally sintered at temperatures ranging from 1100° to 1250°C. During the test they were held at 1000°C for fifty hours. There was no significant change in appearance or dimensions of the tested pieces, except for very slight shrinkage in some cases. The shrinkage was probably due to some residual porosity in these particular samples. These results confirmed the lack of any significant degree of reaction between the yttrium and uranium.

D. Hot Hardness

The effects of composition and particle size on the Vickers hardness values of various alloys at room and elevated temperatures are shown in Table VI. The room temperature hardness values were obtained with a diamond pyramid indenter and a 10-kg load, and the elevated temperature values, with a sapphire indenter and a 1-kg load. Although some doubt exists as to the significance of hardness values of a dispersion-type material, the following conclusions can be drawn from these data:

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TABLE V

THERMAL STABILITY OF 65 w/o URANIUM-35 w/o YTTRIUM HELD FOR 50 HOURS AT 1000°C[a]

Sample No.	Uranium particle size, mesh	Original sintering conditions		Sintered density, gm/cc	Dimensional change after sintering	
		Time, hr	Temp., °C		Diam., in.	Length, in.
20-1	-140 +270	2	1250	8.96	-0.005	0
23-2	-140 +270	2	1250	9.10	0	-0.001
24-6	-270 +325	2	1250	8.40	0	0
26-5	-140 +270	10	1100	8.68	0	0
27-4	-40 +140	10	1100	8.65	-0.001	0

[a] Sample size approximately 3/8" in diameter x 3/8" long.

TABLE VI

VICKERS HARDNESS VALUES OF YTTRIUM-URANIUM ALLOYS

Sample No.	Composition, w/o	Uranium particle size, mesh	Yttrium particle size, μ	Density, gm/cc	Vickers hardness values R.T.	300°C	600°C
58	5U/95Y	-40 +140	30	4.45	50	69	20
57	5U/95Y	-140 +270	30	4.44	48	65	21
59	5U/95Y	-270 +325	30	4.43	52	70	25
				Av:	50	68	22
30A	25U/75Y	-40 +140	30	5.25	62	56	34
31A	25U/75Y	-140 +270	30	5.24	65	-	20
32A	25U/75Y	-270 +325	30	5.33	72	59	31
				Av:	66	58	28
46	50U/50Y	-140 +270	30	6.9	73	82	24
47	50U/50Y	-325	30	6.1	78	80	23
				Av:	76	81	24
19	65U/35Y	-40 +140	30	8.9	105	97	25
20	65U/35Y	-140 +270	30	8.9	102	89	30
24	65U/35Y	-270 +325	30	8.3	91	73	26
				Av:	99	86	27
33B	85U/15Y	-40 +140	30	11.3	111	59	25
34A	85U/15Y	-140 +270	30	12.1	130	79	24
35A	85U/15Y	-270 +325	30	12.1	140	104	30
				Av:	127	81	26
67	65U/35Y	-325	11.5	-	94	83	26
68	65U/35Y	-140 +270	11.5	-	83	87	25
84	65U/35Y	-140 +270	5.6	-	91	96	28
85	65U/35Y	-325	5.6	-	95	103	25
				Av:	91	91	26
S-1	Wrought 100% U	-	-		308	128	16

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1. An increase in the uranium content of the alloy increased its room temperature hardness.

2. Tests at 300°C indicated a hardness increase in some of the lower uranium-containing specimens.

3. The hardness at 600°C did not appear to be affected significantly by composition.

4. The use of fine uranium particles resulted (for most of the test series) in an increase in the alloy hardness.

5. Whereas wrought uranium has a considerably higher hardness than any alloy at room temperature, this is no longer true at 600°C. At the higher temperature, the alloys have greater hardness than wrought uranium.

6. In general, it can be stated that all alloys are sufficiently hard to be used in the 300°-600°C temperature range.

E. Tensile Properties

The effects of composition and particle size on the tensile properties of yttrium-uranium alloys at both room and elevated temperatures are indicated in Table VII. The following conclusions can be drawn from these results:

1. Uranium particle size had little effect on the tensile properties of the alloys in the range from 300° to 600°C.

2. A decrease in the size of the yttrium particles from an average of 30μ to an average of 5.6μ increased the tensile strength, and especially the elongation, at elevated temperatures.

3. Variations in ductility were probably due to residual porosity in the sintered material.

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TABLE VII

TENSILE PROPERTIES OF YTTRIUM-URANIUM ALLOYS

Composition, w/o	Uranium particle size, mesh	Yttrium particle size, μ	Tensile properties at 300°C Y.S., psi	T.S., psi	Elong., %	Tensile properties at 600°C Y.S., psi	T.S., psi	Elong., %
5U/95Y	-40 +140	30	13,900	16,300	6	5,140	5,950	12
5U/95Y	-140 +270	30	12,800	17,100	11	5,780	6,410	25
5U/95Y	-270 +325	30	13,100	14,800	3	5,050	7,010	17
5U/95Y	-140 +270	5.6	20,200	23,800	18	7,150	8,400	29
5U/95Y	-325	5.6	19,400	24,900	18	6,780	7,920	43
25U/75Y	-40 +140	30	13,800	18,500	8	6,750	7,850	14
25U/75Y	-140 +270	30	12,400	17,100	7	6,950	7,800	11
25U/75Y	-270 +325	30	15,500	19,400	9	7,800	8,300	14
50U/50Y	-140 +270	30	22,600	28,400	2	8,000	8,900	26
50U/50Y	-325	30	17,100	20,600	6	8,000	8,600	9
65U/35Y	-40 +140	30	20,000	21,600	2	8,300	9,000	5
65U/35Y	-140 +270	30	-	-	-	-	7,600	3
65U/35Y	-140 +270	11.5	20,500	24,600	7	7,020	8,600	15
65U/35Y	-325	11.5	20,400	25,200	15	6,840	7,700	29
85U/15Y	-140 +270	30	19,000	23,600	5	8,300	9,200	12

F. Thermal Expansion

A plot of elongation versus temperature for several yttrium-uranium alloys is shown in Figure 10. The tests were performed in a quartz dilatometer under vacuum. The results indicated that the coefficient of thermal expansion increased with increasing uranium content, but that the uranium particle size had no apparent effect on the coefficient. Table VIII lists thermal expansion values over various temperature ranges.

TABLE VIII

COEFFICIENT OF THERMAL EXPANSION[a]

Composition, w/o	Temperature range, °C			
	25-100	25-300	25-600	25-700
5U/95Y	-	6.6	8.6	9.1
25U/75Y	11.0	11.0	11.0	11.0
50U/50Y	11.5	11.6	12.6	13.2
65U/35Y	11.5	14.2	17.0	17.3

[a] All values x 10^{-6} per °C.

G. Corrosion in Sodium

A series of corrosion tests was carried out to determine the compatibility of the alloys with sodium at elevated temperatures. The samples (3/8" in diameter x 1/2" long) were immersed in sodium, sealed in stainless steel capsules, and held at 650°C for 300 hours. The results, which are shown in Table IX, indicate that there is no significant degree of reaction between any of the alloys and sodium at 650°C.

H. Thermal Cycling

All thermal cycling samples were sealed under vacuum in Vycor capsules and cycled 500 times, at four cycles per

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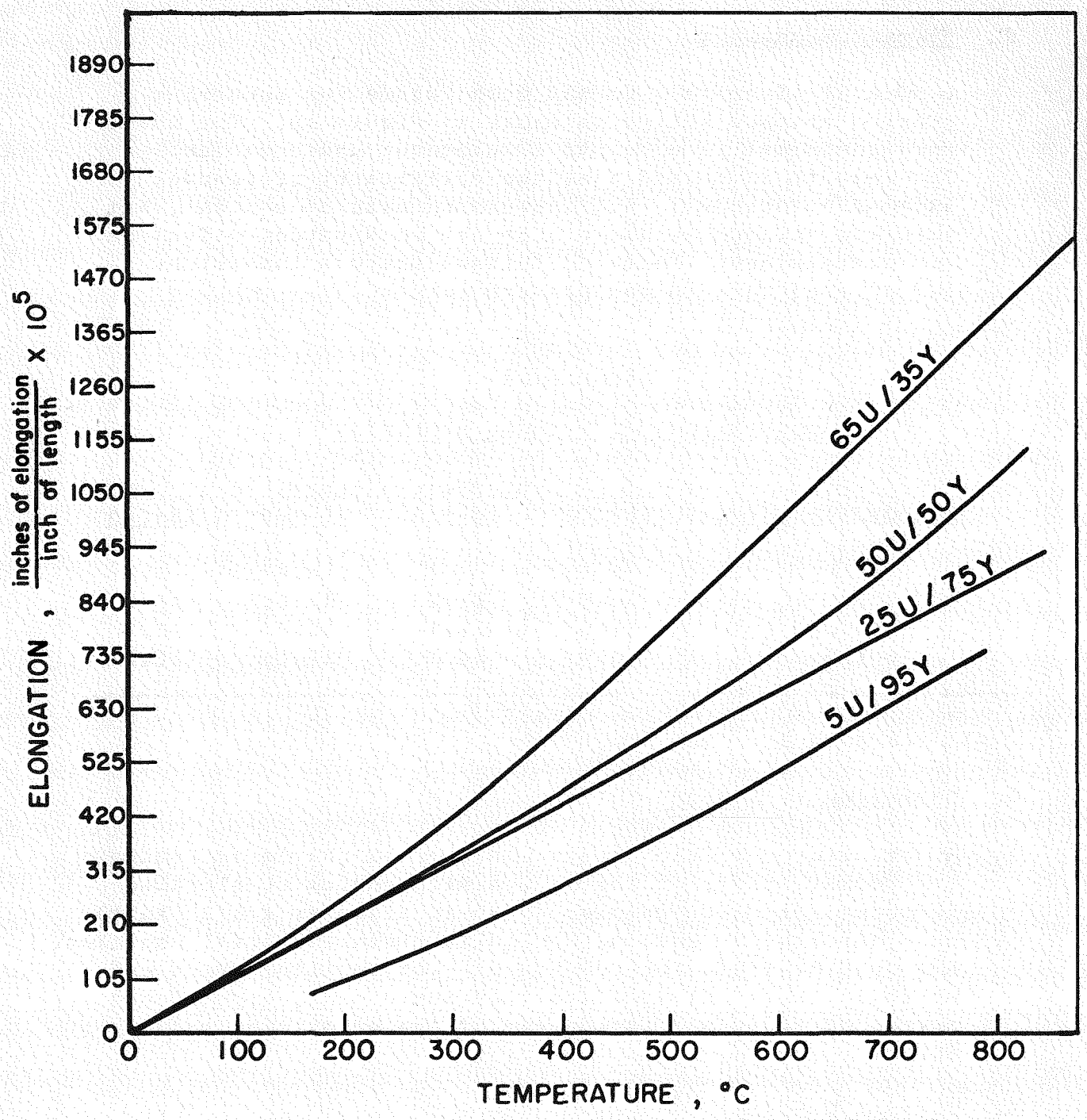

Fig. 10. Elongation versus temperature.

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TABLE IX

STABILITY OF YTTRIUM-URANIUM ALLOYS IN SODIUM[a]

Composition, w/o	Uranium particle size, mesh	Yttrium particle size, μ	Sintering temp., °C	Dimensional change, in. Diam.	Dimensional change, in. Length
5U/95Y	-40 +140	30	1100	0.002	0.0025
5U/95Y	-140 +270	30	1100	0.0004	0.0008
5U/95Y	-270 +325	30	1100	0.0005	0.0002
5U/95Y	-40 +140	30	1200	0.0014	0.0028
5U/95Y	-140 +270	30	1200	0.0026	0.0039
5U/95Y	-270 +325	30	1200	0.0009	0.0007
50U/50Y	-40 +140	30	1100	0	0
50U/50Y	-270 +325	30	1100	0	0
65U/35Y	-40 +140	30	1100	0	0.001
65U/35Y	-40 +140	30	1100	0	0
65U/35Y	-270 +325	30	1250	0.002	0
65U/35Y	-270 +325	30	1250	0	0
65U/35Y	-140 +270	11.5	1100	0.002	0.0017
65U/35Y	-325	11.5	1100	0.0011	0.0018
65U/35Y	-140 +270	5.6	1100	0.001	0.0016
65U/35Y	-325	5.6	1100	0.0007	0.0006

[a] Samples held at 650°C for 300 hours.

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hour, between the limiting temperatures. The tests were performed in the following ways: (1) within the alpha-temperature range, in which case, the samples were cycled between 550° and 150°C; or (2) across the beta-transformation temperature, in which case, the limiting temperatures were 725° and 225°C.

After being cycled 500 times, the samples were checked to determine dimensional changes and the degree of surface roughening. The surface condition of the samples after thermal cycling was measured with a profilometer. This instrument measures surface variations in RMS microinches and has been very satisfactory for obtaining a quantitative measurement of surface roughening. Most of the thermal cycling data have indicated that changes in the alloy are more likely to be reflected in surface roughening than in distortion.

Table X shows the results of some thermal cycling tests which were performed early in the program. Most of these samples had been sintered at temperatures above the melting point of uranium; this was subsequently found to result in a nonhomogeneous material. In this series, samples of the 65U/35Y alloy, approximately 3/8" in diameter x 1/2" long, were cycled through the beta-transformation temperature. These alloy samples showed considerably less distortion than pure uranium, but somewhat more distortion than wrought 3% molybdenum-uranium alloy, under similar cycling. The surfaces of the cycled samples were quite rough, which accounted for a large part of the distortion in these small samples. The data in Table X indicate that sintering at temperatures below the melting point of uranium results in less distortion and better surfaces.

The results of thermal cycling of larger samples, approximately 3/8" in diameter x 2" long, are shown in Tables XI and XII. All the alloys were sintered for twelve hours at 1100°C. The material described by the data in Table XI was alpha cycled; the effects of composition and uranium particle size on distortion and surface roughening

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TABLE X

THERMAL CYCLING DATA FOR 65 w/o U-35 w/o Y ALLOYS[a]

Sample No.	Uranium particle size, mesh	Sintering conditions Temp., °C	Time, hr	Sintered density, gm/cc	Distortion, % Diam.	Length	Surface, RMS micro-inches
18-1	-40 +140	1250	2	8.9	+3.2	-	300
18-3	-40 +140	1250	2	8.7	+6.6	+2.0	275
19-4	-40 +140	1250	2	8.9	-2.5	+0.8	350
19-6	-40 +140	1250	2	8.5	+5.6	+3.7	300
20-3	-140 +270	1250	2	8.7	Capsule broke		
20-4	-140 +270	1250	2	8.6	Capsule broke		
21-6	-140 +270	1250	2	8.1	-1.5	+8.0	175
21-5	-140 +270	1250	2	9.1	+5.5	-1.0	175
23-9	-140 +270	1250	2	9.1	+5.3	-2.6	275
23-10	-140 +270	1250	2	9.2	+5.6	-2.3	225
23-12	-140 +270	1250	2	9.2	0	-3.3	225
24-1	-270 +325	1250	1-1/2	7.7	+4.8	+2.6	225
24-3	-270 +325	1250	1-1/2	7.8	+2.9	+3.4	200
24-5	-270 +325	1250	1-1/2	8.4	+5.5	-	250
26-3	-140 +270	1100	10	8.94	+2.9	+0.5	150
26-4	-140 +270	1100	10	8.84	+3.0	+2.1	190
26-6	-140 +270	1100	10	8.94	+2.9	-1.1	140
S-1	Wrought 100% uranium				Disintegrated		
S-2	Wrought 100% uranium				Disintegrated		
S-3	Wrought 100% uranium				Disintegrated		
S-4	Wrought 3% Mo-U Alloy				-0.8	+2.6	
S-5	Wrought 3% Mo-U Alloy				-1.0	+1.7	

[a] 500 cycles, 725°-200°C.

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TABLE XI

THERMAL CYCLING DATA[a]

Sample No.	Composition, w/o	Uranium particle size, mesh	Yttrium particle size, μ	Density, gm/cc	Distortion, % Diam.	Distortion, % Length	Surface, RMS micro-inches
30A	25U/75Y	-40 +140	30	5.2	0	0	35
31B	25U/75Y	-140 +270	30	5.2	0	0	32
32B	25U/75Y	-270 +325	30	5.2	0	0	30
46-1	50U/50Y	-140 +270	30	7.07	0	0	35
47-1	50U/50Y	-325	30	6.7	0	0	30
44-1	65U/35Y[b]	-140 +270	30	8.45	0	0	40
44-2	65U/35Y	-140 +270	30	8.44	0	0	37
45-1	65U/35Y	-325	30	8.58	0	0	40
48-2	62U/3Mo/35Y	-140 +270	30	8.52	0	0	43
48-3	62U/3Mo/35Y	-325	30	8.56	0	0	40
33A	65U/35Y[b]	-40 +140	30	11.3	2.8	2.6	70 distorted
34B	65U/35Y	-140 +270	30	12.1	4.4	2.6	Distorted

[a] 500 cycles, 550°-150°C.

[b] Sample beta quenched.

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TABLE XII

THERMAL CYCLING DATA[a]

Sample No.	Composition, w/o	Uranium particle size, mesh	Yttrium particle size, μ	Density, gm/cc	Distortion, % Diam.	Distortion, % Length	Surface, RMS micro-inches
63	5U/95Y	-40 +140	30	-	0.8	1.0	Pimples 35
64	5U/95Y	-140 +270	30	-	0.6	0.7	50
65	5U/95Y	-270 +325	30	-	0.6	0.7	60
30B	25U/75Y	-40 +140	30	-	1.3	1.2	150
31A	25U/75Y	-140 +270	30	-	0.6	0.8	130
32A	25U/75Y	-270 +325	30	-	0.9	1.1	55
46-2	50U/50Y	-140 +270	30	6.94	2.0	2	190
47-2	50U/50Y	-325	30	6.03	0	0.5	110
44-2	65U/35Y	-140 +270	30	8.43	6	4	Ripples 220
45-2	65U/35Y	-325	30	8.54	4	4	Ripples 210
48-1	62U/3Mo/35Y	-140 +270	30	8.58	4.0	-3.0	200
49-2	62U/3Mo/35Y	-325	30	8.62	2.5	1.5	130
74	65U/35Y	-140 +270	11.5	-	6.4	2.3	275
77	65U/35Y	-325	11.5	-	4.9	3.0	225
86	65U/35Y	-140 +270	5.6	-	7.3	0.8	250
87	65U/35Y	-325	5.6	-	5.0	1.8	175

[a] 500 cycles, 725°-225°C.

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are indicated. The material described in Table XII was beta cycled; the data shown the effects of composition and uranium and yttrium particle sizes. The following conclusions can be drawn from these results:

1. Reduction in the particle size of the uranium reduced surface roughening and distortion due to thermal cycling in all alloys up to and including 65U/35Y. This effect was not noticeable in the 85U/15Y alloy in which the uranium particles were interconnected. In the 5U/95Y alloy, the only surface roughening occurred when comparatively coarse -40 +140 mesh uranium powder was used. This roughening was noted in the form of small pimples which apparently formed where individual particles of uranium were present on the surface. Roughening did not occur, however, when the same alloy was prepared with finer uranium. The use of coarse powder in alloys with higher uranium contents resulted in an over-all coarse surface rather than individual pimples.

2. Alloys containing up to and including 65 w/o of uranium showed essentially no distortion or surface roughening when thermal cycled in the alpha-temperature range. Alloys containing 85 w/o of uranium were distorted by alpha cycling. Metallographic examination indicated that the uranium particles in this alloy were interconnected, which probably accounts for most of this distortion.

3. Additions of 3% molybdenum to the 65U/35Y alloy resulted in reduced distortion and improved surface after cycling through the transformation temperature.

4. Reduction in the particle size of yttrium from 30μ (-200 mesh) to 11.5μ and 5.6μ resulted in improved material, mostly by the elimination of blister formation during thermal cycling.

Further thermal cycling data which indicate the effects of using a fine yttrium powder and a spherical uranium powder

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are given in Table XIII. The yttrium powder was ground in a pulverizing mill to an average size of 5.6μ. The spherical uranium powder, with densified particles, was prepared by repeatedly passing the powder through a pulverizing mill until the apparent density of the powder remained constant. By this method the apparent density was raised from 4 gm/cc to 9 gm/cc. Microscopic examination indicated that the larger particles were spheroidized, but that the finer particles in the range used for making the alloys were not spheroidized. The data in Table XIII indicate that reduction of the yttrium particle size to 5.6μ improved alloy behavior during thermal cycling; this was probably due to the increased strength of this material. Spheroidizing the uranium powder in the above manner did not result in any apparent improvement.

I. Cold Rolling and Coining

All the alloys could be cold rolled to total reductions of 25% without difficulty. After being annealed at 1000°C, the material returned to its original sintered hardness. These operations did not have any significant effect on mechanical properties or on response to thermal cycling.

65U/35Y alloys were compacted at 50 tsi and sintered for fifteen hours at 1100°C to a density of 8.8 gm/cc. The samples were then coined at 75 tsi and resintered for fifteen hours at 1100°C to a density of 8.9 gm/cc. Although the coining and resintering operations did increase the density of the material, there was no improvement in the stability to thermal cycling.

J. Thermal Conductivity

Thermal conductivity values for several yttrium-uranium alloys are given in Table XIV. All the alloys were prepared from -200 mesh yttrium and -270 +325 mesh uranium. From the plot of thermal conductivity at 200°C versus uranium content given in Figure 11, it can be seen that the conductivity increases with increasing uranium content.

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TABLE XIII

THERMAL CYCLING DATA[a]

Sample No.	Uranium particle size, mesh	Yttrium particle size, μ	Density, gm/cc	Distortion, % Diam.	Distortion, % Length	Surface, RMS microinches
125	-325	30	8.64	2.94	1.3	325, blisters
126	-140 +325	30	8.48	2.73	4.12	310, very good
127	-325	5.6	8.79	2.1	2.83	300, excellent
128	-140 +325	5.6	8.76	1.51	4.26	310, blisters
129	-140 +325, spher.	5.6	8.96	3.74	4.04	350, blisters
130	-325, spher.	5.6	8.94	2.27	3.02	220, excellent
131	-140 +325, spher.	2.00	8.6	3.71	4.96	375, good

[a] 500 cycles, 725°-225°C.

TABLE XIV

THERMAL CONDUCTIVITY OF YTTRIUM-URANIUM ALLOYS -270 +325 MESH URANIUM, -200 MESH YTTRIUM

Composition, w/o	Density, gm/cc	Thermal conductivity, watts/cm-°C	
		150°C	200°C
100Y	4.27	0.118	-
5U/95Y	4.41	-	0.117
25U/75Y	5.26	0.119	0.122
50U/50Y	6.92	0.137	0.140
65U/35Y	8.89	0.160	0.168
85U/15Y	12.9	-	0.214
100U	18.9	0.28	0.30
100Y[a]		-	0.132

[a] Cast yttrium.

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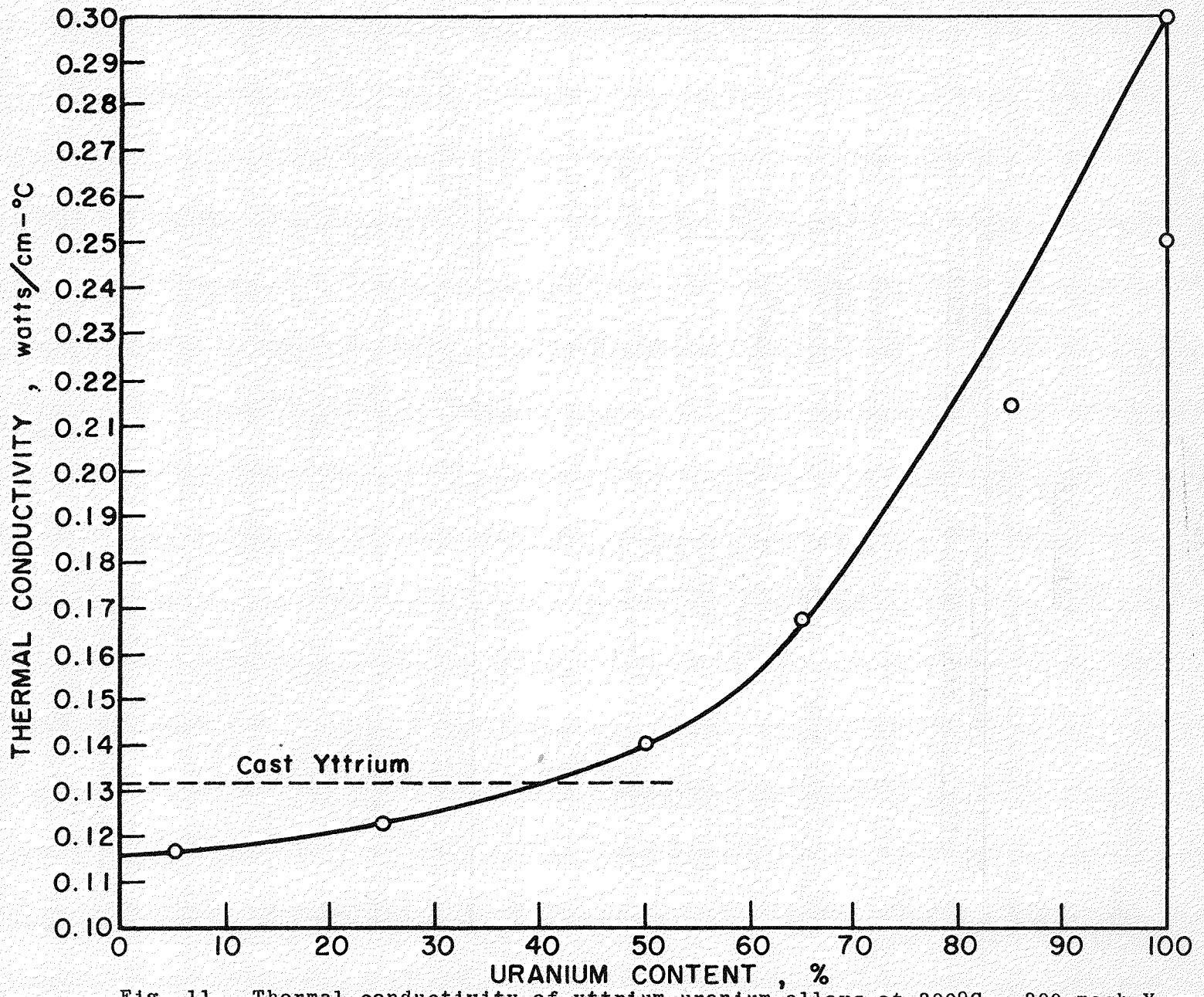

Fig. 11. Thermal conductivity of yttrium-uranium alloys at 200°C, -200 mesh Y, -270 +325 mesh uranium.

V. CONCLUSIONS

Hot pressing of mixtures of yttrium hydride and uranium powders in a graphite die does not provide a satisfactory method of making yttrium-uranium alloys. The metals react with the graphite to yield an impure product; the material is not held at the elevated temperature long enough to reduce the hydrogen content to a satisfactory level.

A complete range of yttrium-uranium alloys can be made by cold pressing and sintering the mixed powders. In this process, mixtures of yttrium hydride powder and uranium powder are compacted and then sintered under vacuum at a temperature slightly below the melting point of uranium until full density is reached. Alloys made by this process have a hydrogen content less than 50 ppm; by handling the powders in an inert-atmosphere dry box, the level of other contaminants can be kept to a minimum. Unless the sintering temperature is kept below the melting point of uranium, the molten uranium tends to bleed out to the surface, and the uranium particles flow together and agglomerate.

The particle sizes of the uranium and yttrium powders do not have any significant effect on the density of the sintered material. In all cases, essentially full density is obtained. Neither the particle sizes nor the compositions have a significant effect on the nitrogen and hydrogen contents of the sintered alloys. The high yttrium alloys and the finer yttrium powders have higher oxygen contents.

Alloys containing up to 65 w/o of uranium in yttrium consist of discrete particles of uranium in a matrix of yttrium. Increasing the uranium content to 85 w/o yields an alloy in which particles of yttrium are dispersed in a matrix of uranium.

All the alloys are stable for extended periods of time at 1000°C. They show essentially no dimensional changes after being held for fifty hours at 1000°C.

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The uranium particle size has little effect on the tensile properties of the alloys at elevated temperatures. The use of finer yttrium powders, however, increases strength and ductility at elevated temperatures. This is probably due to the better sintering characteristics of the finer powder.

Measurements of elongation versus temperature made in a quartz dilatometer have shown that the coefficient of thermal expansion increases with increasing uranium content. The uranium particle size does not have a measurable effect on the coefficient.

Alloys containing 5 to 65 w/o of uranium are compatible with sodium at 650°C. Samples immersed in sodium for 300 hours at 650° do not show any significant changes in dimensions.

Alloys containing up to 65 w/o of uranium show essentially no distortion when thermal cycled in the alpha-temperature range. Alloys containing 85 w/o of uranium are distorted after cycling in this range. This is probably due to the fact that these high-uranium alloys have a structure in which yttrium particles are dispersed in a matrix of uranium.

Alloys containing up to 50 w/o of uranium are quite stable after thermal cycling through the beta-transformation temperature. Alloys containing 65 w/o of uranium show considerable surface roughening as a result of cycling through the transformation temperature.

Reduction of the particle size of the dispersed uranium results in improved surfaces after beta thermal cycling. Reduction of the particle size of the yttrium reduces blistering during beta thermal cycling. This is probably due to the increased strength of material made from the finer yttrium powder.

Alloying the uranium with 3% molybdenum improves the surface after beta thermal cycling.

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Cold rolling the sintered alloys to a total reduction of 25%, followed by annealing at 1000°C, does not affect alloy behavior under thermal cycling. Coining and re-sintering do not affect alloy behavior.

The results obtained during this program indicate that a wide range of yttrium-uranium dispersion-type alloys can be prepared with the following characteristics: (1) good mechanical properties at elevated temperatures, (2) stability after long intervals at temperatures up to 1000°C, and (3) stability to thermal cycling. 50 w/o of uranium appears to be the maximum uranium content for a binary alloy to retain stability under thermal cycling. In order to increase the uranium content and still retain this stability, more complex alloys will have to be used. The binary alloys which contain up to 50 w/o of uranium appear very favorable for use as nuclear fuel materials.

VI. RECOMMENDATIONS FOR FURTHER WORK

The results obtained in thermal cycling and other tests indicate that the most promising alloys for use as nuclear fuel materials are 50U/50Y, and 25U/75Y. For more complete evaluation, irradiation tests should be carried out.

Results to date indicate that the amount of uranium which can be dispersed in yttrium and still form a stable alloy is limited by the formation of a network of uranium rather than a dispersion of discrete particles, and also by the limited strength of these alloys at elevated temperatures. Alloys with high uranium contents which are stable to thermal cycling at elevated temperatures can be achieved by the following techniques: (1) strengthening the yttrium network by alloying the yttrium powder prior to mixing, thus restraining deformation of the uranium particles; (2) alloying the uranium to increase it resistance to radiation; and (3) alloying the yttrium powder prior to mixing, resulting in an alloy having limited solubility and miscibility with uranium. During the sintering operation, the yttrium alloy will diffuse to the surface of

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the uranium and, in effect, leave uranium particles coated with a film of yttrium alloy.

By using dense, spherical uranium particles instead of irregular porous particles, it may be possible to increase the uranium contents of the stable alloys.

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