UNCLASSIFIED

Photostat Price \$ 4.80

Microfilm Price \$ 2.70

Available from the
Office of Technical Services
Department of Commerce
Washington 25, D. C.

BMI-1113(DEL.)

Contract No. W-7405-eng-92

THE PROPERTIES OF URANIUM
CONTAINING MINOR ADDITIONS
OF ALUMINUM OR ZIRCONIUM

by

Henry A. Saller
Frank A. Rough
Walston Chubb

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, express or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission to the extent that such employee or contractor prepares, handles or distributes, or provides access to, any information pursuant to his employment or contract with the Commission.

July 16, 1956

BATTELLE MEMORIAL INSTITUTE
505 King Avenue
Columbus 1, Ohio

This document is
PUBLICLY RELEASABLE

8 Stalo
Authorizing Official

Date: 4-17-07

UNCLASSIFIED

UNCLASSIFIED

56A-1

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

~~CONFIDENTIAL~~

3 and 4

TABLE OF CONTENTS

	<u>Page</u>
ABSTRACT	5
INTRODUCTION	5
EXPERIMENTAL PROCEDURES AND RESULTS	8
SUMMARY	26
REFERENCES	27

~~CONFIDENTIAL~~

DECLASSIFIED

5010 5

094020 1731

THE PROPERTIES OF URANIUM CONTAINING MINOR
ADDITIONS OF ALUMINUM OR ZIRCONIUM

Henry A. Saller, Frank A. Rough, and Walston Chubb

The tensile properties, hot hardness, and heat-treatment characteristics of uranium alloys containing up to 2.9 a/o aluminum or 4.0 a/o zirconium are described. The data show that zirconium has a favorable effect upon the grain size, hardness, and tensile strength of uranium without materially affecting ductility. Additions of 3.8 a/o zirconium raised the tensile strength of uranium at room temperature from about 100,000 psi to about 200,000 psi, while the elongation in 2 in. increased from 14 to 22 per cent and the grain size decreased from about 0.20 mm to less than 0.01 mm. The data show that aluminum raises the yield strength, that it has very little effect on either the grain size, hardness, or tensile strength of uranium, and that it decreases ductility. The addition of 2.6 a/o aluminum decreased the room-temperature ductility of uranium from 14 to 4 per cent.

INTRODUCTION

A number of uranium-base alloys have been investigated in the hope that alloying additions will improve the behavior of slug-type fuel elements. As irradiation proceeds, slug-type fuel elements have a tendency to grow in size, warp, bump, and crack. After a small amount of irradiation, this type of fuel element becomes brittle and ultimately develops microcracks and porosity. The tendency to crack and the brittleness of irradiated uranium represent a loss of ductility; however, it is generally assumed that increasing the purity of uranium will improve its performance by decreasing the number of inclusions which act as stress raisers during irradiation. Alloying to increase the strength of uranium also has been proposed as a means of improving performance in the reactor. The present study is a part of the effort to develop high-strength alloys of good ductility that may be useful for slug-type fuel elements.

Figure 1 compares the effects of several elements upon the reactivity of a slug-type fuel element. No consideration is given to the loss of reactivity due to dilution of the uranium by alloying, or to the increase in reactivity due to moderation and other effects. It is assumed that the decrease in reactivity is a linear function of the thermal-neutron-capture cross section of the slug.

FIGURE 1. EFFECT OF ALLOYING ELEMENTS UPON THE THERMAL-NEUTRON-CAPTURE CROSS SECTION OF URANIUM

The points plotted in Figure 1 represent the compositions considered initially for fuel slugs; the results of the investigation on these alloys are reported in BMI-1068.

On the basis of reactivity loss, alloys with aluminum, silicon, and zirconium appear most promising and were investigated further.

A uranium-1.5 a/o silicon alloy has been examined in considerable detail^{(1, 2)*}. Alpha uranium has negligible solubility for silicon even at 660 C, so that even though the silicide exerts an important strengthening effect upon uranium, the 1.5 a/o silicon alloy is somewhat difficult to fabricate and has lower ductility than pure uranium.

Uranium-aluminum alloys were examined in 1943 by Gordon⁽³⁾. It was found that a uranium-2.5 a/o aluminum alloy was readily forgeable in the upper alpha region and that it could be age hardened at 500 C after quenching from 1040 C. It was found that a uranium-5 a/o aluminum alloy was unforgeable and brittle, and it was suggested that an optimum alloy would be found between 2.5 and 5 a/o aluminum.

In 1950, Grobecker, Arnold, Taub, and Doll⁽⁴⁾ attempted to prepare uranium-aluminum alloys in a high vacuum. Since the vapor pressure of aluminum is in the neighborhood of 10 to 100 microns at 1100 C, losses of aluminum by volatilization were observed, and porous and inhomogeneous ingots were produced. As a result of an evaluation of these ingots, it was concluded that aluminum is "generally undesirable" as an alloying element in uranium alloys. This opinion of aluminum has persisted to the present day.

Because of the strong tendency of zirconium to stabilize gamma uranium, uranium alloys containing 5 a/o or more zirconium exhibit pronounced heat-treatment effects and considerable attention has been given to such alloys^(5, 6). Relatively little attention has been given to uranium alloys containing less than 5 a/o zirconium. Possibly one reason for this neglect has been the fact that zirconium tends to scavenge the carbon and oxygen present in impure or graphite-melted uranium. Zirconium replaces uranium in UC, and the net effect is a purification of the uranium and elimination of zirconium from solution. The effects of less than 5 a/o zirconium upon uranium can be properly determined only in low-carbon alloys. Two-tenths weight per cent carbon will eliminate 4 a/o zirconium as ZrC.

In 1954, Shober⁽⁷⁾ demonstrated that zirconium in amounts less than 2.5 a/o (1 w/o) had a pronounced beneficial effect upon the high-temperature hardness of uranium.

* References are at the end of the report.

8

~~CONFIDENTIAL~~

TABLE 1. ANALYSES OF URANIUM-ALLOY INGOTS

Nominal, a/o	Composition, balance uranium	
	Analyzed	
	a/o	w/o
0.5 aluminum	0.3 aluminum	0.03 aluminum
1.0 aluminum	1.0 aluminum	0.11 aluminum
1.0 aluminum	1.1 aluminum	0.12 aluminum
1.0 aluminum	1.1 aluminum	0.12 aluminum
2.0 aluminum	1.8 aluminum	0.21 aluminum
2.0 aluminum	1.9 aluminum	0.22 aluminum
2.0 aluminum	2.6 aluminum	0.30 aluminum
3.0 aluminum	2.9 aluminum	0.34 aluminum
1.0 zirconium	1.0 zirconium	0.41 zirconium
2.0 zirconium	1.6 zirconium	0.66 zirconium
2.0 zirconium	1.8 zirconium	0.70 zirconium
4.0 zirconium	3.8 zirconium	1.50 zirconium
4.0 zirconium	4.0 zirconium	1.60 zirconium

~~CONFIDENTIAL~~

TABLE 2. HARDNESS AND GRAIN SIZE OF CAST AND HOT-ROLLED URANIUM ALLOYS

Alloy Composition (Balance Uranium), a/o	Cast Hardness, BHN (3000-Kg Load)	As Hot Rolled at 620 C	
		Hardness (DPH), kg/mm ²	Grain Size, Average Diameter, mm
0.3 aluminum	--	270	0.04(a)
1.0 aluminum	--	287	0.02(a)
1.1 aluminum	224	243	2.0
1.1 aluminum	240	260	0.5
1.8 aluminum	232	274	0.5
1.9 aluminum	--	322	0.02(a)
2.6 aluminum	282	285	0.04
2.9 aluminum	265	283	0.2
1.0 zirconium	--	309	<0.01(a)
1.6 zirconium	232	281	0.01
1.8 zirconium	--	297	<0.01(a)
3.8 zirconium	268	294	<0.01
4.0 zirconium	--	345	<0.01(a)

(a) 100-g ingots.

~~CONFIDENTIAL~~

specimens for study. For each ingot size, however, the data show a definite trend toward higher hardness and smaller grain sizes with increasing alloy content. The superior grain-refining effect of zirconium is evident in the hot-rolled products.

All heat treatments were performed either in a vacuum furnace or in evacuated Vycor tubes. The effects of beta quenching and the effects of subsequent alpha annealing are shown in Table 3. These data show that aluminum and zirconium both increase the hardness of uranium, and that this increase occurs at about the same rate for equal amounts of each alloying element. However, the data show that for each ingot size the grain-refining effect of aluminum upon uranium is almost insignificant, whereas the grain-refining effect of zirconium is quite evident.

It should be noted that no appreciable softening occurred as a result of the 600 C treatment in any alloys except the 3.8 and 4.0 a/o zirconium alloys. It is believed that the 600 C treatment does produce a stabilizing effect on all alloys of this type, but that the effect on hardness is so small that it is masked by random errors except where the effect is large as in the case of the two uranium-zirconium alloys.

Typical microstructures are shown in Figures 2 through 5. The polarized-light photomicrographs were taken on electropolished surfaces. The bright-field photomicrographs were taken on the same samples electro-etched in chromic acid. The polarized-light photomicrographs show the effect of aluminum and zirconium upon the conventional grain size of uranium. The grain size of a uranium-1.1 a/o aluminum alloy is not greatly different from that of pure uranium given the same treatment; however, 2.6 a/o aluminum produces a mild grain-refining effect upon uranium. On the other hand, 1.6 a/o zirconium produces a mild grain-refining effect and 3.8 a/o zirconium produces a very striking grain-refining effect upon uranium.

The bright-field photomicrographs show the fine precipitates of UAl_2 and "delta" uranium-zirconium which are responsible for many of the grain size and mechanical-property effects found in these alloys.

Tensile specimens were prepared from 1/2-in. -diameter rod. This rod was heated to 730 C for 30 min, water quenched, reheated to 600 C for 1 hr, and furnace cooled prior to machining. The test specimens had a 1/4-in. -diameter, 2-1/4-in. -long reduced section. Tests up to 150 C were performed in air; tests at higher temperatures were performed in helium. The results of the tensile tests are summarized in Figures 7, 8, 9, and 10. In addition, Figure 6 shows results obtained previously on arc-melted biscuit uranium⁽²⁾. These results show that increasing amounts of aluminum produce only a small increase in the strength of uranium at room temperature. Aluminum has a somewhat greater strengthening effect upon uranium

TABLE 3. HARDNESS AND GRAIN SIZE OF HEAT-TREATED URANIUM ALLOYS

Alloy Composition (Balance Uranium), a/o	Heated to 730 C for 30 Min and Water Quenched		730 C, Water Quench, Plus 1 Hr at 600 C and Furnace Cooled	
	Hardness (DPH), kg/mm ²	Grain Size, Average Diameter, mm	Hardness (DPH), kg/mm ²	Grain Size, Average Diameter, mm
0.3 aluminum(a)	236	0.14	232	0.07
1.0 aluminum(a)	260	0.07	262	0.04
1.1 aluminum	268	0.10	276	0.14
1.1 aluminum	260	0.2	260	0.3
1.8 aluminum	268	0.10	285	0.14
1.9 aluminum(a)	289	0.07	274	0.04
2.6 aluminum	302	0.2	280	0.04
2.9 aluminum	302	0.10	279	0.10
1.0 zirconium(a)	245	0.07	260	0.05
1.6 zirconium	264	0.05	260	0.04
1.8 zirconium(a)	251	0.05	264	0.02
3.8 zirconium	366	<0.01	300	<0.01
4.0 zirconium(a)	351	0.02	304	0.02

(a) 100-gram ingots.

~~CONFIDENTIAL~~

13

100X

Polarized

N31549

500X

Bright Field

N31550

FIGURE 2. URANIUM-1.1 a/o ALUMINUM ALLOY HOT ROLLED AT 620 C, HEATED 30 MIN AT 730 C AND WATER QUENCHED, REHEATED 1 HR AT 600 C AND FURNACE COOLED

Hardness = 260 DPH

Grain size = 0.3 mm

~~CONFIDENTIAL~~

UNCLASSIFIED

560

11

~~CONFIDENTIAL~~

15

100X

Polarized

N31553

500X

Bright Field

N31554

FIGURE 4. URANIUM-1.6 a/o ZIRCONIUM HOT ROLLED AT 620 C, HEATED 30 MIN AT 730 C AND WATER QUENCHED, REHEATED 1 HR AT 600 C AND FURNACE COOLED

Hardness = 260 DPH

Grain size = 0.04 mm

~~CONFIDENTIAL~~

UNCLASSIFIED

560

10

16

100X

Polarized

N31555

500X

Bright Field

N31556

FIGURE 5. URANIUM-3,8 a/o ZIRCONIUM HOT ROLLED AT 620 C, HEATED 30 MIN AT 730 C AND WATER QUENCHED, REHEATED 1 HR AT 600 C AND FURNACE COOLED

Hardness = 300 DPH

Grain size = less than 0.01 mm

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

17

FIGURE 6. TENSILE PROPERTIES OF ARC-MELTED BISCUIT URANIUM

~~CONFIDENTIAL~~

UNCLASSIFIED

564 19

~~CONFIDENTIAL~~

18

FIGURE 7. TENSILE PROPERTIES OF URANIUM-1.1 a/o ALUMINUM ALLOY

~~CONFIDENTIAL~~

560 11

073700 1730

FIGURE 8. TENSILE PROPERTIES OF URANIUM-2.6 a/o ALUMINUM ALLOY

~~CONFIDENTIAL~~

20

FIGURE 9. TENSILE PROPERTIES OF URANIUM-1.6 a/c ZIRCONIUM ALLOY

~~CONFIDENTIAL~~

073700 1030

~~CONFIDENTIAL~~

21

FIGURE 10. TENSILE PROPERTIES OF URANIUM-3.8 a/o ZIRCONIUM ALLOY

~~CONFIDENTIAL~~

DECLASSIFIED

SCD 15

22

The results of the tensile tests show that small amounts of zirconium produce a large increase in the strength of uranium both at room temperature and at temperatures as high as 300 C. This increase in strength is accompanied by no appreciable loss in ductility; nor is there any noticeable effect upon the tensile-ductility transition temperature.

The hot-hardness curves in the temperature range 600 to 800 C demonstrate a degree of correspondence with the established constitution diagrams. The uranium-aluminum hot-hardness curves show the appearance of hard, beta uranium between 650 and 750 C. The uranium-zirconium hot-hardness curves demonstrate the gradual replacement of hard, beta uranium by soft, gamma uranium with increasing zirconium content and increasing temperature between 600 and 800 C.

The results of end-quench tests on uranium alloys containing 1.1, 1.8, and 2.9 a/o aluminum are shown in Figure 13. The data show that the hardness of the 1.1 and 1.8 a/o aluminum alloys is not affected by rate of cooling, at least in the range of rates available in the end-quench test. The data also show that the hardness of the 2.9 a/o aluminum alloy is affected by rate of cooling from 900 C. This effect, occurring rather suddenly at 2.9 a/o aluminum alloy content, suggests that the addition of aluminum has resulted in the onset of some new reaction involving retention of aluminum

~~CONFIDENTIAL~~

A 3x10 grid of dots forming the words "MATHS" and "MATHS". The first row contains 10 dots, the second row contains 10 dots, and the third row contains 10 dots. The dots are arranged to form the letters M, A, T, H, S in the first row and M, A, T, H, S in the second row.

FIGURE 11. HOT HARDNESS OF URANIUM-ALUMINUM ALLOYS

FIGURE 12. HOT HARDNESS OF URANIUM-ZIRCONIUM ALLOYS

A-19245

FIGURE 13. END-QUENCH TESTS ON URANIUM-ALUMINUM ALLOYS

Bars quenched from 900 C.

in supersaturated solution at the fastest quenching rates achieved in the end-quench bar. At the slower quenching rates a transformation process and simultaneous precipitation of UAl_2 could easily account for the age-hardening-type curve shown in Figure 13. Microstructural examination of the 2.9 a/o aluminum end-quench bar disclosed quench cracks at $1/4$ to $1/2$ in. from the quenched end. This is also the area of maximum hardness. No precipitate or second phase was observed for $3/4$ in. from the quenched end. At about $3/4$ in. from the quenched end, precipitation, or transformation and simultaneous precipitation, was observed to begin at gamma grain boundaries. This process progressed across the grains and at 2 in. from the quenched end the old gamma grains were completely filled with precipitate or transformation products. The tendency to produce quench cracks does not make this type of reaction desirable.

As has been found in end-quench bars of pure uranium, columnar grains were found to extend a considerable distance from the quenched end of all of the uranium-aluminum alloys. It is suggested that these columnar grains are formed from gamma uranium by a martensite-like shear mechanism. The columnar grains were 1 in. long in the 1.1 a/o aluminum alloy, $1/2$ in. long in the 1.8 a/o aluminum alloy, and $1/4$ in. long in the 2.9 a/o aluminum alloy. In the 2.9 a/o aluminum alloy, it would appear that the martensite-like reaction producing columnar grains is displaced by the pre-precipitation stages of the reaction which ultimately results in transformation of gamma and precipitation of UAl_2 .

No end-quench tests were performed on uranium-zirconium alloys. However, it should be noted that heat-treatment effects similar to those found in the uranium-2.9 a/o aluminum alloy are to be expected in uranium-zirconium alloys when they are end quenched from the gamma-phase field. These effects in uranium-zirconium alloys have been described in considerable detail elsewhere^(5, 6).

SUMMARY

A series of uranium-aluminum alloys and uranium-zirconium alloys suitable for slug-type fuel elements have been investigated. It has been demonstrated that aluminum produces a small increase in the strength and hardness of uranium, but results in a marked decrease in the ductility of uranium. Aluminum produces a grain-refinement effect in uranium, but the effect is not very pronounced even at the 2 a/o aluminum level.

For low-carbon uranium alloys only (less than 0.03 w/o carbon), it has been demonstrated that zirconium produces a large increase in strength and hardness. Practically no change in ductility accompanies this increase

in strength. Tensile data show that uranium-zirconium alloys maintain their relatively high strength up to 300 C. Hardness data show that uranium-zirconium alloys maintain their relatively high hardness up to about 600 C. Zirconium has a pronounced grain-refining effect upon low-carbon uranium. A grain size much smaller than 0.010 mm was produced in a 4 a/o zirconium alloy.

A heat treatment effect has been observed in a uranium-2.9 a/o aluminum alloy when end quenched from the gamma-phase field (900 C). It is surmised that a similar effect occurs in uranium-zirconium alloys during end quenching.

REFERENCES

- (1) Ryan, M. A., "Evaluation of Uranium-Silicon Alloy", HW-33821 (December, 1954).
- (2) Saller, H. A., Rough, F. A., Chubb, W., "The Properties of Uranium Containing Minor Additions of Chromium, Silicon, and Titanium", BMI-1068 (February, 1956).
- (3) Gordon, P., "Precipitation Hardening of Uranium-Aluminum Alloys", CT-1101 (December, 1943). See also NNES-IV-12A.
- (4) Grobecker, D. W., Arnold, W. F., Taub, J. M., Doll, D. T., "The Preparation and Physical Properties of Uranium Binary Alloys Containing Aluminum and Chromium", LA-1177 (September, 1950).
- (5) Aronin, L. R., Kaufman, A. R., "Development of Structures in Clad 2 w/o Zirconium-Uranium Alloys", TID-7502 (I), p 178 (January, 1956).
- (6) Chiswik, H. H., Dwight, A. E., "Heat Treatment of Some Uranium-Zirconium and Uranium-Zirconium-Niobium Alloys for Radiation", TID-7502 (II), p 581 (January, 1956).
- (7) Shober, F. R., Marsh, L. L., Muehlenkamp, G. T., "Development of Dimensionally Stable Uranium Alloys", BMI-961 (October, 1954).
- (8) Saller, H. A., Rough, F. A., "The Properties of High-Uranium Alloys Containing Zirconium or Chromium", BMI-984 (February, 1955).
- (9) Chubb, W., Muehlenkamp, G. T., "The Effects of Chromium, Iron, and Nickel on the Mechanical Properties of Zirconium", BMI-938 (August, 1954).

- [Faint, illegible text from bleed-through]*