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URANIUM ALLOY DEVELOPMENT FOR NON-NUCLEAR APPLICATION

PROGRESS REPORT NO. 2

by

N. S. SPENCE, H. M. SKELLY & C. F. DIXON

PHYSICAL METALLURGY DIVISION

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SUMMARY

The melting and alloying procedure used in the preparation of three uranium-base alloys (viz. U-2% Mo, U-6% Zr-2% Nb and U-2% Mo-2% Zr-2% Nb-0.5% Ti) is described. Details are given of heat treatment and swaging operations carried out on the alloys, and their mechanical properties are listed. Results of microexaminations are included.

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1. INTRODUCTION

High density materials are of interest for applications where inertia or kinetic energy properties are important, and work is being carried out elsewhere on the development of alloys based on tungsten and tungsten carbide for such purposes. Uranium-base alloys possess properties that make them attractive in this field, and at current prices uranium and tungsten are competitive. Depending upon their alloying content, uranium alloys may have densities as high as, or even higher than those of tungsten alloys or cemented tungsten carbide and, whereas the properties of the two latter materials are substantially unalterable, a wide range of physical and mechanical properties can be obtained with uranium by varying the alloy composition, amount of mechanical work, and the heat treatment.

An earlier report[1] describes the evaluation of various uranium alloy compositions prepared as small (100 g) melts in a tungsten-arc furnace, and also gives details of a heat treatment investigation of the U-2% Mo alloy which exhibits promising properties.

This report gives details of the preparation and properties of three alloys prepared for testing elsewhere[2,3]. Of the three compositions, one (U-2% Mo) - as mentioned above - has already been investigated in these laboratories[1], and the other two compositions have been studied by investigators in the U.S.A.

2. EXPERIMENTAL PROCEDURE AND RESULTS

2.1 Preparation of Alloys

Table 1 lists the compositions and melt identifications of the alloys. In order to provide sufficient material for testing, two melts were made for each alloy.

TABLE 1

Melt Identifications and Nominal Alloy Compositions

Melt Identifications	Nominal Composition (wt %)				
	Mo	Zr	Nb	Ti	U
R-AK, R-AL	2.0	-	-	-	balance
R-AM, R-AN	-	6.0	2.0	-	balance
R-AH, R-AJ	2.0	2.0	2.0	0.5	balance

Details of the uranium and alloying additions are given in Table 2.

TABLE 2

Uranium and Alloying Additions

Metal	Purity	Source
Uranium	Reactor grade	AECL, Chalk River
Molybdenum	99.5% min	Sylvania Electric Products Inc.
Zirconium	Reactor grade	Wah Chang Corporation
Niobium	99.7%	Fansteel Metallurgical Corporation
Titanium	99.5% min	Osaka Titanium Corporation

To facilitate solution of the alloying additions in the molten uranium, hardeners were first prepared in a tungsten-arc furnace fitted with a water-cooled copper hearth. The compositions of the hardeners are given in Table 3, together with the identifications of the melts in which they were used.

TABLE 3

Hardener Alloys

Nominal Composition (wt %)					Melts in Which Used
Mo	Zr	Nb	Ti	U	
15.0	-	-	-	balance	R-AK, R-AL
-	30.0	10.0	-	balance	R-AM, R-AN
16.0	16.0	16.0	4.0	balance	R-AH
14.0	14.0	14.0	3.5	balance	R-AH, R-AJ

The quantity of hardener prepared at one time varied from 109 to 195 g and the hardeners were re-melted three times to render them homogeneous. No chemical analyses were carried out on the hardeners as it was considered that weighing before and after melting was a satisfactory check on the composition. The hardeners were broken into pieces about 1/4 in. or less in size before being added to the melts.

The alloys were prepared in a vacuum resistance furnace capable of temperatures up to 2000°C at an absolute pressure of 0.1 micron mercury or less. Table 4 gives some alloy preparation details. Temperatures were determined with an optical pyrometer. "Holding Time" in Table 4 refers to the total time that the melt was completely molten. In all melts the pressure was less than 1 micron of mercury during alloying and casting. Crucibles of two materials were used -- fused stabilized zirconia and graphite; the latter was coated on the inside with a wash of zirconite to minimize carbon pick up.

The melts were cast into unheated, uncoated graphite moulds of two sizes. The first four melts listed in Table 4 (R-AH, R-AJ, R-AK and R-AL) were cast into a mould containing two cavities measuring 3/4 in. in diameter by 4-3/4 in., and the other two melts (R-AM and R-AN) were cast into a mould containing two cavities 7/8 in. in diameter by 4-1/2 in. The average diameters of the bars obtained from the two moulds were 0.736 in. and 0.853 in. respectively. All bars but one were about 4-3/8 in. long after cutting off the feeder-head; in the case of melt R-AM one of the cavities did not fill completely and the resulting bar was about 3 in. long. Metal recovery ranged from 90.5% to 97.2%.

TABLE 4

Melt Details

Melt Ident.	Melt Size (g)	Crucible	Holding Time (min)	Max Temp (°C)	Casting Temp (°C)
R-AH	1500	ZrO_2	124	1420	1310
R-AJ	1500	ZrO_2	110	1510	1350
R-AK	1500	Graphite	108	1360	1355
R-AL	1500	Graphite	92	1355	1355
R-AM	1700	ZrO_2	130	1515	1350
R-AN	1700	ZrO_2	85	1545	1400

Table 5 lists the results of chemical analyses of samples taken from each casting. The samples from melts R-AK, R-AL, R-AH and R-AJ were turnings machined from the bars after swaging and the samples from melts R-AM and R-AN were turnings machined from the as-cast bars. Extra analyses were carried out on some of the castings to determine how much segregation existed in the castings, and in those cases average figures are quoted, with the actual number of determinations following in brackets. Where there was considerable scatter, the range is given in brackets below the average figure.

TABLE 5

Chemical Analyses of Castings*

Sample Ident.	Chemical Analyses (wt %)					
	Mo	Zr	Nb	Ti	C	Si
R-AK	1.99	<0.07	-	-	0.06	0.02
R-AL	1.89 (2)	<0.07 (2)	-	-	0.04 (2)	0.04 (2)
R-AM	-	5.6 (4) (4.9-6.9)	2.0 (4) (1.8-2.5)	-	0.02 (2)	-
R-AN	-	7.3 (4) (6.0-8.7)	2.4 (4) (1.9-2.8)	-	0.04 (2)	-
R-AH	1.46	1.10	1.0	0.45	0.02	-
R-AJ	1.64 (2) (1.19-2.10)	1.36 (2) (1.00-1.73)	1.6 (2) (1.2-2.0)	0.43 (2)	0.02 (2)	-

*Carried out by Extraction Metallurgy Division.

2.2 Swaging

The cast material was mechanically worked by swaging to improve the properties. The bars cast in the small-cavity mould were swaged with the cast surface on them, but the larger bars were cleaned up by machining them to 0.736 in. diameter - the same as that of the smaller bars.

The bars were heated to swaging temperature in an argon atmosphere and swaged down to final size in six passes as follows:

Start (in.)						Finish (in.)
0.736	→ 0.700	→ 0.630	→ 0.560	→ 0.500	→ 0.460	→ 0.430

The final pass was made through a "cold" die to straighten the bars. The dies were lubricated with a high temperature grease containing colloidal graphite and mica.

The U-2% Mo alloy was swaged at 650°C (1200°F), in the alpha plus gamma phase region, but the other two alloys were swaged at 800°C (1470°F), in the gamma phase region, since other specimens broke up when worked at the lower temperature. The average diameter of the bars after swaging was 0.42 in., representing a reduction of about 67%.

2.3 Heat Treatment and Hardness

The swaged bars were required to be heat treated to maximum hardness, and details of the heat treatment and hardness of each alloy are given separately below. Houghton 980 salt was used for the heat treatments carried out in molten salt, and the specimens so treated were coated with graphite to protect them from the action of the salt, and molten lead when it was used as a quenching medium. The hardness values are the averages of four or more determinations.

Uranium-2% Molybdenum Alloy

Work had already been carried out on this alloy and a hardening heat treatment developed[1]. This treatment was used in the present work, details being as follows: 800°C (1470°F) for 30 min in salt bath and then transfer quickly to lead bath at 400°C (750°F), holding at that temperature for 5 min and then quenching into water. Table 6 gives the hardness of the alloy in the as-cast and swaged and heat treated conditions. (The latter is referred to hereafter as the "heat treated" condition).

TABLE 6

Hardness (VHN) of U-2% Mo Alloy

Melt Identification	As-cast	Heat Treated
R-AK	384	534
R-AL	362	540

Uranium-2% Mo - 2% Zr - 2% Nb - 0.5% Ti Alloy

The following two possible heat treatments were suggested for this alloy[2]: (a) 870°C (1600°F), cool to 480°C (900°F) and hold for 1 hr, water quench; (b) 900°C (1650°F) then water quench, age at 205°C (400°F). The latter treatment was said to harden to 51 R_C[2], but the maximum hardness obtained from it was 250 VHN (<30 R_C). The following treatment was therefore given to the swaged bars: 870°C for 30 min in salt bath then into lead bath at 480°C for 1 hr, followed by water quench. Table 7 gives the hardness of the alloy in the as-cast and heat treated conditions.

TABLE 7

Hardness (VHN) of U-2% Mo-2% Zr-2% Nb-0.5% Ti Alloy

Melt Identification	As-cast	Heat Treated
R-AH	492	598
R-AJ	541	582

Uranium-6% Zr-2% Nb Alloy

The recommended treatment for this alloy was to water quench from 955°C (1750°F) followed by ageing at 427°C (800°F), and this treatment was said to produce a hardness of 54 R_C[2]. It was found that heat treating at 955°C for 30 min and water quenching, followed by ageing at 430°C for 1 hr and then water quenching produced a hardness of 576 VHN (about 55 R_C). However,

the alloy was very soft at 955°C, which is well into the gamma phase region, and it was therefore decided to try heat treating at 850°C (1560°F) which is also in the gamma region but less likely to make the alloy unduly soft; when this treatment was followed by ageing at 430°C for 1 hr then water quenching, a hardness of 562 VHN (about 54 R_C) was obtained. As the latter treatment produced a satisfactory hardness without running risk of damage due to softening during solution heat treatment, all of the swaged bars were given this treatment. The bars were sealed in quartz tubes for the solution heat treatment. Table 8 gives the hardness of the alloy in two conditions.

TABLE 8

Hardness (VHN) of U-6% Zr-2% Nb Alloy

Melt Identification	As-cast	Heat Treated
R-AM	410	572
R-AN	397	555

2.4 Mechanical Properties

Tensile and compression tests were carried out on material that had been swaged and then heat treated as described above. Two of the tensile specimens (R-AL and R-AH) were machined to a modified Hounsfield specimen (0.12 in. gauge diameter and 0.75 in. gauge length) and the other specimens (R-AM and R-AN) were machined to Physical Metallurgy Division Drawing No. 107 (0.179 in. gauge diameter and 0.632 in. gauge length). The specimens for determining compressive properties were 0.34 to 0.38 in. diameter by 1.1 to 1.2 in. long and, in the case of R-AM and R-AN, they were cut in two for determination of the ultimate compressive strength (UCS). The alloys were too hard to lathe-machine in the heat treated condition and the test specimens were therefore first machined to a few thousandths of an inch oversize, heat treated to harden, and then ground to finished size.

The mechanical test results are listed in Table 9. Where more than one test was made the number of tests is in brackets following the average value.

TABLE 9

Results of Tensile and Compression Tests

Melt Identification	Nominal Composition (wt %)				Tensile Properties				Compressive Properties	
	Mo	Zr	Nb	Ti	UTS (kpsi)	0.2% YS (kpsi)	% El. $4\sqrt{A}$	% RA	UCS (kpsi)	0.2% CYS (kpsi)
R-AK, R-AL	2.0	-	-	-	153.3	none	0	0	295.2 (2)	ND
R-AM, R-AN	-	6.0	2.0	-	197.4 (2)	none	0	0	332.0 (4)	285.5 (2)
R-AH, R-AJ	2.0	2.0	2.0	0.5	182.4	none	1.0	0	240.5 (2)	ND

ND = not determined

2.5 Density

Density determinations were carried out on various specimens in accordance with ASTM Designation B311-58 which is based on the displacement of water by the specimen. The results are given in Table 10.

TABLE 10

Densities of Alloys

Nominal Composition (wt %)				Condition	Density (g/cc)
Mo	Zr	Nb	Ti		
2.0	-	-	-	As-cast	18.49
"	-	-	-	Heat treated	18.48
-	6.0	2.0	-	As-cast	16.70
-	"	"	-	Heat treated	16.59
2.0	2.0	2.0	0.5	As-cast	17.35
"	"	"	"	Heat treated	17.50

2.6 Microexamination

Metallographic examinations were carried out on samples of each alloy in order to make an assessment of the quality of the castings with respect to alloying efficiency and melt contamination.

The samples were first mechanically polished on silicon carbide abrasive paper up to a 600 grit, then with 1 and 1/2 micron size diamond polishing compound on miracloth and selvyt respectively. This was followed by a 3 sec electropolish in a solution containing 4 parts acetic acid and 1 part of a solution of 118 g of chromium trioxide in 100 cc of distilled water. The current density was 2 amp/sq cm and a water-cooled stainless steel container was used as the cathode.

Figure 1 is a photomicrograph of the U-2% Mo alloy (melt R-AL) in the as-cast condition, and most of the inclusions present in it are uranium carbide.

Figures 2, 3 and 4 are photomicrographs of the U-2% Mo-2% Zr-2% Nb-0.5% Ti alloy (melt R-AH) in the as-cast and heat treated conditions. Most of the inclusions showing here are uranium oxide, and there is little evidence of the presence of uranium carbide.

Figures 5, 6 and 7 are photomicrographs of the U-6% Zr-2% Nb alloy (melt R-AM) and, as in melt R-AH, most of the inclusions are uranium oxide.

3. DISCUSSION

Table 5 (page 4) shows that the composition of the uranium-molybdenum alloy is satisfactory but that the other two alloys are off-composition and show signs of segregation of the alloying constituents. It might be possible to improve the alloying procedure by applying some form of mechanical stirring to the melt, but this is not easy with the furnace available. Table 5 also indicates that there is a slight pick-up of carbon in these melts (R-AK and R-AL) made in graphite crucibles as compared to the other melts which were made in zirconia crucibles; the silicon content in the former melts is also slightly above normal and this could have been caused by the zirconite wash used.

The photomicrographs (see Figures 1 to 7) show that most of the inclusions in the melts made in a graphite crucible are uranium carbide, whereas the main impurity in the melts made in zirconia crucibles is uranium oxide. The presence of uranium oxide is attributed to oxygen pick-up from the zirconia. The amount of oxide appears to be greater in melt R-AM (see Figure 5) than in melt R-AH (see Figure 2) and this may be due to the higher temperature attained in the former melt (1515°C vs 1420°C). The uranium oxide was broken up and dispersed to some extent on swaging, as can be seen by comparing Figures 5 and 6. There was no evidence of the presence of undissolved alloy additions in any of the specimens examined.

The swaging operation was carried out satisfactorily except that considerable oxidation and roughening of the surface of the bars occurred. It is possible that a better surface finish would be obtained if the bars were protected during swaging by cladding them with copper or some such suitable material.

It was requested[2] that the three alloys be supplied in as fully hardened condition as possible but it should be borne in mind that such a condition is not necessarily the optimum one for the application involved and that a less hard but tougher condition might give a better performance. The heat treatment given to the U-6% Zr-2% Nb alloy involved quenching into water, and this could cause cracking if it had to be carried out on material of larger cross-section than that of the swaged bars.

The most noticeable feature of the mechanical properties of the alloys is their lack of ductility (Table 9, page 8). The possibility of varying the heat treatment to produce a condition of greater ductility with the sacrifice of some hardness is to be investigated.

As expected, the U-2% Mo alloy showed the highest density (Table 10, page 9) and this is a point in favour of this composition. The U-6% Zr-2% Nb alloy had the lowest density.

4. FUTURE WORK

Further work is being carried out on the development of high-strength uranium-based alloys. It is planned to investigate the properties of binary and ternary alloys of uranium with molybdenum, niobium, vanadium, zirconium and titanium.

5. REFERENCES

1. N.S. Spence, H.M. Skelly and C.F. Dixon - "Uranium Alloy Development for Non-Nuclear Application: Progress Report No. 1A" - Mines Branch Investigation Report IR 64-15 (March 3, 1964).

2. Letter - H.P. Tardif, Canadian Armament Research and Development Establishment, Valcartier, Quebec to S.L. Gertsman, Mines Branch, Ottawa (February 21, 1963).

3. Letter - N.S. Spence, Mines Branch, Ottawa, to H.P. Tardif (April 5, 1963).

X150

Figure 1. U-2% Mo Alloy. Melt R-AL.
As-Cast, As-Polished.

X150

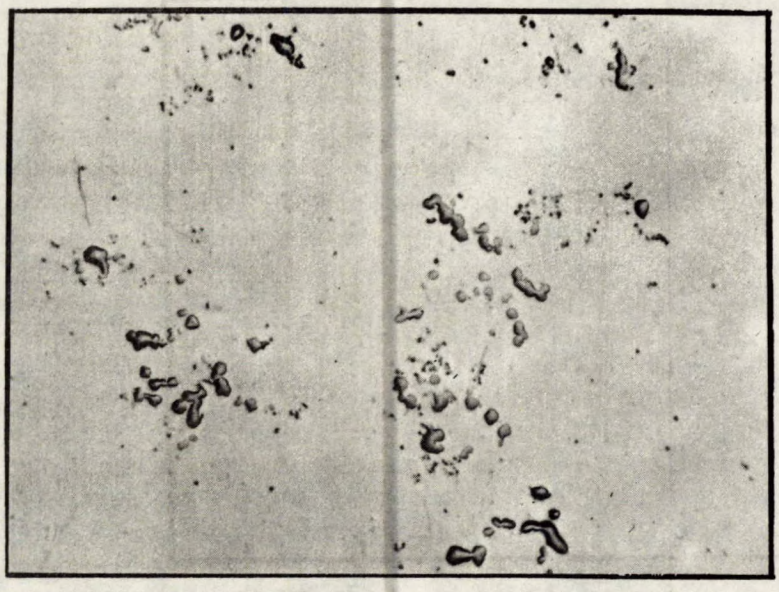
Figure 2. U-2% Mo-2% Zr-2% Nb-0.5% Ti Alloy.
Melt R-AH. As-Cast, As-Polished.

X150

Figure 3. U-2% Mo-2% Zr-2% Nb-0.5% Ti Alloy.
Melt R-AH. Heat Treated, As-Polished.

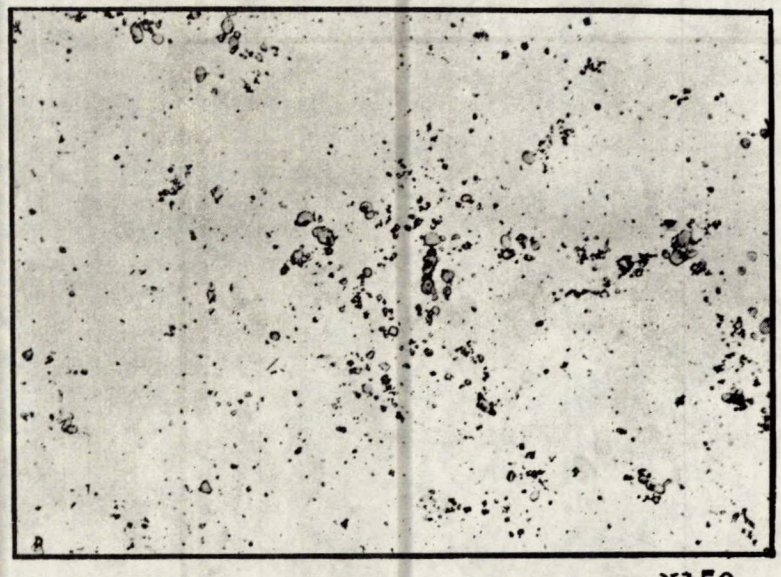
X500

Figure 4. U-2% Mo-2% Zr-2% Nb-0.5% Ti Alloy.
Melt R-AH. Heat Treated, As-Polished.



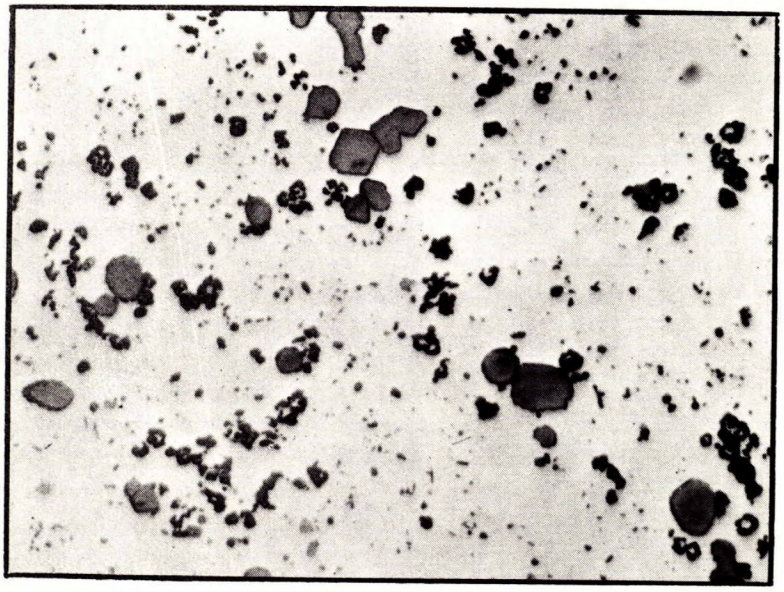
X150

Figure 5. U-6% Zr-2% Nb Alloy. Melt R-AM. As-Cast, As- Polished.



X150

Figure 6. U-6% Zr-2% Nb Alloy. Melt R-AM. Heat Treated, As-Polished.



X500

Figure 7. U-6% Zr-2% Nb Alloy. Melt R-AM. Heat Treated, As-Polished.