

10/325

BMI-1500

MASTER

DEVELOPMENT OF CONTAINER MATERIALS
FOR LAMPRE APPLICATIONS

BATTELLE MEMORIAL INSTITUTE

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.

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, expressed 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, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

Printed in USA

Price \$1.25

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

Report No. BMI-1500

UC-25 Metals, Ceramics, and Materials
(TID-4500, 16th Ed.)

Contract No. W-7405-eng-92

DEVELOPMENT OF CONTAINER MATERIALS
FOR LAMPRE APPLICATIONS

by

David C. Drennen
Merritt E. Langston
Charles J. Slunder
Joseph G. Dunleavy
A. M. Hall

February 14, 1961

BATTELLE MEMORIAL INSTITUTE
505 King Avenue
Columbus 1, Ohio

TABLE OF CONTENTS

	<u>Page</u>
ABSTRACT	1
INTRODUCTION	1
PREPARATION OF EXPERIMENTAL ALLOYS	2
Alloy Selection	2
Materials	3
Arc Melting and Casting	3
Equipment	3
Melting Procedure	5
Radiography	7
Sectioning	9
Fabrication	9
Cleaning	9
Pressing	9
Machining	10
Cold Rolling	11
Vacuum Annealing	12
Intermediate Annealing	12
Final Annealing	12
EVALUATIONAL PROCEDURES	13
Surface Tests for Iron	13
Chemical Analyses	14
Metallography	14
Hardness Measurements	15
Tensile Testing	15
EXPERIMENTAL RESULTS AND DISCUSSION	16
Arc-Casting Behavior	16
Fabrication Behavior	16
Annealing Studies	16
Chemical Analyses	22
Microstructure	27
Hardness	35
Tensile Properties	39
Corrosion Testing	45
CONCLUSIONS	45
ACKNOWLEDGMENT	46
REFERENCES	46

DEVELOPMENT OF CONTAINER MATERIALS FOR LAMPRE APPLICATIONS

David C. Drennen, Merritt E. Langston, Charles J. Slunder,
Joseph G. Dunleavy, and A. M. Hall

Some 83 high-purity binary tantalum-base alloys were prepared as candidate materials of construction for the Los Alamos Molten Plutonium Reactor Experiment (LAMPRE). The alloys were evaluated metallurgically at Battelle and then forwarded to LASL for evaluation in the plutonium-alloy fuel mixtures.

Although the corrosion tests are not complete, preliminary data indicate that good resistance to attack by the fuels can be obtained by alloying tantalum. Alloys containing additions of rhenium and tungsten have shown good corrosion resistance in polythermal (1382 to 1022 F) tilting-furnace exposures. Tantalum-yttrium alloys also have displayed good corrosion resistance, even though the yttrium apparently was lost during arc melting. Most of the alloys, including those which showed good corrosion resistance, were amenable to arc melting and casting and fabrication at room temperature.

INTRODUCTION

Tantalum has been considered for the reactor-core structural material in the Los Alamos Molten Plutonium Reactor Experiment (LAMPRE) because of its resistance to attack by plutonium-alloy fuels, its good forming properties at room temperature, and its possession of adequate strength at the proposed operating temperature of 1200 F. However, data obtained at LASL indicated that the resistance of commercial grades of tantalum to corrosion by the molten plutonium-alloy fuel mixtures was inconsistent and had to be improved to eliminate (1) intergranular corrosion, which seems to be affected by the degree of cleanliness in the grain boundaries, and (2) general surface roughening. (1) Although sufficient data are not available to determine precisely the chemical phenomena associated with the first effect, it is believed that a high oxygen content in commercial tantalum contributes to the second type of attack.

To assist LASL in the LAMPRE project, a research program directed toward the development of container materials suitable for use in LAMPRE service was undertaken at Battelle Memorial Institute. The objective of the program was to develop a tantalum-base alloy which would be acceptable with respect to corrosion resistance in the plutonium-alloy fuel mixtures. A satisfactory combination of elevated-temperature strength, ductility, and weldability was desired, but it was not imperative that the alloy be superior to tantalum except from a corrosion standpoint.

(1) References are at end of report.

The basic precept of the research program was that only a high-purity material held maximum promise from the viewpoint of compatibility with the plutonium-alloy fuels. The program involved (1) selection of alloy systems, (2) preparation of materials as high-purity alloys, (3) evaluation of the alloys with reference to purity, homogeneity, soundness, fabricability, hardness, and strength, and (4) preparation of specimens for fuel-corrosion tests at LASL.

PREPARATION OF EXPERIMENTAL ALLOYS

Alloy Selection

Because of the large number of alloy systems to be investigated, the addition of alloying elements to tantalum was limited to a maximum of 6 w/o. For the most part, the alloying elements were of two general types. One consisted of elements which were expected to form solid-solution alloys of tantalum. Single-phase alloys were considered preferable because the presence of a secondary phase in the alloy might detract from corrosion resistance. The other consisted of elements which were added to serve primarily as scavengers of interstitial impurities, which might be responsible for certain types of corrosion in tantalum. The expected mechanisms operative in the alloys containing scavenger elements were (1) the formation of a more stable carbide, hydride, nitride, or oxide in the alloy, and/or (2) the removal of interstitial impurities from the tantalum during melting.

The tantalum-tungsten system was selected first for investigation for several reasons:

- (1) Tungsten is known to resist attack by the plutonium-alloy fuels. (1)
- (2) Tungsten forms a complete series of solid-solution alloys with tantalum. (2)
- (3) Tungsten is known to increase the strength of tantalum by solid-solution hardening. (3)
- (4) The tantalum-tungsten system was of interest because of the transmutation of tantalum to tungsten upon irradiation.

The other elements alloyed with tantalum were expected to show considerable solid solubility. These included hafnium, rhenium, titanium, yttrium, and zirconium.

Elements which were added for the purpose of acting as scavengers of interstitial impurities were aluminum, beryllium, carbon, cerium, lanthanum, scandium, thorium, titanium, uranium, yttrium, and zirconium.

Alloys containing additions of either boron, iron, or silicon were of special interest to Los Alamos personnel and were prepared at their request.

Some 83 unalloyed tantalum and binary tantalum-base alloys, which were divided into four series as indicated in Table 13, were prepared for plutonium-alloy-fuel corrosion tests at LASL. The first series of alloys included a large number of unalloyed tantalum buttons, for the purpose of evaluating the arc-melting operation. In addition, subsequent series of alloys contained at least one unalloyed tantalum button for control purposes.

Materials

All of the unalloyed tantalum and tantalum-base alloys prepared for this study were made from various lots of ultrahigh-purity electron-beam-melted tantalum purchased from the Temescal Metallurgical Corporation either by Battelle or by LASL. The tantalum processed by Battelle originally had been capacitor-grade scrap which was purified to the analyses indicated in Table 1. The tantalum sheet obtained from LASL had been high-purity powder, provided by the National Research Corporation, which was also purified by electron-beam melting to the purity level indicated in Table 1.

LASL began supplying tantalum to Battelle after the first two series of alloys had been prepared. The purpose was to provide both the LASL and the Battelle program with the same base material and, thus, eliminate it as a factor in evaluating the corrosion behavior of LASL and Battelle alloys. Tantalum was received from LASL in the form of 0.030-in.-thick sheet scrap obtained from punching out blanks for test thimbles.

Alloying additions of the highest purity available were used in preparing the tantalum alloys. Chemical analyses were performed on some of the alloying additions as indicated in Table 2.

Arc Melting and Casting

Preliminary studies had been conducted at the end of a previous program for LASL to establish a suitable basic arc-melting procedure with respect to (1) maintaining the high-purity levels of the melting stock, (2) obtaining homogeneous alloys, (3) obtaining sound button specimens, (4) determining the optimum button weight, and (5) obtaining information on alloy-addition recoveries. From these preliminary studies, a basic melting procedure was developed. Also, the chemical analyses and hardness data obtained therefrom indicated that arc melting under conditions to be discussed later, had no significant effect on the purity of unalloyed tantalum and tantalum alloys, unless difficulties were encountered during melting.

Equipment

Button specimens, weighing from 100 to 250 g, were prepared by arc melting in a flip-type arc furnace with a tungsten electrode and a water-cooled copper crucible under 1/3 atm of AEC helium. The specially designed crucible had a small cavity located adjacent to the casting cavity to hold a small charge of either zirconium or tantalum, for the purpose of getting residual impurities in the furnace.

TABLE 1. ANALYSES AND HARDNESS OF HIGH-PURITY ELECTRON-BEAM-MELTED TANTALUM USED AS MELTING STOCK

Lot	Alloy Series in Which Used	Hardness, VHN	Analyses, ppm									
			C	H ₂	N ₂	O ₂	W	Nb	Si	Cu	Fe	Zr
B9269-1	1, 2	92	<10	1.7	20	11	<50	300	10	15	7	<10
B9269-2	2	90	10	0.4	10	11	<50	100	<10	5	30	<10
LASL-1	3	91	10	0.3	<10	46	<100	50	<10	10	3	<10
LASL-2	3	--	20	1.0	20	14	200	<10	<10	100	<3	<10
LASL-3	4	--	10	0.20	20	14.5	50	(a)	5	10	8	<10
LASL-4	4	--	10	<0.6	30	8	<100	(a)	3	<0.5	1.0	<10

(a) Not detected.

TABLE 2. ANALYSES OF ALLOYING ELEMENTS USED TO MAKE HIGH-PURITY TANTALUM-BASE ALLOYS

Element	Lot Number	Supplier's Analysis w/o	Alloy Series in Which Used	Form	Analyses, ppm										
					C	H ₂	N ₂	O ₂	W	Nb	Si	Cu	Fe	Zr	Hf
Al	Al-1	99.99	4	Briquette	--	--	--	--	--	--	10	20	10	--	--
B	B-1	99.14	4	Powder	--	--	--	--	--	--	--	--	--	--	--
Be	Be-1	--	4	Sintered balls	1300	210	166- 280	773	<50	<200	1000	20	1500	50	--
C	C-1	--	2	Spectrographic-grade rod	--	--	--	--	--	--	--	--	--	--	--
Ce	D6906-2	99.9	4	Ingot	--	--	--	--	--	--	--	--	--	--	--
Fe	Fe-1	--	4	Ingot	10	<0.1	<2	15	<10	--	10	2	--	<0.5	--
Hf	Hf-1	--	3	Strip	10	17	20	1020	3000	--	30	30	250	8000	--
Hf	E3873	--	4	Crystal bar	300	--	90	--	(a)	--	--	--	--	1300	--
La	D7656	99.5 (min)	4	Ingot	--	--	--	--	--	--	--	--	--	--	--
Re	C2295	--	2	Sintered strip	40	6	<10	55	--	<50	<10	<5	5	<50	--
Sc	D6906-1	99+	4	Wafer	--	--	--	--	--	--	--	--	--	--	--
Si	Si-1	--	4	Single crystal	--	--	--	--	--	--	--	--	--	--	--
Th	D2974	--	3, 4	Crystal bar	<10	27	<10	231	<120	(a)	<10	2	10	(a)	--
Ti	D2975-1	--	3, 4	Crystal bar	<10	300- 326	<10	227- 238	<50	<100	100	40	50	<30	--
U	U-1	99.999	4	Biscuit	--	--	--	--	--	--	--	--	--	--	--
W	C3043-1	--	1	Undoped rod	<10	0.6	10	8	--	--	100	10	5	<10	--
W	C3043-2	99.999	3	Zone-refined rod	--	--	--	--	--	--	--	--	--	--	--
Y	Y-1	--	2, 3, 4	Ingot	110	5.2	80	2670	--	<50	10	<10	<10	10	--
Zr	Zr-2	--	3, 4	Crystal bar	60	54	10	340	500	<300	200	20	300	--	--

(a) Not detected.

Melting Procedure

Prior to each melting, the furnace was cleaned as follows:

- (1) Swabbed with paper tissues
- (2) Vacuumed with sweeper
- (3) Wire brushed
- (4) Casting cavity abraded with silicon carbide abrasive disks
- (5) Swabbed again with paper tissue
- (6) Vacuumed again with sweeper.

The getter material, which was changed periodically, and the charge, which usually consisted of small sheared pieces of tantalum and the alloy addition, were then placed in the proper cavities in the crucible. The alloy addition was placed between two layers of tantalum stock to prevent vaporization of the alloying element when the arc was started.

The basic melting cycle for 100 to 150-g buttons was as follows:

- (1) The furnace was charged and evacuated to an operating pressure of 10μ or less.
- (2) The system was flushed three times with AEC helium, and pumped down to the operating pressure after each flushing.
- (3) With the cold water flowing around the crucible and the lowest pressure obtained, the leak rate was checked and was limited to a maximum of 2μ per min.
- (4) The furnace was evacuated again to the lowest pressure possible.
- (5) The vacuum valve was closed and $1/3$ atm of helium was introduced.
- (6) The arc was started on the getter charge and held for 5 to 10 sec and then swung over to the charge on the first melt after the furnace was closed. On subsequent melts the arc was started directly on the charge if the high-frequency arc starter was used; if the arc starter was not used, the arc was again initiated on the getter by touching it.
- (7) Each 100 to 150-g alloy button was melted at least six times at 30 v, 800 amp for 30 sec; buttons were flipped after each melt by rapping on the bottom of the furnace.
- (8) The buttons were cooled 3 min before removal from the furnace.

For buttons weighing 200 and 250 g it was necessary, due to equipment limitations, to first prepare two 100- and 125-g buttons according to the procedure above. These were then placed one on top of the other and consolidated into one large button. The first six steps of the melting-cycle procedure used for the small buttons were also used in the consolidation. However, the remainder was different and the best procedure was as follows:

- (1-6) The procedure was the same as for the 100 to 150-g buttons.
- (7) The buttons were melted four times at 30 v, 800 amp for 60 sec. The buttons were flipped after the first three meltings. At the end of the fourth melting, the power was reduced to 30 v, 400 amp for about 30 sec to smooth the surface. The buttons were then flipped and melted again at 30 v, 400 amp for about 60 sec.
- (8) The buttons were cooled 8 min before removal from the furnace.

A summary of the arc melting of the unalloyed tantalum and tantalum-base alloys; according to alloy series (see Table 13), is given in Table 3. Figure 1 shows the surface of a typical arc-cast 200-g button, in this case, of unalloyed tantalum.



FIGURE 1. TYPICAL ARC-CAST 200-G BUTTON OF UNALLOYED TANTALUM

Zirconium was used as the gettering material in the arc melting of the unalloyed tantalum buttons of the first series. However, in an attempt to eliminate foreign materials in the furnace during melting, the zirconium was replaced by tantalum during the preparation of materials thereafter.

After the first series of alloys had been prepared, it was believed that a zirconium liner in the furnace might improve the purity of the alloys by gettering gaseous impurities during melting. Therefore, a 1/16-in. -thick liner was fitted to the inside of the crucible above the casting cavity. However, later chemical analyses, which are contained in Table 11, on the alloys of the second series failed to show any improvement in the purity levels. Because of the apparent ineffectiveness and also the operating problems associated with the zirconium liner, it was decided to discontinue its use.

At the initiation of arc melting of the fourth series of alloys, difficulty was encountered from severe erosion of the tungsten electrode which resulted in contamination of the alloys with tungsten. Melting was discontinued temporarily to determine the source of trouble and to eliminate it if possible. It was believed that the electrode material might be a factor in the unusual tip erosion. Also, it was suggested that the erosion problem might be modified by conditioning the electrode with yttrium. In order to investigate these possibilities, the following series of arc-melting experiments was conducted:

- (1) Unalloyed tantalum was melted with electrodes prepared from normal sintered, forged, and thoriated tungsten.
- (2) Unalloyed tantalum was melted with the three types of electrodes, but with 0.2 w/o yttrium placed on top of the tantalum charge.
- (3) Unalloyed tantalum was melted with the three types of electrodes, but the electrodes were "conditioned" by having been spattered with yttrium from the melting done in (2).

The results of these tests indicated that (1) an addition of 0.2 w/o yttrium to the tantalum charge was far more than was needed to eliminate tip loss, (2) tip loss could be significantly reduced by conditioning the tip with yttrium as in (3) above, and (3) tip loss for unconditioned electrodes increased in the order: thoriated tungsten, forged tungsten, and normal sintered tungsten. The loss for the latter was almost eight times that of the thoriated tungsten. As a result of this finding, yttrium was either added to some of the tantalum and tantalum-base alloy charges or an electrode conditioned previously with yttrium was used. Some of the alloying elements of the fourth series were similar to yttrium chemically and, therefore, were expected to behave like yttrium in controlling tip loss. In these cases, yttrium was not used. In other cases, yttrium was not added because there were indications of a possible interaction between the alloying element and the yttrium. Table 4 contains a list of the materials in which yttrium was either added to the charge or the tip was previously conditioned by melting a charge that had yttrium added to it. In these cases, the yttrium addition was 0.10 w/o rather than the 0.2 w/o used in the tip-loss control experiments, because the latter had been found to be much more than necessary.

Radiography

The soundness of the arc-cast buttons was determined by means of radiographic examination. The radiographic negatives were studied, and if a button was sound or contained voids that were not expected to prevent the preparation of good quality strip material, the button was processed. However, if the button contained too many voids or a fairly large void, the button was remelted.

TABLE 3. SUMMARY OF ARC-MELTING PROCEDURES FOR THE PREPARATION OF TANTALUM AND TANTALUM-BASE ALLOYS

Alloy Series	Unalloyed Tantalum		Tantalum-Base Alloys	
	Button Weight, g	Number of Melts	Button Weight, g	Number of Melts
1	100	2	100	6
	200(a)	2	200(a)	2
2	150	7	125	6
			150	7
			250(b)	2
3	150	7	125	6
			150	7
			250(b)	4
4	100	6	100	3(c)
	200(a)	4-5	100	6
			200	4-5

(a) Two 100-g buttons were consolidated to form one 200-g button.

(b) Two 125-g buttons were consolidated to form one 250-g button.

(c) This represents the melting of the tantalum-yttrium alloys (109 and 110), in which the objective was to retain yttrium rather than to obtain homogeneity.

TABLE 4. TANTALUM AND TANTALUM-BASE ALLOYS ARC MELTED WITH YTTRIUM OR YTTRIUM-CONDITIONED TUNGSTEN ELECTRODES

Alloys in Which 0.1 w/o Yttrium Was Added to Charge		Alloys Melted With Electrode Previously Conditioned With 0.1 w/o Yttrium	
Alloy	Composition (Balance Tantalum), w/o	Alloy	Composition (Balance Tantalum), w/o
102	100 tantalum	76	2.0 beryllium
103	100 tantalum	78	2.0 cerium
104	100 tantalum	80	2.0 lanthanum
105	100 tantalum	82	1.5 scandium
86	2.0 titanium(a)	84	2.0 uranium
101	6.0 titanium	89	0.5 thorium
92	3.0 iron	91	1.0 iron
94	3.0 silicon	93	1.0 silicon
95	0.5 hafnium		
96	1.5 hafnium		
97	3.0 hafnium		
98	0.5 zirconium		
99	1.5 zirconium		
100	3.0 zirconium		

(a) Only 0.05 w/o yttrium was added to this alloy during the consolidation of the two 100-g buttons.

Sectioning

The arc-cast buttons were sectioned to provide specimens for metallographic examination and hardness measurements, chemical analyses, and stock for processing to strip. Two sections about 1/4 in. wide were sawed from opposite edges of a button, thus leaving the bulk of the material for fabrication. Care was taken to prevent overheating during sawing, which could have resulted in contamination of the material by oxygen, hydrogen, and nitrogen from the air.

Fabrication

The ultimate objective was to fabricate the sectioned arc-cast buttons to 0.030-in.-thick strip without introducing any impurities into the materials and in such a manner that a fine-grained structure could be obtained after annealing. As time progressed, procedures were improved with these two objectives in mind.

Cleaning

Special cleaning procedures were employed (1) between fabrication steps to remove surface contamination picked up during processing and (2) to prepare the alloys for vacuum annealing. The general cleaning procedure was as follows:

- (1) Wash in a boiling Alconox (wetting agent) solution (about 10 g of Alconox per liter) for 20 min⁽⁴⁾
- (2) Rinse with water and cp acetone
- (3) Pickle in aqua regia (2 parts HCl-1 part HNO₃) for 20 min
- (4) Rinse with water, alcohol, and then cp acetone.

There were some variations used in the cleaning procedure from time to time. For example, in the first series of alloys, the materials were pickled in a "Bright Dip" solution (55 parts H₂SO₄-25 parts HNO₃-20 parts Hf) rather than in aqua regia. However, tests conducted at LASL indicated that exposures greater than 15 sec in the Bright Dip solution resulted in gas pickup in the tantalum. ⁽¹⁾ On the other hand, exposures up to 30 min in aqua regia did not cause any contamination. As a result of their finding, LASL advised against use of the Bright Dip solution for exposures greater than a few seconds. The cold-rolled materials of the third series were given the general cleaning treatment indicated previously plus a 5-sec immersion in the Bright Dip solution, for the purpose of improving the surface finish.

Pressing

The arc-cast buttons were pressed to break up the large-grained cast structure, and to flatten the convex surfaces in order to facilitate machining and to cut down

material losses. The pressing was carried out on a 700-ton hydraulic press. The buttons were placed between two hardened steel plates that had been cleaned thoroughly with acetone and coated with melted Palma Shield lubricant (an artificial substitute for palm oil) to prevent iron pickup. The buttons were pressed at room temperature at an approximate rate of 8 in. per min. In general, a load of about 100 to 150 tons was required to press the buttons from a thickness of about 1/2 to 1/4 in.

Table 5 contains a tabulation of the thicknesses of the various size buttons in each alloy series after pressing and the amount of reduction. The per cent reduction is not accurate since the buttons had a convex surface to begin with. The figures merely serve as an indication of the amount of reduction given the buttons.

TABLE 5. THICKNESS AND PER CENT REDUCTION OF TANTALUM AND TANTALUM ALLOYS ACCORDING TO ALLOY SERIES AFTER PRESSING

Alloy Series	100-g Buttons		150-g Buttons		200-g Buttons		250-g Buttons	
	Thickness, in.	Reduction, per cent	Thickness, in.	Reduction, per cent	Thickness, in.	Reduction, per cent	Thickness, in.	Reduction, per cent
1	Buttons were not pressed							
2	--	--	0.160-0.215	28-46	--	--	0.200-0.255	33-65
3	--	--	0.208-0.270	19-45	--	--	0.274-0.292	22-37
4	0.170-0.202	19-28	--	--	0.227-0.266	19-46	--	--

Machining

All of the arc-cast buttons were machined to remove surface contamination picked up during melting and pressing and to improve the surface finish for subsequent cold rolling. To obtain the desired smooth finish on the buttons a special high-speed tool was developed in a brief machining study. This tool was used with a hydraulic shaper having an automatic clapper. Machining conditions were: (1) 60-stroke-per-min surface speed, (2) 0.003 to 0.005-in. feed, and (3) 0.005-in. cut. A water-soluble oil was used as a spray-mist lubricant during machining. The edges of a button, where sections had been removed for various evaluations, were machined parallel to facilitate easier chucking in the shaper. Then each face of a button was machined with a minimum amount of material removed. However, not less than 0.010 in. was removed from each face. Table 6 lists the thicknesses of the alloys by series after being machined.

TABLE 6. THICKNESSES OF TANTALUM AND TANTALUM-BASE ALLOYS (BY SERIES) AFTER MACHINING

Alloy Series	Thickness, in., of Button Size Indicated			
	100 G	150 G	200 G	250 G
1	--	--	0.229-0.339	--
2	--	0.146-0.196	--	0.160-0.205
3	--	0.194-0.261	--	0.254-0.272
4	0.139-0.189	--	0.211-0.255	--

All of the alloys machined very well. In fact, the surfaces were generally so smooth that large grains were visible with the naked eye.

Cold Rolling

With the exception of the first series of alloys which were rolled directly without intermediate annealing to 0.030-in. -thick strip, the alloys were taken through two rolling steps. The first step consisted of rolling the machined alloy buttons to thicknesses ranging from about 0.150 to 0.200 in. , followed by stress relief or intermediate vacuum annealing. Then, after the vacuum-annealing treatment, the second rolling step was to reduce the materials to 0.030-in. -thick strip. The cold-rolled strip was used for (1) providing LASL with cold-rolled specimens for corrosion testing, (2) conducting annealing studies, and (3) providing material for vacuum annealing.

The alloys were rolled on an 8 by 12-in. mill using flat rolls. The rolls were cleaned thoroughly with acetone and then coated with melted Palma Shield lubricant to prevent iron pickup. (4) The surfaces of the alloys were also coated with the melted lubricant prior to each pass. A nominal reduction of 10 per cent was taken per pass.

Significant changes were made in the rolling procedure from one series of alloys to the next. These changes were made for the purpose of obtaining the desired microstructure in the 0.030-in. annealed material. Therefore, the rolling procedures are discussed in greater detail in the following paragraphs.

The alloys of the first series were rolled in one direction to 0.030-in. strip after the machining operation. A total reduction of 90 per cent was made. Work-hardening effects were studied and are discussed later.

Alloys in the second series were alternately cross rolled and rolled lengthwise to a thickness of from 0.163 to 0.193 in. with total reductions up to 15 per cent. Then, the alloys were given an intermediate anneal to relieve stresses. Finally, the alloys were unidirectionally (lengthwise) cold rolled to 0.030-in. strip with total reductions of from 80 to 85 per cent.

The machined alloy buttons of the third series were unidirectionally (lengthwise) cold rolled to a thickness of from 0.142 to 0.148 in. with total reductions of from 25 to 47 per cent. After a stress-relief anneal, these alloys were alternately cross rolled and rolled lengthwise to 0.030-in. strip, the total reduction being about 80 per cent. Considerable difficulty was experienced in obtaining the desired microstructure in this series of alloys, so the procedure was changed slightly for the fourth and final series of alloys.

The machined arc-cast buttons of the fourth series were rolled lengthwise and crosswise alternately, using equal reductions in both directions, down to a thickness of from 0.143 to 0.154 in. This represented a total reduction of 33 to 41 per cent. Alloy 85A was not rolled, since it was already less than 0.150 in. thick. All of the alloys of this series were given a stress-relief anneal and then alternately cross rolled and rolled lengthwise to 0.030-in. strip, a total reduction of 80 per cent. Alloys 85, 87 to 100, 102, and 103 were rolled with equal reductions in both directions. The remaining alloys of this series were not rolled with equal reductions per direction.

The rolling behavior of the alloys was observed and is discussed later. After evaluation at Battelle, cold-rolled specimens were sent to LASL for corrosion testing.

Vacuum Annealing

Vacuum annealing of the tantalum and tantalum-base alloys was necessary in order to prevent contamination from the environment. For example, tantalum absorbs hydrogen, nitrogen, and oxygen from the air at high rates, especially in the annealing-temperature range of 2600 F. (5) The objectives of the annealing treatments were (1) to relieve stresses at an intermediate stage of processing, (2) to investigate the effects of a high-temperature homogenizing treatment at an intermediate stage of processing and (3) to obtain recrystallized fine-grained materials for plutonium-alloy-fuel corrosion testing.

Prior to annealing, the materials were cleaned thoroughly, as indicated previously, and then handled with clean tongs to prevent contaminating with grease, which would be absorbed during heating. Three types of vacuum furnaces were used during the course of the program: a tantalum-element resistance furnace, a tungsten-element resistance furnace, and a high-frequency induction furnace having a clear fused-quartz work tube. The furnaces were cleaned thoroughly prior to each annealing run. The lowest possible furnace pressure was obtained and the leak rate was checked before heating the specimens. The leak rate was limited to a maximum of 0.2μ (2×10^{-4} mm of mercury) per min. The materials were annealed at the selected temperature for specified periods of time. Temperature of the specimens was measured by means of an optical pyrometer. Corrections were made for the emissivity of tantalum(6) and for the sight glass.

Intermediate Annealing. As mentioned earlier, the objective of this annealing treatment was to relieve stresses introduced from the cold-pressing and cold-rolling operations, and to investigate the effects of a high-temperature homogenizing treatment. Table 7 summarizes the intermediate annealing treatments performed on the various series of alloys.

Final Annealing. In order to provide recrystallized, fine-grained materials for plutonium-fuel corrosion tests LASL and evaluational tests at Battelle, it was necessary to vacuum anneal specimens of the cold-rolled 0.030-in. strip material. Studies to determine optimum annealing temperatures were conducted on the first and fourth series of alloys. On the other hand, the actual corrosion-test specimens of the second and third series were annealed at various temperatures until the desired structures were obtained. Although an annealing study was conducted on the fourth alloy series, only the corrosion-test specimens of Alloys 95 to 100 were annealed at Battelle because of the termination of the program. Therefore, the remainder of these materials were sent to LASL for final annealing.

A complete breakdown of the final annealing temperatures and times is given in Table 14. Pressures ranged from 3.5×10^{-6} to 7×10^{-5} mm of mercury and leak rates were in the order of from 0.003 to 0.04μ per min. The results of annealing studies are presented and discussed later. Annealed materials were evaluated at Battelle by means of (1) metallographic examination, (2) hardness measurements, (3) chemical analyses, and (4) tensile tests. Then, specimens were forwarded to LASL for corrosion testing in plutonium-alloy fuel mixtures.

TABLE 7. SUMMARY OF INTERMEDIATE VACUUM-ANNEALING TREATMENTS PERFORMED ON THE VARIOUS SERIES OF ALLOYS

Alloy Series	Alloy Thickness, in.	Temperature, F	Time, hr	Starting Pressure, mm of mercury	Leak Rate, μ per min
<u>Stress-Relief Anneal</u>					
1(a)	--	--	--	--	--
2	0.163-0.193	2600	2	$1.5-4.5 \times 10^{-6}$	0.02-0.045
3	0.142-0.148	2600	1	7.5×10^{-6} 2×10^{-5}	0.035-0.12
4(b)	0.143-0.154	2600	1	$3.5-5 \times 10^{-5}$	0.02
<u>High-Temperature Homogenizing</u>					
2(c)	0.165-0.175	4000	2	6×10^{-6}	0.02

(a) The alloys of this series were not intermediate annealed because they were processed directly to 0.030-in. strip.

(b) The pressure and leak-rate data apply only to Alloys 85, 87 to 100, 102, and 103. The remaining alloys of this series were annealed at LASL and the pressure and leak-rate data are not included here.

(c) The high-temperature homogenizing treatment was conducted only on portions of Alloys 30, 35, and 39.

EVALUATIONAL PROCEDURES

Surface Tests for Iron

Experience at Battelle and at LASL has shown that tantalum becomes contaminated superficially during fabrication, even though extreme care is taken to avoid it. The contaminant is principally iron. The special cleaning technique described previously was used to eliminate these surface-imbedded impurities. In order to determine whether the surface iron picked up during fabrication of the alloys was completely removed by the cleaning operation, it was necessary to employ some type of inspection test.

The first inspection test used was an X-ray fluorescence test. In this test the iron K-alpha intensity was measured on both sides of a 0.030-in.-thick specimen in two different areas. However, the results of these tests were rather questionable, because the internal iron count on the specimen was the same as that on the surface. It was felt that a simpler and faster qualitative test was needed to indicate the presence of surface iron contamination.

Therefore, a search was made for another test and it was learned that a potassium ferrocyanide test had been used in another program to detect the presence of iron on the surface of tantalum. (4) The procedure for this test was as follows:

- (1) A dilute solution of potassium ferrocyanide was acidified with HCl.

- (2) The tantalum or tantalum-alloy specimen was made the anode in the solution.
- (3) A carbon rod was made the cathode in the solution.
- (4) A potential of 6 v was applied to the circuit.

The presence of iron on the surface of a specimen was indicated by bright blue spots or streaks at the iron-containing areas. This test was considered satisfactory and was used on the last three series of alloys. If iron was detected, the specimens were pickled in aqua regia until free of the iron. Thus, all specimens sent to LASL for corrosion testing were free of surface iron contamination, based on this ferrocyanide test.

Chemical Analyses

Chemical analyses were performed on the materials to determine the recovery of the alloying additions and to determine the impurity levels. The analyses indicated the purity of the melting stock, which was given earlier, and the effectiveness of the arc-melting and the vacuum-annealing operations.

Chemical samples generally consisted of small sheared pieces of the material to be analyzed. These samples were usually cleaned by degreasing with cp acetone, washing in a boiling Alconox solution for 20 min, rinsing with water, alcohol and then cp acetone, and dried thoroughly by heating to vaporize the acetone.

Carbon, nitrogen, and a few of the alloying additions were determined by wet-analysis techniques. Oxygen and hydrogen contents were determined by vacuum-fusion analysis. Spectrographic techniques were used to determine the trace elements and most of the alloying additions. Special techniques were developed to improve the results of the vacuum-fusion and spectrographic analyses.

Metallography

Cross sections of the tantalum and tantalum-base alloys were generally mounted in bakelite for microstructural examination and hardness determinations. Then the sections were wet ground successively with 240-, 400-, and 600-grit silicon carbide grinding papers. Final polishing was done on a high-speed (1750 rpm) polishing wheel covered with a Buehler Mira cloth, using a slurry composed of 15 g of Linde B (Al_2O_3), 5 cm³ of 20 per cent chromic acid, and 35 cm³ of water. The specimens were then satisfactorily etched with one of the following etchants:

- (1) 30 parts lactic acid - 10 parts HNO₃ - 10 parts HF.
- (2) An electrolytic etchant consisting of 90 parts H₂SO₄ - 10 parts HF.

The microstructural characteristics of the materials were observed by means of an optical microscope. In some alloys, a second phase was observed; however, no

attempt was made to identify these phases. The large grain size of the materials in the as-cast condition was determined by (1) counting the total number of grains on the polished cross section of the specimen, (2) measuring the polished area with a polar planimeter, (3) calculating the number of grains per square millimeter, and (4) calculating the diameter of equivalent spherical grains.

The grain size of materials in the annealed condition was determined by direct comparison at 100 diameters with a standard ASTM grain-size chart. Then it was converted to average grain diameter in millimeters.

Hardness Measurements

The hardness of the alloys was determined to gain information on alloying effects, work-hardening characteristics, and response to annealing. Hardness was used also to indicate the purity of the unalloyed tantalum control specimens. Mounted sections of the alloys used for microstructural examination were also used in hardness determinations. Five to ten Vickers hardness impressions, using a 5- or 10-kg load, were taken on a polished or etched surface. The average hardness was determined from these impressions. The Vickers hardness of tensile specimens was determined from impressions taken on the surface outside the reduced section. The surface condition was that present after cold rolling and/or annealing.

Tensile Testing

The room-temperature tensile properties of the materials were determined to study the effects of alloying on strength and ductility, to investigate the influence of cold rolling and of annealing, and to provide data with which to evaluate the effect of irradiation. Tensile-specimen blanks were sheared from cold-rolled 0.030-in. strip and then ground to the following dimensions, using water as a coolant to prevent overheating and concomitant contamination:

Length, in.	$4.50 \pm 1/64$
Width, in.	$11/32 \pm 1/64$
Gage Length, in.	1.0 ± 0.002
Gage Width, in.	0.250 ± 0.002
Radius at Gage Section, in.	1.0

Most of the materials were tested in only the annealed condition. However, the tantalum and tantalum-tungsten alloys were also tested in the as-rolled condition. All of the specimens were tested with a Baldwin-Southwark universal testing machine using a strain rate of 0.005 in. per in. Templin grips were used to hold the specimens during testing. A clip-on extensometer was used with a recorder to obtain load-deformation curves.

EXPERIMENTAL RESULTS AND DISCUSSION

Arc-Casting Behavior

In general, the casting behavior of the unalloyed tantalum and tantalum-base alloys was good. Notable exceptions were the alloys containing charged additions of cerium, lanthanum, 3 w/o yttrium, 1.5 to 6 w/o thorium and 3 w/o silicon. The binary alloys containing the first three elements were difficult to form into a symmetrical button, due to the formation of "fingers". In addition, the cerium- and lanthanum-containing alloys had several voids. However, they were not remelted for fear of further vaporization of the alloy additions. The thorium-containing alloys contained a great deal of porosity which was not eliminated by remelting the buttons. The alloy containing 3 w/o silicon broke into several pieces during the cooling period after casting.

Fabrication Behavior

The relative fabrication behavior of the materials is indicated in Table 13. In general, the tantalum and tantalum-base alloys could be fabricated at room temperature very easily into good quality 0.030-in. strip. The poorest behavior of the fabricable alloys was exhibited by those containing charged additions of 6 w/o hafnium, 6 w/o rhenium, 0.5 to 6 w/o thorium, and 3 w/o zirconium. The poorest quality strip was that of the thorium-containing alloys.

As indicated in Table 13, the alloys with additions of from 0.5 to 1.0 w/o boron, 0.1 to 0.25 w/o carbon, and 1.0 w/o silicon were not fabricable. The extremely hard alloys with boron and silicon cracked into several pieces when the load was applied during the pressing operation. Although the alloys with carbon survived the pressing stage, they cracked catastrophically during the first cold-rolling step with a total reduction of about 32 per cent. Their failure was attributed to the presence of excessive amounts of a grain-boundary phase, presumably a carbide.

The work hardening of a few of the unalloyed tantalum buttons and tantalum-tungsten alloys of the first series was studied. The results are shown in Table 8 and in Figure 2. These data show that, in general, the materials had a relatively low rate of work hardening during reductions of from 30 to 60 per cent. However, this rate increased fairly rapidly with reductions of 60 per cent and greater. It is interesting to note that the rates of work hardening were about the same for the unalloyed tantalum specimens. The rates of work hardening of the tungsten-containing alloys seemed, in some cases, to be slightly higher than those for the unalloyed tantalum.

Annealing Studies

As mentioned previously, studies were conducted on the first and fourth series of alloys to determine an optimum annealing temperature, i. e., a temperature at which a fine-grained structure of about minimum hardness could be obtained. In these studies, observations were made of the effect of annealing temperature on the hardness, and on the grain size, and also the effect of alloying on the recrystallization temperature. The

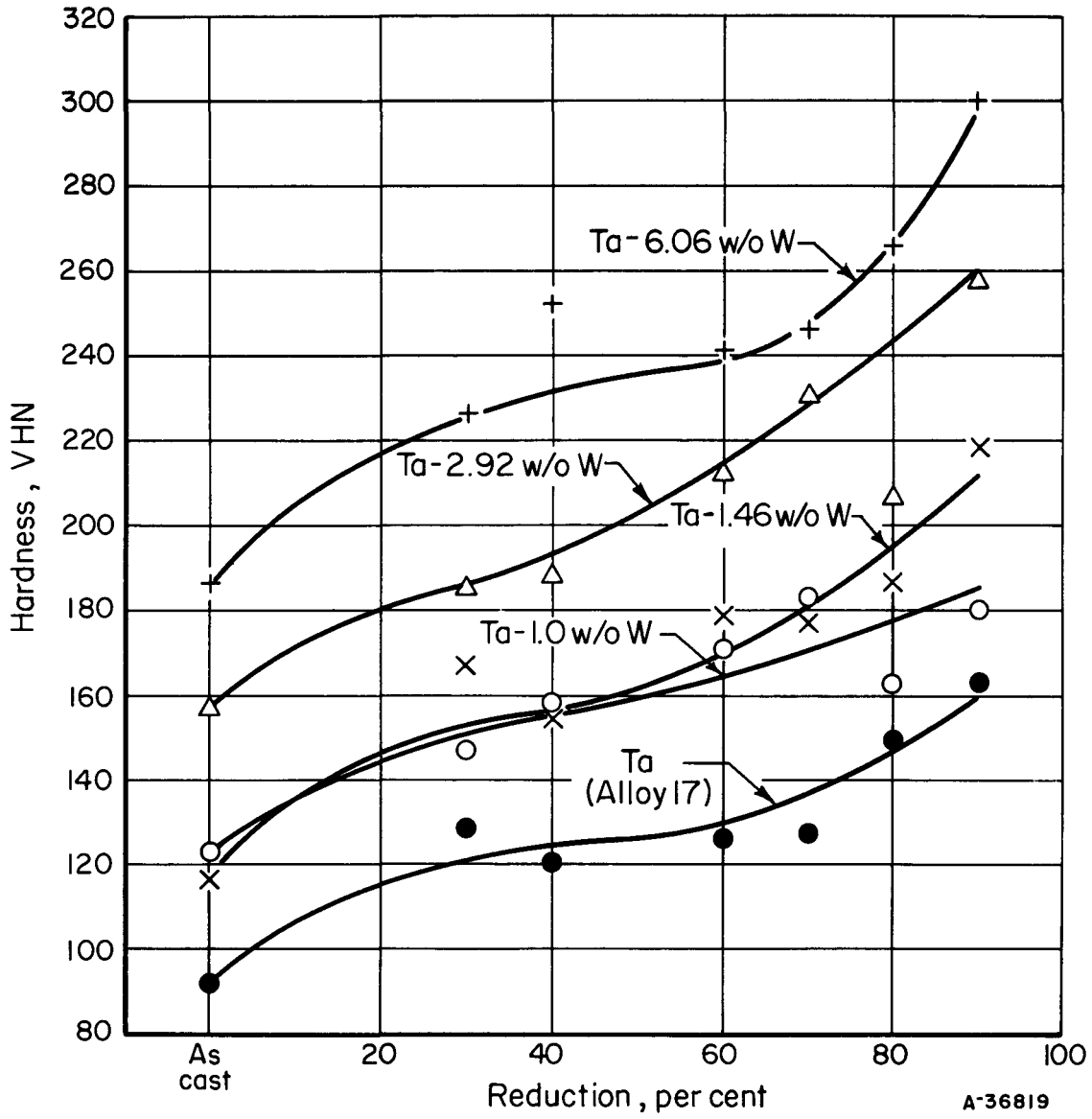


FIGURE 2. EFFECT OF COLD DEFORMATION ON THE HARDNESS OF TANTALUM AND TANTALUM-TUNGSTEN ALLOYS

results of the annealing studies are shown in Tables 9 and 10 and Figures 3 and 4. In Tables 9 and 10, the optimum annealing temperature is indicated for the materials and a time of 1 hr at temperature is suggested. In general, an increase in the annealing temperature produced softer materials and larger grains.

TABLE 8. EFFECT OF COLD DEFORMATION ON THE HARDNESS OF TANTALUM AND TANTALUM-TUNGSTEN ALLOYS

Alloy	Composition (Balance Tantalum), w/o	Hardness, VHN						
		As Cast	After Total Percentage Reduction Shown					
			30	40	60	70	80	90
11	100 Ta	106	181	135	161	146	201	171
17	100 Ta	92	129	121	127	128	150	163
10	1.0 W ^(a)	123	147	158	171	183	163	180
18	1.46 W	116	166	152	178	177	187	218
4	2.92 W	157	185	188	212	230	206	258
9	6.06 W	186	225	252	241	246	266	300

(a) The tungsten in this alloy was a result of contamination from the electrode during arc melting.

TABLE 9. EFFECT OF ANNEALING TEMPERATURE ON THE HARDNESS AND GRAIN SIZE OF TANTALUM AND TANTALUM-TUNGSTEN ALLOYS

Alloy	Composition (Balance Tantalum), w/o	Hardness, As Cold Rolled, VHN	Hardness, VHN, and Average Grain Diameter, mm, After Annealing at Indicated Temperature ^(a)							Optimum Annealing Temperature ^(b) , F
			2010 F	2205 F	2415 F	2610 F	2815 F	3000 F	3270 F	
			16	100 Ta	171	121	87	75	74	
			<0.02	0.05	0.07	0.07	0.10	0.28	>0.3	
18	1.46 W	227	169	114	92	94	94	97	85	2600
			<0.02	0.05	0.07	0.07	0.10	0.14	0.28	
5	2.71 W	243	220	176	119	116	114	117	109	2600
			<0.02	0.05	0.07	0.07	0.10	0.10	0.14	
7	6.23 W	299	276	268	180	177	177	172	164	2600
			<0.02	0.03	0.05	0.05	0.07	0.07	0.12	

(a) Vacuum annealed 30 min for each alloy, the first line contains hardness data; the second line contains grain-size data.

(b) Temperature required to produce a fine-grained structure of low hardness. Annealing for 1 hr is recommended.

In the case of the unalloyed tantalum and the tantalum-tungsten alloys of the first series, the indications are that all of the materials were completely recrystallized after 30 min at 2400 F. Then, between 2400 and 2800 F, no significant change in the hardness or grain size was observed. However, above 2800 F the grain size increased greatly with only a slight decrease in hardness. The grain growth was more pronounced in the unalloyed tantalum. Increases in tungsten content had a progressively retarding effect on the grain growth. An optimum annealing temperature of 2600 F was selected for all the materials of the first series of alloys.

TABLE 10. EFFECT OF ANNEALING TEMPERATURE ON THE HARDNESS OF BINARY TANTALUM-BASE ALLOYS

Alloy	Composition (Balance Tantalum), w/o	Hardness, VHN							Optimum Annealing Temperature ^(a) , F
		As Cold Rolled	After Vacuum Annealing 30 Min at Indicated Temperature						
			2200 F	2400 F	2600 F	2800 F	3000 F	3200 F	
<u>Fourth Series, Group 1</u>									
85	0.005 Y	147	98	83	80	79	89	--	2600
89	0.4 Th	172	161	158	140	125	115	--	3200 ^(b)
90	1.2 Th	171	160	156	145	133	121	--	3200 ^(b)
87	2.7 Th	203	169	167	146	135	119	--	3200 ^(b)
88	5.6 Th	192	157	155	139	129	110	--	3200 ^(b)
91	<0.01 Fe	165	108	94	88	77	88	--	2600
92	0.015 Fe	198	133	116	117	111	119	--	2600
95	0.5 Hf	192	161	124	118	105	107	--	3000
96	1.5 Hf	214	169	136	123	124	124	--	3000
97	3.0 Hf	251	212	188	144	144	144	--	3000
98	0.43 Zr	197	195	134	119	114	117	--	3000
99	1.3 Zr	257	192	140	139	135	135	--	3000
100	2.9 Zr	293	188	174	182	190	193	--	3000
102	100 Ta (control)	165	87	81	75	73	76	--	2600
103	100 Ta (control)	190	115	93	88	87	113	--	2600
<u>Fourth Series, Group 2</u>									
71	<0.001 Al	177	--	122	95	92	92	95	2800
72	<0.001 Al	171	--	127	105	102	100	108	2800
76	0.01 Be	208	--	154	127	118	109	109	3200
78	<0.10 Ce	139	--	115	100	96	100	109	3200
80	<0.03 La	162	--	132	98	107	105	103	3000
82	<0.10 Sc	179	--	160	130	122	118	129	3000
84	1.2 U	186	--	132	118	111	112	115	2800
86	0.01 Ti	168	--	104	105	107	103	106	3200
101	0.34 Ti	187	--	140	118	120	125	131	2800
109	0.001 Y	161	--	151	157	146	117	139	2800
110	<0.001 Y	181	--	140	165	173	179	173	2800
104	100 Ta (control)	193	--	126	118	114	117	114	3200
105	100 Ta (control)	120	--	83	95	91	89	95	2600

(a) Temperature required to produce a five-grained structure. Annealing for 1 hr is recommended.

(b) Estimated because recrystallization was not complete at the highest temperature used in the study.

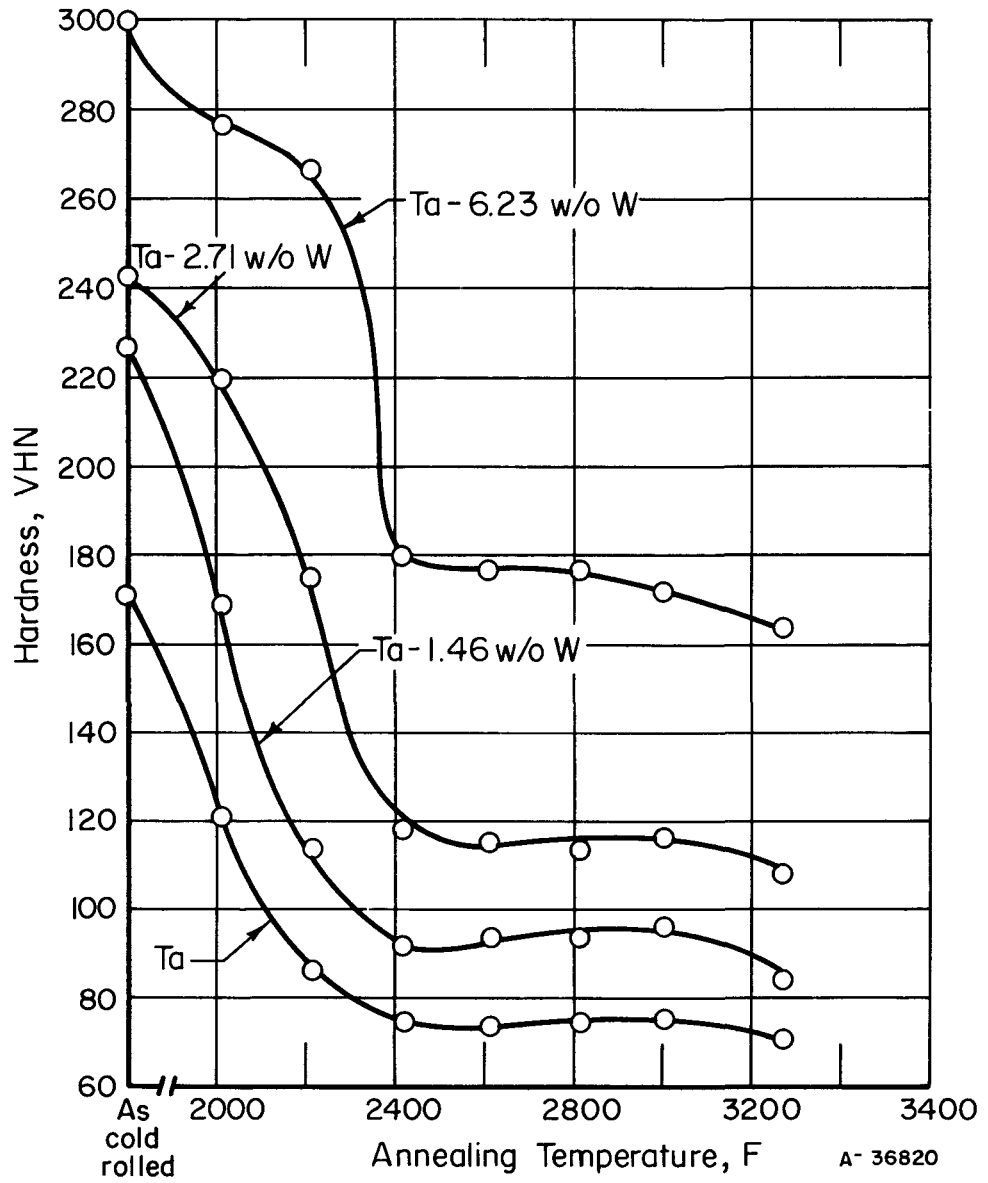


FIGURE 3. EFFECT OF ANNEALING TEMPERATURE ON THE HARDNESS OF TANTALUM AND TANTALUM-TUNGSTEN ALLOYS

Specimens cold rolled (90 per cent reduction) prior to annealing in vacuo for 30 min.

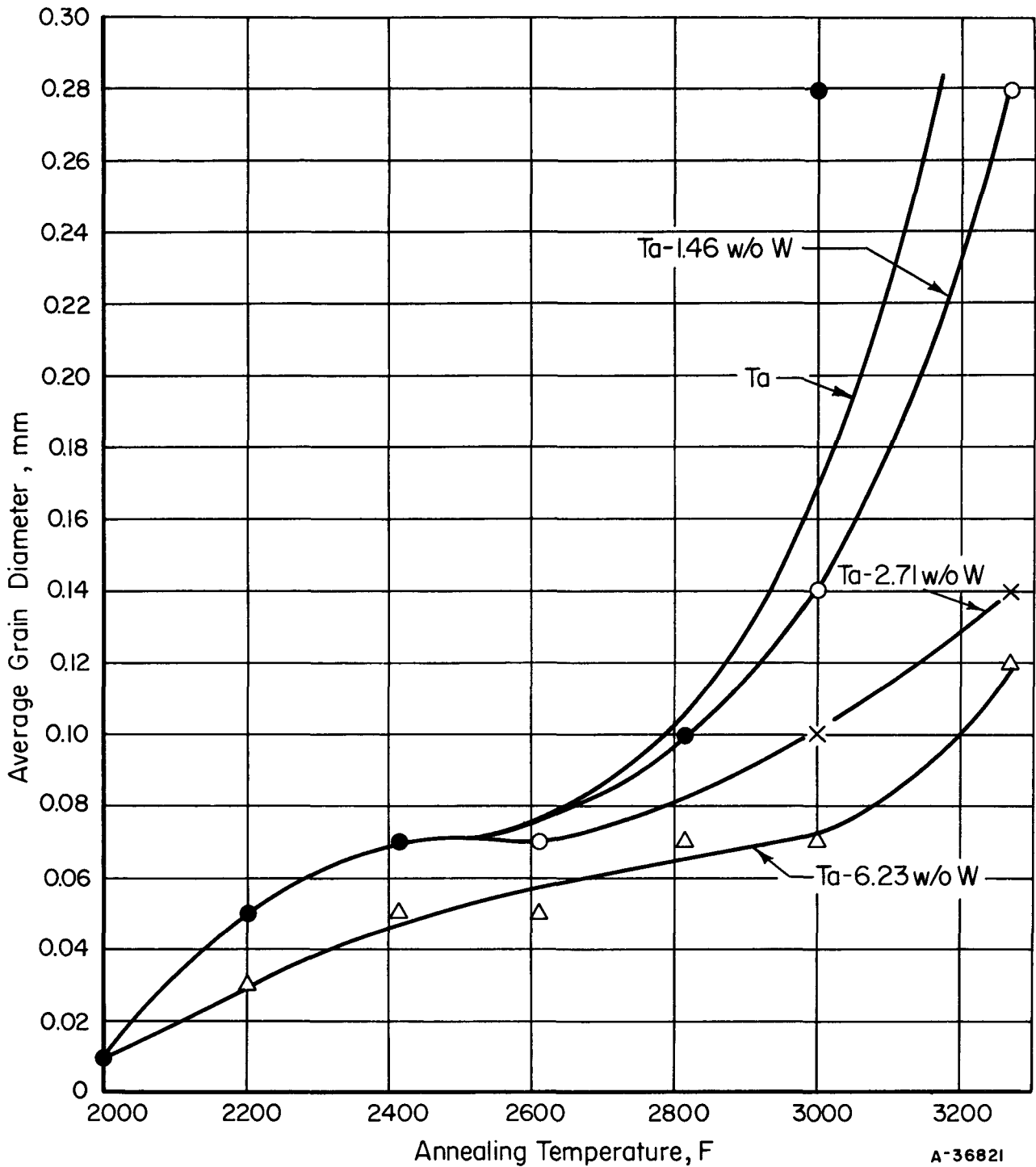


FIGURE 4. EFFECT OF ANNEALING TEMPERATURE ON THE GRAIN SIZE OF TANTALUM AND TANTALUM-TUNGSTEN ALLOYS

Specimens cold rolled (90 per cent reduction) prior to annealing
in vacuo for 30 min.

Three of the unalloyed tantalum control specimens of the fourth series were also satisfactorily recrystallized at 2600 F. However, one of the tantalum specimens (Alloy 104) was not completely recrystallized until treated at 3200 F. The cause of its resistance to recrystallization was not determined. The variations in the hardness of the tantalum specimens was probably due to differences in impurity levels. However, chemical analyses were not performed to substantiate this.

It was difficult to select an optimum annealing temperature for two of the yttrium-containing alloys (109 and 110) and those containing scandium and titanium, because of elongated grain structures and/or excessive banding (inhomogeneity).

The data in Table 10 indicate that optimum annealing temperature was increased significantly in alloys containing additions of beryllium, cerium, hafnium, lanthanum, thorium, and zirconium. For the tantalum-hafnium alloys, there was no significant change in hardness between 2600 and 3000 F. The alloys containing thorium, as indicated in Table 10, were not completely recrystallized at 3000 F. For these alloys, the optimum temperature was estimated as 3200 F, because the hardness was still decreasing at 3000 F. In the case of the tantalum-zirconium alloys, the data indicate that there was no significant change in hardness for the 0.43 w/o zirconium alloy between 2600 and 3000 F and for the 1.3 w/o zirconium alloy between 2400 and 3000 F. The hardness of the 2.9 w/o zirconium alloy increased from a minimum of 174 VHN to a maximum of 193 VHN at 3000 F. The cause of this is not known, but it may have been due to the gettering action of the alloy during annealing.

Chemical Analyses

Because purity was considered as being extremely important in reference to the corrosion behavior of tantalum and its alloys in the plutonium fuels, most of the alloys were subjected to complete analyses in one or more of the following conditions: as cast, as rolled, and as annealed. Table 11 contains the results of the analyses performed on the materials.

In general, recovery of the following additions to tantalum was good: carbon, hafnium, rhenium, thorium, 0.025 w/o titanium, tungsten, uranium, and zirconium. However, the data show that the recovery of titanium in alloys containing charged additions of from 0.05 to 6 w/o was very poor. Also, practically all of the additions of aluminum, beryllium, cerium, iron, lanthanum, scandium, and yttrium were lost during arc melting. Several attempts were made to recover yttrium by using special melting practices, but without success.

In general, as indicated by the analytical data, the arc-melting procedures were effective in maintaining purity of the melting stock. However, some difficulty was experienced with tungsten contamination from the tungsten electrode. This is shown in the results for the unalloyed tantalum alloy, 10, the hafnium-containing alloys (46 through 50, and 56) and the zirconium-containing alloys (53 and 54). LASL was especially interested in hafnium and zirconium and wanted to evaluate the effect of each, per se, on the corrosion resistance of tantalum. It was for this reason that another group of tantalum-hafnium and tantalum-zirconium alloys (95 through 100) was prepared later. The difficulty with tungsten contamination in the alloys was eliminated by conditioning the electrode with yttrium. A discussion of the use of yttrium to prevent tip loss was presented in a previous section concerned with arc melting.

TABLE 11. (Continued)

Alloy	Composition (Balance Tantalum), w/o			Analyses, ppm													
	Element	Charged	Actual	C	H	N	O	Fe	Si	Cu	W	Nb	Zr	Al	Mo	Ni	Cr
91	Fe	1.0	<0.01	--	--	--	--	--	<10	<10	<300	<50	<10	--	--	--	<10
92	Fe	3.0	0.015	--	--	--	--	--	<10	<10	<300	<50	<10	--	--	--	<10
95	Hf	0.5	0.5	--	--	--	--	<10	<10	<10	<300	<50	<10	--	--	--	<10
96	Hf	1.5	1.5	--	--	--	--	<10	<10	<10	<300	<50	<10	--	--	--	<10
97	Hf	3.0	3.0	--	--	--	--	<10	<10	<10	<300	<50	<10	--	--	--	<10
98	Zr	0.5	0.43	--	--	--	--	<10	<10	<10	<300	<50	--	--	--	--	<10
99	Zr	1.5	1.3	--	--	--	--	<10	<10	<10	<300	<50	--	--	--	--	<10
100	Zr	3.0	2.9	--	--	--	--	<10	<10	<10	<300	<50	--	--	--	--	<10
<u>Fourth Series (Group 2)^(d): As Rolled</u>																	
104	--	100 Ta (control)	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--
105	--	100 Ta (control)	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--
71	Al	0.5	<0.001	--	--	--	--	--	--	--	--	--	--	--	--	--	--
72	Al	1.0	<0.001	--	--	--	--	--	--	--	--	--	--	--	--	--	--
76	Be	2.0	0.01	--	--	--	--	--	--	--	--	--	--	--	--	--	--
78	Ce	2.0	<0.10	--	--	--	--	--	--	--	--	--	--	--	--	--	--
80	La	2.0	<0.03	--	--	--	--	--	--	--	--	--	--	--	--	--	--
82	Sc	1.5	<0.10	--	--	--	--	--	--	--	--	--	--	--	--	--	--
84	U	2.0	1.20	--	--	--	--	--	--	--	--	--	--	--	--	--	--
86	Ti	2.0	0.01	--	--	--	--	--	--	--	--	--	--	--	--	--	--
101	Ti	6.0	0.34	--	--	--	--	--	--	--	--	--	--	--	--	--	--
109	Y	3.0	0.001	--	--	--	--	--	--	--	--	--	--	--	--	--	--
110	Y	3.0	<0.001	--	--	--	--	--	--	--	--	--	--	--	--	--	--

(a) Value is in w/o.

(b) Annealed 2 hr at 2600 F.

(c) Refer to Table 13 for annealing conditions.

(d) Analyses for trace elements and interstitials were not performed on this group of alloys.

The high carbon content in two of the hafnium alloys (49 and 50) was probably caused by overheating the Wilson vacuum seal on top of the arc-melting furnace. The cause of the unusually high nitrogen levels, 100 and 220 ppm, in Alloys 49 and 51 was not determined, but it was thought to have been due to a slight air leak during melting.

An interesting phenomenon is seen on comparing the oxygen contents of the cold-rolled materials of the third series of alloys with the amount of alloying element present. Generally speaking, the oxygen content decreased significantly as the amount of the alloying element - hafnium, thorium, titanium, yttrium, zirconium - increased. This suggests that all of these elements were effective scavengers of oxygen in tantalum.

A comparison of the analytical data for the rolled and the annealed materials shows that vacuum annealing at 2600 to 2750 F resulted in a slight increase in nitrogen and oxygen levels and a decrease in hydrogen. Annealing at 2900 F caused a substantial increase in gaseous contamination. The increase in carbon levels in some of the annealed materials was believed to have been caused by the back flow of diffusion-pump oil.

Microstructure

All of the materials prepared in this program were examined metallographically to observe the characteristics of the alloys in reference to the type structure (single phase or two phase), homogeneity, and grain size.

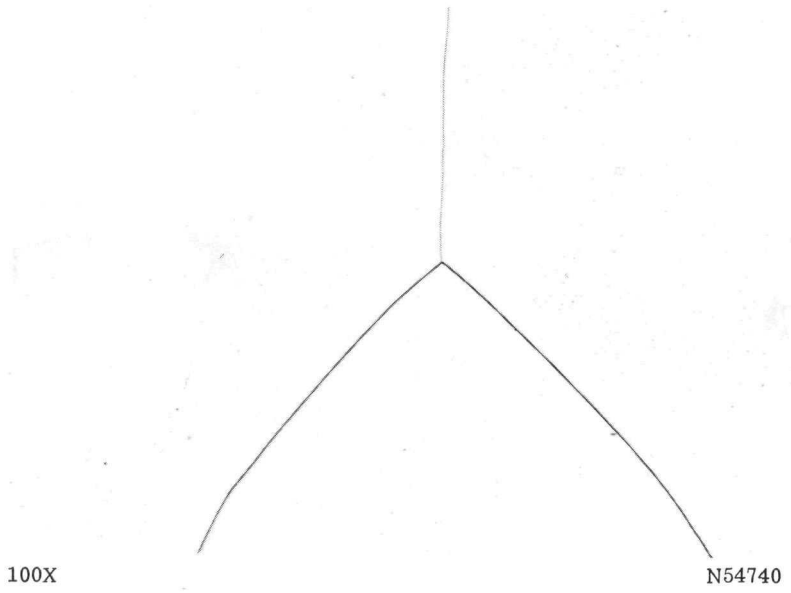
Table 12 summarizes the microstructures observed for the unalloyed tantalum and all of the binary tantalum-base alloys. Figure 5 shows the microstructure of a typical unalloyed high-purity tantalum control specimen (Alloy 13) in the as-cast, as-rolled, and as-annealed conditions. The photomicrograph of the as-cast specimen is also representative of the single-phase tantalum-base alloys except for grain size, which varied considerably depending upon the alloy.

TABLE 12. SUMMARY OF MICROSTRUCTURES OBSERVED IN
AS-CAST TANTALUM-BASE ALLOYS

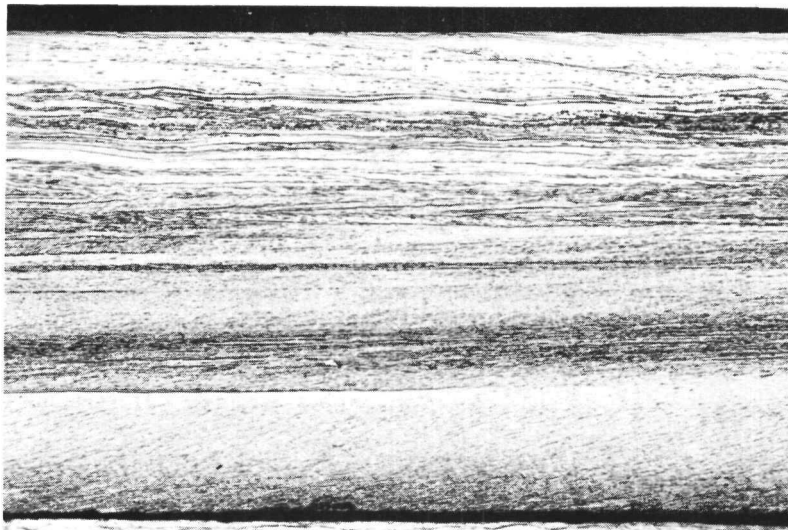
Actual Composition (Balance Tantalum), w/o	
Single-Phase Structures	Two-Phase Structures
Unalloyed Tantalum	--
<0.001 Al	--
--	0.5-1.0 B ^(a)
--	0.01 Be
--	0.028-0.031 C
--	0.10-0.25 C ^(a)
<0.10 Ce	--
<0.01-0.015 Fe	--
0.025-5.55 Hf	--
--	<0.03 La
1.5-6.2 Re	--
--	<0.10 Sc
--	1.0-3.0 Si ^(a)
0.018-0.066 Th	0.4-5.6 Th
0.015-0.34 Ti	--
1.2 U	--
1.45-6.23 W	--
0.005 Y	0.001 Y ^(b)
0.021-0.43 Zr	1.24-2.9 Zr

(a) Nominal composition.

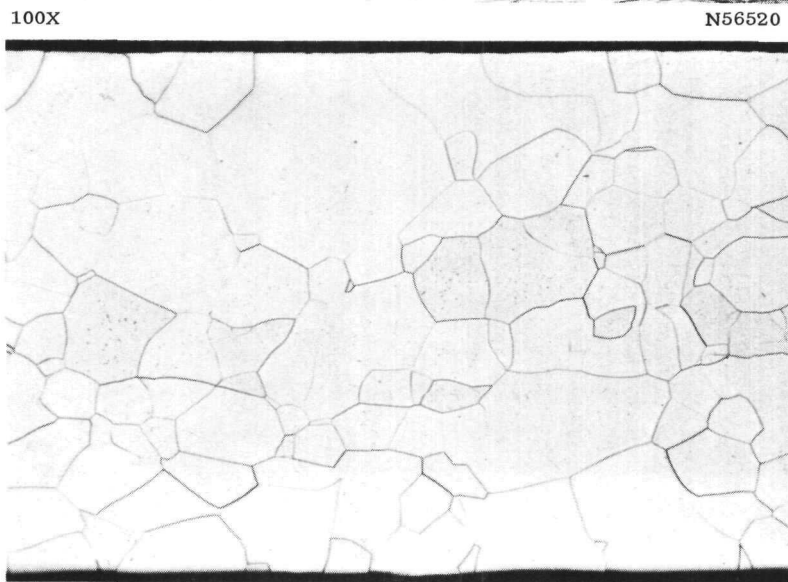
(b) Alloy 109 which showed inhomogeneity.



a. As Cast



b. Cold Rolled (90 Per Cent Reduction)



c. Vacuum Annealed 2 Hr at 2600 F After Cold Rolling

FIGURE 5. TYPICAL MICROSTRUCTURES OF ARC-MELTED HIGH-PURITY TANTALUM (ALLOY 13)

Several grain boundaries and areas in the matrix of the tantalum 1.2 w/o uranium alloy contained a second phase, although most of the alloy was a single-phase structure.

Thorium and zirconium were found to have limited solubility in tantalum. In material arc cast and rapidly cooled, the solubility limit of thorium was greater than 0.061 w/o and less than 0.40 w/o. The solid solubility of zirconium in material with a similar history was shown to be greater than 0.43 w/o and less than 1.3 w/o. The microstructures of the tantalum-thorium alloys, illustrated in Figure 6, contained an inter- and intragranular phase which increased with increased amounts of thorium from 0.4 to 5.6 w/o. Figures 7 and 8 show the microstructures of the tantalum-zirconium alloys in both the as-cast and the as-annealed conditions, respectively. Although the tantalum-0.43 w/o zirconium alloy had a single-phase structure in the as-cast condition (Figure 7a), after vacuum annealing at 3000 F for 1 hr a discontinuous phase was precipitated at the grain boundaries (Figure 8a). The 1.3 w/o zirconium alloy, shown in Figures 7b and 8b, contained a slight discontinuous inter- and intragranular phase in the as-cast condition and significantly more in the annealed condition. Similarly, the as-cast 2.9 w/o zirconium alloy contained more of what appeared to be the same discontinuous grain-boundary phase plus a very slight amount of a phase within the grains (Figure 7a). After annealing at 3000 F for 1 hr, the quantity of grain-boundary phase remained about the same. However, there was an increase in the amount of the intragranular phase (Figure 8c).

The alloy containing 0.01 w/o beryllium had a relatively small spherical-shaped phase distributed both inter- and intragranularly, as shown in Figure 9a. Also, this alloy contained an area which was segregated and this area appears in Figure 9a. Additions of from 0.5 to 1.0 w/o boron to tantalum resulted in a eutectic boride phase in the tantalum matrix. The amount of the second phase increased with the boron content. Figure 9b illustrates the microstructure of the 1 w/o boron alloy. As indicated previously, these extremely hard boron alloys could not be fabricated at room temperature. Alloys with from 0.03 to 0.25 w/o carbon contained fine particles, presumably carbides, at the grain boundaries and within the grains. The quantity of the second phase increased significantly with increasing carbon additions. Figure 9c shows the structure of the 0.03 w/o carbon alloy, which was the only one that was fabricable, in the as-cast condition. The lack of fabricability of the alloys with more than 0.03 w/o carbon was attributed to the presence of excessive amounts of intergranular phase.

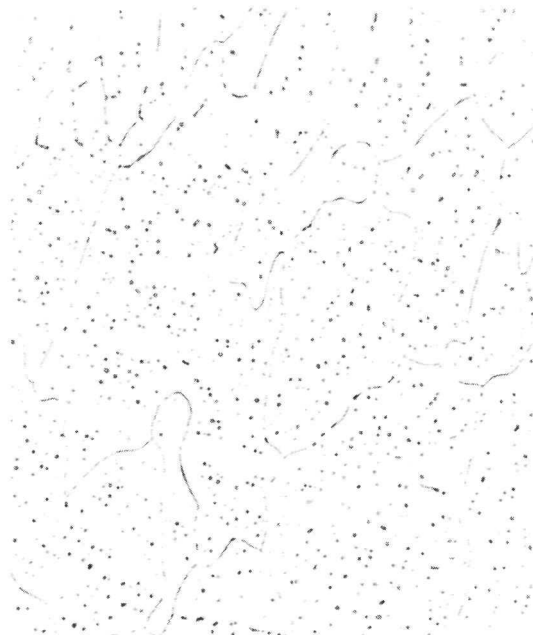
Although chemical-analysis data showed a recovery of less than 0.03 w/o lanthanum in the alloys with an intended lanthanum content of 1 w/o, the microstructure contained a large quantity of an acicular phase in the matrix and a discontinuous grain-boundary precipitate with a depleted zone near the grain boundaries (Figure 10a). Similarly, the tantalum-1.0 w/o scandium alloy, which was analyzed as less than 0.10 w/o scandium, had a large amount of a needlelike phase in the matrix and a discontinuous intergranular precipitate with a depleted zone adjacent to the grain boundaries (Figure 10b). The addition of 1.0 and 3 w/o silicon to tantalum produced structures, as shown in Figure 10c, having a very large amount of a eutectic phase similar to that of the boron alloys. The 3 w/o silicon alloy contained more of this eutectic phase, but the size was smaller. The microstructure of the inhomogeneous yttrium alloy (109) contained areas, as shown in Figure 10d, with a second phase in the matrix and a discontinuous grain-boundary precipitate.



100X

N67615

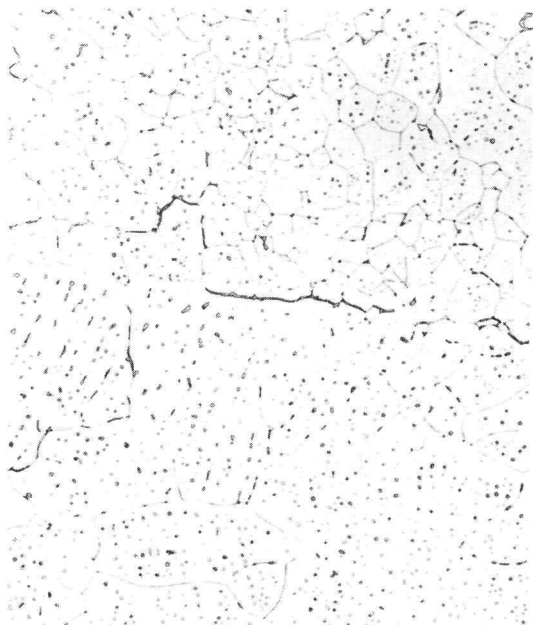
a. Tantalum-0.4 w/o Thorium (Alloy 89)



100X

N67616

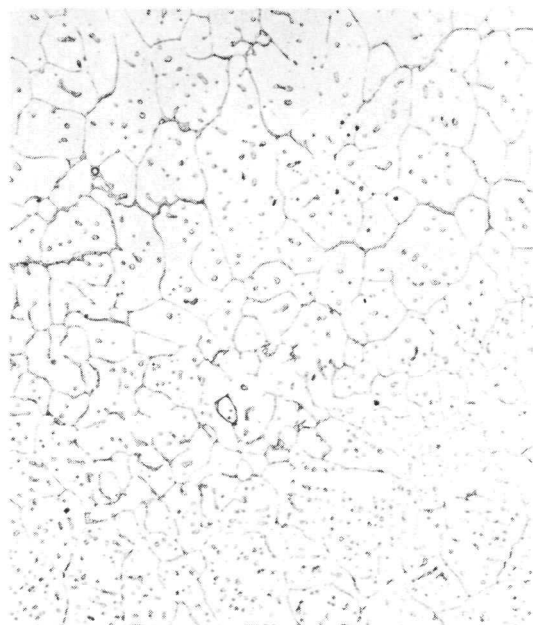
b. Tantalum-1.2 w/o Thorium (Alloy 90)



100X

N67613

c. Tantalum-2.7 w/o Thorium (Alloy 87)

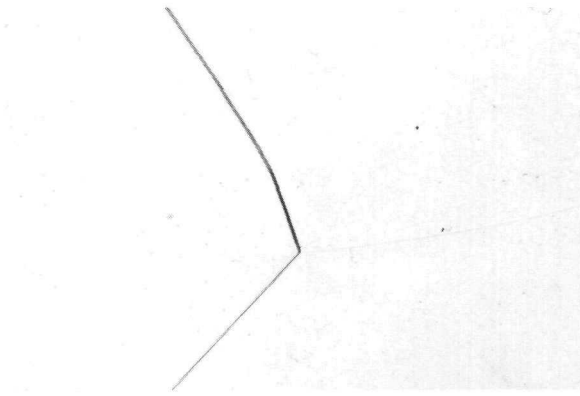


100X

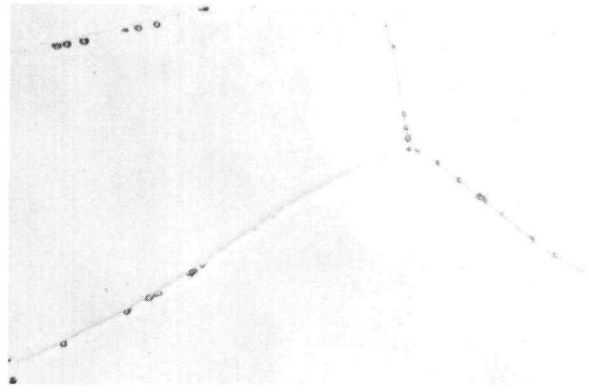
N67614

d. Tantalum-5.6 w/o Thorium (Alloy 88)

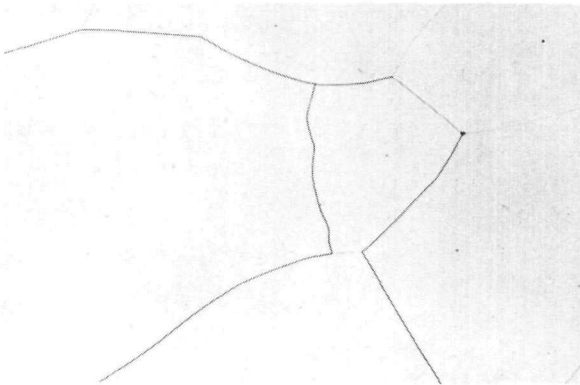
FIGURE 6. MICROSTRUCTURES OF AS-CAST TANTALUM-THORIUM ALLOYS



100X N67624
a. Tantalum-0.43 w/o Zirconium (Alloy 98)



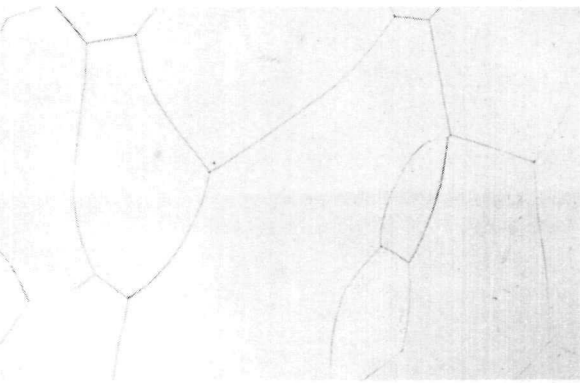
1000X N72257
a. Tantalum-0.43 w/o Zirconium (Alloy 98)



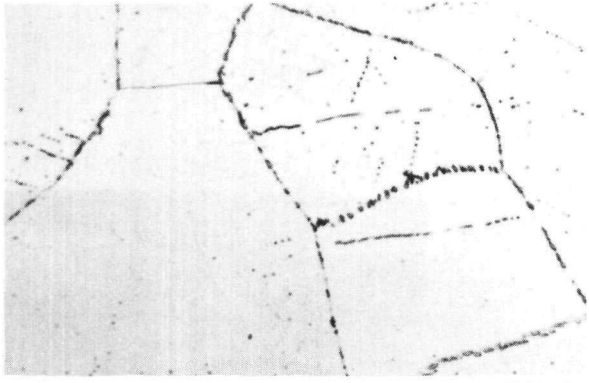
100X N67625
b. Tantalum-1.3 w/o Zirconium (Alloy 99)



1000X N72258
b. Tantalum-1.3 w/o Zirconium (Alloy 99)



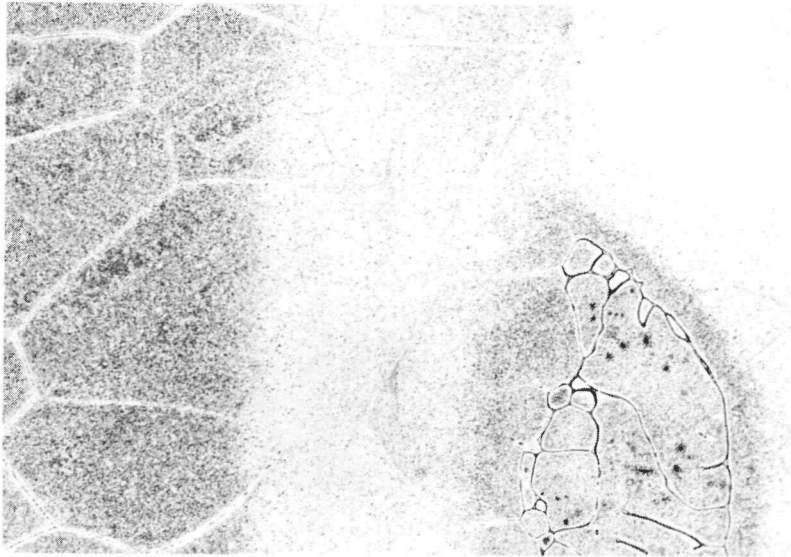
100X N67626
c. Tantalum-2.9 w/o Zirconium (Alloy 100)



1000X N72259
c. Tantalum-2.9 w/o Zirconium (Alloy 100)

FIGURE 7. MICROSTRUCTURES OF AS-CAST TANTALUM-ZIRCONIUM ALLOYS

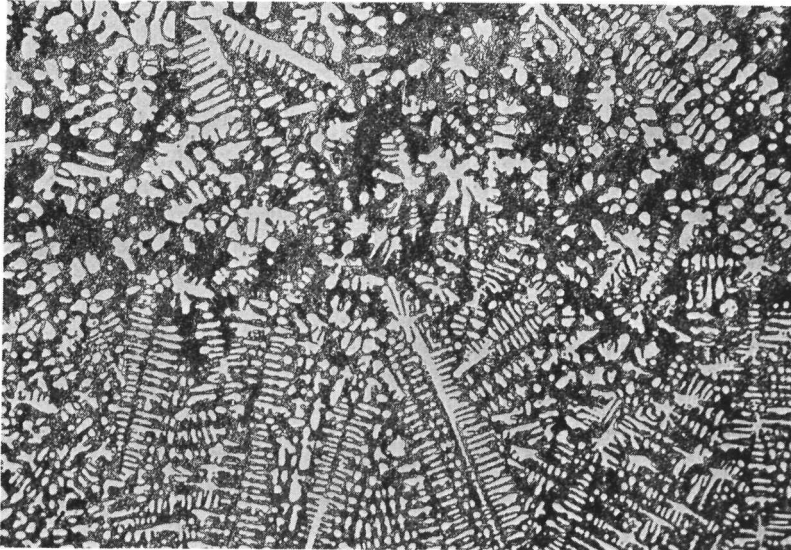
FIGURE 8. MICROSTRUCTURES OF TANTALUM-ZIRCONIUM ALLOYS AFTER VACUUM ANNEALING 1 HR AT 3000 F



a. Tantalum-0.01 w/o Beryllium

100X

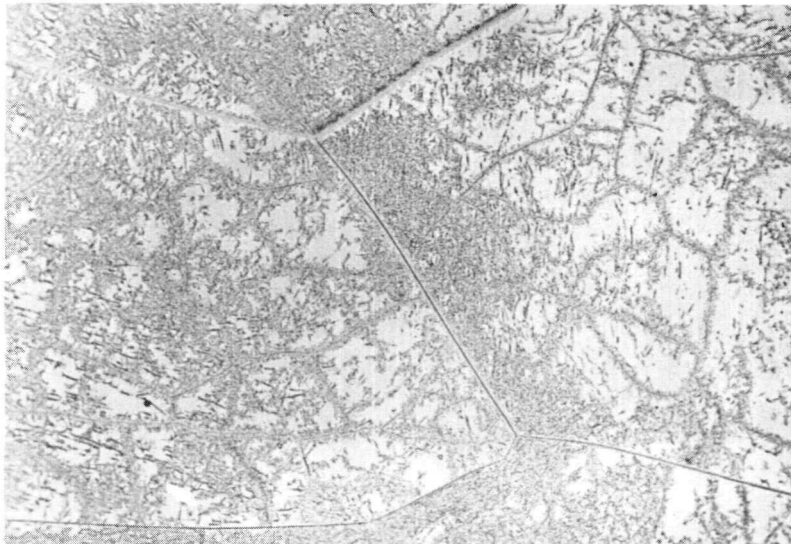
N72224



b. Tantalum-1 w/o Boron (Nominal Composition)

100X

N67611

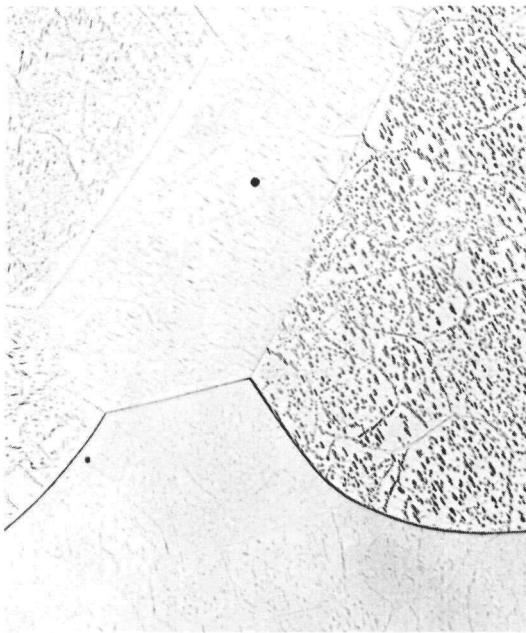


c. Tantalum-0.03 w/o Carbon

100X

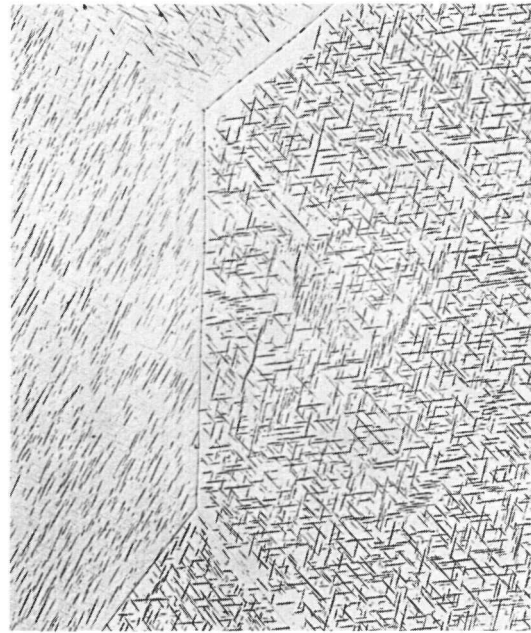
N57327

FIGURE 9. MICROSTRUCTURES OF AS-CAST TANTALUM-BASE ALLOYS



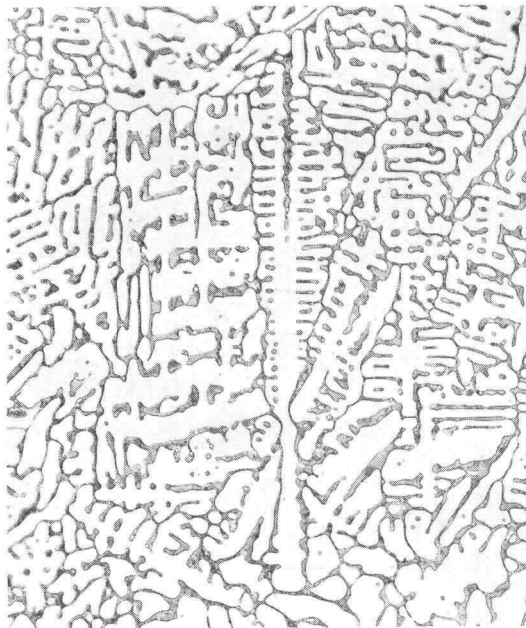
100X

N72226

a. Tantalum- <0.03 w/o Lanthanum

100X

N72227

b. Tantalum- <0.10 w/o Scandium

100X

N67619

c. Tantalum-1 w/o Silicon (Nominal Composition)



100X

N72233

d. Tantalum-0.001 w/o Yttrium (Alloy 109)

FIGURE 10. MICROSTRUCTURES OF AS-CAST TANTALUM-BASE ALLOYS

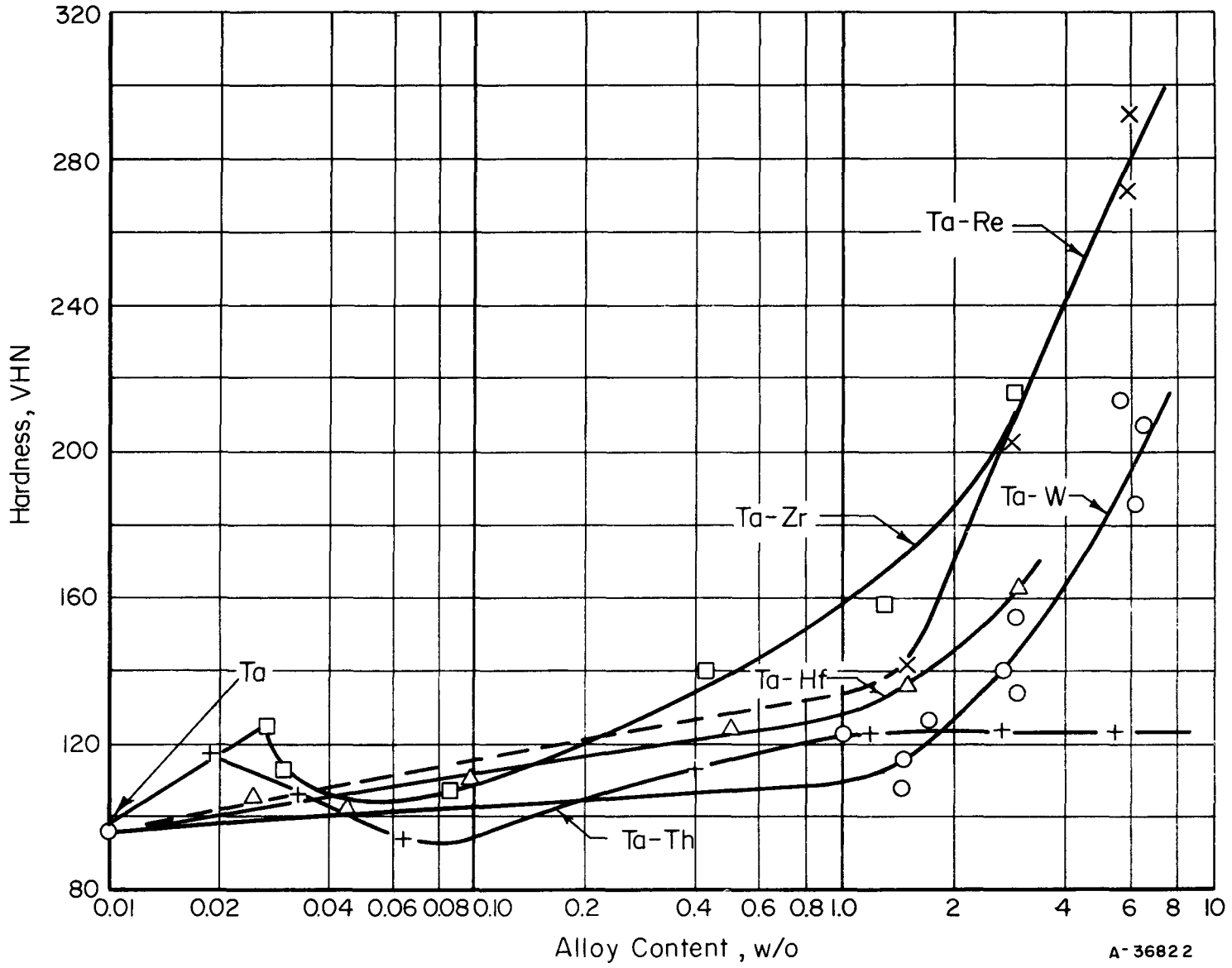


FIGURE 11. EFFECTS OF ALLOYING ON THE HARDNESS OF AS-CAST TANTALUM

In general, the alloys had homogeneous microstructures when melted by the standard procedure given earlier. However, alloys containing from 1.26 to 2.9 w/o zirconium and from 1.2 to 5.55 w/o hafnium had cored structures at the surfaces of the arc-cast buttons. This coring was attributed to the rapid cooling following arc casting. Also, the tantalum 0.01 w/o beryllium and the tantalum-1.2 w/o uranium alloy contained areas of inhomogeneity. The inhomogeneous portion of the tantalum-0.01 w/o beryllium alloy is shown in Figure 9a. As mentioned previously, one of the yttrium alloys (109) was inhomogeneous, as shown in Figure 10d. However, this alloy was melted mainly for the purpose of recovering a portion of the yttrium rather than for obtaining a homogeneous alloy.

The grain-size data for all of the materials in the as-cast and as-annealed conditions are given in Table 13. A comparison of the effect of alloying on the grain size of the annealed material cannot be made since different annealing treatments were employed. Therefore, all of the following comments on grain size refer to the alloys in the as-cast condition. In general, alloying additions resulted in a refined grain structure. Exceptions were alloys containing additions of <0.001 w/o aluminum (Alloy 72) <0.01 to 0.015 w/o iron, 0.015 to 0.03 w/o titanium, and <0.001 w/o yttrium (Alloys 37, 66, 67, 68, and 69). The grain size of the yttrium alloys was generally slightly larger than that of unalloyed tantalum. Increasing additions of rhenium, tungsten, and additions of thorium from 0.018 to 0.066 w/o produced corresponding decreases in grain size. In the case of the hafnium alloys, the grain size decreased with increasing hafnium additions from 0.5 to 1.5 w/o. For the zirconium alloys the grain size remained about the same from 0.021 to 0.10 w/o zirconium. Then from 0.10 to 2.9 w/o zirconium the grain size behaved similarly to that of the tungsten and rhenium alloys.

Hardness

Table 13 contains the hardness data for most of the materials in three conditions: as cast, as rolled, and as vacuum annealed. In the case of unalloyed tantalum and tantalum-base alloys having the same composition, the differences in hardness were attributed to variations in the impurity levels. A very good illustration of this can be seen in comparing the hardness and analytical data (Table 12) of the unalloyed tantalum specimens of the first series.

Generally speaking, the alloying of tantalum resulted in harder materials. However, the alloys to which the following additions were made had about the same hardness as unalloyed tantalum: 0.5 w/o aluminum, 2 w/o cerium, 1.0 w/o iron, 2 w/o lanthanum, 1.5 w/o scandium, 2 w/o uranium, 0.025 to 2 w/o titanium, and 0.025 to 3 w/o yttrium. It is possible and probable that the hardness of tantalum would have been increased by the above elements, excepting uranium which was recovered, if any significant quantities of them had been retained in the tantalum. Additions of boron and silicon to tantalum produced extremely hard alloys, as indicated in Table 13, which could not be fabricated at room temperature. In fact, as mentioned earlier, the 3 w/o silicon alloy broke into several pieces during casting. The hardness of these alloys increased with the amount of the addition. Figure 11 shows the effect of alloying on the hardness of tantalum. The hardness of alloys with from 0.028 to 0.25 w/o carbon was about the same, but significantly greater than that of unalloyed tantalum. The high hardness of the tantalum-0.1 w/o carbon alloy was believed to have been caused by contamination during melting. Alloys containing from 0.025 to 0.46 w/o hafnium had about the

TABLE 13. HARDNESS, GRAIN SIZE, AND FABRICATION BEHAVIOR OF ARC-CAST TANTALUM AND TANTALUM-BASE ALLOYS

Alloy	Actual Composition ^(a) (Balance Tantalum), w/o	Hardness, VHN				Average Grain Diameter, mm			Relative Fabrication Behavior ^(b)
		As Cast	Cold Rolled	Vacuum Annealed	Homogenized, Cold Rolled and Annealed	As Cast	Vacuum Annealed	Homogenized, Cold Rolled and Annealed	
<u>First Series</u>									
11	100 Ta	106	171	112 ^(c)	--	2.4	--	--	E
13	100 Ta	93	166	100 ^(c)	--	2.9	0.10	--	E
16	100 Ta	94	171	111 ^(c)	--	--	--	--	E
17	100 Ta	92	163	-- ^(c)	--	3.1	--	--	E
10	100 Ta	123	174	140 ^(c)	--	2.6	0.042	--	E
3	1.71 W	126	233	145 ^(c)	--	2.0	--	--	E
18	1.46 W	116	227	-- ^(c)	--	2.3	--	--	E
19	1.45 W	108	203	135 ^(c)	--	2.1	0.060	--	E
4	2.92 W	157	254	184 ^(c)	--	2.0	--	--	E
5	2.71 W	140	243	-- ^(c)	--	1.7	--	--	E
6	2.94 W	134	266	176 ^(c)	--	1.9	0.040	--	E
7	6.23 W	208	299	-- ^(c)	--	1.0	--	--	E
8	5.80 W	214	306	243 ^(c)	--	1.2	--	--	E
9	6.06 W	186	297	222 ^(c)	--	1.6	0.025	--	E
<u>Second Series</u>									
27	100 Ta	89	129	86 ^(c)	--	3.5	0.100	--	E
28	1.5 Re	141	239	145 ^(d)	--	2.1	0.060	--	E
29	1.5 Re	141	245	139 ^(d)	--	2.5	0.050	--	E
30	2.85 Re	208	317	--	206 ^(d)	1.6	--	0.036	E
31	3.0-2.75 Re	198	290	204 ^(d)	--	2.0	0.060	--	E
32	6.0 Re	293	387	--	--	1.5	--	--	P
33	5.5-6.2 Re	271	383	264 ^(d)	--	1.6	0.036	--	P
35	0.031 C	152	187	97 ^(e)	93 ^(c)	2.8	0.036	0.042	E
36	0.10 C ^(f)	267	338	--	--	1.5	--	--	NF
34	0.25 C ^(f)	143	219	--	--	1.3	--	--	NF
43	0.25 C ^(f)	157	239	--	--	0.6	--	--	NF
38	<0.001 Y	86	153	85 ^(c)	--	2.2	0.060	--	E
39	<0.001 Y	90	125	80 ^(d)	86 ^(d)	2.5	--	--	E
37	<0.001 Y	117	203	114 ^(d)	--	5.8	0.120	--	E
<u>Third Series</u>									
44	100 Ta	100	178	119 ^(e)	--	2.4	0.06	--	E
46	0.025-0.026 Hf	106	154	112 ^(g)	--	2.0	0.05	--	E
47	0.043-0.046 Hf	102	150	102 ^(g)	--	1.6	0.04	--	G
52	0.096-0.10 Hf	111	176	100 ^(e)	--	1.7	0.04	--	E
49	1.20-1.30 Hf	103	280	170 ^(e)	--	0.8	0.04	--	F
50	2.70 Hf	222	267	150 ^(g)	--	0.4	0.04	--	E
56	5.25-5.55 Hf	170	255	148 ^(e)	--	0.4	0.05	--	P

TABLE 13. (Continued)

Alloy	Actual Composition ^(a) (Balance Tantalum), w/o	Hardness, VHN				Average Grain Diameter, mm			Relative Fabrication Behavior ^(b)
		As Cast	Cold Rolled	Vacuum Annealed	Homogenized, Cold Rolled and Annealed	As Cast	Vacuum Annealed	Homogenized, Cold Rolled and Annealed	
<u>Third Series (Continued)</u>									
53	0.033-0.021 Zr	125	198	100 ^(e)	--	1.7	0.05	--	E
54	0.027-0.033 Zr	113	164	99 ^(h)	--	2.0	0.06	--	E
55	0.10-0.071 Zr	107	175	82 ^(e)	--	1.9	0.06	--	E
51	1.26-1.24 Zr	198	246	196 ^(l)	--	0.4	0.04	--	E
57	2.75-2.65 Zr	245	267	168 ^(e)	--	0.4	0.04	--	F
59	0.024-0.015 Ti	86	159	82 ^(e)	--	2.0	0.14	--	E
60	0.027-0.004 Ti	111	143	72 ^(e)	--	2.2	0.14	--	E
61	0.030 Ti	98	156	73 ^(e)	--	3.1	0.20	--	G
62	0.019-0.018 Th	117	163	131 ^(j)	--	1.8	--	--	E
63	0.031-0.034 Th	106	163	132 ⁽ⁱ⁾	--	1.5	0.10	--	E
64	0.066-0.061 Th	94	151	109 ^(j)	--	1.2	0.06	--	E
65	2.90-2.50 W	129	219	138 ^(e)	--	1.5	--	--	E
66	<0.001 Y	100	154	89 ^(h)	--	2.9	0.10	--	E
67	<0.001 Y	94	129	86 ^(h)	--	3.1	0.07	--	E
68	<0.001 Y	100	152	131 ^(h)	--	2.5	0.14	--	E
69	<0.001 Y	100	152	106 ^(h)	--	3.5	0.06	--	E
<u>Fourth Series (Group 1)</u>									
102	100 Ta (control)	94	165	--	--	3.4	--	--	E
103	100 Ta (control)	105	190	--	--	4.2	--	--	E
85	0.005 Y	94	147	--	--	1.6	--	--	E
89	0.4 Th	113	172	--	--	--	--	--	P
90	1.2 Th	123	171	--	--	--	--	--	P
87	2.7 Th	124	203	--	--	--	--	--	P
88	5.6 Th	123	192	--	--	--	--	--	P
91	<0.01 Fe	98	165	--	--	3.8	--	--	F
92	0.015 Fe	123	198	--	--	3.9	--	--	F
93	1.0 Si ^(f)	596	--	--	--	--	--	--	NF
94	3.0 Si ^(f)	720	--	--	--	--	--	--	NF
95	0.5 Hf	124	192	112 ^(k)	--	2.4	--	--	E
96	1.5 Hf	136	214	110 ^(k)	--	1.2	--	--	E
97	3.0 Hf	163	251	149 ^(k)	--	1.2	--	--	E
98	0.43 Zr	140	197	135 ^(k)	--	1.3	--	--	E
99	1.30 Zr	158	257	166 ^(k)	--	0.33	--	--	E
100	2.90 Zr	216	293	210 ^(k)	--	0.23	--	--	P

TABLE 13. (Continued)

Alloy	Actual Composition ^(a) (Balance Tantalum), w/o	Hardness, VHN				Average Grain Diameter, mm			Relative Fabrication Behavior ^(b)
		As Cast	Cold Rolled	Vacuum Annealed	Homogenized, Cold Rolled and Annealed	As Cast	Vacuum Annealed	Homogenized, Cold Rolled and Annealed	
<u>Fourth Series (Group 2)</u>									
104	100 Ta (control)	107	193	--	--	3.2	--	--	G
105	100 Ta (control)	78	120	--	--	3.0	--	--	G
71	<0.001 Al	98	177	--	--	2.3	--	--	E
72	<0.001 Al	134	171	--	--	3.0	--	--	G
73	0.5 B ^(f)	288	--	--	--	--	--	--	NF
74	1.0 B ^(f)	376	--	--	--	--	--	--	NF
76	0.01 Be	147	208	--	--	--	--	--	F
78	<0.10 Ce	81	139	--	--	2.1	--	--	F
80	<0.03 La	100	162	--	--	0.88	--	--	F
82	<0.10 Sc	105	179	--	--	1.7	--	--	G
84	1.2 U	109	186	--	--	1.65	--	--	G
86	0.01 Ti	85	168	--	--	0.82	--	--	G
101	0.34 Ti	116	187	--	--	1.3	--	--	G
109	0.001 Y	96	161	--	--	--	--	--	F
110	<0.001 Y	150	181	--	--	0.68	--	--	E

(a) In cases where two compositions are given, the first is the analysis of annealed material and the second is that of as-rolled material.

(b) E = excellent; G = good; F = fair; P = poor; NF = not fabricable.

(c) Vacuum annealed 2 hr at 2600 F.

(d) Vacuum annealed 1 hr at 2600 F and 1 hr at 2700 F.

(e) Vacuum annealed 1 hr at 2600 F.

(f) Nominal composition.

(g) Vacuum annealed 2 hr at 2600 F and 1 hr at 2750 F.

(h) Vacuum annealed 2 hr at 2600 F, 1 hr at 2750 F, 1 hr at 2900 F, cold rolled specimens from 0.030 to 0.025 in., and annealed 1 hr at 2900 F.

(i) Vacuum annealed 2 hr at 2600 F, 1 hr at 2750 F, and 1 hr at 2900 F.

(j) Vacuum annealed 2 hr at 2600 F, 1 hr at 2750 F, 1 hr at 2900 F, cold rolled specimens from 0.030 to 0.025 in., and annealed 2 hr at 2900 F.

(k) Vacuum annealed 1 hr at 3000 F.

same hardness as tantalum. However, additions of from 0.096 to 5.55 w/o hafnium produced progressively harder alloys. The effect of thorium and zirconium on the hardness of tantalum was very interesting. The initial addition of 0.018 w/o thorium and 0.027 w/o zirconium caused an increase in hardness. Then, increasing the additions to 0.064 w/o thorium and 0.085 w/o zirconium produced a decrease in hardness. Further additions of from 0.4 to 1.2 w/o thorium or from 0.43 to 2.9 w/o zirconium resulted in progressively harder materials. Thus, it appeared that thorium and zirconium acted first as scavengers of impurities and then as hardeners.

The hardness of alloys with either rhenium or tungsten increased almost linearly with increasing amounts of these elements, as shown in Figure 11.

The effect of cold deformation on the hardness of unalloyed tantalum and tantalum-tungsten alloys was shown previously in Table 8 and Figure 2. Also, the effect of annealing temperature on the hardness of tantalum and some of the tantalum-base alloys was illustrated in Tables 9 and 10 and Figure 3.

Tensile Properties

The room-temperature tensile properties were determined on alloys for which sufficient material was available. The tensile tests were generally performed on vacuum-annealed material, but some tests were conducted on tantalum and tantalum-tungsten alloys in the cold-rolled condition to determine the effect of rolling.

Table 14 summarizes the tensile properties of all the unalloyed tantalum and the tantalum-base alloys, and Figures 12, 13, and 14 show the effect of a few of the alloying additions on the tensile properties. These data indicate that, in general, the strength of tantalum was increased by alloying, accompanied by slight to large losses in ductility. Low yield and ultimate tensile strengths and high ductility were characteristic of the high-purity annealed tantalum. The ultimate strength of the cold-rolled tantalum (90 per cent reduction) was about twice that of the annealed material, but the ductility was reduced about six to seven times. The addition of from 1.46 to 6.23 w/o tungsten increased the strength linearly, as shown in Figure 12. The yield and ultimate strengths of vacuum-annealed material were more than doubled by the addition of 6.23 w/o tungsten and the ductility was reduced by half. The tensile strength of tungsten alloys in the cold-rolled condition was about twice that of the annealed material as it was in the case of the unalloyed tantalum. The ductility of the cold-rolled tungsten alloys was reduced about 11 times. For example, the ductility of one of the 1.46 w/o tungsten alloys was reduced from 44 to 4 per cent. Zirconium was found to be the most effective strengthener of tantalum. The tensile strength of the zirconium alloys increased significantly and almost linearly with increasing amounts of zirconium from 0.43 to 2.9 w/o, while the ductility decreased only slightly. As indicated in Table 14, the strength of the 1.3 w/o zirconium alloy was about 1-3/4 times that of tantalum. It should be pointed out that the annealing temperatures for the two groups of zirconium alloys were different as indicated in Table 14. The high strength of Alloy 51 is probably attributable to the fact that it was only partially recrystallized.

The alloys containing rhenium were found to have the second highest strength. The tensile strength of the annealed 1.5 w/o rhenium alloy was about 1-1/2 times that of tantalum. There was a linear increase in the ultimate strength and a linear decrease in the ductility as the rhenium content was increased from 1.5 to 6 w/o.

TABLE 14. ROOM-TEMPERATURE TENSILE PROPERTIES OF ARC-MELTED HIGH-PURITY TANTALUM AND TANTALUM-BASE ALLOYS

0.030-In. Strip Specimens

Alloy	Actual Composition (Balance Tantalum), w/o	<u>Annealing Conditions</u>		Hardness, VHN	0.2 Per Cent Offset Yield Strength, 1000 psi	Ultimate Tensile Strength, 1000 psi	Elongation in 1 In., per cent
		Temperature, F	Time, hr				
<u>First Series^(a)</u>							
<u>As-Cold-Rolled Specimens (90 Per Cent Reduction)</u>							
16	100 Ta	--	--	171	72.6	74.7	5
17	100 Ta	--	--	163	71.9	75.3	5
18	1.46 W	--	--	227	97.9	100.5	4
5	2.71 W	--	--	243	95.7	99	2
7	6.23 W	--	--	299	116	139	3
<u>Vacuum-Annealed Specimens</u>							
11	100 Ta	2600	1	--	30.5	38.2	44
13	100 Ta	2600	1	--	22.8	34	35
17	100 Ta	2600	1	--	24.7	33.7	30
3	1.71 W	2600	1	--	34.6	47	35
18	1.46 W	2600	1	--	31.5	45	44
19	1.45 W	2600	1	--	27.1	41.8	38
4	2.92 W	2600	1	--	43.4	54.8	29
5	2.71 W	2600	1	--	36.8	51.5	38.5
6	2.94 W	2600	1	--	36.4	51	37
7	6.23 W	2600	1	--	63.1	74.8	23
8	5.80 W	2600	1	--	69.1	79.5	10
9	6.06 W	2600	1	--	59.7	71	15
<u>Second Series^(b)</u>							
29	1.5 Re	2600	1	--	43.4	57.2	26
30	2.85 Re	2600	1	--	76.3	86.5	14
32 ^(c)	6.0 Re	2600	1	--	115.3	117.6	5
<u>Third Series^(b)</u>							
49	1.46 Hf	2600	1	159-179	56.3	75.5	19
50	2.65 Hf	2600	1	149-170	52.8	73.1	22
56	6.7 Hf	2600	1	154-165	55.6	69	31
51 ^(c)	1.26 Zr	2600	1	151-177	53.9	74.6	16
57	2.75 Zr	2600	1	171-187	63.3	77.6	26

TABLE 14. (Continued)

Alloy	Actual Composition (Balance Tantalum), w/o	Annealing Conditions		Hardness, VHN	0.2 Per Cent Offset Yield Strength, 1000 psi	Ultimate Tensile Strength, 1000 psi	Elongation in 1 In., per cent
		Temperature, F	Time, hr				
<u>Fourth Series</u>							
103 ^(b)	100 Ta	2600	1	99-107	32	37.2	21
90 ^(a)	1.2 Th	(d)	(d)	115-128	32.9	49.6	14
87 ^(a)	2.7 Th	(d)	(d)	113-127	33.8	55	13
91 ^(a)	<0.01 Fe	2600	1	89-96	29.3	36.5	15
92 ^(a)	0.015 Fe	2600	1	109-113	34.5	40	17
95 ^(b)	0.5 Hf	3000	1	119-122	37.7	48.1	24
96 ^(b)	1.5 Hf	3000	1	107-116	34.8	49.3	25
97 ^(b)	3.0 Hf	3000	1	143-178	48.8	62.1	22
98 ^(b)	0.43 Zr	3000	1	122-126	39.9	50.9	18
99 ^(b)	1.3 Zr	3000	1	138-153	46.7	61.8	24
100 ^(a)	2.9 Zr	3000	1	198-226	71.4	86.3	18

(a) Results based on average of duplicate specimens per composition.

(b) Results based on average of triplicate specimens per composition.

(c) Not fully recrystallized.

(d) Annealed 1 hr at 2900 F plus 1 hr 3200 F.

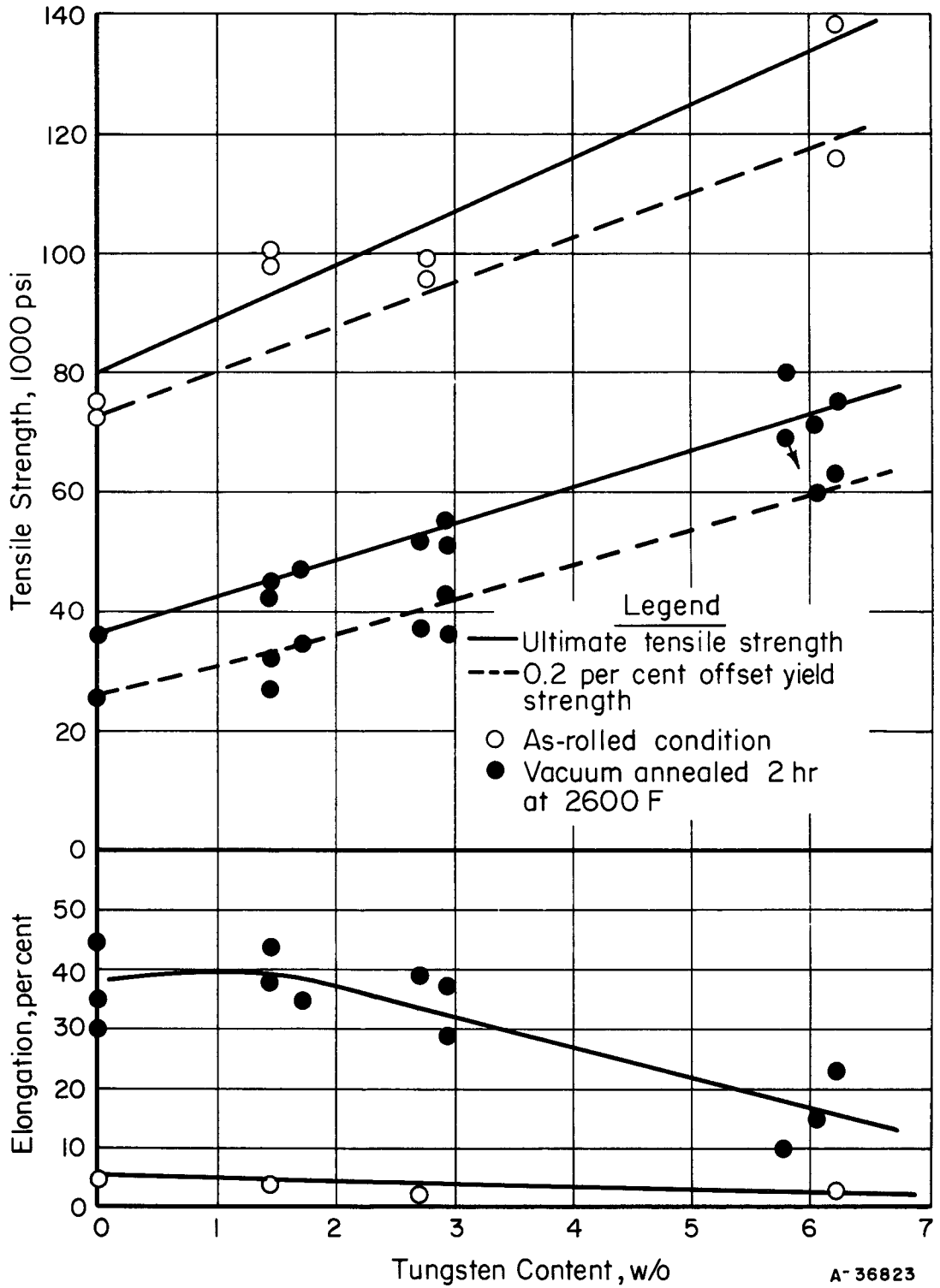


FIGURE 12. TENSILE PROPERTIES OF TANTALUM AND TANTALUM-TUNGSTEN ALLOYS

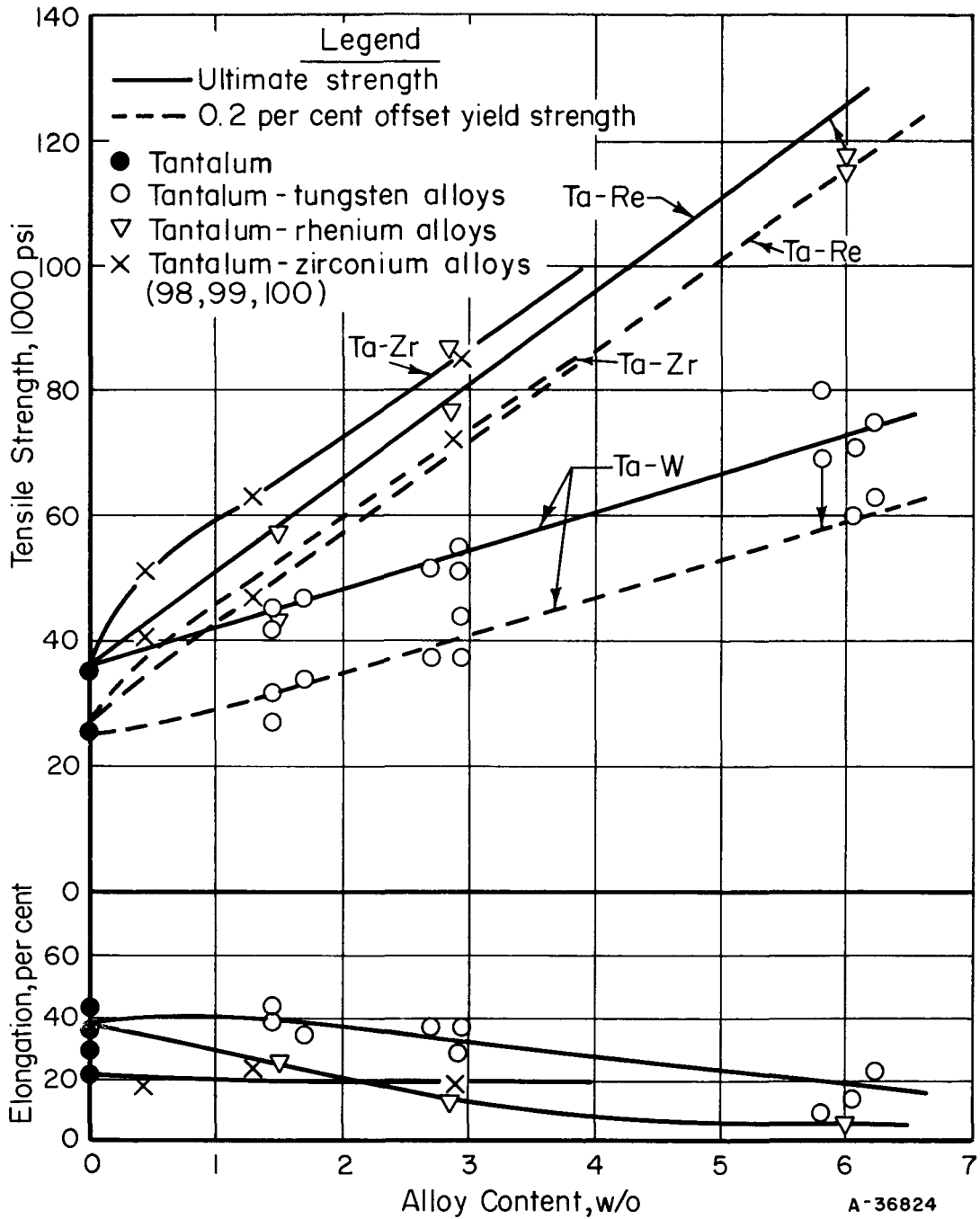


FIGURE 13. EFFECT OF RHENIUM, TUNGSTEN, AND ZIRCONIUM ADDITIONS ON THE TENSILE PROPERTIES OF ARC-MELTED TANTALUM

Specimens vacuum annealed as indicated in Table 15.

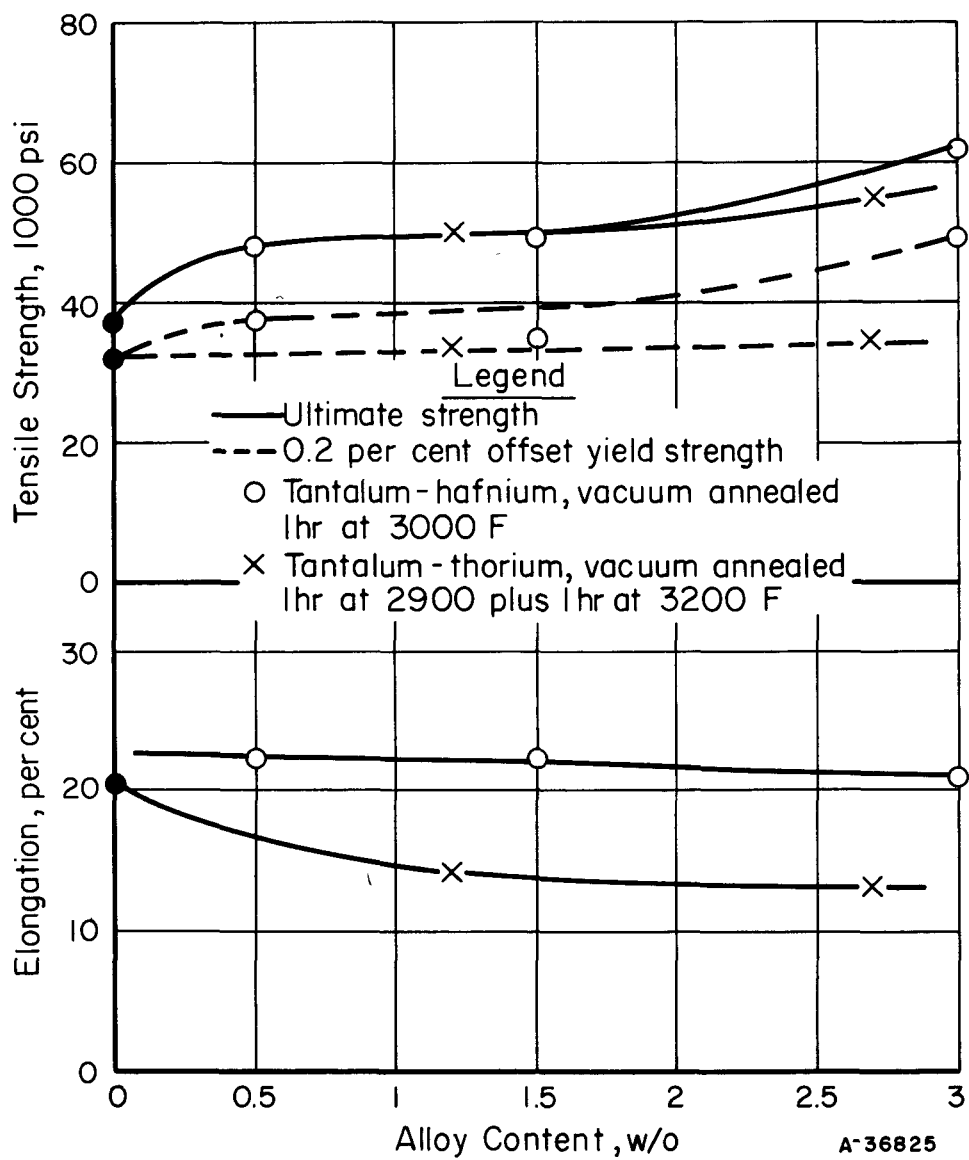


FIGURE 14. EFFECT OF HAFNIUM AND THORIUM ON THE TENSILE PROPERTIES OF ARC-MELTED TANTALUM

Increasing additions of both hafnium and thorium produced stronger alloys with slightly lower ductilities. The higher strengths of two of the hafnium alloys, 49 and 50, compared with those of two other hafnium alloys, 96 and 97, were believed to have been caused by tungsten contamination (See Table 11) and the lower temperatures used to anneal the former alloys.

Corrosion Testing

Plutonium-alloy corrosion tests on the alloys prepared at Battelle were conducted at LASL. At present, not all of the materials have been evaluated. However, preliminary corrosion data were obtained on a few of the Battelle alloys and are encouraging. For example, alloys of the tantalum-rhenium and tantalum-tungsten systems have shown good corrosion resistance in polythermal (1382 to 1022 F) tilting-furnace exposures. The tantalum-yttrium alloys also displayed good corrosion resistance, although all of the yttrium was apparently lost during arc melting. The tantalum-0.025 w/o carbon alloy was severely attacked by the fuel. On the basis of the preliminary data, there is reason to believe that the resistance of tantalum to corrosion by plutonium-alloy fuels can be improved. A more complete and detailed report on the corrosion resistance of the Battelle alloys can be given by LASL, and is reserved for presentation in a LASL report.

CONCLUSIONS

The following conclusions were made regarding the use of tantalum and binary tantalum-base alloys as container materials in LAMPRE applications:

- (1) Preliminary plutonium-alloy corrosion tests indicate that good resistance to attack can be obtained by suitably alloying tantalum.
- (2) Generally, high purity, which seems to be important in promoting resistance to plutonium-fuel alloy attack, can be maintained during arc melting.
- (3) The hardness and tensile properties of tantalum can be increased significantly by moderate additions of alloying elements.
- (4) In general, alloying additions had a refining effect on the grain size of tantalum. Also, there was a marked tendency to increase the resistance to recrystallization.
- (5) The oxygen content in tantalum apparently can be reduced by the addition of scavenging elements such as thorium, yttrium, and zirconium.
- (6) In general arc-cast binary tantalum-base alloys containing additions not greater than 6 w/o, can be fabricated to strip at room temperature. Notable exceptions are alloys of 0.5 to 1.0 w/o boron, greater than 0.025 w/o carbon, and 1.0 to 3.0 w/o silicon.

ACKNOWLEDGMENT

The authors wish to express their appreciation to the Battelle staff members who contributed to the work, particularly Messrs. E. L. Foster and W. J. Hildebrand who provided helpful consultations, and Messrs. R. D. Tenaglia and R. R. Walker, who rendered technical assistance.

REFERENCES

- (1) Private communication with LASL personnel.
- (2) Hansen, M. , editor, Constitution of Binary Alloys, Second Edition, McGraw-Hill Book Co. , New York (1958), p 1226.
- (3) Myers, R. H. , "Some Properties of Tantalum-Rich Alloys With Wolfram and Molybdenum", Metallurgia, 42, 3-9, (June, 1950).
- (4) Stewart, J. C. , BMI, Unpublished Data.
- (5) Perkins, R. , "Tantalum Annealing and Degassing and Hardness Effects of Dissolved Gases", LA-2136 (September 30, 1957).
- (6) Malter, L. , and Langmuir, R. B. , "Resistance, Emissivities, and Melting Points of Tantalum", Phys. Rev. , 55, 743-47 (1939).

DCD:MEL:CJS:JGD:AMH/ajk