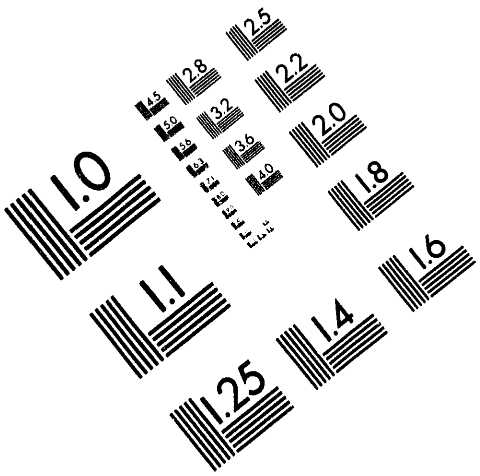

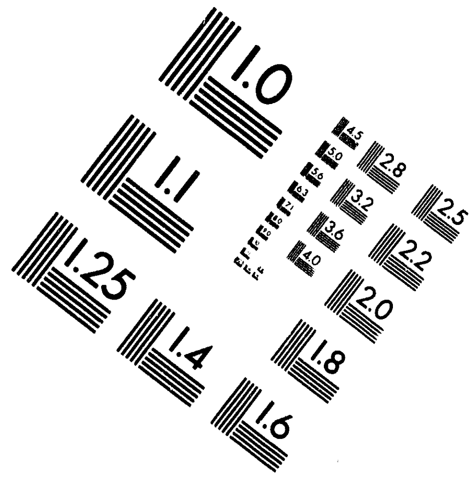

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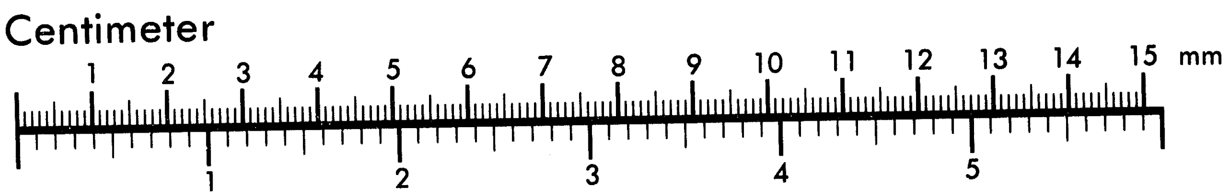

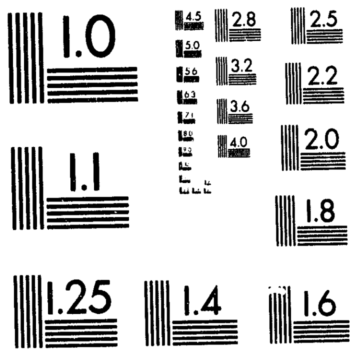

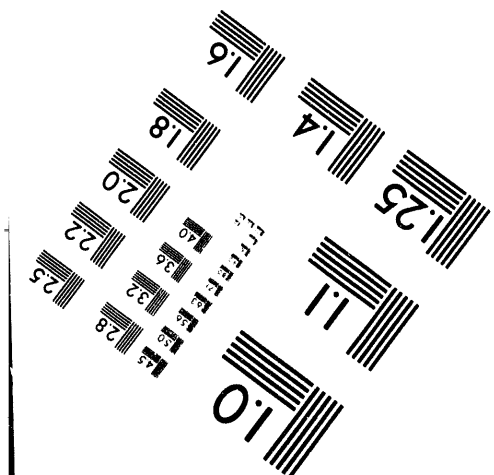

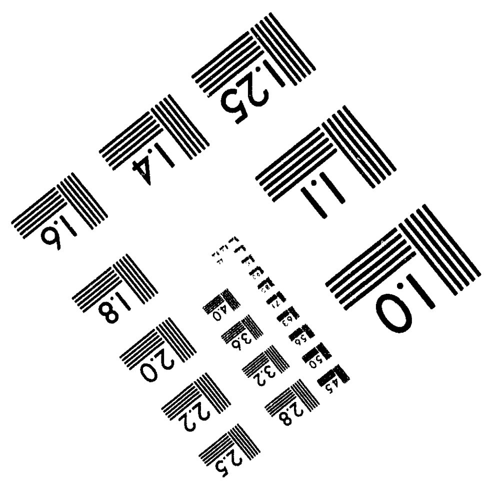

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Title: PLASMA ARC MELTING OF TITANIUM-TANTALUM ALLOYS

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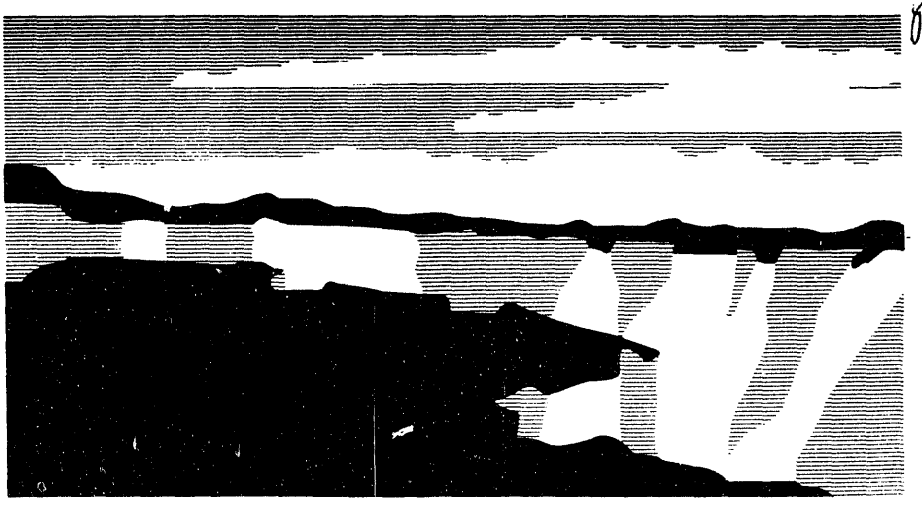

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PLASMA ARC MELTING OF TITANIUM-TANTALUM ALLOYS

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Abstract

Los Alamos has several applications for high temperature, oxidation and liquid-metal corrosion resistant materials. Further, materials property constraints are dictated by a requirement to maintain low density ; e.g.. less than the density of stainless steel. Liquid metal compatibility and density requirements have driven the research toward the titanium-tantalum system with an upper bound of 60 wt% Tantalum - 40 wt% Titanium.

The initial melting of these materials was performed in a small button arc melter with several hundred grams of material, however ingot quantities were soon needed. But, refractory metal alloys whose constituents possess very dissimilar densities, melting temperatures and vapor pressures pose significant difficulty and require specialized melting practices. The Ti-Ta alloys fall into this category with the density of tantalum 16.5 g/cc and that of titanium 4.5 g/cc. Melting is further complicated by the high melting point of tantalum(3020 C) and the relatively low boiling point of titanium(3287 C). Previous electron beam melting experience with these materials resulted in extensive vaporization of the titanium and poor chemical homogeneity. Vacuum arc remelting(VAR) was considered as a melting candidate and discarded due to density and vapor pressure issues associated with electron beam. Plasma arc melting offered the ability to supply a cover gas to deal with vapor pressure issues as well as solidification control to help with macrosegregation in the melt and has successfully produced high quality ingots of the Ti-Ta alloys.

Introduction

An alloy development program was initiated at Los Alamos to produce a material with somewhat unusual thermophysical and physical properties compared to standard materials. The primary requirements for the alloy included good corrosion resistance to liquid plutonium and oxidation resistance over a range of temperatures (800 to 1200 C) in air for extended times (several hours). In addition, the material required good room-temperature ductility and reasonable high-temperature strength. The acceptable material was further constrained by the requirement that the new alloy maintain a low density (6 to 8 g/cc).

The initial work centered on alloys with tantalum since tantalum is known to resist liquid plutonium attack and in fact is used for plutonium foundry crucibles. While tantalum works well in the vacuum induction melting of plutonium, the oxidation resistance of tantalum at 1000 C in air is very poor. Metallographic analysis of the liquid plutonium attack on electron beam welded titanium-tantalum couples indicated that this alloy system may possess the desired characteristics. A preliminary alloy composition of Ti - 40 wt% Ta was chosen based on these preliminary corrosion tests and the fact this material had a density of 6.1 g/cc. A small quantity (100g) of this material was successfully melted in a nonconsumable button melter. The material was then screened for depth of molten plutonium attack using small coupons and found to be acceptable.

Figure 1 shows the plutonium and oxidation attack depth based on titanium-tantalum alloy composition. Review of these curves, reveals that a composition of Ti- 60 wt% Ta would possess improved liquid-metal corrosion resistance and only marginal losses in oxidation resistance when compared to the Ti - 40 wt% Ta alloy. In addition, the Ti - 60 wt% Ta alloy density approached the upperbound of our specified range (8 g/cc). As such, our alloy development effort was expanded to included the Ti - 60 wt% Ta composition. Results of the Ti - 60 wt% Ta alloy melting were found to be similar to that of the Ti - 40 wt% Ta. Therefore, we will only present specifics for Ti - 40 wt% Ta within this report.

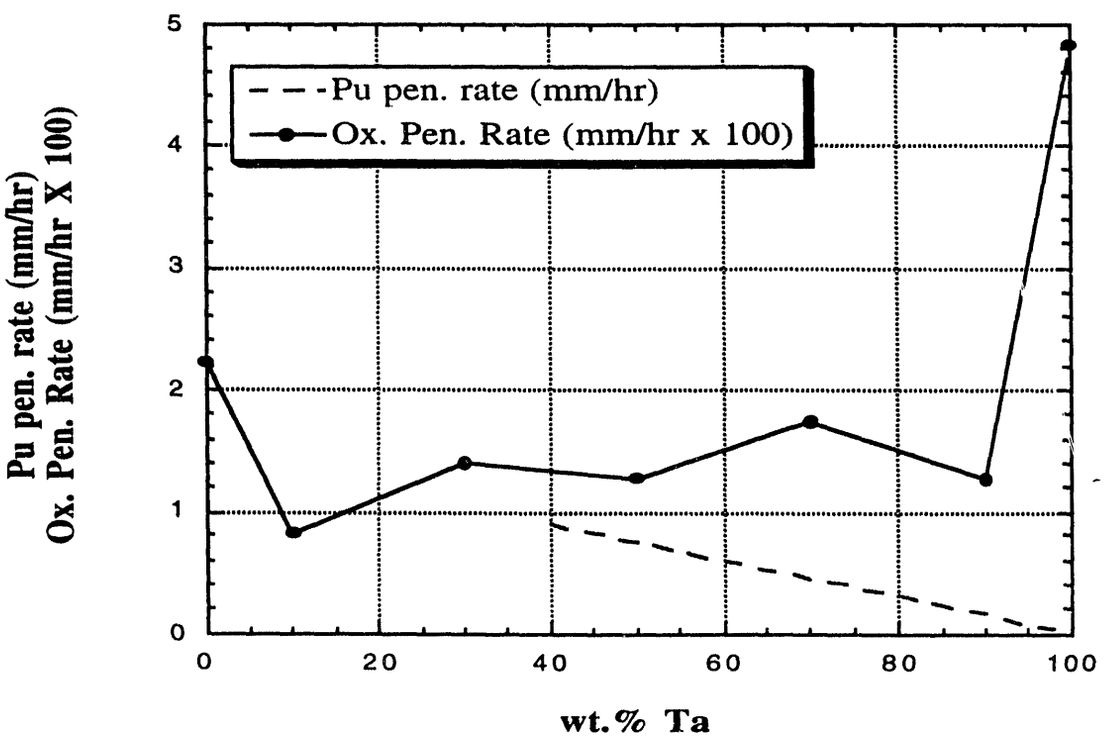

Figure 1 - Oxidation and molten plutonium attack compared to various compositions in the titanium-tantalum system.

The Ti-Ta binary phase diagram, Figure 2, shows the melting point of the two compositions of interest, (40 wt% Ta =1960 C and 60 wt% Ta = 2180 C). These high melting temperatures combined with the known reactivity of titanium quickly narrowed the possible melting techniques. Three melting techniques were examined for producing the ingots. These included vacuum arc remelting(VAR), electron beam cold hearth (EBCH) and plasma arc melting.

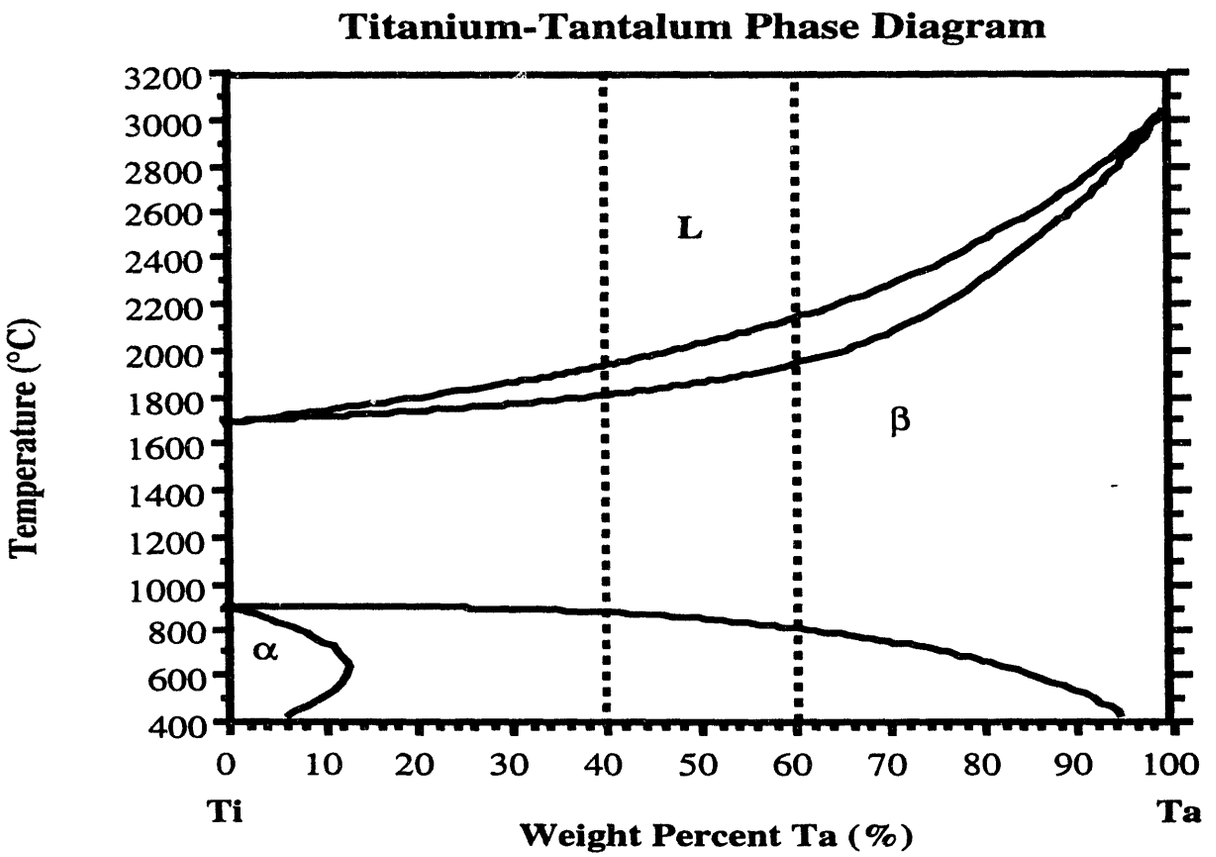

Figure 2 - Binary phase diagram for the titanium - tantalum system (Ref. 1)

Vacuum arc remelting was first examined since this process is used for ingot production of niobium-titanium superconductor alloys (Ref. 2), as well as, for melting titanium alloys and pure tantalum (Ref. 3-6). Information on the melting of superconductor alloys was of particular interest since the Ti - 50 wt% Nb alloy and the desired Ti - 40 wt% Ta alloy have similar melting points. However with further analysis the VAR process was eliminated from consideration because of two concerns. The first was the large density variation between titanium and tantalum (4.54g/cc vs. 16.65g/cc) compared to titanium and niobium (4.54g/cc vs. 8.65g/cc). Large elemental density differences in alloys have been shown to produce extensive macrosegregation in the final ingots made by the VAR process (Ref. 3). The second concern was the difference in melting point between the two elements. The large temperature variation, Ti 1662 C versus Ta 3020 C, would make electrode design difficult to produce uniform melt back rates and a homogeneous ingot. Additionally, titanium loss during melting due to vaporization under typical VAR vacuum levels was also a possible area of concern.

Electron beam cold hearth melting was next considered since this technique is used extensively for melting refractory metals as well as titanium sponge (Ref. 7-9). The process offers an added benefit of purifying the material during melting by vaporizing interstitial inclusions. The purification process is the result of the relatively high vacuum levels, as well as, the highly concentrated energy input from the electron beam. This purification process allows pure tantalum to be processed with interstitial levels in the low ppm level. However, the same attributes that

help EBCH purify metal would make the alloying of titanium and tantalum very difficult. Figure 3 (Ref. 10) shows the vaporization pressure of titanium versus temperature. Imposed on the graph are the melting points of pure titanium, pure tantalum, the two Ti-Ta alloys, and the typical operating vacuum levels for VAR, EBCH and plasma arc melting.

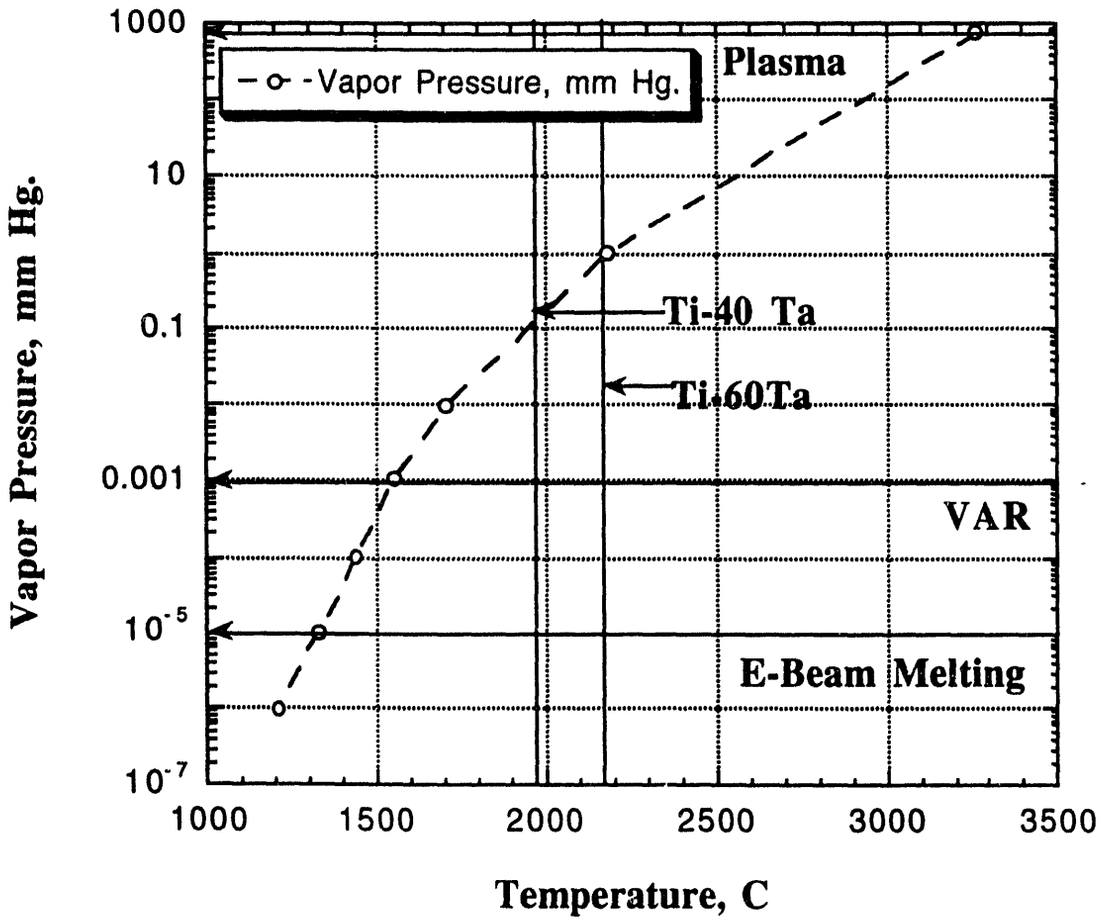

Figure 3. Vapor pressure versus temperature for titanium. Imposed on this graph are the melting points of two select alloys and the typical operating pressures for VAR, EBCH and plasma arc melting

Electron beam cold hearth melting typically operates at vacuum levels of 10^{-3} to 10^{-5} mm Hg, which translates into a vaporization temperature of approximately 1350 C. Therefore, at the lower vacuum levels, the vaporization temperature is approximately 300 C below the melting point of titanium resulting in suspected alloy composition changes due to titanium vaporization. The vaporization problem is further compounded when we considered the two Ti-Ta alloys of interest in this study. The melting points of these alloys are 610 C and 830 C above the vaporization temperature of titanium promoting extensive loss of titanium during melting.

Plasma arc melting (Ref. 11-13) appeared to solve many of the problems associated with the other techniques, but, introduced other uncertainties in the area of interstitial content. Plasma arc melting typically operates at ambient pressures or above and would minimize titanium vaporization, Figure 3. In addition, the ability to manipulate the heat source in the plasma arc furnace was considered to be beneficial for minimizing the macrosegregation resulting from density differences discussed in the evaluation of the VAR process. One potential negative aspect of this technique was that the high chamber pressure relative to VAR and EBCH would

preclude interstitial purification of the melt indicating a need for clean feed materials and careful foundry practices. After evaluating all the options, plasma arc melting was chosen as the basis for this experimental program.

Experimental Approach

Screening Experiments

Preliminary investigations were performed using small ingots (100g) of the Ti-Ta alloys which were produced by nonconsummable arc melting using a Retech button melting system. Feed materials consisted of sponge titanium and commercially pure tantalum sheet. Alloy constituents were weighed using an electronic balance to the nearest gram, and chemically cleaned. Melting was accomplished with high-purity reagent grade argon cover gas at a pressure of 500mm Hg. Each button was melted 4 times and flipped in orientation preceding each melt. Buttons were then used as feed for a 10 cm by 10 cm by 1.7 cm thick plate that was also melted and flipped 5 times to achieve homogeneity. Electron beam surface melting was used to further improve plate homogeneity and to smooth the plate surfaces in preparation for subsequent thermo-mechanical processing. A representative melted plate is shown in Figure 4.

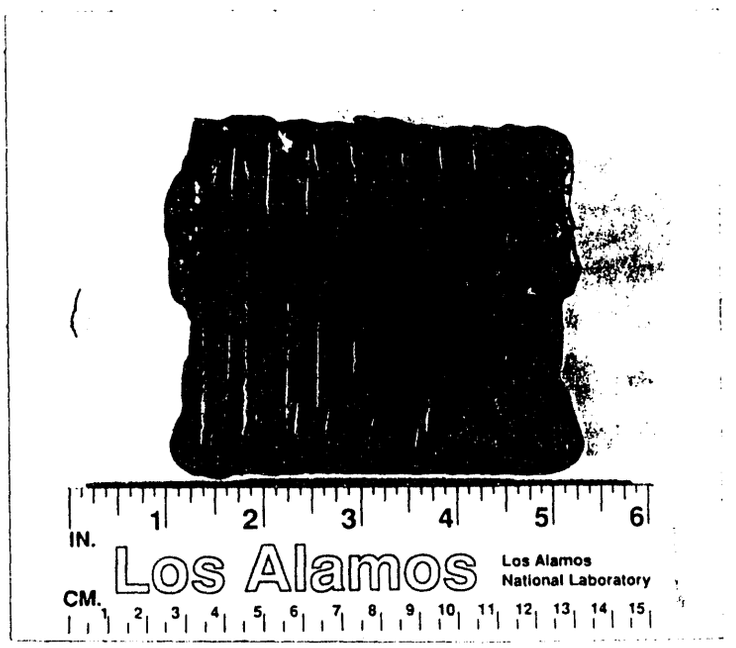

Figure 4. Non-consumable arc melted plate of Ti - 40 wt% Ta alloy after electron beam surface smoothing

Initial results with the button melted plates indicated that homogeneous alloys could be produced and that these alloys could be fabricated by conventional thermo-mechanical techniques into sheet form. Further, liquid plutonium compatibility and air oxidation experiments, proved that the Ti-40 wt% Ta alloy possessed the desired corrosion resistance and warranted examination on a larger scale. This next step required a scale-up in product quantities to 200 Kg ingots.

Ingot Scale-up and Plasma Arc Melting

Two approaches were taken with regard to the morphology of the starting feed stock. The first series of castings were similar to the initial arc melted plates in that titanium sponge and tantalum sheet material were used a feed stocks. A second series of castings were made, but this time,

with tantalum powder instead of solid pieces. The titanium sponge, tantalum sheet and tantalum powder were all commercial purity materials, Table I.

The desired alloy compositions were pressed into discs 7.6 cm diameter by 6.4 cm thick, for feeding into the plasma torch. The individual disks were fabricated by preweighing samples to match the desired alloy composition then pressing the material at a uniaxial load of 100 Mtons in a steel die. All pressings were made at room temperature. Figure 5 shows the starting materials and the final pressings.

Table I: Chemical Analysis of Interstitials in Starting Materials

Material	C	H	O	N
Titanium Sponge	50	210	430	100
Tantalum Sheet	11	1	84	20
Tantalum Powder	9	9	599	12

* All quantities in ppm

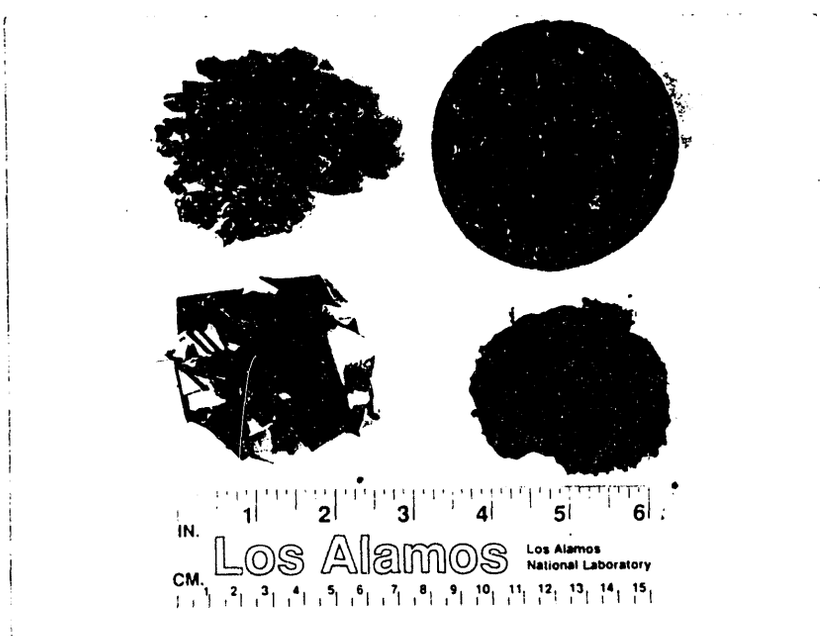

Figure 5. Morphology of starting material: titanium sponge, tantalum sheet and tantalum powder and pressing used as feed material for first ingot melting.

The alloy ingots were prepared by placing the pressed disks into the ingot withdrawal collar on the plasma arc furnace and sequentially feeding them into the molten pool until a 15.25 cm dia by 33 cm long ingot was produced. This starting ingot was remelted three additional times to maximize homogeneity. The plasma torch parameters are outlined in Table II.

Table II: Plasma Torch Parameters for Melting Titanium-Tantalum Alloys

Chamber pressure (Torr): 800	Ingot Withdrawal: 1.69 cm/min.
Volts: 175 V	Melt Rate: 3.6 Kg/min.
Amps: 1375 A	Kilowatts Input: 250

Results and Discussion

Ingot Melting Using Tantalum Sheet

Initial ingot fabrication was accomplished using the titanium-tantalum sheet compacts. The tantalum sheet had a low interstitial content compared to the titanium sponge or tantalum powder and we anticipated that this would help to minimize alloy impurities. Compacts were successfully melted and the ingot remelted without significant difficulty. However, during the remelt operations, we observed residual pieces of tantalum sheet protruding from the melt region as the ingot was consumed by the plasma arc. In some instances, entire pieces of apparently solid tantalum sheet fell into the molten pool. In other cases, molten islands of segregated material presumed to be tantalum rich could be observed due to an increased melting sluggishness as the torch passed over the feed ingot. The frequency of these occurrences decreased with repetitive melting, and the final melting trial appeared to produce a homogeneous product. However, due to the low density of titanium and the titanium-tantalum alloy, the possibility of residual high density tantalum segregation was considered to be high. As such, the ingot was machined to remove surface roughness and examined by X-Ray Radiography. Figure 6 shows the X-Ray Radiograph of the Ti - 40 wt% Ta ingot after four melts. Note the high density areas at the centerline of the ingot.

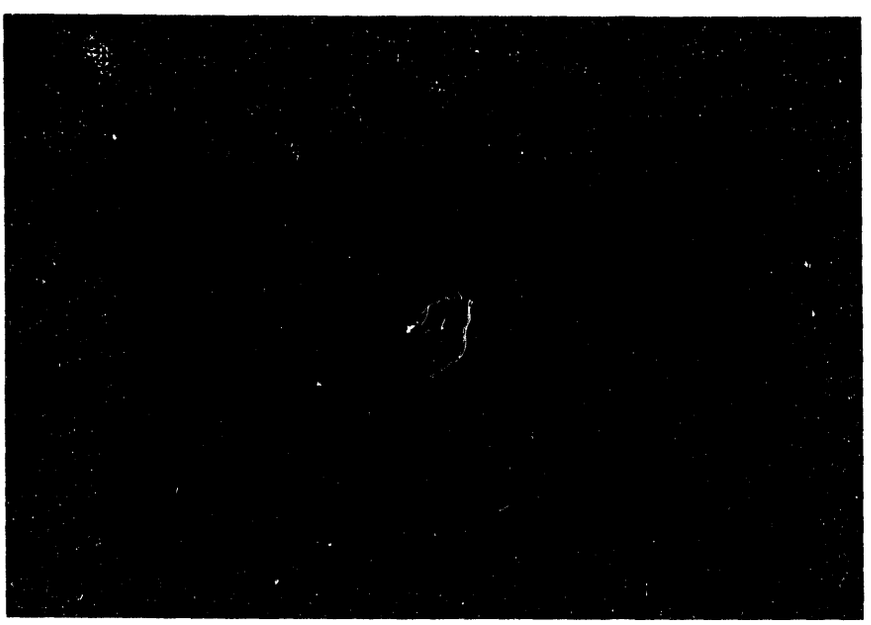

Figure 6. X-Ray Radiograph of Ti - 40 wt% Ta ingot made with titanium sponge and tantalum sheet as the starting material and plasma arc melted four times. Note the occurrence of high density regions (light areas) at the center line of the ingot indicating the presence of tantalum segregation.

Following X-Ray Radiography, the ingot was then sectioned longitudinally in preparation for metallographic examination and to extract chemical analysis samples. Figure 7 shows that the solidification front, as revealed by macroscopic etching contrast, is relatively flat which is the desired condition to minimize chemical segregation within the ingot. Tantalum rich islands are also evident thus confirming the X-Ray Radiography results and indicating that the use of tantalum sheet as feed is not an acceptable procedure.

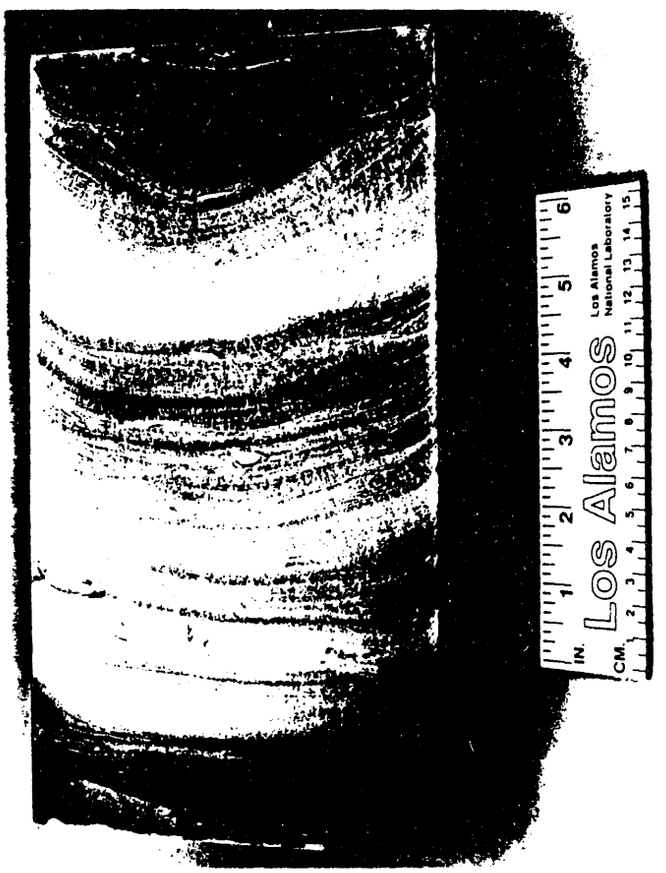

Figure 7. Macrograph of Ti - 40 wt% Ta ingot solidification front. Gray islands are tantalum rich regions.

Chemical analysis samples were taken longitudinally and radially to determine overall homogeneity and resultant alloy composition. Table III shows the final chemical analysis with standard deviations compared to expected levels based on the rule of mixtures for the starting elements. Carbon and oxygen levels increased while the hydrogen and nitrogen levels decreased. These results were consistent with our expectations since the argon cover gas apparently precluded the vaporization of interstitial compounds in the Ti-Ta system.

Metallographic examination revealed a predominate colony two-phase alpha plus beta morphology in the majority of the ingot, which was to be expected based on the composition of the material and the relatively slow solidification rates. Tantalum rich islands were also observed and when examined by scanning electron energy dispersive analysis revealed that localized compositional variations transitioned from the matrix composition (Ti- 40 wt% Ta) to a final composition of approximately Ti - 90 wt% Ta at the island center.

Table III: Chemical Analysis of Ti - 40 wt% Ta Ingot

	Ingot	Expected Levels
Titanium	58.94 +/- 1.58	60.0
Tantalum	40.44 +/- 1.43	40.0
Oxygen	436 +/- 46	291
Carbon	80 +/- 10	34
Nitrogen	24 +/- 5	68
Hydrogen	12 +/- 2	126

Figure 8 shows a representative metallographic cross-section of the ingot showing the tantalum rich areas at higher magnification. These tantalum rich islands were determined to be fully beta phase based on the etching contrast, composition, and microstructural morphology. Extensive characterization of these titanium-tantalum alloy microstructures has been reported previously and will not be repeated within the scope of this study (Ref. 15).

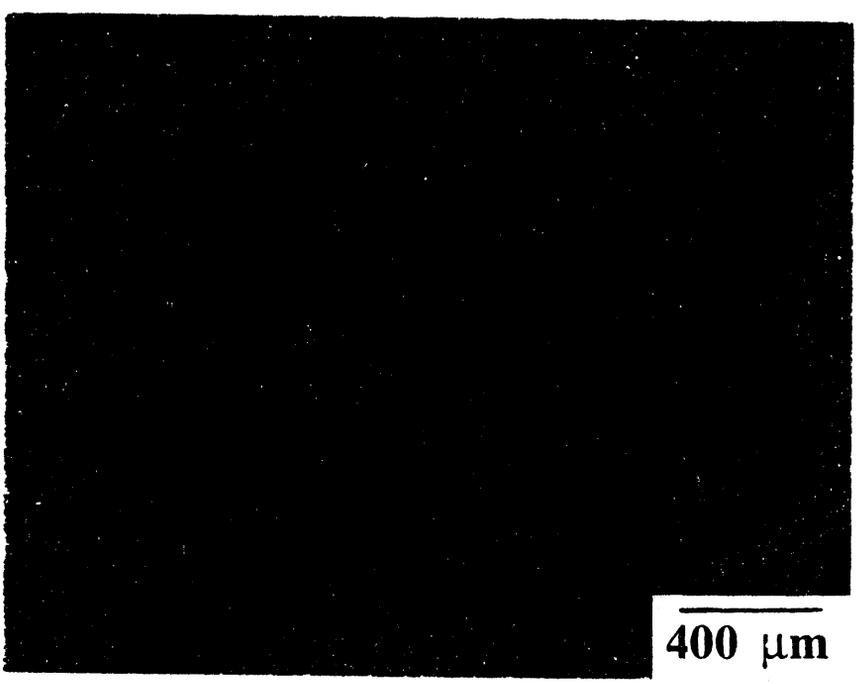

Figure 8. Typical ingot microstructure with tantalum rich islands.

Ingot Melting Using Tantalum Powder

After the initial melting series failed to produce a homogeneous ingot, subsequent alloys were fabricated using tantalum powder instead of tantalum sheet. The main drawback expected with the tantalum powder was an increased interstitial content compared to the sheet. Pre-alloyed

disks of the target composition were made using the same technique described previously and the ingots were remelted four times. The material melted very well in all the runs and we did not observe any of the anomalies encountered with the tantalum sheet. The ingots were machined to remove surface roughness and examined by X-Ray Radiography. Unlike the first series of ingots, the ingots fabricated from tantalum powder showed no gross density variations, Figure 9.

Figure 9. X-Ray radiograph of Ti - 40 wt% Ta melted with Ta powder and remelted four times.

Chemical samples were taken from the ingot in the same locations as the first series for direct comparison. Table IV shows the actual chemistries compared to the expected levels.

Table IV: Chemical Analysis of Ti - 40 wt% Ta Ingot

	Ingot	Expected Levels
Titanium	60.10 +/- 0.0	60.0
Tantalum	39.60 +/- 0.0	40.0
Oxygen	660 +/- 10	498
Carbon	120 +/- 6	35
Nitrogen	30 +/- 3	67
Hydrogen	19 +/- 2	130

The trend in the chemical analysis for these ingots was similar to the first series with a slight increases in oxygen and carbon and a decrease in nitrogen and hydrogen.

Metallographic examination revealed the same colony two-phase alpha plus beta morphology as in the first ingot, but no evidence of tantalum rich islands. Figure 10 shows a representative metallographic cross-section of the ingot.

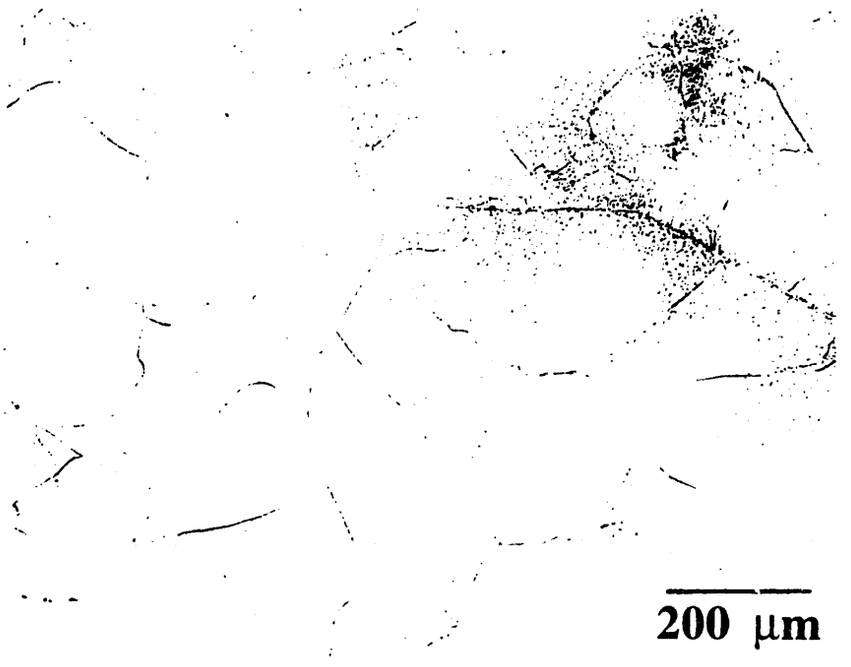

Figure 10. Micrograph of the Ti - 40 wt% Ta ingot fabricated with tantalum powder.

Conclusion

1. Plasma melting provides an excellent technique for fabricating high melting temperature alloys whose constituents possess greatly dissimilar melting temperatures and vaporization pressures. Vaporization is controlled by the positive pressure cover gas used in the melting process.

2. Care must be taken in choosing the feed materials for melting so as to assure good homogeneity in the final melt. Powder compacts were found to be produce the best alloy homogeneity while maintaining relatively low O, N and C levels.

3. Interstitial content in the final ingots was relatively low with slight increases in oxygen and carbon and decreases in nitrogen and hydrogen levels when compared to the feed material compositions. These interstitial levels would be acceptable for most titanium base commercial alloys.

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