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Title: TITANIUM-TANTALUM ALLOY DEVELOPMENT

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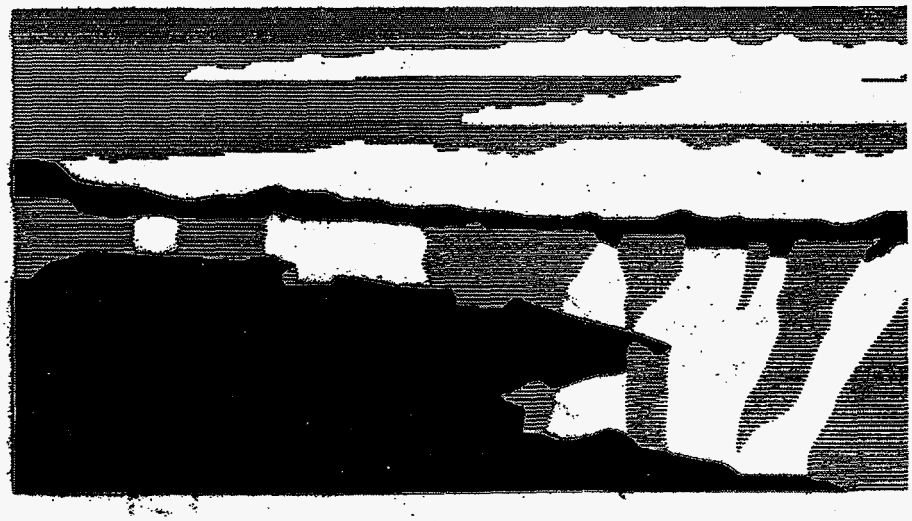

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TITANIUM-TANTALUM ALLOY DEVELOPMENT

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Abstract

Research has been underway at Los Alamos National Laboratory for several years to develop an alloy capable of containing toxic materials in the event of a fire involving a nuclear weapon. Due to their high melting point, good oxidation resistance, and low solubility in molten plutonium, alloys based on the Ti-Ta binary system have been developed for this purpose. The course of the alloy development to-date, along with processing and property data, are presented in this overview.

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Introduction

Fire resistance is an important aspect of the safety of nuclear weapons during storage and handling. One method by which to enhance the fire resistance is to provide reliable containment to prevent the release of toxic materials in the event of a fire. The development of effective containment may be accomplished by an appropriate alloy which must display several attributes: liquid metal (typically Pu) corrosion resistance, oxidation resistance, sufficient strength and ductility, moderate density and processability. This paper describes how each of these attributes have been addressed within the context of an unusual alloy development program.

Service Design Goals

Survival of 1000 °C for 2 hours in the presence of molten Pu on and air on opposing sides of the containment wall have been presented as the initial design goals. In addition, the material must support its own weight at these conditions, and have a preferred density of 6 to 8 g/cc, Processability requirements necessitate the ability to cast, form and weld the containment material with a minimum of difficulty. These constraints were used to guide the alloy development.

Approach

Preliminary work indicated that the most challenging aspect of this program would be liquid metal corrosion resistance. In the absence of intermetallic compound formation, liquid metal corrosion is dominated by dissolution of the containment material into the liquid. This process proceeds at a rate proportional to the solubility of the containment material in the liquid phase at the temperature of interest [1]. Thus, it is logical to choose a containment material which has a low solubility in liquid Pu. A comparison of various metals with melting points above 1000 °C indicates that tungsten (W) and Ta are two of the best metals in this regard, having 1000 °C solubilities in liquid Pu of 0.05 and 0.5 at.%, respectively [2]. Based on this metric alone, W is clearly the best choice for containment, and historically, this has been practiced for liquid Pu refining hardware. However, it's high density (19.3 g/cc) and poor processability make Ta the better choice in the current program.

Another advantage of Ta is that it's oxidation resistance can be improved by alloying with Ti [3]. In fact, the oxidation rate in air is reportedly a minimum at approximately 5 wt.% Ta, with a moderate increase to about 90 wt.% Ta and a marked increase at higher Ta contents. The addition of Ti to Ta also decreases the melting point, as shown by the phase diagram in Figure 1 [4, after 5]. This facilitates casting and welding, and has been shown to have no detrimental effect on ductility [6]. Furthermore, the Ti-Ta system is one of a class of β-isomorphous Ti alloys which are generally ductile and heat treatable, which would provide some measure of property control. Finally, the addition of Ti to Ta reduces the density in an efficient manner, due to the low density of Ti (4.5 g/cc). The density requirement essentially confines the Ta contents to between 30 to 60 wt.% Ta, using the constant volume assumption of:

$$\rho_{Ti-Ta} = \frac{100}{\frac{wt.\%Ti}{\rho_{Ti}} + \frac{wt.\%Ta}{\rho_{Ta}}}$$

However, Ti is extensively soluble in liquid Pu, displaying a solubility limit of 37.1 at.% at 1000 °C [2]. This behavior is counter to the oxidation resistance, which improves at Ti-rich compositions, and the density requirements. Thus, a need to optimize the Ta content for these properties exists, as well as to verify the processing characteristics of this alloy system. Additionally, microstructural characterization is required to understand and control the alloys adequately.

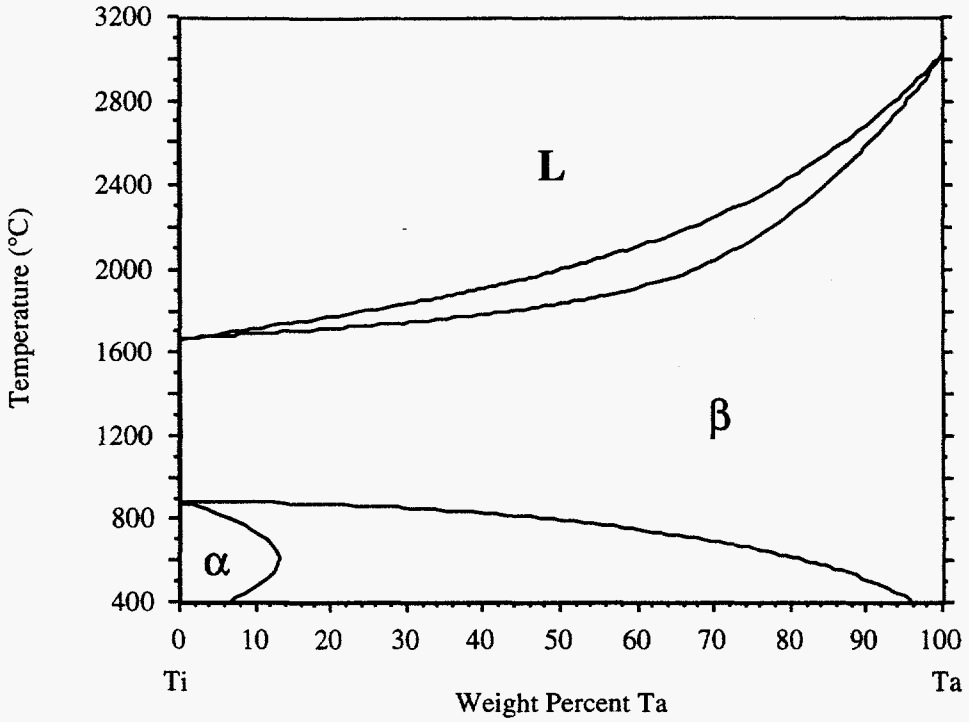

Figure 1. Ti-Ta Binary Phase Diagram.

Previous Work

Research of Ti-Ta alloys has been relatively limited. One study concerning liquid metal corrosion behavior of Ti-Ta alloys has been published in the open literature [7], and this is discussed below in the context of this program. The mechanical properties of Ti-Ta alloys have been reviewed in a previous paper [8] and are not repeated here except as required. Other work in this system is confined to phase equilibria [5,9-17], and oxidation behavior [3,18-20]. Some of the issues relevant to casting Ti-Ta alloys are described in a related paper [21].

Experimental Procedures

The experimental procedures are described in detail elsewhere [4,7,8,19,21]. In general, however, the initial castings were produced by arc melting 5N pure Ti plate with commercial purity Ta sheet in proportions to attain 5, 10, 20, 40, 60 and 80 wt.% Ta in the alloys. Extensive remelting was employed to promote homogeneity. To produce sufficient material for hot rolling to sheet, several buttons were melted together to produce slabs approximately 100 x 100 x 21 mm in size. Later, the plasma arc melting process was used to create billets up to 152 mm in dia. and 300 mm in length [21]. Sheet product was used for most experiments and was produced by hot rolling in air at 900 °C to appropriate reductions.

Liquid Pu corrosion resistance was usually measured by exposure experiments in vacuum or argon at 700 to 1100 °C for two hours, and also for various liquid volume/contact area (V/A) values. These were evaluated by measurement of time to perforation and metallography. Oxidation resistance was measured in both laboratory air, and in 20%O_2-80%Ar at a variety of temperatures and times, by thermogravimetric (TGA) to record weight losses. Metallography was also utilized to ascertain the amount of structural alloy lost to oxidation and α-case formation.

Mechanical properties were evaluated by tensile testing of sheet specimens according to ASTM E8 for a one-inch gauge length. In addition, tensile strength at 1000 °C was determined on a thermomechanical test system for selected alloys in vacuum. Density

was measured by the Archimedes' method.

Results and Discussion

A portion of the TGA data are summarized in Figure 2 and compared with an oxidation-resistant Ti alloy, β21S. It can be observed that below 60 wt.% Ta the alloys are approximately equivalent in their total weight gain, and competitive with β21S. However, the more pertinent method to evaluate material losses due to oxidation and α case formation is by metallography, which was combined with like data for liquid Pu corrosion and presented in Figure 3. Experimentally measured densities are given in the Table, and match closely with the theoretical values. Since the design thickness of the alloy varied with density (Ta content), the data are plotted as % thickness remaining in the test specimen. These results indicate that while the maximum material lost due to liquid Pu dissolution occurs at 60 wt.% Ta, material lost to oxidation increases at an increasing rate with Ta content. The sum of these two processes results in the dark curve, which indicates a worst case composition at 80 wt.% Ta and increasing goodness with Ti content. For the results presented here, the best performance occurs at 40 wt.% Ta. This may improve further with increasing Ti content, although it is anticipated that at some point the curve will reach a maximum due to increasing solubility in the molten Pu.

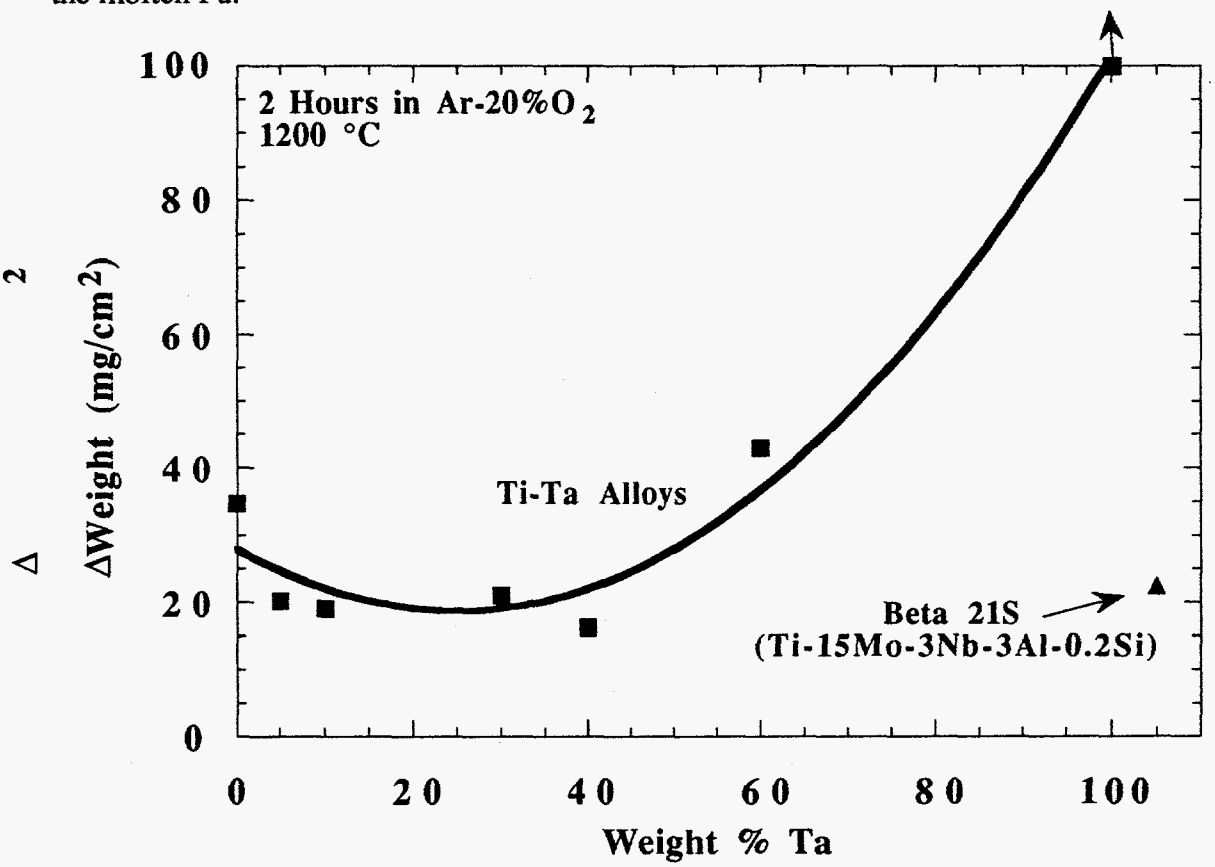

Figure 2. TGA Data for the Oxidation of Ti-Ta Alloys (and β21S) in 20% O_2-80% Ar.

The room-temperature tensile properties are given in Table II for β-annealed (900 °C/1 hour + water-quench) heat treatment conditions, and also the 1000 °C tests [8]. These mechanical property values were deemed adequate for the application.

In the course of this development, investigations of the weldability (electron beam) and machinability were undertaken, and no unusual behavior was noted, with the exception of a highly tenacious surface oxide which required bead blasting plus electrochemical means to remove. Both cold and hot forming of sheet into small cups was possible, the latter in air at

• 800-850 °C with minimal interstitial element pickup. In general, these alloys behaved similar to other β-Ti alloys in their response to processing and heat treatment.

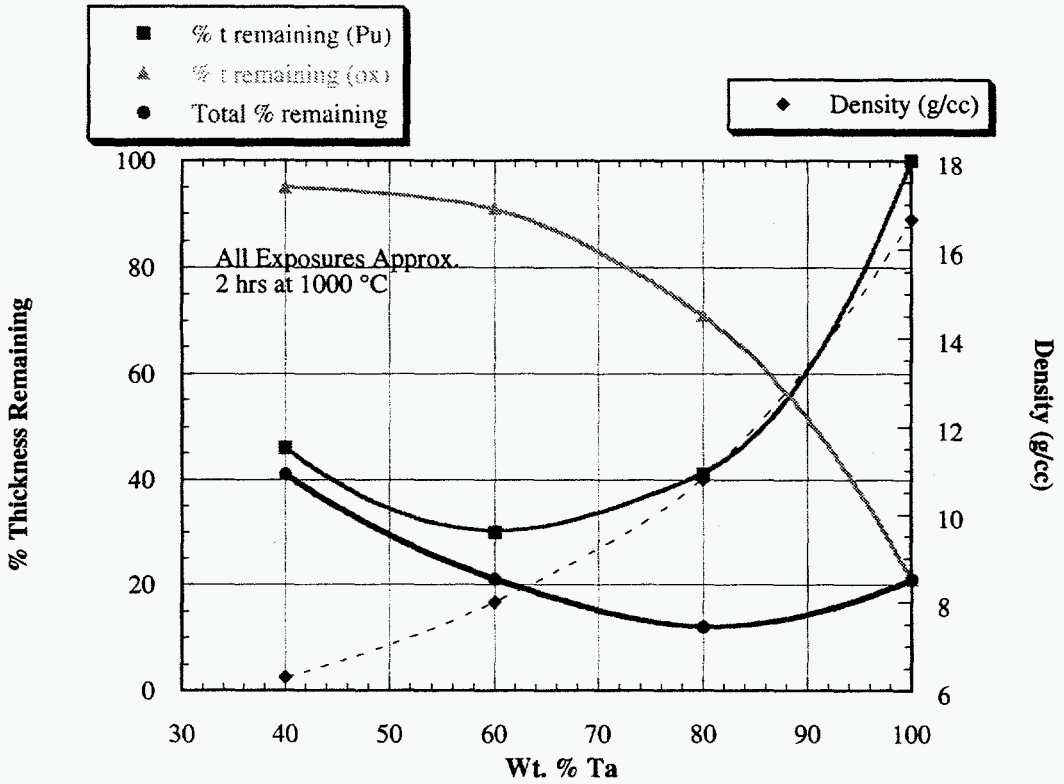

Figure 3. Combined Effects of Oxidation and Liquid Pu Corrosion on Material Thickness Losses for Ti-Ta Alloys.

Table. Measured Densities of Ti-Ta Alloys Compared to Pure Ti and Ta

Composition (wt.%)	Density (g/cc)
Ti	4.54
60Ti-40Ta	6.3
40Ti-60Ta	8.0
20Ti-80Ta	10.8
Ta	16.6

Summary

A program was undertaken to develop an alloy resistant to liquid Pu corrosion, and to oxidation, at 1000 °C. This has been accomplished within the constraints of the program.

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