106
5-4-92 JS (2)

UCRL-LR-109230

Hazards of Using Tantalum Parts in a Molten Plutonium Environment

James S. Furr

October 19, 1991

Lawrence
Livermore
National
Laboratory

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

This document was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor the University of California 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 own rights. Reference herein to any specific commercial products, 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 the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or the University of California, and shall not be used for advertising or product endorsement purposes.

This report has been reproduced
directly from the best available copy.

Available to DOE and DOE contractors from the
Office of Scientific and Technical Information
P.O. Box 62, Oak Ridge, TN 37831
Prices available from (615) 576-8401, FTS 626-8401

Available to the public from the
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Rd.,
Springfield, VA 22161

UCRL-LR--109230

DE92 012565

Hazards of Using Tantalum Parts in a Molten Plutonium Environment

James S. Furr

Manuscript date: October 19, 1991

LAWRENCE LIVERMORE NATIONAL LABORATORY
University of California • Livermore, California • 94551

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

ED

Hazards of Using Tantalum Parts in a Molten Plutonium Environment

James S. Furr

Abstract

Tantalum experiences severe intergranular attack (IGA) when in contact with molten plutonium. This IGA of tantalum has produced part failures in crucibles used during Trident and molten salt extraction (MSE) runs and in two stirring rods used in MSE runs. These parts were used at temperatures ranging from 750°C to 900°C for a minimum time of 12 hours to a maximum time of 20 hours.

Introduction

This report discusses our findings on two Ta-10W stirring rods with Ta paddles (identified as A and B) used during MSE runs. Stirrer A was made by inserting the paddle into a notch in the rod and welding it in place (Fig. 1). Stirrer B was made by milling a notch in the paddle and welding it to the Ta-10W rod. Welding was done by laser. Both stirring rods were used to stir molten Pu with CaCl_2 , KCl, and PuCl_3 salts included in the melt. The temperature range varied from 750°C to 850°C and the stirring times varied from 12 to 20 hours. Figures 2(a) and 2(b) show stirring rod A after one run and after failure.

Sectioning and Mounting

The stirrers were sectioned transverse to the rod and longitudinal to the paddles (Fig. 3). The sectioned parts were mounted in a low-temperature-setting epoxy resin.

Metallographic Preparation

The samples were ground to a level below the deformation produced by sectioning. To produce a surface suitable for metallographic polishing the samples were ground with 240, 400 and 600 grit SiC grinding paper. The samples were polished in a vibratory polisher using 2-micron diamond paste and a nylon cloth. The final polish was with 1-micron diamond paste on a nap cloth. The samples were cleaned after polishing and examined on the metallograph. After the examination was completed, the samples were swab-etched using a solution of equal parts H_2O , HNO_3 , and HF.

Examination of Rod A—Polished

Macro examination of the rod, Fig. 4, shows the IGA of the paddle and weld. Pu can be seen in the gap (the gap is caused by a poor fit of the parts) between the paddle and the rod. This gap was sealed with the molten weld metal. Finding Pu in this gap suggested that there was a major failure of the weld. Figure 5 shows the fractured surface of the paddle. Pu can be seen on the surface of the fracture. This fracture was probably initiated by the Pu migrating along the grain boundaries of the Ta, and thus reducing its tensile strength to that of the liquid Pu. In the presence of large volumes of molten Pu, the Ta grains are free to migrate away from the Ta and into the liquid Pu. Over time, this reduces the width and total strength of the paddle and leads to eventual failure. In Fig. 6, a Ta grain is migrating from the matrix into the molten Pu. The other grain boundaries in this section of the paddle also show Pu in them. Due to the large grains and possible stresses induced by rapid cooling, the weld zones are prime targets for rapid IGA by molten Pu. The long straight grain boundaries of the weld metal give the Pu very little resistance and produce rapid embrittlement of the weld metal. The residual stresses in the Ta weld may help the IGA by applying a separating force between the grains. A type of liquid metal stress corrosion cracking could be produced. Figure 7 shows the weld with major IGA. Pu is in the weld metal and in the Ta-10W rod. The long straight grain boundaries of the weld metal are quite clear. Figure 8 is the same weld at a higher magnification and shows Pu throughout the weld and its heat-affected zone (HAZ); the gray phase is PuH_2 , which is a commonly observed when Ta and molten Pu come in contact with each other.

Examination of Rod A—Etched

Etching the sample revealed a large increase in the grain size of the paddle just beyond the weld. The grain sizes, $168\mu\text{m}$ for the base metal and $300\mu\text{m}$ for the enlarged grains, can be attributed to the high heat produced by welding. The increased grain size is believed to make the part more susceptible to liquid metal embrittlement in that location. The Ta-10W rod has a smaller grain size, $26\mu\text{m}$, and is embrittled at a much slower rate. The smaller grain size increases the distance and time needed for the Pu to advance through the rod. Figure 9 shows the structure of the Ta paddle, Ta-10W rod, the weld and its HAZ. Examination of the the rod shows very deep cracks penetrating the rod with many other smaller cracks reaching to the center of the rod (Fig. 10). These cracks are the results of Pu penetration along the grain boundaries. Figure 11 shows a network of many small cracks, all intergranular. This photomicrograph was taken near the HAZ of the weld.

Examination of Rod B—Polished

Stirring rod B shows the same type of major IGA by Pu. Figure 12 is a macro of the cross section showing the damage to the stirring rod by molten Pu. The IGA by Pu was more severe at the HAZ and bond line of the weld in this sample. When the sample was sectioned, one side of the weld fell apart. Figure 13 shows major cracks penetrating to

the center of the rod and weld HAZ. Examination of these cracks at high magnification shows Pu in the grain boundaries. This sample also has many larger cracks along the circumference.

Examination of Rod B—Etched

Etching has shown the most damaging Pu attack to be along the interface of the HAZ and the base metal. Figure 14 shows the area where the paddle and weld metal fell apart on sectioning. Some of the damage seen in these photomicrographs was produced by mechanical stresses applied during sectioning. The etched structure of this zone shows a fracture that has occurred along the interface of the HAZ and the base metal. Figure 15 shows a major crack along the bond line of the weld. Some cracking can be seen in other areas of the weld. The fracture tip seen in Fig. 16 is at the paddle and rod joint and is a brittle fracture attributed to embrittlement by liquid Pu penetration along the grain boundaries. At the paddle-rod joint the Pu has penetrated completely through the paddle to the base metal. The weld along this joint created large grains in the weld metal and the paddle, and these large grains are more susceptible to rapid attack and subsequent catastrophic failure. The lines in the structure are the flow patterns of the weld metal. Pu intergranular attack has brought this stirring to a point of almost total failure.

Microprobe Analysis of Rods A and B

A microprobe analysis for Pu was taken across the weld to the bond line on stirrer A. Figure 17 shows the location chosen for analysis. Figure 18 is a scanning electron image (SEI) taken from the selected location. Some grain boundary cracking can be seen. Figure 19 is an x-ray map for Pu at the same location as the SEI shot. The Pu x-ray map emphasizes the extent of the liquid metal embrittlement throughout the weld. An area near the weld zone was chosen for Pu x-ray mapping on stirrer B. Figures 20, 21 and 22 are a scanning electron image, a compositional image, and a Pu x-ray map, in that order, of the same area. The SEI image shows some separation of the Ta grains. In the compositional image, Pu is white in the micrograph and can be seen in the grain boundaries of the Ta. The Pu x-ray map of this area shows Pu in all the grains of this zone. Although some of these grains could not be seen in the SEI photo, they are visible in the Pu x-ray map. The Pu x-ray mapping of the metallographic samples taken from the stirring rods provides positive proof that Pu has penetrated the Ta grain boundaries.

Conclusion

The photomicrographs and the microprobe analysis of the Ta stirring rod used in MSE runs give conclusive evidence of the destructive nature of liquid Pu on Ta metal. The welds between the paddles and stirring rods contain large grains that have little resistance to molten Pu. Another possible contributing factor to the rapid failure of the weld is the residual stresses produced by rapid cooling of the weld metal. The stresses can be high and are usually near the elastic limit of the metal and can force the metal

components apart.¹ These high stresses can also enhance the molten Pu attack in the Ta. The combination of long straight grain boundaries in the weld, molten Pu, and stresses have led to a catastrophic failure of the weld. To minimize O₂ embrittlement of the weld metal the stirrers were laser welded in a vacuum. The IGA on the Ta-10W rods is due to liquid metal embrittlement by Pu. No stress risers were found in the rods but the stirring action induces a torque load on the stirrer which leads to shear stresses. These stresses could have some effect on the Pu attack of the rods. PuH₂ has been identified in the grain boundaries of the Ta. What effect this phase has on the failure of the stirrer is not known. The effect of the salts used in the MSE runs has not been studied. The failure of Ta parts used in a molten Pu environment has been documented in *Results of the Metallographic Examination of the Ta Crucible Used in the MSE Runs*,² a metallographic report on the failure of the Ta crucible used in the Trident runs,³ and a metallographic report on the destruction of a Ta container used in a molten Pu and hydriding atmosphere.⁴ In each case listed above, the part failed because the IGA was severe enough to reduce the effective strength of the part.

Solution

A possible solution is the addition of Ta₂C to the Ta grain boundaries that are in contact with molten Pu. The addition of Ta₂C three to four grains deep in the Ta base metal will give the Ta better resistance to Pu and should not effect its mechanical properties.⁵

Fig. 1. Stirring rod construction.

Fig. 2(a). Stirring rod after one run.

Fig. 2(b). Stirring rod after failure.

Fig. 3. Cross-section of stirrer.

Fig. 4. 6.8x Stirring rod A, as polished.

Fig. 5. 32× Stirring rod A, as polished, paddle showing the IGA.

Fig. 6. 100× Stirring rod A, as polished, paddle fracture tip.

Fig. 7. 40× Stirring rod A, as polished, weld failure.

Fig. 8. 100× Stirring rod A, as polished, failed weld with Pu in the Ta boundaries.

Fig. 9. 20× Stirring rod A, etched, cross section.

Fig. 10. 32× Stirring rod A, etched, showing sub cracking in the Ta-10W rod.

Fig. 11. 100 $\times$ Stirring rod A, etched, showing the IGA in the Ta-10W rod

Fig. 12. 6.8 $\times$ Stirring rod B, as polished, cross section.

Fig. 13. 20× Stirring rod B, as polished, failed weld and major crack.

Fig. 14. 16× Stirring rod B, etched, IGA cracking in the HAZ of the weld and rod.

Fig. 15. 16× Stirring rod B, etched, crack along the HAZ of the weld.

Fig. 16. 40× Stirring rod B, etched, weld joint of the paddle and the rod.

Fig. 17. Location of microprobe analysis for stirring rod A.

Fig. 18. 100 $\times$ Stirring rod A, scanning electron image of the weld.

Fig. 19. 100× Stirring rod A, x-ray map for Pu.

Fig. 20 400× Stirring rod B, scanning electron image in the weld metal.

Fig. 21 400× Stirring rod B, compositional image.

Fig. 22 400× Stirring rod B, x-ray map for Pu.

References

1. Dr. Mars G. Fontana, *Corrosion*, Metals Institute, The American Society for Metals, 1968.
2. James S. Furr, *Results of the Metallographic Examination of the Ta Crucible Used in the MSE Runs*, UCRL-ID-105713, Oct. 22, 1990.
3. James S. Furr and Rick Randich, Metallographic report sample number ME2835.
4. James S. Furr and Ralph Condit, Metallographic report sample number ME3143
5. Jenny H. De Pruneda, Private conversation.