Results of the Metallographic Examination of the Ta Crucible used in the M.S.E. Runs

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INTRODUCTION

A cross section from a Ta crucible used in numerous M.S.E. runs was submitted to metallography to determine the soundness of the crucible wall, type of Pu attack, depth of wall penetration by the Pu and general microstructure. The crucible contained molten Pu and Am, with $CaCl_2$, KCl and $PuCl_3$ salts ran at temperatures of 750°C to 900°C for approximately 10 to 12 hours.

SAMPLING

Two samples were sectioned from the Ta crucible. One from above the meniscus and one from the bottom of the crucible. Figure 1 shows the location of the samples and their metallographic sample numbers.

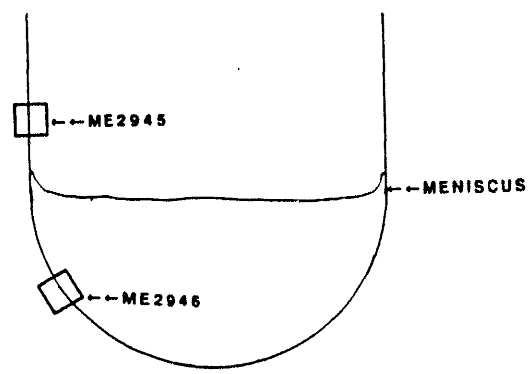

Figure 1.

Sample ME2945 taken from above the meniscus is the control sample, this part of the Ta crucible should retain it's original wall thickness and microstructure. Sample ME2946 taken from the bottom of the crucible will be examined for the soundness of the Ta wall, type of attack by the molten Pu, depth of penetration by the Pu and general microstructure.

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MOUNTING AND POLISHING

The samples starting with were mounted in a low temperature setting epon. The samples were ground with 120 grit grinding paper thru 600 grit paper. The first polish step was 2μ diamond paste on a nylon cloth in a vibratory polisher. Final polishing was done with 1μ diamond on a napcloth. The samples were cleaned with alcohol after each polishing step. These samples were hard to polish due to the large amount of salts left in sample ME2946. The salt reacted with the epon and was not contained by it. During the polishing steps the salt would fall out and contaminate the wheel and leave large gouges in the surface of the Ta. The salt contains some Pu oxides which are much harder than the Ta.

AS POLISHED EXAMINATION OF ME2945

The metallographic examination of this sample in the as polished condition revealed some wicking of the Pu up the wall of the crucible, Figure 2 shows Pu on the inside wall of the Ta shell. Note the wavy surface of the wall at this point, the wavy surface indicates some dissolution of the Pu. The wall thickness of the Ta crucible above the meniscus measured 1.4mm. Figure 3 shows the cross section of the crucible.

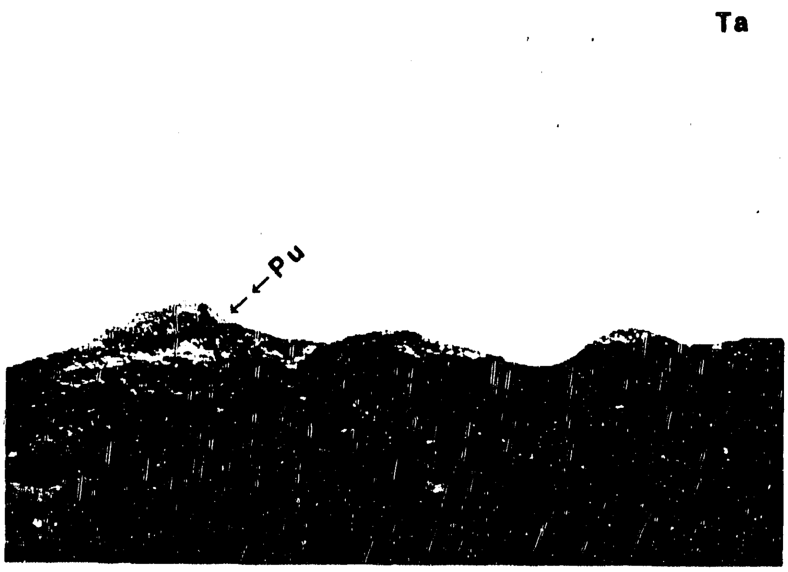

Figure 2. 200X Pu on the inside surface of the Ta.

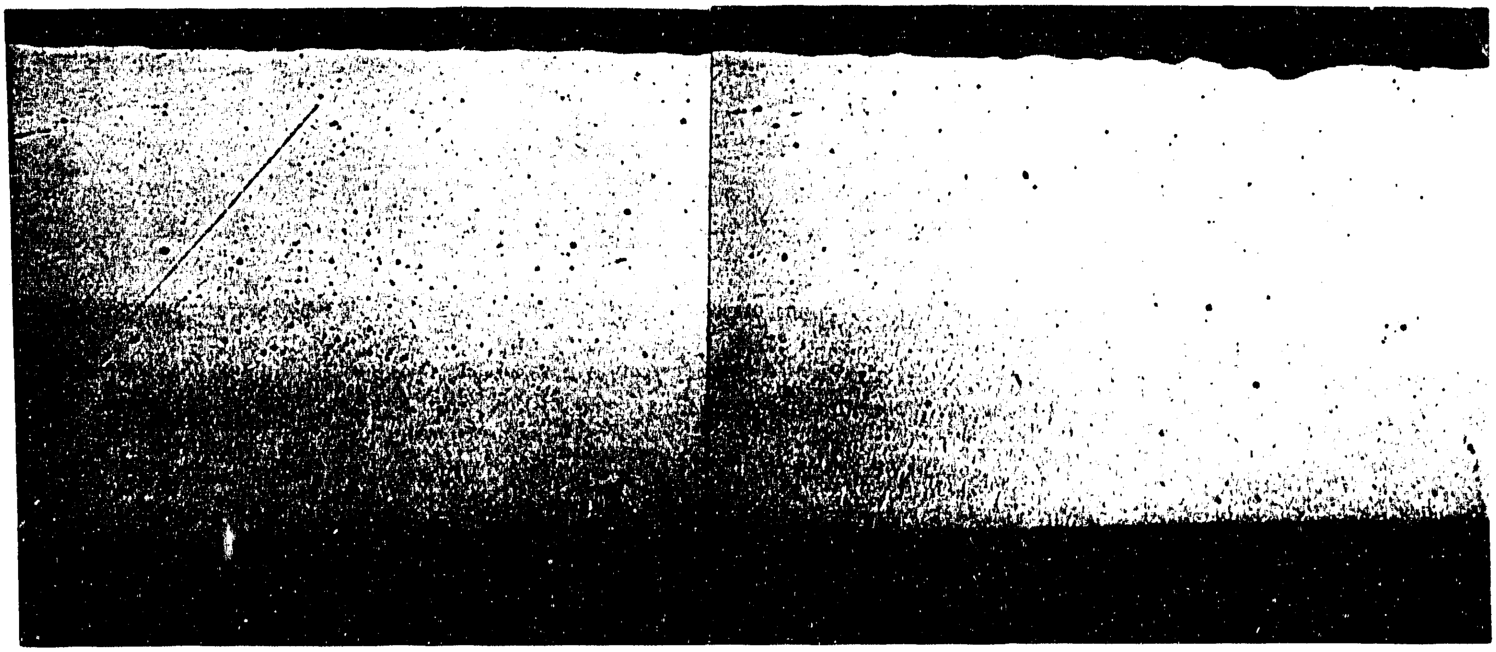

Figure 3. 50X Montage of the crucible wall.

AS POLISHED EXAMINATION OF ME2946

Metallographic examination of this sample revealed a heavy intergranular attack (IGA) of the crucible wall by the molten Pu Figure 4 is a montage taken at the location of the heaviest attack.

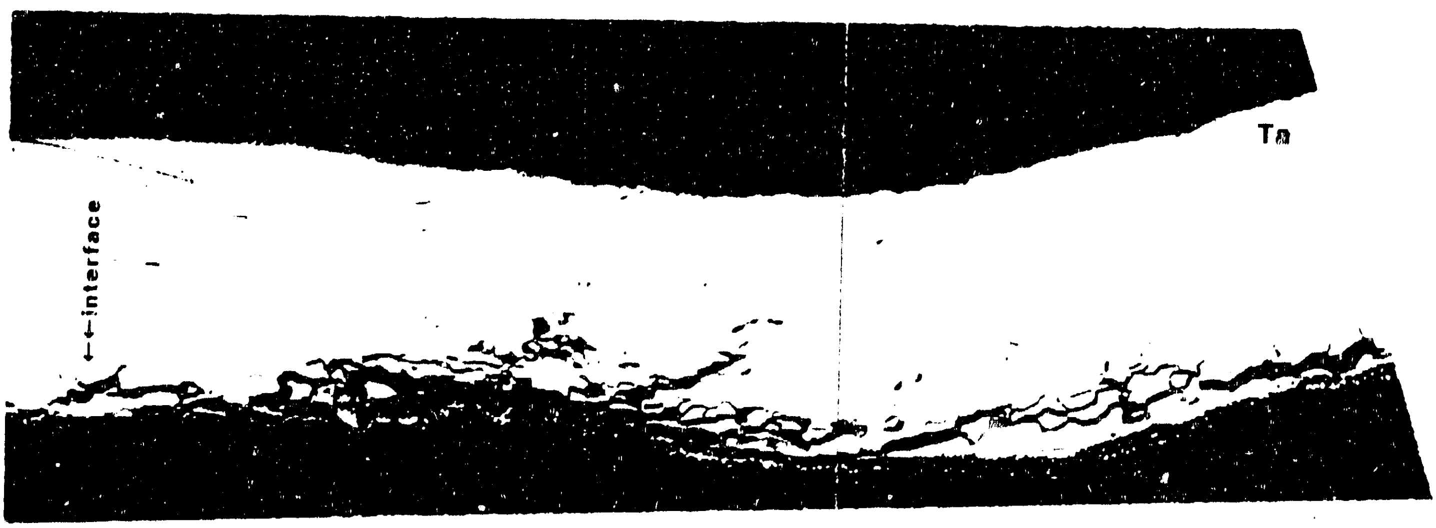

Figure 4. 50X Montage of sample ME2946 heaviest attack.

Figures 5, 6 and 7 show the major mode of attack. These photomicrographs show Pu in the grain boundaries of the Ta. The Pu penetrates along the grain boundaries until the grain becomes detached. When this happens the grain migrates into the melt. In Figure 5 the Pu molten path produces cracks several grains deep, when these cracks join a large section of the wall separates from the crucible resulting in reducing the wall thickness at a rapid rate.

Figure 5. 200X IGA of the Ta by Pu.

Figure 6. 500X Ta grains that have been completely wetted and detached by the molten Pu.

Figure 7. 500X IGA of the Ta by molten Pu. The accular phase is TaC when the free Ta reacts with the carbon in the Pu.

The crucible wall was examined to determine the depth of penetration by the molten Pu. The crucible wall showed full penetration, Pu was found on the surface of the outer wall. Figure 8 shows Pu in the grain boundaries at the midpoint of the wall. Figures 9 and 10 shows Pu in the Ta boundaries near the outside edge and Pu on the outside surface of the crucible wall. Calcium was also found on the outside surface of the crucible wall as shown in Figure 11.

In Figure 8 the Pu appears as thin gray grain boundaries.

In Figure 9 the Pu can be seen on the Ta surface as a gray mass, indicated by the arrows. Below the surface Pu can be seen in an area of IGA one grain deep along a series of grains ending when the Pu reached the surface.

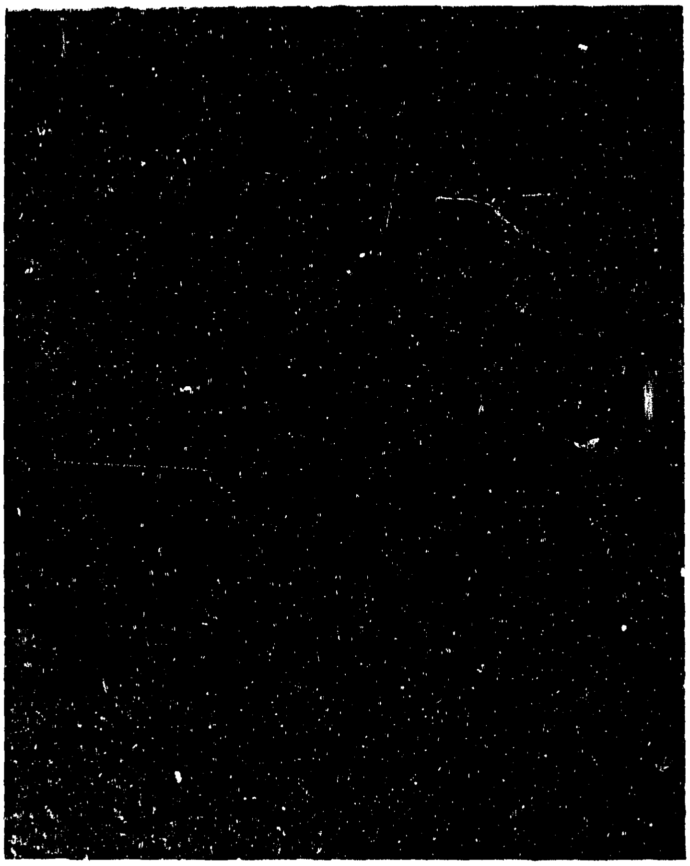

Figure 8. 500x Pu in the Ta boundaries center

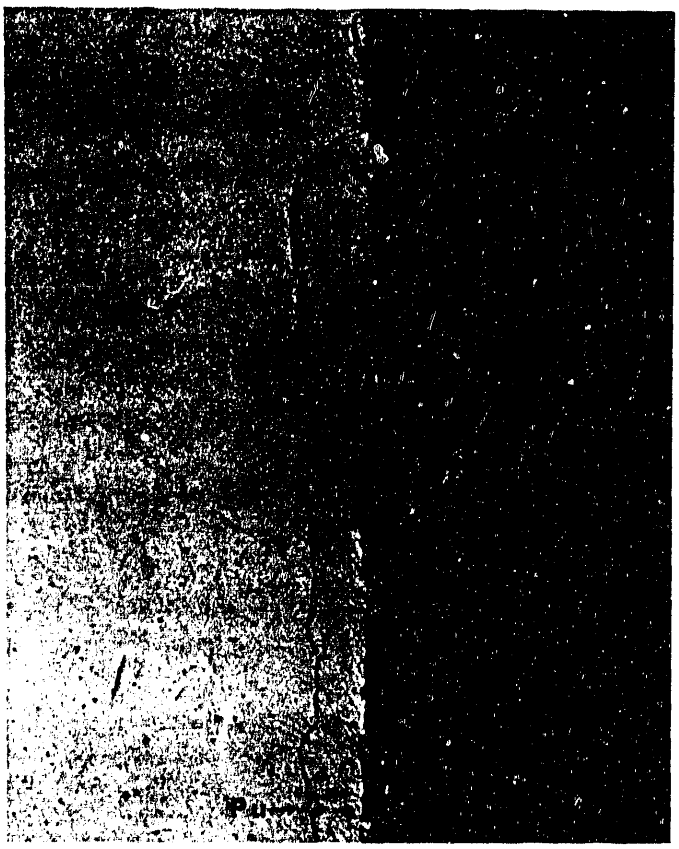

Figure 9. 200x Pu at the outer surface.

In Figure 10 Pu can be seen on the surface and in the grain boundaries of the Ta.

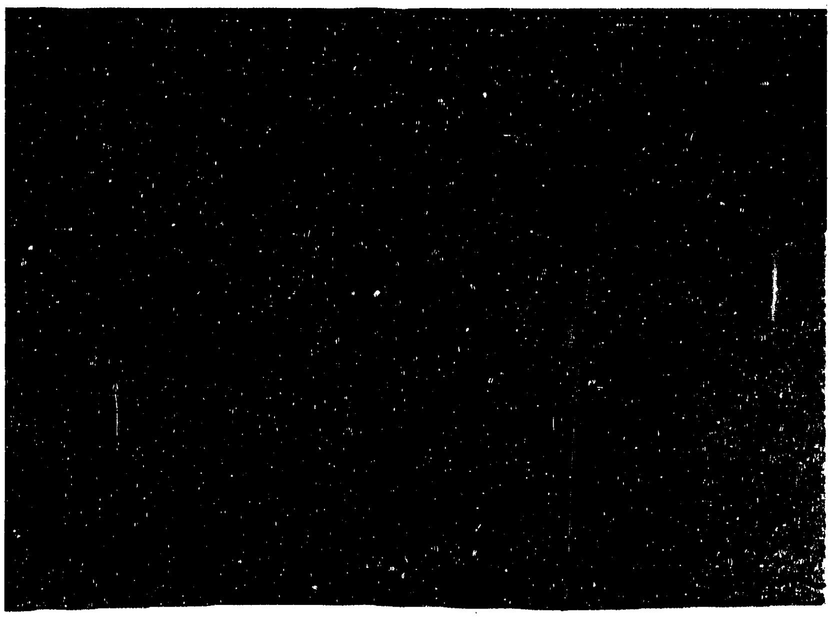

Figure 10. 500x Pu at the outer surface.

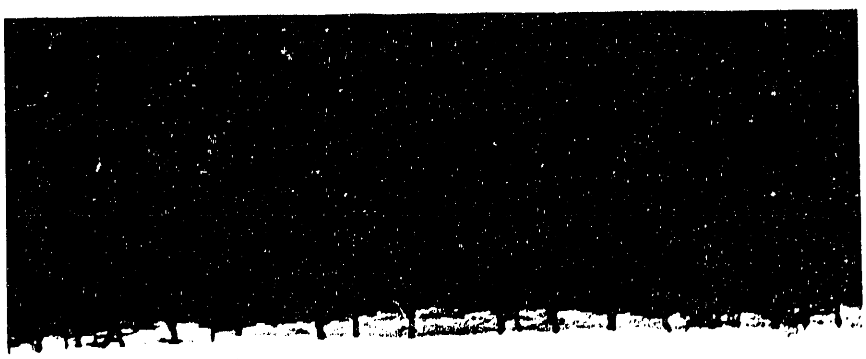

Figure 11. 200x Ca on the surface outer wall.

The uneven attack of molten Pu convection currents in the molten Pu could produce such an attack by removing Ta saturated Pu from the Ta wall allowing unreacted Pu to come in contact with the Ta wall allowing more Ta to go into solution producing an erosion corrosion type attack on the Ta crucible wall as seen in figure 4 is an unexplained occurrence. Convection currents in the molten Pu could produce such an attack by removing Ta saturated Pu from the interface and allowing unreacted Pu to come in contact with the Ta at the interface allowing more Ta to go into solution an erosion corrosion type of attack. Measurements taken along the Ta wall of figure 4 illustrates the rate of attack along the length of this section. Table 1 gives these measurements with the amount of dissolution that has occurred. At the point of deepest penetration the Pu has breached the Ta wall in some grain boundaries See Figure 10 but the crucible wall is still solid at this point.

Table 1. Wall Thickness

Location	Wall Thickness	% Dissolution
Control	1.4mm	0
1	.74mm	47
2	.70mm	50
3	.56mm	60
4	.32mm	77

MICROPROBE ANALYSIS FOR Pu

Microprobe analysis was done on sample ME2946 for the presence of Pu within the grain boundaries of the Ta crucible and on the outside wall. Figure 12 shows the location, as indicated by the arrows, where the microprobe analysis was done. This area is the area of maximum attack.

Figure 12. 50x Location of the microprobe analysis

The microprobe analysis of the Ta Pu interface was made in this area because of the IGA at the interface, the large internal crack, and the spheroidized Ta at the interface. Figure 13 is a secondary electron image (SEI) view at the Ta Pu interface including the crack.

Figure 13. 200x SEI at the interface

A backscatter image (Figure 14) was taken in the same location as Figure 13. In a backscatter (also called compo) image the larger the Z of the element the whiter it shows in the image, Pu appears white and Ta dark gray. The compo image of the Ta Pu interface shows Ta spheres in the Pu melt, IGA of the Ta grain boundaries creating a large subsurface crack. Pu can be seen thru out the compo image which is a clear indication of Pu penetration along the Ta grain boundaries.

Figure 14. 400x backscatter image of the Ta Pu interface.

The Ta spheres are probably produced by dissolution of some of the Ta grains which are detached. (The dissolution of the hexagonal grains results in rounding the edges giving a spherical appearance by the Pu. As described before the Pu wets the Ta grain boundaries and lifts the grain out and into the melt where some of it goes into solution.

A x-ray map for Pu was made at the interface. Figure 15 show the x-ray map of a triple point in the Ta. The x-ray map gives clear evidence of the presence of Pu in the boundaries of the Ta.

Figure 15. 1000x X-ray map for Pu.

Figure 16 shows SEI image taken in the center of the crucible wall. Pu can be seen as the grain boundary phase pointed out by the arrow. At this location the Pu has not penetrated many of the grains. Figure 17 is a backscatter image of the same location as Figure 16. The Pu (white phase) occurs at the same grain boundaries as in Figure 16. These images confirm the penetration of Pu to the center of the crucible wall.

Figure 16. SEI 400x center of the crucible wall

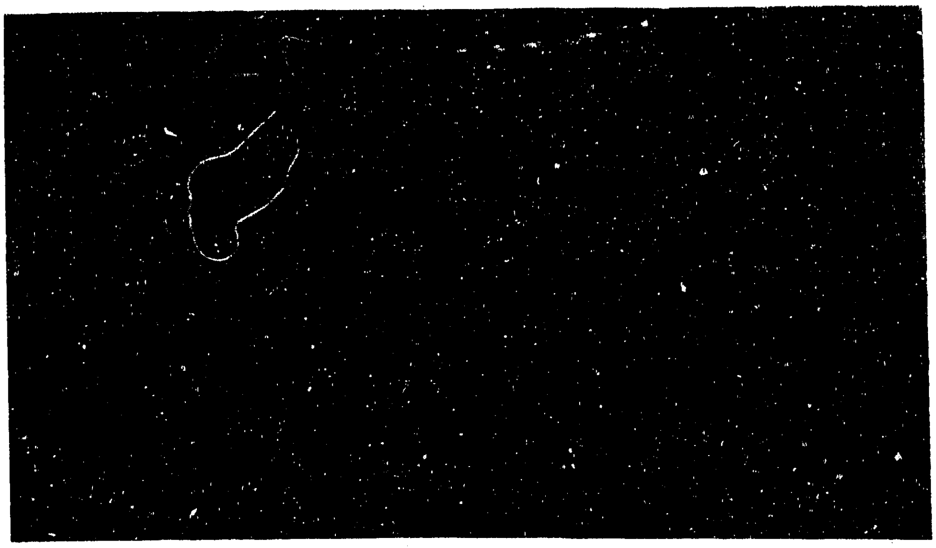

Figure 17. compo 400x at the center of the crucible wall

Examination of the outer wall of the crucible with a low magnification backscatter image revealed the presence of Pu on that surface, see Figure 18. An analysis at a higher magnification was done to confirm the presence of Pu and its' mode of attack. Figure 19 is a SEI image showing the area of interest. The Pu on the surface is not obvious but Pu in the grain boundaries shows up well, it appears as a surface crack leading to IGA in the interior of the wall. A backscatter image, Figure 20, of this area shows the Pu (white phase) on the surface and in the grain boundaries x-ray mapping for Pu was also done in this area. The results can be seen in Figure 21, the white dots represents the pattern for Pu. The x-ray map clearly shows Pu in the boundaries and on the surface. Figure 22 is a x-ray map of Pu made on the outside surface of the wall again showing a concentration of Pu there. Ca was also present on the outer surface of the crucible, as determined by x-ray mapping as shown in Figure 23.

The microprobe data indicates the presence of Pu throughout the grain boundaries and on the outside surface of the Ta crucible. The data also supports IGA as the main mode of attack.

Figure 18. 200x Compo outside surface Ta crucible

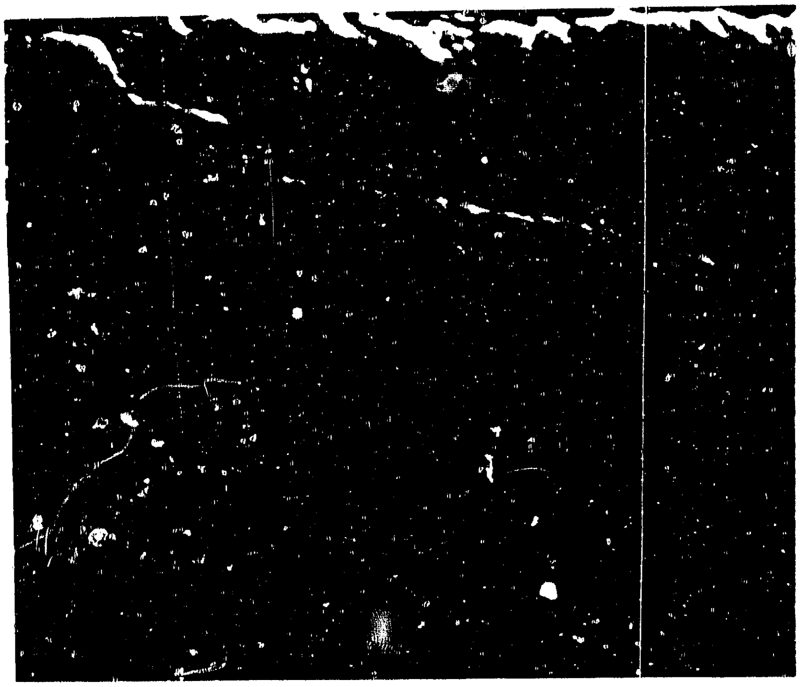

Figure 19. 1000x SEI of the outside wall of the Ta crucible.

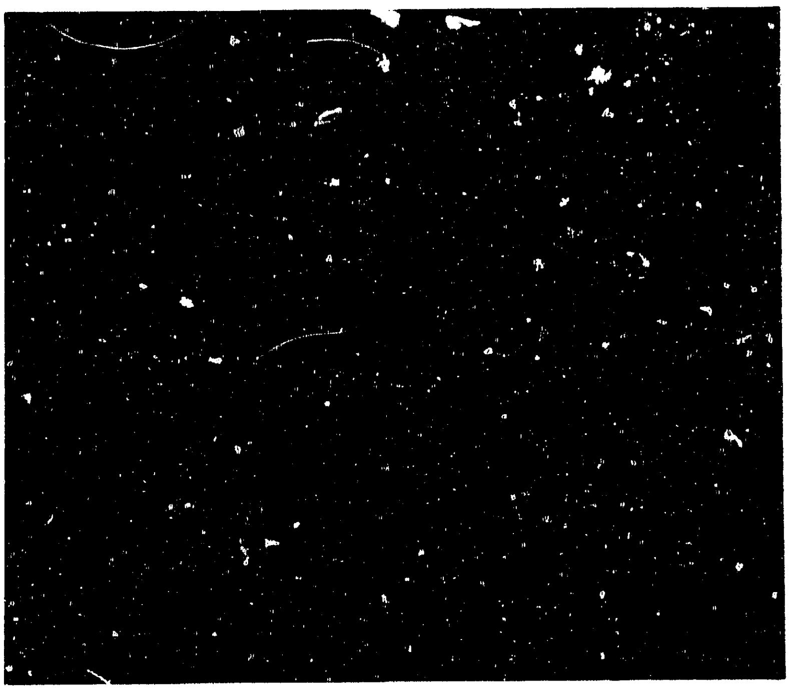

Figure 20. 1000x Backscatter image of the outside wall of the Ta crucible.

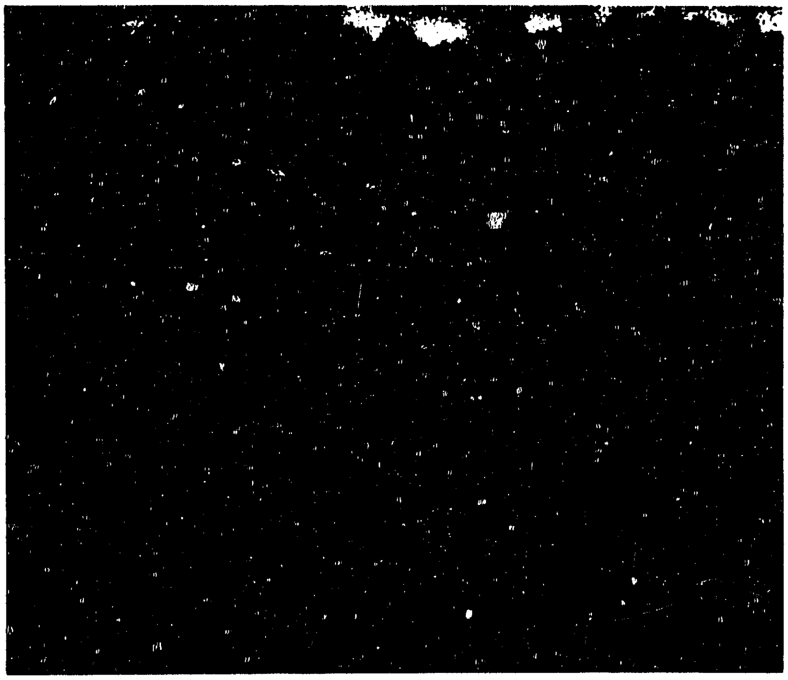

Figure 21. 1000x X-ray mapping for Pu.

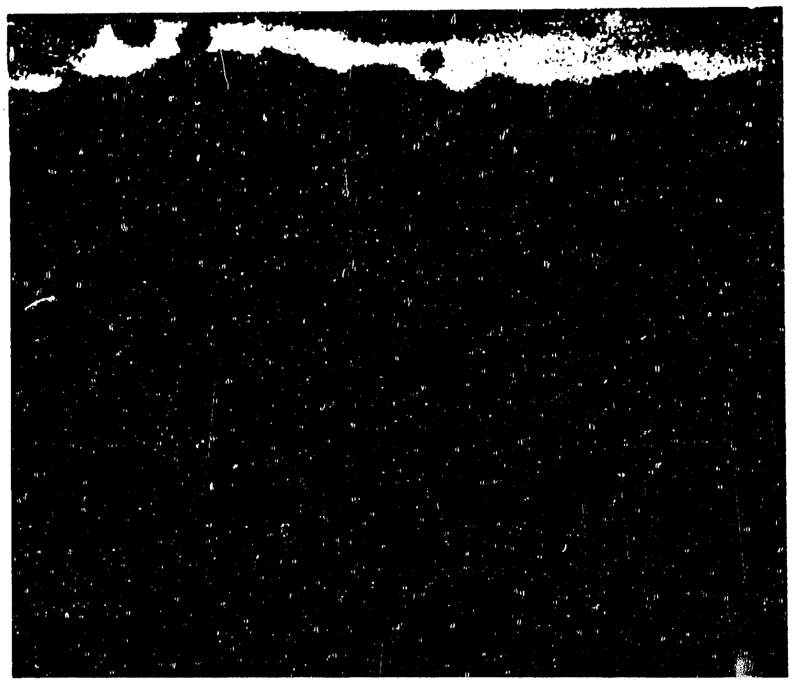

Figure 22. 1000x X-ray map for Pu at the outer surface of the crucible.

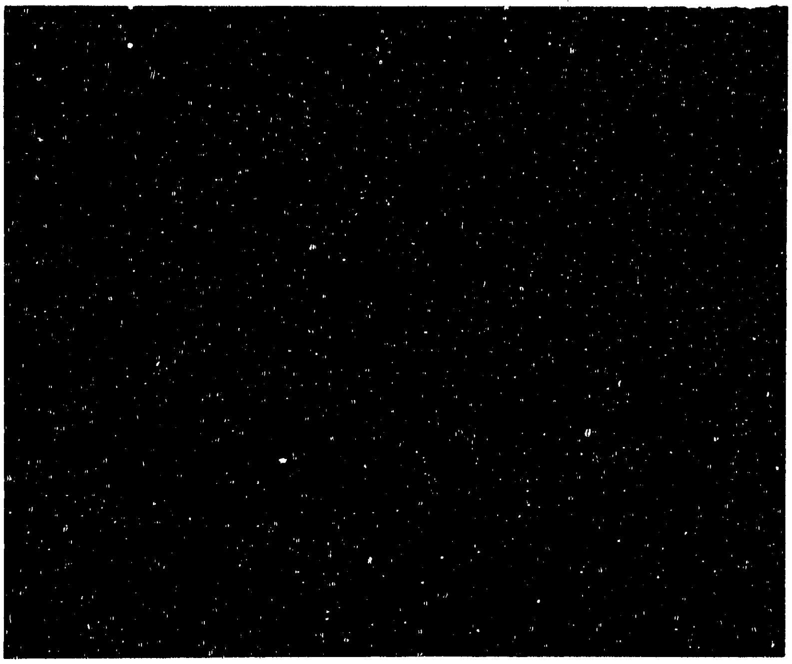

Figure 23. 1000x X-ray map for Ca at the outer surface of the crucible.

ME2945 ETCHED STRUCTURES

The samples were etched with a solution of 30cc lactic acid, 30cc HNO_3 and 30cc HF cleaned and photomicrographs taken. Figure 24 shows the etched microstructure of the Ta from above the meniscus. The microstructure shows a cold rolling reduction of 50 to 60%. No Pu attack was seen. Figure 25 shows the etched microstructure from below the meniscus.

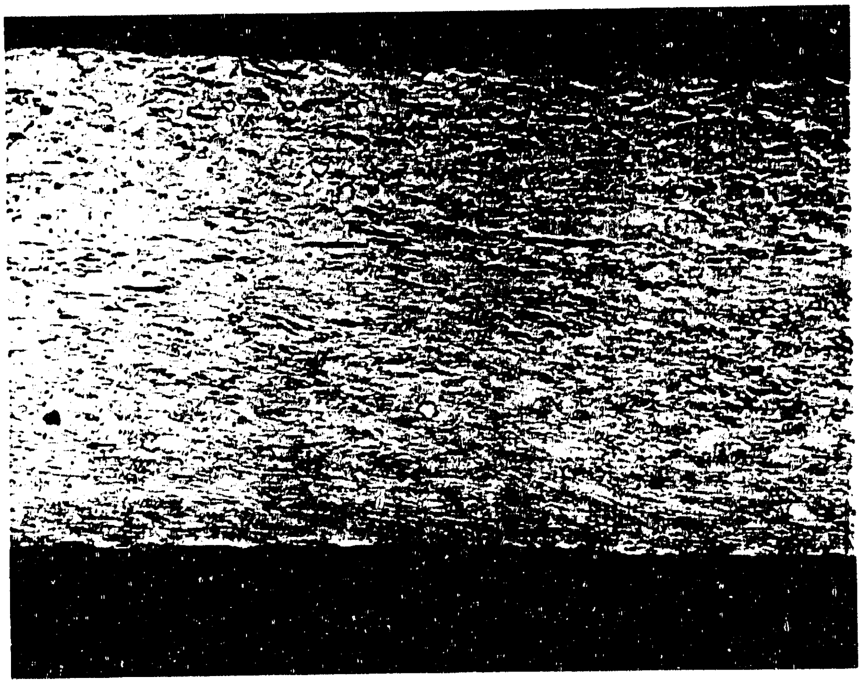

Figure 24. 50x as etched crucible wall

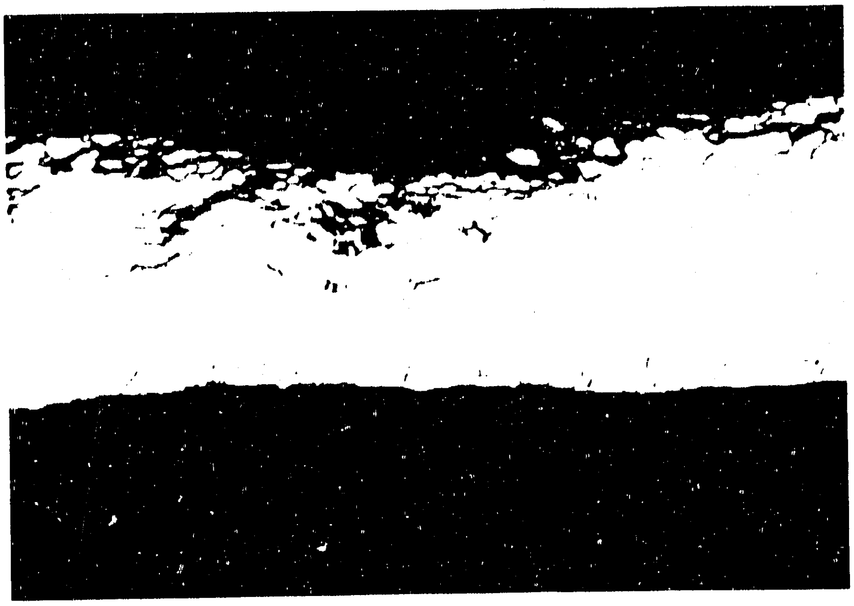

Figure 25. 50x as etched below meniscus.

Figure 26. 200x Etched structure, inside surface with some free floating grains and a major crack.

Figure 27. 500x Etched structure center of wall, Pu can be seen in the grain boundaries at the center of the photomicrograph.

Figure 28. 200x etched outside edge. Pu can be seen on the surface and in the grain boundaries. The Pu in the boundaries has been removed by the etchant.

CONCLUSION

Metallographic examination of the Ta crucible used in the MSE runs showed extensive intergrainular attack by the liquid metal. The crucible wall was breached by the liquid Pu at one point; Pu was found on the out side of the crucible wall and in the grain boundaries of the Ta. This IGA reduced the wall thickness by 77% in some areas. The steps on the attack appear to be: (1) the molten Pu wets the grain boundaries of a Ta grain, (2) Ta become detached from the crucible wall, and (3) the Ta migrates in the liquid Pu. As more and more grains are lifted out, the wall thickness is reduced. Some dissolution of the Ta also takes place but the solubility of Ta in Pu is low. The localized attack that reduces the wall thickness of some areas, see Figure 31, at a faster rate than others is not clearly understood. The presence of a convection current in this area could bring unreacted Pu into contact with the Ta wall and thus speeding up the reaction of Pu with Ta.

The IGA of Ta by liquid Pu is well documented. An example of this can be seen in photomicrographs and microprobe data taken from a Ta crucible used in a Trident run. Figure 31 is a cross section of this crucible. Figure 29 shows Pu on the inside wall and in the grain boundaries. Figure 30 shows Pu in the triple points of the Ta grain boundaries.

Figure 29. 50x As polished

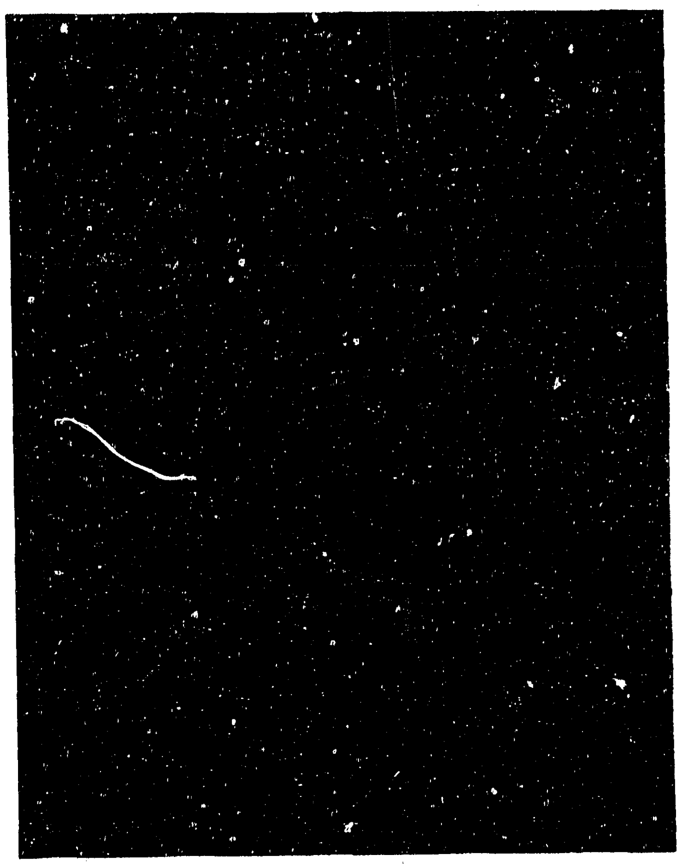

Figure 30. 500X As polished

Figure 31. 50x As polished

Figure 32 shows Pu in the grain boundaries of the Ta, at the marked triple point a x-ray mapping for Pu was done; Figure 33 shows the results of the x-ray map. Pu appears as white dots, the x-ray map clearly shows Pu in the triple point of the Ta grain boundary.

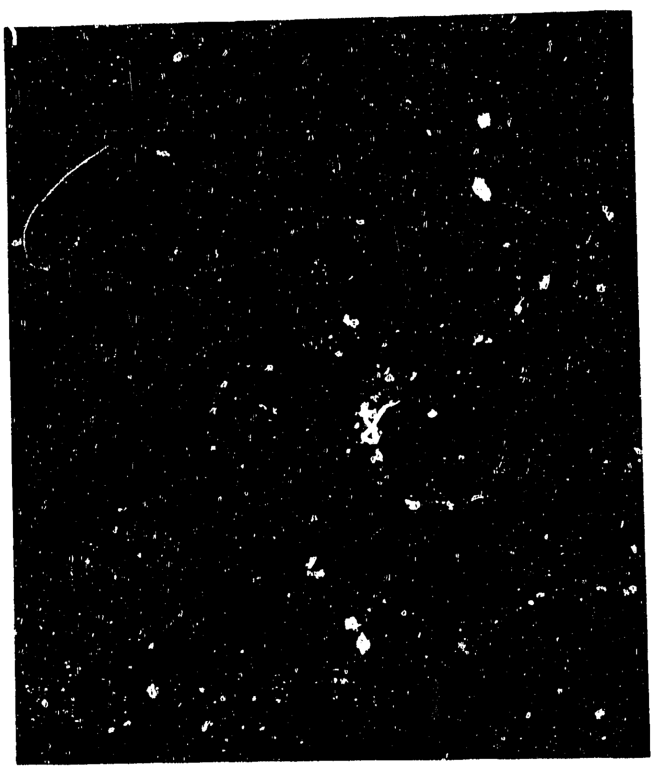

Figure 32. 100x As polished

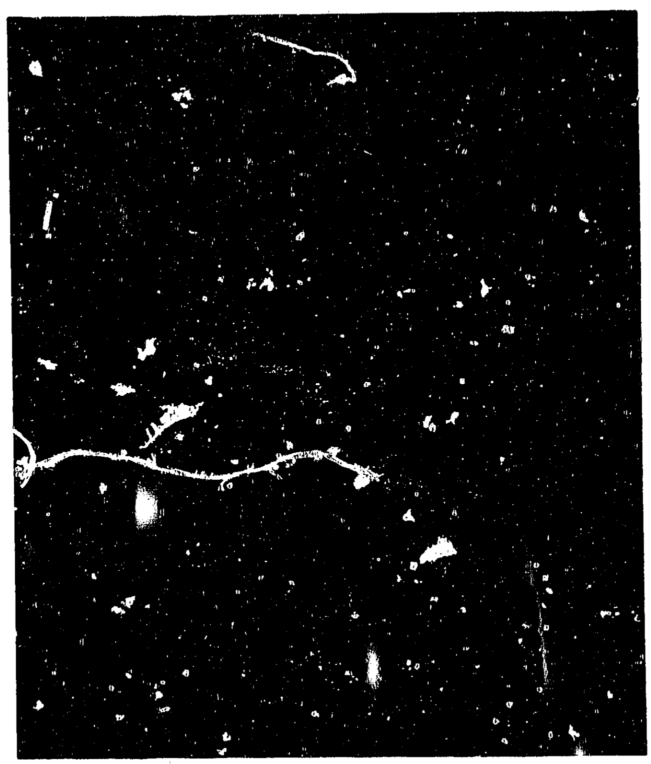

Figure 33. 1000x X-ray map Pu.

Due to the rapid rate of intergrainular attack of the Ta by liquid Pu it is not recommended for prolonged use as a container for molten Pu.

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