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MATERIALS TECHNOLOGY
INFORMATION BRIEF

Title: Failure Analysis of Cracked FS-85 Tubing and ASTAR-811C End Caps

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Date: FEB 9 2006

Abstract: Failure analyses were performed on cracked FS-85 tubing and ASTAR-811C end caps which had been fabricated as components of biaxial creep specimens meant to support materials testing for the NR Space program. During the failure analysis of cracked FS-85 tubing, it was determined that the failure potentially could be due to two effects: possible copper contamination from the EDM (electro-discharge machined) recast layer and/or an insufficient solution anneal. To prevent similar failures in the future, a more formal analysis should be done after each processing step to ensure the quality of the material before further processing.

During machining of the ASTAR-811C rod to form end caps for biaxial creep specimens, linear defects were observed along the center portion of the end caps. These defects were only found in material that was processed from the top portion of the ingot. The linear defects were attributed to a probable residual ingot pipe that was not removed from the ingot. During the subsequent processing of the ingot to rod, the processing temperatures were not high enough to allow self healing of the ingot's residual pipe defect. To prevent this from occurring in the future, it is necessary to ensure that complete removal of the as-melted ingot pipe is verified by suitable non-destructive evaluation (NDE).

Background: Prior to the termination of the NR program effort to deliver a space reactor for the Prometheus Project, efforts were in progress to fabricate biaxial creep specimens for the evaluation of material properties. The destination of these biaxial specimens was either the JOYO reactor in O-arai, Japan for irradiation testing or Oak Ridge National Laboratory in Oak Ridge, TN to serve as unirradiated experimental controls. The major refractory metal alloys in this campaign were FS-85 (Nb-27.5%Ta- 11%W-0.85%Zr), ASTAR-811C (Ta-8%W-1%Re-0.7%Hf-0.025%C), and Mo-47.5Re. T-111 (Ta-8%W-2%Hf) and Ta-10W were designated as back-up materials.

Tube hollows for biaxial creep specimens were fabricated from rod stock by electro-discharge machining (EDM), which creates a re-cast layer on the tube ID that must be removed. A two-step procedure was used to remove the re-cast layer. This procedure consisted of mechanically honing the ID surface to remove a minimum of 0.002" from the ID surface, followed by acid pickling to chemically remove an additional 0.001"; this procedure had been demonstrated previously by Pittsburgh Materials Technology Inc. (Large, PA) to completely remove the recast layer typically resulting from the EDM process. The chemically-cleaned tube hollows were then vacuum-annealed to remove any hydrogen pick-up that resulted from the pickling process.

True Tube (Paso Robles, CA), an experienced vendor in refractory metal tube drawing, processed ASTAR-811C, FS-85, and T-111 tube hollows into their final dimensions (OD 0.25 inches, ID 0.20 inches, 0.025 inch wall thickness). During the first week of June 2005, two tube hollows of each refractory metal alloy, ASTAR-811C, FS-85, and T-111, were drawn into tubing. Each material was formed into final sized tubing except

for the niobium-based alloy FS-85. Although one FS-85 tube hollow was processed successfully, the second FS-85 tube hollow exhibited severe cracking along a portion of its length. This behavior was unexpected and metallography was performed to determine its cause.

End caps for biaxial creep specimens were fabricated from rods by machining to the proper shape and dimensions. The rod stock was extruded to a 1-inch diameter from a 3-inch diameter double arc-melted ingot and brought to its final 0.25" diameter by swaging. The material was heated to 250 °C (482 °F) after each pass until the critical 50% reduction point was reached and received an in-process anneal after 90% reduction. During the machining of ASTAR-811C end caps, cracking was observed in the center of the material. This cracking behavior was unexpected and SEM (scanning electron microscopy) imaging was used to evaluate the extent of the cracking and determine its cause.

Experimental: Processing details for the FS-85 tubing and ASTAR-811C end caps can be found in Reference 1.

FS-85 Tubing

The starting (pre-drawn) blank tube and failed FS-85 materials (Figures 1 and 2 respectively) were mounted and examined metallographically to compare microstructures.

ASTAR-811C End caps

The initial inspection on the cracked end caps was done by low magnification examination using the SEM.

Results: *FS-85 Tubing*

Due to the tight processing schedule for these samples, the materials could not be examined metallographically after each processing step. Based on prior experience, the defined annealing time and temperature (1 hour at 1300 °C) for this material were assumed to have recrystallized the material. However, after analysis of the failed drawn tube, it was noticed that the material had not been annealed properly. The annealing conditions for the tubes in Figures 1 and 2 were 1300°C/1 hour. After discovering the improperly annealed microstructure in the failed FS-85 tube, another FS-85 tube was annealed at 1400°C/1 hour. This annealing time/temperature combination yielded the proper, recrystallized structure seen in Figure 3. This microstructure has equiaxed grains, indicating that the annealing temperature/time combination was successful.

Based on the theory that a 1400 °C/1 hour anneal is necessary for proper recrystallization, the successful drawing of one of the two FS-85 tubes at True Tube may have been an anomaly. Because both of the tubes were annealed at 1300 °C/1 hour, they both should have failed. Although the successfully drawn tube is not shown below, the failed drawn tube and successfully drawn tube both are similar in microstructure.

To address concerns that the FS-85 tube failure may have been due to material contamination, the failed FS-85 tube was evaluated via SEM and Energy Dispersive Spectroscopy (EDS) to map the surface chemistry of the sample. No contamination was found by the surface mapping.

Figure 1.
Honed, pickled, and annealed
(1300°C/1 h) FS-85 starting, pre-
drawn blank tube
(as-etched, 200x)

Figure 2.
FS-85 failed tube
(as-etched, 200x)

Figure 3.
Drawn and annealed (1400 °C) FS-85 tube microstructure
(as-polished, 100x)

It was also observed that both samples had black pits throughout their microstructures. The microstructures in Figure 1 and 2 are from the same blank, pre- and post-draw. FS-85 should have dispersed second phase particles of ZrC, Zr(CN), and ZrO₂, but if these second phase particles are removed as a result of metallographic preparation, pores, or pits, will remain in these areas (as seen in Figures 1 and 2).

The pores were then examined under the SEM to determine their origin. Figure 4 shows the porosity at 500x as well as the crack path that was generated during tube drawing.

Figure 4. Failed tube of FS-85 (500x). Circled regions show crack propagation.

SEM and EDS spot chemistry in the pores did not detect remnants of contamination or precipitates. The EDS spectrum in Figure 5 shows only tantalum, niobium, and tungsten was found inside the pores.

Figure 5. Spot chemistry analysis of pore feature in failed FS-85 tube sample

The results shown in Figure 5 may indicate that the result of failure was not due to contamination or precipitates. However, this theory cannot be ruled out completely because the etchant may have been strong enough to completely remove any surface contamination.

EDS was also performed on a bulk area to determine if any contamination was present on the surface of the failed material (Figure 6).

Figure 6. EDS spectrum of bulk FS-85 failed tube sample

Figure 6 shows that the elements found on the surface of the material were the primary

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alloying constituents (niobium, tantalum, and tungsten) and indicates no contamination.

While SEM results revealed no contamination on the surface of the material, the etchant could have been extremely efficient in removing any contamination or precipitates from the surface. A wet chemistry analysis was done on the extruded FS-85, failed FS-85, and successfully drawn FS-85 to determine if any unexpected elements could be detected. The tubes were evaluated to determine if further processing of this material (extrusion to tube formation) caused a change in chemistry. If a drastic change in chemistry was shown between these two tubes, chemical analysis would be performed on a sample taken after each processing step to determine during which step the contamination/precipitation occurred. The extruded piece was used as the beginning of this processing analysis.

The chemistry results indicate there was no contamination and the material was within specification. The chemistries for each sample are in Table 1.

Element	FS-85 Extrusion	FS-85 Un-cracked Tube	FS-85 Cracked Tube
C	<20 ppm	68 ppm	26 ppm
H	2 ppm	6 ppm	2 ppm
N	<10 ppm	<0.10 ppm	<10 ppm
Nb	60.1%	60.2%	60.5%
O	7 ppm	4 ppm	1 ppm
Ta	28.05%	27.79%	27.66%
W	11.01%	11.12%	10.99%
Zr	0.79%	0.82%	0.82%

Table 1. Chemistries of the extruded ingot and un-cracked and cracked FS-85 tubes.

When comparing the main elements of the alloy in each sample, it can be seen that niobium, tantalum, and tungsten are similar in value, and each sample meets the specification required for FS-85. When comparing values in the table, the only element that is questionable is carbon, however, it still meets specification. If carbon levels remain below 100 ppm, not only does the material meet specification, but it also suggests that any problem that should arise in the mechanical properties is not due to the carbon content. The low carbon levels in this material eliminated the notion that excess carbon was the cause of the tube failure.

Micrographs were taken of the FS-85 material after the first melt. Figure 7 shows a typical as-cast FS-85 microstructure and has been documented as uncontaminated FS-85². When compared, no distinct difference is apparent between the ideal microstructure (Figure 7) and that of the failed microstructure (Figure 8) described herein. In each case, second phase precipitates are dispersed throughout the microstructure.

Figure 7. FS-85: Typical as-cast, uncontaminated microstructure² (100X)

Figure 8. As-cast microstructure of failed FS-85 material (100x)

After additional investigation into the possible causes of the FS-85 tube failure, it was determined that one reason for the failure of the FS-85 tube material may have been that the recast layer on the tube ID from the EDM procedure used to form the starting FS-85 tube blanks may not have been completely removed. Copper is generally used for EDM electrodes. After the EDM process, if the recast layer is not totally removed, copper contamination could occur and result in tube drawing failure. Copper is generally insoluble in tantalum and niobium but zirconium and hafnium form multiple intermetallic compounds with copper. These intermetallic compounds would form along the grain boundaries because these intermetallic compounds form low melting eutectics (895 °C). A similar phenomenon was observed during the processing of a tantalum alloy T-222 (Ta-10W-2Hf-0.01C) for the Atomic Vapor Laser Isotope Separation (AVLIS) program³. In the AVLIS program, a slight amount of copper contamination occurred during the final ingot melting operation, but chemical analysis indicated that the copper contamination was determined to be less than 50 wppm and thus was considered acceptable. Subsequent processing of the AVLIS program ingot resulted in failures which could be traced directly to the copper contamination because hafnium and zirconium form series of intermetallic compounds with low melting eutectics. These intermetallic compounds are located on the grain boundaries and being very narrow, are not possible to detect with basic analytical tools. During the SEM as well as the wet chemistry analysis, no copper contamination was evident; however, as stated above, there could have been trace amounts in the material without it being detected.

Further analysis of this tubing was planned to determine the extent, if any, of copper contamination when the NR effort to deliver a space reactor for the Prometheus Project was terminated. However, due to the processes that are necessary for the formation of this tubing and the past AVLIS program results, it is possible that copper contamination is responsible, at least in part, for this failure.

ASTAR-811C End Caps

The initial inspection on the cracked end caps was performed by SEM and EDS; EDS results showed no contamination in the material. Figure 9 shows the surface of a cracked end cap and highlights the crack surface.

Figure 9. Cracked ASTAR-811C end cap

The yellow line in Figure 9 highlights the smooth un-bonded linear defect which was formed during extrusion and swaging of the portion of the ingot which contained the secondary solidification pipe. The end cap was then separated along the linear defect for examination of the crack surface (Figure 10); the red line in Figure 9 indicates the crack path that was created during this separation.

Figure 10. Crack interface of ASTAR-811C end cap

The area outlined in red shows the original crack interface (reference yellow line in Figure 9). The smooth texture of this area indicates that it was formed during solidification and never bonded to another region. The area outlined in blue resulted from the dissection of the end cap. The expected failure surface of the end cap is the ductile cup cone type fracture seen in the blue area. It should be noted that the defect is confined to the center of the material. No cracks were found on the outer rim of the end cap. If improper machining were the cause of the failure, there would be defects on the edges of the end cap-the tooling has the majority of its contact with the circumference of the end cap. Therefore, improper machining of the end caps was ruled out as a cause of the defect.

It can also be seen in Figure 10 that there are no precipitates, voids, or pores in the cross-section of the end cap. Because SEM results show that the crack propagated from the center of the material, it can be assumed that the defect was formed during a process that would have the potential to create holes or cracks. One process that is notorious for the creation of holes in the center of the product is melting. During the melting process, a pipe (large void) may form during solidification. The smooth texture of the area outlined in red in Figure 10 gives support to the presence of a pipe defect; therefore, further chemistry analysis was not performed.

Figure 11. Cross-sectional view: Solidification of an alloy not properly hot-topped

Figure 11 shows the different pipes that can form in a solidified ingot of a refractory metal. The primary reason for a pipe to form is the volume change that occurs during solidification. The ingot is melted using an AC arc furnace. When the arc is turned off, the top of the surface of the ingot solidifies rapidly. The change in volume occurs when the remainder of the ingot begins to solidify, leaving a void between the rapidly cooled top surface and the inner top portion of the ingot. Usually the primary pipe is easily detected by visual inspection and removed; however, because the center region of the ingot is the last to solidify, another pipe can form within this area due to shrinkage. The secondary pipe can be overlooked because it is encased within the ingot. When an electrode is consumed almost completely during melting, the power is turned down to where the electrode stops melting. However, the molten metal is still being stirred allowing for a slower solidification rate. This technique is called hot-topping and if not done properly, it can cause the molten metal to solidify rapidly creating an opportunity for voids to form within the melt. The likely cause for the cracking observed in the ASTAR-811C end caps was the extrusion of a crack-like defect formed at a secondary pipe. The secondary pipe could have been a result of the melt not being hot-topped properly.

After the ingot was released from the mold, it was machined, extruded, and swaged. Figure 12 illustrates how a linear defect evolves. Frame 1 shows the pipe formed during ingot solidification, Frame 2 shows the pipe during extrusion, and Frame 3 shows the pipe during the final swaging operation. The swaging operation creates unbonded surfaces in the center of the end cap. The hole is compressed creating folds that lead to the linear defect in the material. Figure 10 highlights the area in the ASTAR-811C material likely created by the swaging operation.

Figure 12. Illustration of the Progression of a Piping Defect

Only those end caps taken from rod which was formed from the top portion of the ingot contained this type of defect. The ingot was cut in a similar fashion as that displayed in Figure 11.

Conclusions: *FS-85 Tubing*

One potential cause for the failure of the FS-85 tube material may have been the lack of recrystallization in the material. Another potential cause for the failure of the FS-85 tube material may have been the incomplete removal of the recast layer on the tube ID from the EDM procedure used to form the starting FS-85 tube blanks. If the recast layer is not totally removed, a small amount of copper contamination could result in failure during tube drawing. Copper is generally insoluble in tantalum and niobium but zirconium and hafnium form multiple intermetallic compounds with copper. A similar occurrence happened in the Atomic Vapor Laser Isotope Separation (AVLIS) program³. While EDS did not reveal copper contamination, at the time of the restructuring of the Prometheus Project, additional chemistry had been planned to determine the extent, if any, of copper contamination in this material.

ASTAR-811C End Caps

During the melting process, an ingot solidification pipe can extend farther into the ingot than expected. Detecting where it ends can be challenging if it forms similar to that displayed in Figure 11. It appears this issue was the reason for failure of the ASTAR-811C end caps. To prevent this occurrence in the future, a more detailed NDE is necessary to ensure complete removal of the ingot solidification pipe prior to further processing.

Significance to the NR Program:

This investigation exposed potential reasons for failure of the materials in question and provided knowledge 1) on the susceptibility of FS-85 to become contaminated during processing and 2) on the importance of understanding the pipe regions of arc-melted ingots.

References:

- (1) B-MT(SRME)-49, dated October 31, 2005.
- (2) NASA Contractor Report (NASA CR-1609)
- (3) LLNL- Atomic Vapor Laser Isotope Separation (AVLIS) Program (1990-1998)

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