ORNL/CP-99076

CONF-980921--

MATERIALS COMPATIBILITY STUDIES FOR THE SPALLATION NEUTRON SOURCE

J. R. DiStefano, S. J. Pawel, and E. T. Manneschmidt
Oak Ridge National Laboratory
P.O. Box 2008
Oak Ridge, TN 37831-6157
(423) 574-4452

I. INTRODUCTION

The Spallation Neutron Source (SNS) is a high power facility for producing neutrons that utilizes flowing liquid mercury inside an austenitic stainless steel container as the target for a 1.0 GeV proton beam. The energy deposited in the target is transported by two separate mercury flow streams: one to transport heat in the interior target region and one to cool the stainless steel container. Three-dimensional computational fluid dynamics simulations have been performed to predict temperature, velocity, and pressure distributions in the target. Results have generally shown that the power deposited in the bulk mercury can be effectively transported with reasonable flow rates and the bulk mercury temperature should not exceed 160°C. Assuming good thermal contact, the maximum stainless steel wall temperature should be 130°C. Type 316 SS has been selected as the container material for the mercury and consequences of exposure of 316 SS to radiation, thermal shock, thermal stress, cavitation and hot, flowing mercury are all being addressed by R&D programs. In addition, corrosion studies also include evaluation of Inconel 718 because it has been successfully used in previous spallation neutron systems as a window material.

Two types of compatibility issues relative to 316 SS/mercury and Inconel 718/mercury are being examined: (1) liquid metal embrittlement (LME) and (2) temperature gradient mass transfer. Although the phenomenon of LME has been known for over 100 years, it is still not well understood.[1] For LME to occur, a tensile-stressed metal must be wetted by the liquid. Without wetting LME is unlikely, but extensive chemical corrosion by the liquid usually means no LME. In fact, LME is usually most severe near the melting temperature of the liquid and decreases as the temperature is increased. Mercury is known to embrittle several metals, e.g., aluminum, nickel, zinc, and titanium.[2] Hayden and Floreen[3] also found that Fe-Ni was embrittled by Hg-0.1% In, and Krupowicz[4] indicated that types 304 and 304L SS were somewhat embrittled by Hg in slow strain rate tests at room temperature, but types 316 and 316L SS were not.

Temperature gradient mass transfer is a form of corrosion that results from dissolution by the liquid in the higher temperature regions of the system and subsequent deposition of solute in colder regions because of super-saturation. Nickel in 316 SS or Inconel 718 has the highest relative solubility of the elements in these alloys and should be the most susceptible to mass transfer; however, if solid state diffusion limits the supply of nickel at the surface, preferential corrosion of nickel would be suppressed.

II. PRELIMINARY EVALUATION OF WETTING BY MERCURY

Some form of wetting by mercury is required for cooling of the target container. Therefore, prior to conducting LME or mass transfer studies, some qualitative wetting experiments were conducted. In these studies, we found that both stainless steels and nickel-based alloys were resistant to wetting by mercury, particularly at or near room temperature. For example, U-bend samples of type 304L SS were ultrasonically cleaned and then treated as follows:

1. no additional treatment,
2. gold plated (10 μm),
3. acid cleaned with acid subsequently displaced by mercury, and
4. specimen abraided under mercury.

The samples were then exposed to mercury in air for 72 h at 25, 150, 225, and 275°C. There was no discernible wetting until 225°C (very slight) and 275°C. Gallium is a highly active liquid metal that has some solubility in mercury even at room temperature. When small amounts

DISCLAIMER

of gallium (100 wppm) were added to the mercury, some type of "apparent" wetting was noted even at room temperature. However, the mercury-gallium solution that was attached to the surface of the stainless steel after exposure was scum-like in appearance. Further experiments showed that when gallium was added to mercury at 100°C, there was rapid formation of a crusty, gray scum on the surface.

Further experiments were then conducted in a vacuum-inert gas glove box in order to exclude air from the environment. With a small hot plate, the mercury was first heated to 95-125°C, prior to the addition of gallium. After stirring, the gallium went into solution, which remained bright and shiny. Inconel 625 and 316L SS specimens were placed in the mercury-gallium-solution and then visually examined for evidence of wetting under several conditions. Results can be summarized as follows:

1. No scum was observed for additions of up to 0.5 wt. % gallium in mercury, even after cooling the solution to room temperature.
2. No visual evidence of wetting of Inconel 625 or type 316L SS was noted up to 230°C for mercury containing up to 2 wt. % gallium.
3. When the 2 wt. % solution was cooled to room temperature a liquid second phase formed. Droplets of this second phase tended to agglomerate and floated on the surface. Heating to >70°C dissolved the second phase into the solution once again.

Subsequent testing for LME or mass transfer corrosion was conducted in "pure" mercury or mercury-gallium solutions as noted. For the LME tests, the test assembly, consisting of the specimen container and the mercury-gallium solution, was ultrasonically agitated for thirty minutes prior to testing.

III. RESULTS

A. Liquid Metal Embrittlement (LME)

Both type 316 SS and Inconel 718 were evaluated for LME in mercury. Except for small areas on the fracture surface that were in mercury, no wetting was observed on specimens in "pure" mercury. In mercury-100 wppm gallium most of the specimen, including fracture faces and sides of the specimen visually appeared wetted by the solution.

Figures 1 and 2 show the results of constant extension rate tensile tests at room temperature for type 316 SS and Inconel 718 in air, mercury, and mercury-gallium. For the type 316 SS, one set of specimens was tested in the mill annealed (MA) condition; another was heat treated to produce a microstructure sensitive to stress corrosion cracking (sensitized); and the third was autogenously welded using the gas-tungsten arc process (weld). Note that for the same material condition, there was no significant change in tensile properties as a result of the test environment. For type 316 SS the fracture mode was unchanged as well and remained ductile (Fig.3).

Tensile data for Inconel 718 did not show any significant changes as a result of testing in the mercury or mercury-gallium environments, however, there was some difference in the appearance of the fractures. Scanning electron microscope fractographs showed typical microvoid coalescence and dimpling over large portions of the central region of the specimens, particularly those tested in air (Fig. 4). For specimens tested in mercury or mercury-gallium, both ductile and brittle features were found (Figs. 5 and 6).

Additional tests have recently been completed to investigate LME effects at 100°C as well as to evaluate an additional method to achieve wetting.

In tests at 100°C in which the procedure was the same as previously used at room temperature (23°C), all three specimens (air, mercury, mercury-gallium) exhibited similar tensile properties. The specimen tested in mercury was wet on most of both fracture surfaces but not on its side surface. In mercury-gallium the specimen was wet on both fracture surfaces and was partially wet on its side surface as well. However, lightly shaking the samples dislodged most of the mercury or mercury-gallium.

A test was also performed on a specimen coated with silver solder along its gage length. The coated specimen was first pre-exposed to mercury at room temperature to permit some dissolution of the tin/silver and then it was transferred to the test container with fresh mercury. When tested at room temperature, the specimen was wet on both fracture surfaces as well as its side surface. Tensile test results were similar to that previously obtained indicating once again no LME effects.

B. Mass Transfer

Initially, screening tests were conducted in which mercury was cycled between hot and cold regions of a sealed quartz capsule containing a specimen in each region (Fig. 7). These tests were conducted at temperatures from 150-200°C above those expected in the target system in order to accelerate the mass transfer process. Weight change results in Table 1 indicate small losses in type 316 SS specimens from both hot and cold regions. Losses increased when magnesium or gallium was added to the

Table 1. Weight change (mg/cm^2) from rocking test of type 316SS and Alloy 718 in mercury after 2800 h (10,000 Cycles: 15 min/2 min)

	Hot zone 350°C			Hot zone 375°C
Material	Hg	Hg-100 ppm Mg	Hg-100 ppm Ga	Hg-100 ppm Ga
Type 316SS				
Hot zone	-0.04	-1.84	-2.81	-7.0
Cold zone	-0.41	-0.07	-0.21	-1.62
Alloy 718				
Hot zone			+1.24	
Cold zone			+0.12	

mercury and when the maximum temperature was increased from 350 to 375°C. In contrast, in a comparison test, Inconel 718 showed small weight increases in both hot and cold zone specimens when exposed to mercury-100 wppm gallium. Metallographic results are shown in Fig. 8. There was no evidence of internal corrosion of either material, only surface dissolution or deposition.

In the above experiments, the quartz containers were partially filled with mercury and the air atmosphere was replaced with helium. After approximately 2500 h of operation, flashes of blue light began to appear inside the containers that appeared to be associated with an electrical discharge as indicated in Fig. 9. A recent publication[5] explained that the repetitive emission of light from mercury moving over glass results from a net charge transfer between mercury and glass. Relative movement of the mercury against the glass drags mercury against gravity until eventually the mercury slips relative to the glass resulting in an electrical discharge and a flash of light.

More prototypic testing for mass transfer effects is continuing in two type 316 SS thermal convection loops as shown in Fig. 10. Each loop contains specimens in both hot (300°C) and cold (240°C) vertical legs. Mercury or mercury-0.1 wt. % gallium is being circulated at approximately 1 m/min for a scheduled period of 5000 h.

IV. SUMMARY AND CONCLUSIONS

Type 316 SS has been selected to contain the mercury target of the SNS. Two types of compatibility issues have been examined: LME and temperature gradient mass transfer. Studies have shown that mercury does not easily wet type 316 SS below 275°C. In the LME experiments, attempts were made to promote wetting of the steel by mercury either by adding gallium to the mercury or coating the specimen with a tin-silver solder that the mercury easily wets. The latter proved more reliable in establishing wetting, but there was no evidence of LME in any of the constant extension rate tensile tests either at 23 or 100°C. Inconel 718 also showed no change in room temperature properties when tested in mercury or mercury-gallium. However, there was evidence that the fracture was less ductile.

Preliminary evaluation of mass transfer of either type 316 SS or Inconel 718 in mercury or mercury-gallium at 350°C (maximum temperature) did not reveal significant effects. Two 5000 h thermal convection loop tests of type 316 SS are in progress, with specimens in both hot and cold test regions, at 300 and 240°C, respectively.

ACKNOWLEDGMENTS

Research sponsored by the Division of Material Sciences, U.S. Department of Energy under contract DE-AC05-96OR22464 with Lockheed Martin Energy Research Corporation.

REFERENCES

1. V. I. Igoshev, L. G. Rogova, L. I. Trusov, and T. P. Khvostantseva, "Embrittlement of Nanocrystalline Nickel by Liquid Metals," *J. of Mat'ls. Sci.*, 29 (1994) 1569-1574.

2. M. G. Nicholas, "A Survey of Literature on Liquid Metal Embrittlement of Metals and Alloys," in Embrittlement by Liquid and Solid Metals, M. H. Kamdar, Ed., *Proceedings of the Metallurgical Society of AIME, Fall Meeting of the Metallurgical Society in St. Louis, Missouri, October 4-28, 1982*, pp. 27-50.
3. H. W. Hayden and S. Floreen, *Phil. Mag.*, 20 (1969), p. 135.
4. J. J. Krupowicz, "Effect of Heat Treatment on Liquid Metal-Induced Cracking of Austenitic Alloys," *Slow Strain Rate Testing for the Evaluation of Environmentally Induced Cracking: Research and Engineering Applications*, ASTM STP 1210, R. D. Crane, Ed., ASTM, Philadelphia, 1993, pp. 193-201.
5. R. Budkian, K. Weninger, R. A. Hiller, and S. J. Putterman, *Nature*, Vol. 391/15, January 1998, pp. 266-68.

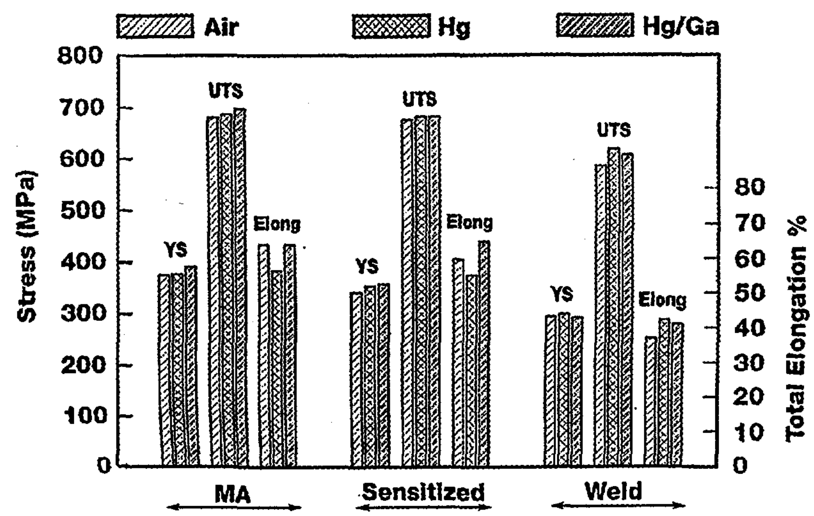

Fig. 1. Mercury or mercury-gallium did not change the room temperature tensile properties of type 316 SS in LME tests.

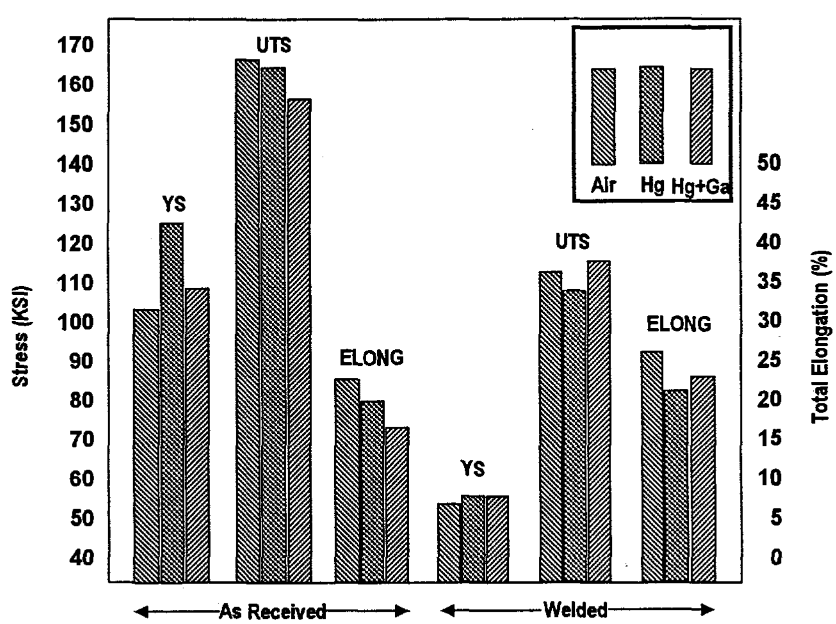

Fig. 2. Mechanical property results from constant extension rate tests for Alloy 718.

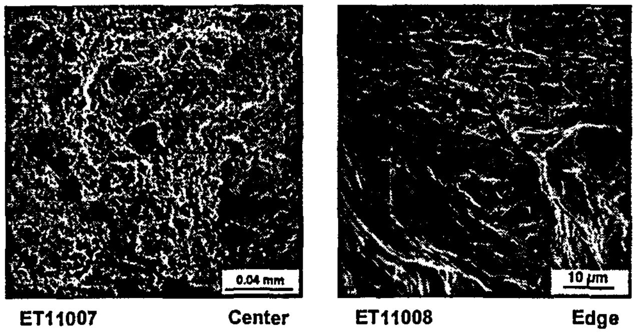

Fig. 3. SEM micrographs showing the appearance of type 316 SS tested in Hg-Ga at room temperature.

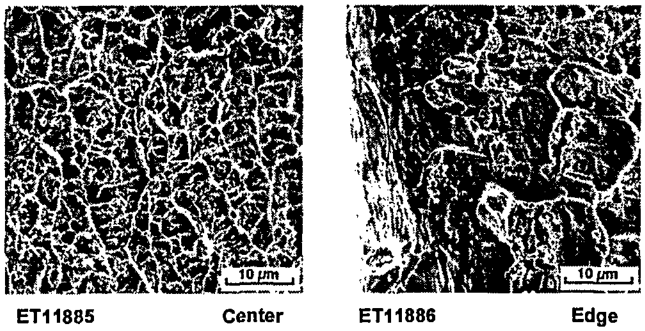

Fig. 5. SEM photograph of the center and edge of the fracture surface of the welded In 718 specimen tested in Hg.

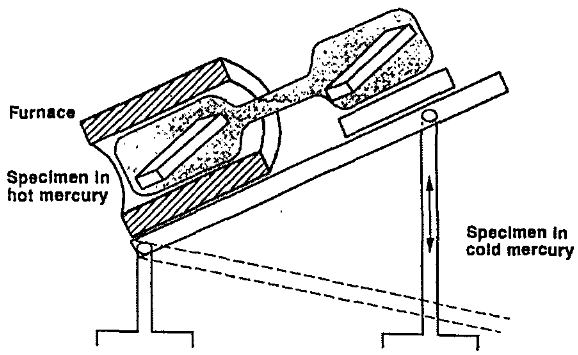

Fig. 7. Rocker apparatus used to screen for thermal gradient mass transfer.

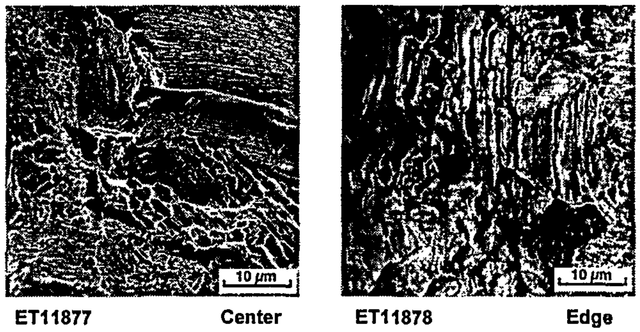

Fig. 4. SEM photographs of the center and edge of the fracture surface of the as-received In 718 specimen tested in air.

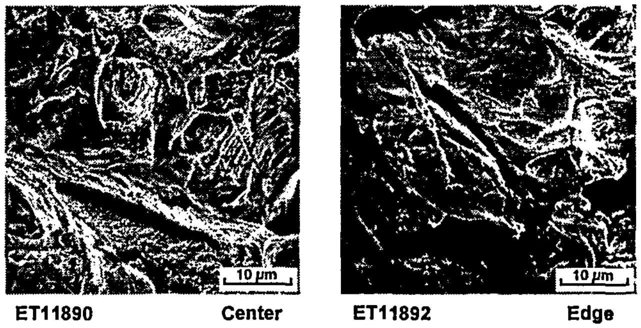

Fig. 6. SEM photographs of the center and edge of the fracture surface of the welded In 718 specimen tested in Hg-Ga.

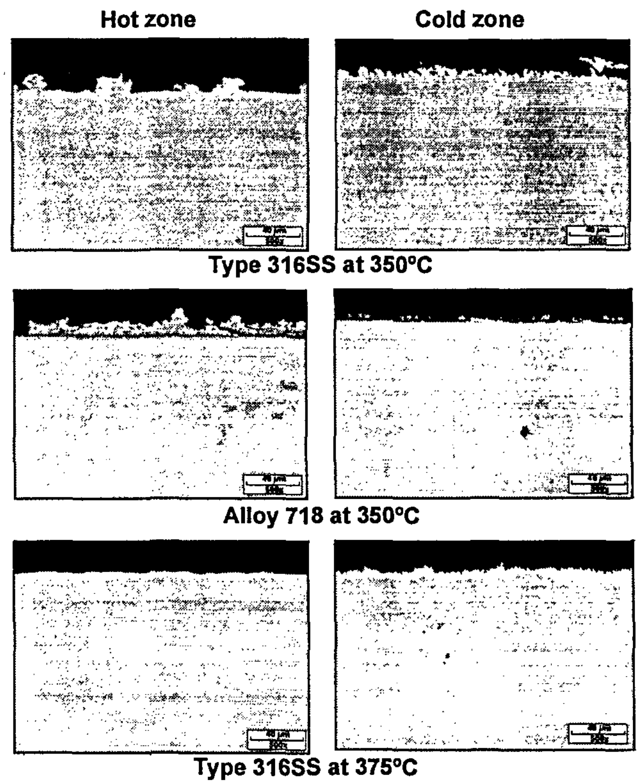

Fig. 8. Behavior of type 316 SS with Alloy 718 exposed to Hg-Ga.

Fig. 9. Light observed in rocking tests after ~2500 h of testing.

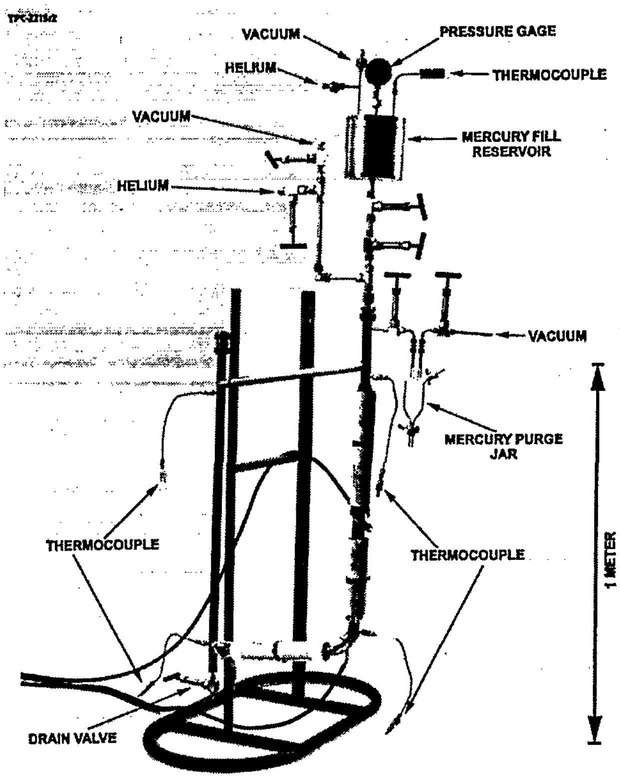

Fig. 10. Mercury thermal convection loop.