Short-Term Solubility of Eight Alloys Circulating in Mercury at Room Temperature

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Because mercury is a dense liquid at room temperature, it has found application in particle-accelerator targets with high energy-deposition rates such as proton spallation targets. Such targets usually require an active heat-removal system to remove the energy deposited by the particle beam, energy deposition rates that can be as much as several megawatts (Weeks, 1996). Solid targets generally require a water-cooling system that can introduce significant complexities into the system. However, a mercury target has several advantages over a solid target, one of which is that the mercury itself can be circulated to remove the deposited heat. Although the service lifetime of a mercury target container may be limited by corrosion or erosion caused by the flowing mercury, the present authors found it difficult to retrieve clear guidance from the literature for our application on the compatibility of various alloys with circulating mercury and even less guidance on the effects of radiation. Although one approach to a potential mercury incompatibility problem could be to schedule periodic removal of a target assembly for examination and possible replacement, it would be useful to have experimental evidence of the performance of a material under representative conditions prior to actual service.

In order to be useful for application in a mercury-wetted environment, a material should form no solid alloys with mercury (Weeks, 1967). Since nickel is known to form intermetallic compounds with mercury as well as with lead and bismuth, it has been recommended that austenitic steels or other alloys with more than a trace of nickel be avoided for use with mercury (Weeks, 1997). Also, chromium is soluble in heavy liquid metals. Therefore, chromium depletion at surface grain boundaries of seasoned steels can be a concern when in contact with mercury. In spite of these concerns, the Spallation Neutron Source project under construction at the Oak Ridge National Laboratory is using 316L Stainless Steel for the mercury target container, and no such difficulties have been observed in laboratory and proton irradiation experiments with 316L Stainless Steel vessels containing mercury (National Spallation Neutron Source, 1997; Haines, 2002). The muon collider project is also proposing to use mercury for their production target in the configuration of a free jet; in very limited tests conducted to date, no mercury incompatibility issues have been encountered (Simos et al., 2001). Although the potential exists for the mercury to become saturated with an alloying constituent, it has been suggested that the soluble components could precipitate or plate out in the cold sections of a flowing loop, thereby promoting the continued dissolution process. It has been suggested that molybdenum, tantalum, or carbide coatings may protect underlying alloys from corrosion by mercury (Weeks, 1997); however, the subject of coatings is beyond the present scope.

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Experimental Studies

The motivation for this work was to investigate the short-term metallurgical effects of mercury on eight alloys to evaluate their suitability for service in a mercury-wetted environment for short durations at room temperature, such as in an experiment of perhaps weeks or months. Although the effects of radiation are of considerable interest, these tests did not include radiation effects. The alloys tested were 316L Stainless Steel, Inconel 600, Inconel 718, TiAlV (all purchased from the Goodfellow Corporation, Berwyn, PA), MP35N (provided at no cost by the Carpenter Technology Corporation, Reading, PA), Havar (provided at no cost by Hamilton Precision Metals, Inc., Lancaster, PA), Aluminum 3004-H19, and Copper B152-ETP. These alloys, with the exception of copper and aluminum, were procured with the mill certified compositions listed in Table 1.

The materials used in this investigation were prepared as described below. The metal samples, with the exception of MP35N, were sheared into rectangular coupons nominally 1-in. wide by 2-in. long. After cutting, the sample lengths and widths were measured with a dial-indicating caliper to a precision of 0.001 in., and the thicknesses were measured with a micrometer to a precision of 0.0001 in. The MP35N was only available in the form of rod stock, and, hence, was used as such. After cutting and measuring, the samples were cleaned with detergent and hot water in an ultrasonic bath. They were then rinsed with distilled water and rinsed again in methanol. After air drying, they were weighed to a precision of 0.0001 g

Table 1. Alloy Compositions (weight fractions from mill certifications)

	Alloy							
Metal	316L Stainless	Inconel 718	MP35N	Havar	TiAlV	Inconel 600	Al* 3004	Cu* B152
Fe	0.677	0.185	0.010	0.191		0.080		
Ni	0.130	0.525	0.350	0.127		0.760		
Cr	0.170	0.190	0.200	0.195		0.150		
Mo	0.023	0.030	0.100	0.022				
Mn		0.002		0.016		0.005	0.013	
Ti		0.009	0.010		0.900			
Si		0.002				0.002	0.003	
Al		0.005			0.060		0.960	
Nb		0.052						
C				0.002		0.001		
Co			0.330	0.420				
W				0.027				
V					0.040			
Cu						0.002	0.003	0.999
Zn							0.003	
Mg							0.010	
Other							0.008	0.001

*Compositions of copper and aluminum are not certified.

using a Sartorius model BP121S analytical balance. The samples were then stored in glassine envelopes until they were tested.

In order to rotate the metal specimens in mercury, a rotating-disk sample holder was constructed, as shown schematically in Figure 1. The sample holder had an upper support shaft that could be attached to a stirrer motor assembly to provide circular rotation. On the bottom side of the disk were four clamping fixtures to which individual metal samples could be attached. The entire assembly was mounted vertically and the samples were submerged in a beaker of mercury. All the tests were conducted at room temperature. The cylindrical disk was sized to just fit within the ID of the beaker to ensure vertical orientation of the samples without physical interference with the sides of the beaker while at the same time obscuring the free surface of the mercury to minimize the escape of mercury vapors from the beaker. In addition, a layer of water was placed over the mercury to provide for additional suppression of mercury vapors. The mercury that was used in these tests was distilled in order to ensure that there would be no impurities present at the start of the tests. Fresh mercury was used for the 24-h test and the 50-h test to avoid carrying contaminants from the first test to the second test.

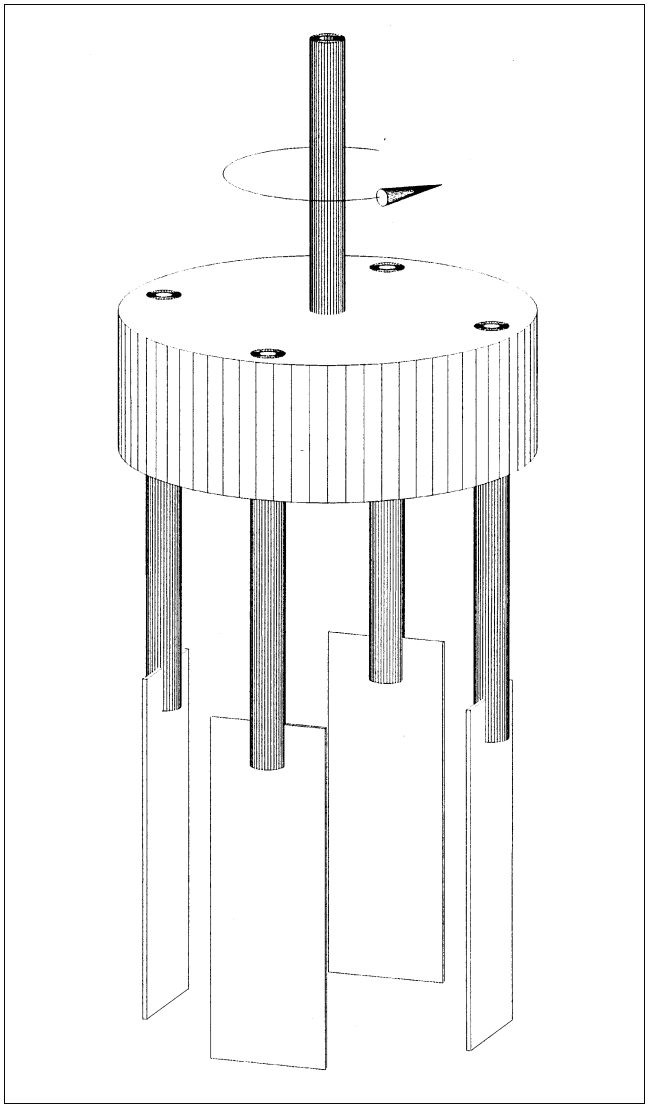

Figure 1. The holder for stirring samples in mercury.

Exposure of the samples to mercury was conducted in a secondary containment pan within a laboratory fume hood. The stirrer motor was energized to the desired rotation rate and the samples were rotated in a circular motion in room-temperature mercury for one to two days at 120 rpm ($\sim$55

Table 2. Test Parameters and Results

Alloy	Width (in.)	Height (in.)	Thickness (in.)	Wetted Area (cm^2)	M_i (g)	M_f (g)	ΔM (g)	Time (h)	Density (g/cm^3)	Mass-Transfer Rate ($\mu g/cm^2 \cdot h$)	Recession Rate ($\mu m/h$)
Inconel 718	0.677	1.368	0.0054	12.066	0.9976	0.9976	0.0000	50	8.19	0	0
316L SS	0.995	1.368	0.0080	17.751	2.0553	2.0553	0.0000	50	8.03	0	0
TiAlV	0.875	1.368	0.0131	15.746	1.4469	1.4469	0.0000	50	4.42	0	0
MP35N**	—	—	—	7.344	23.8589	23.8589	0.0000	24	8.43	0	0
Inconel 600	0.995	1.620	0.0078	21.007	2.1240	2.1240	0.0000	24	8.42	0	0
Havar	1.003	1.368	0.0051	17.823	1.4276	1.4224	0.0052	50	8.30	5.8	0.007
Aluminum	1.010	1.620	0.0487	22.442	4.2221	3.4206	0.8015	24	2.71	1488.1	5.49
Copper	0.830	1.620	0.0098	17.603	2.3496	1.8287	0.5209	24	8.95	1233.0	1.38

*Dimensions listed are mercury-wetted dimensions.
**MP35N was in the form of rod stock.

cm/s). Upon removal from the mercury bath at the conclusion of the tests, it was clearly evident that the copper and aluminum samples were wetted by the mercury over their entire submerged surfaces. The other six metals, however, were not wetted by the mercury and were able to be returned to a shiny condition simply by wiping their surfaces with a swab. The mass of each sample was measured after the mercury exposure to evaluate the effect of the mercury on the alloy. To make the posttest mass measurements, those samples that were compatible with nitric acid were briefly rinsed in 8N nitric acid, washed in water and alcohol, and then dried. This was not possible for the aluminum and copper samples, which had to be cleaned prior to the posttest measurements by mechanical methods. An immersion line that was visible on all of the samples after exposure to the mercury was used to indicate the depth of immersion; a dial-indicating caliper was used to measure from this line to the bottom of the sample to determine the sample's mercury-wetted height. Since these tests were intended to be engineering tests, no metallurgical examinations were conducted.

Discussion of Results

The test parameters and results for the eight alloys tested are listed in Table 2. The 316L Stainless Steel sample, both Inconel samples, and the MP35N sample all exhibited no change in mass over the duration of their respective tests. The only visible evidence of immersion of the samples after cleaning was a barely perceptible immersion line that coincided with the mercury/water interface. This result is consistent with the guidance in Callahan (1985) for 316L Stainless Steel, Inconel, and an alloy of similar composition to MP35N. The results for copper and aluminum were also consistent with the guidance in Callahan (1985). As expected, the copper and aluminum samples both exhibited significant mass loss, at rates of 1233.0 μg/cm^{2}h, and 1488.1 μg/cm^{2}h, respectively. The aluminum sample lost approximately 19% of its mass, while the copper sample lost 22% of its mass during their 24-h exposure. These results support the well-known fact that aluminum and copper are both unsuitable for use with mercury.

It was suspected by the authors that the TiAlV alloy might be unsuitable for use in mercury since Callahan (1985) lists titanium and aluminum as incompatible with mercury (see Table 1). However, the sample that was tested exhibited no mass loss for its 50-h immersion, and exhibited no visible signs of metallurgical attack on its surface. From the measured gravimetric data and visual observations, there was no indication that TiAlV alloy performed any differently than the 316L Stainless Steel sample, the Inconel samples or the MP35N sample. A slight mass loss of 5.2 mg was measured in the Havar sample, which represented a 0.36% mass loss over 50 h. This represents a mass loss rate of 5.8 μg/cm^{2}h. Careful review of the mass measurements for this sample convinced the authors that this mass loss, although unexpected, was real. The authors believe that a machining burr may have been dislodged after initial weighing and resulted in the unexplained mass loss, because no surface corrosion was visible by microscopic examination after cleaning.

Pawel et al. (2001) reported on mass-transfer studies with 316L Stainless Steel and alloy 718 in flowing mercury in thermal convection loops. They measured the mass loss of numerous test coupons after exposure to flowing mercury at temperatures between 250 and 310°C for up to 5000 h. Surface depletion of chromium and nickel from the 316L Stainless Steel coupons was confirmed in these tests; however, no surface corrosion was measured for alloy 718, presumably because the surface was not wetted by mercury. The authors concluded that surface wetting was a necessary condition for metallurgical attack by mercury. For the 316L Stainless Steel coupons that did erode due to the mercury, average surface recession rates in the range 4×10^{-4} μm/h to 5×10^{-3} μm/h were reported. Extrapolation of these rates to the conditions for the present experiments (assuming the following parameters: surface area = 20 cm^2, duration = 50 h, density = 8 g/cm^3), suggests that mass losses in the range 3 mg to 40 mg should have been measured. Differential mass measurements in this range, although easily measurable, were not measured in the present experiments (copper and aluminum are not under consideration here), reinforcing the conclusion that the six alloys tested in this study were immune to metallurgical attack even in flowing mercury at room temperature, in agreement with the observations of Pawel et al. that the cutoff temperature for metallurgical attack on 316L Stainless Steel is above 250°C.

Conclusions and Recommendations

The parameters and results for these tests are listed in Table 2. Although the copper and aluminum experienced considerable mass loss due to dissolution in the mercury as expected, the other six alloys performed well. Five of the six alloys exhibited no mass loss; only Havar experienced any mass loss, and even that was insignificant and not believed to be the result of corrosion by mercury. These six alloys demonstrated satisfactory performance in room temperature mercury for short-duration service. The following conclusions and recommendations are made:

- The reported results should be considered strictly valid for the conditions of these tests only, and extrapolation of the reported performance to radiation environments, long-term mercury service, or elevated temperatures is not recommended.
- Other metals such as beryllium, titanium, and ferritic steels should be studied in future mercury-compatibility investigations, as should epoxies, elastomers, and refractories.
- Investigations of the effects of coatings on mercury erosion and corrosion would be useful, especially coatings of molybdenum and tantalum. Carbide coatings that can improve corrosion and erosion resistance are particularly attractive.
- Studies in radiation environments would be necessary before performance in mercury-wetted radiation environments could be assessed. Life-cycle tests for durations approximating expected service lifetimes in circulating mercury are recommended.
- Metallurgical evaluations of the mercury-wetted surfaces would be useful to identify the depletion of specific elements to distinguish between erosion and corrosion. Such examinations would help to identify the mechanisms of corrosion.
- Materials are now finding application in mercury-wetted environments where they not only must resist chemical solu-

bility by mercury but other challenges such as severe and cyclical shock impact, proton beam-induced pitting, and radiation damage. As materials find applications in increasingly hostile and challenging environments, it will become increasingly more important to know the mercury compatibility of materials prior to assessing their suitability under such challenges. A simple apparatus such as that described in this article could provide useful information on the performance in flowing mercury, lead-bismuth, and other liquid metals.

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