Journal of Nuclear Materials 377 (2008) 174–181

Contents lists available at ScienceDirect

Journal of Nuclear Materials

journal homepage: www.elsevier.com/locate/jnucmat

Cavitation-erosion resistance of 316LN stainless steel in mercury containing metallic solutes

S.J. Pawel *, L.K. Mansur

Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831-6156, United States

ABSTRACT

Room temperature cavitation tests of vacuum annealed type 316LN stainless steel were performed in pure mercury and in mercury with various amounts of metallic solute to evaluate potential mitigation of erosion/wastage. Tests were performed using an ultrasonic vibratory horn with specimens attached at the tip. All of the solutes examined, which included 5 wt% In, 10 wt% In, 4.4 wt% Cd, 2 wt% Ga, and a mixture that included 1 wt% each of Pb, Sn, and Zn, were found to increase cavitation-erosion as measured by increased weight loss and/or surface profile development compared to exposures for the same conditions in pure mercury. Qualitatively, each solute appeared to increase the tenacity of the post-test wetting of the Hg solutions and render the Hg mixture susceptible to manipulation of droplet shape.

Published by Elsevier B.V.

1. Introduction

The spallation neutron source (SNS) generates neutrons via interaction of a pulsed (60 Hz) 1.0 GeV proton beam with a liquid mercury target. The high energy pulses are expected to give rise to thermal-shock induced pressure waves in the Hg which, after reflection from the container surfaces, will result in negative pressure transients and cavitation in the target liquid [1,2]. Some of the energy released during the collapse of the cavitation bubbles will be manifested in a jetting action of liquid at extreme velocity that can potentially erode the adjacent containment material.

Based on a favorable combination of factors, including resistance to corrosion by Hg, well-characterized behavior in a neutron radiation environment, and retention of acceptable ductility after irradiation to moderate displacement doses, 316LN stainless steel was selected as the target container material [3]. However, previous tests have indicated that annealed 316LN is susceptible to potentially significant pitting and erosion damage resulting from cavitation in Hg using a split Hopkinson pressure bar (SHPB) apparatus [4], in-beam exposures [5,6], and a vibratory horn [7–11]. Of particular concern is the observation that pit depths resulting from relatively brief SHPB and in-beam exposures (order of 10–200 cavitation pulses/events), if linearly extrapolated over the anticipated target service life of several hundred million pulses, would prematurely threaten the integrity of the target containment.

It has been shown that the cavitation-erosion resistance of annealed 316LN may be improved by hardening the material via surface treatments and/or cold-working [10,11]. Further, cavitation-erosion resistance of alternate materials may be somewhat superior to 316LN [10], but these alternate materials may exhibit less desirable or less well known properties for other aspects of the target service requirements.

It has also been noted that dissolving a significant amount of metallic elements into Hg leads to changes in surface tension characteristics of the liquid. In one particular experiment [12], it was observed that a 4 wt% addition of alloying elements (primarly 1% each of Pb, Sn, and Zn along with smaller amounts of other elements) increased the surface tension about 35% compared to pure Hg. Further, droplets of the modified Hg formed at the end of a capillary tube were observed to become much larger and to stretch longer prior to breaking compared to the pure Hg counterparts, and the authors suggested the alloyed liquid appeared to be on the path to semi-solid behavior. If this would be the case, cavitation behavior in highly alloyed Hg might be expected to be significantly different than that in pure Hg, particularly if the apparently semi-solid behavior of the alloyed Hg caused cavitation bubbles to collapse at lower velocities, thereby imparting less potential erosion damage to nearby containment surfaces.

In contrast to previous work [10,11], which examined cavitation-erosion in Hg as a function of the physical properties of potential containment materials and various surface treatments, the concept under investigation here considers Hg composition as a potential variable to mitigate cavitation-erosion damage in a given structural material. While there are no doubt practical issues associated with alteration of the composition and fluid properties of the target Hg (such as pumping characteristics, neutron performance, and isotope generation/waste handling), the purpose of this laboratory effort was to compare cavitation damage on 316LN produced by pure Hg with that produced by Hg with various alloying additions.

---

* Corresponding author.
E-mail address: pawelsj@ornl.gov (S.J. Pawel).

0022-3115/$ - see front matter Published by Elsevier B.V.
doi:10.1016/j.jnucmat.2008.02.048

2. Experimental

Binary phase diagrams [13] for Hg in combination with all other elements for which data were available were examined in the search for readily soluble alloying additions to Hg. Diagrams were located for Hg in combination with 74 different elements (although a few of the diagrams were calculated rather than observed/measured). Among these 74 elements, ambient temperature solubility in Hg was essentially nil for a large number of them and at least 1 wt% for only eleven elements. Of these eleven, three (Cs, Rb, and Tl) were dropped from further consideration in these screening tests due to handling dangers in the pure state – primarily the potential for explosive oxidation. Of the remaining eight soluble elements, Bi and Mg were deleted from the list of potential solutes because lab experiments suggested they oxidized too readily and presented very sluggish dissolution kinetics. The remaining six elements appear in Table 1 along with other information about each potential solute.

It has been observed [12,14–16] that metals dissolved in Hg tend to have an extremely high activity in the atomic state and are therefore relatively reactive with even trace oxygen in the environment. As a result, oxides of the solute metal tend to form quickly as a scum on the surface of the host Hg, and these oxides may also catalyze the oxidation of Hg itself. This effect is very readily apparent visually, as nominally clean/pure Hg has a very shiny silver surface whereas alloyed Hg exposed to air exhibits a dull gray surface layer that appears to encase the Hg. The scum may be somewhat self-limiting in that its formation slows or precludes further reaction with air unless regularly disturbed (e.g., by stirring). In the absence of air (for example, within inert gas chambers), the oxidation reaction is drastically retarded and perhaps even eliminated on a practical time scale. To minimize the oxidation of alloying elements in Hg, the test vessel used in these experiments included a feature permitting the surface of the Hg to be flooded with inert gas during solution preparation and testing.

In addition to the cover gas requirement, a test vessel with a relatively small volume of Hg was deemed useful in order to limit the total amount of solute necessary to make the test solutions. Previous cavitation tests in this laboratory utilized $\sim$750 ml pure Hg within a wide-mouth stainless steel dewar, but a glass vessel with a working volume of $\sim$50 ml was fabricated for these experiments. Due to the limited volume of Hg in the test vessel, specimen surfaces were immersed only to a depth of approximately 2 mm for the test exposures. (Immersion depth and solution volume are variables with some influence on the quantitative results of cavitation tests – see Ref. [16] for additional details).

Because of the intense energy/work associated with a vibratory horn cavitation test, the solution test temperature tends to increase during sonication, particularly within a limited solution volume. To maintain a constant temperature of the Hg bath during testing, the glass test vessel was fabricated with a double wall to permit circulation of coolant around the Hg bath to maintain a constant Hg test temperature of 30–31 °C. Taking into consideration the desire for an inert gas cover, reduced volume, and cooling capability, the vessel and test arrangement schematically depicted in Fig. 1 was used for all the tests presented here.

Table 1
Potential solutes for Hg in cavitation-erosion testing

Element	Ambient solubility in Hg (wt%) [13]	Relative dissolution kinetics[a]
Zn	$\sim$1	Sluggish
Ga	$\sim$2	Rapid
Cd	$\sim$5	Rapid
In	$\sim$52	Very rapid
Sn	$\sim$1	Slow
Pb	$\sim$1	Slow

[a] $T \leqslant 30$ °C, modest agitation, argon cover gas.

All cavitation-erosion tests were performed using a titanium vibratory horn and the general test methodology described in ASTM G-32 [17]. The working face of each test specimen had a surface area of 180 mm^2, and the specimen was attached to the vibratory horn via a threaded shank. Photos of the vibratory horn and representative specimens are given in Ref. [10]. The horn tip oscillated at a fixed frequency (20 kHz) and was set to generate a peak-to-peak vibrational amplitude of approximately 25 μm. The rapid reciprocating displacement induces the formation and collapse of cavities in the liquid near the specimen surface, and cavitation-erosion damage from collapsing cavities can be quantified by measurement of specimen weight change and/or erosion depth as a function of exposure time. The baseline test condition included immersion of the working surface of the specimen to a depth of about 2 mm in approximately 30 ml of Hg, and a Hg test temperature maintained at approximately 30 °C.

Nominally pure Hg (filtered through cheesecloth until the luster associated with the Hg surface was very bright/shiny; chemical analysis detected only $\sim$85 ppb Ag and $\sim$100 ppb Si above detection limits) was used for baseline testing and for alloying. Solutes examined in this set of screening tests included pure Hg with 5 wt% In, 10 wt% In, 4.4 wt% Cd (solubility limit), 2% Ga (solubility limit), and a mixture – following the example in Ref. [12] – that included 1 wt% each of Zn, Pb, and Sn. Other details associated with preparation of Hg solutions appear elsewhere [16].

The test specimens were machined from a block of 316LN stainless steel that was also used as a source for specimens in previous testing [10,11,16]. The composition of the 316LN is given in Table 2. All specimens were machined from the original cross-rolled plate material such that the few inclusions present were oriented parallel to the test face. Following machining and light sanding of the test surface on 800 grit paper, the specimens were loosely wrapped in Ta-foil and vacuum annealed at 1020 °C at 10^{-4} Pa (10^{-6} torr) or less for 1 h, followed by cooling to less than 300 °C in about 1 h.

Post-exposure cleaning of the specimens consisted of ultrasonic cleaning sequentially in (1) an aqueous solution containing dissolved sulfur species to chemically bind Hg, (2) distilled water, and (3) reagent grade acetone, followed by forced air drying. After cleaning, specimens were weighed and examined with an optical microscope to determine the average cavitation-erosion profile

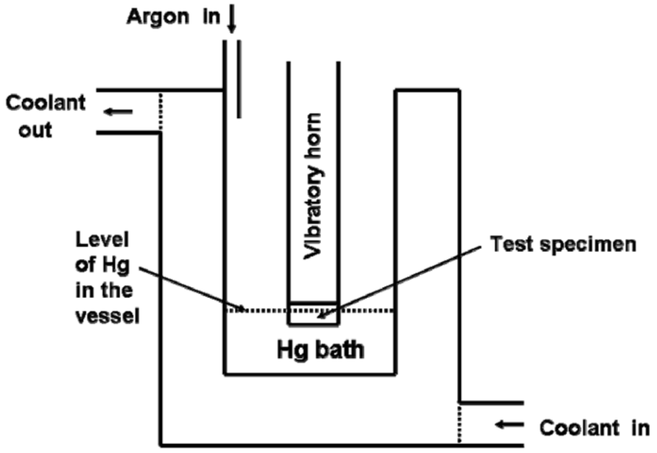

Fig. 1. Schematic drawing of the glass vessel used to contain the Hg in the present experiments. The specimen is on the tip of the vibratory horn with the test surface submerged about 2 mm into the Hg bath. Actual dimensions include an internal vessel diameter of 5 cm and a 1 cm gap between the bottom of the Hg containment and the test specimen surface.