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Journal of Nuclear Materials 318 (2003) 313–324

journal of nuclear materials

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# Influence of mercury velocity on compatibility with type 316L/316LN stainless steel in a flow loop

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**Abstract**

Previous experiments to examine corrosion resulting from thermal gradient mass transfer of type 316L stainless steel in mercury were conducted in thermal convection loops (TCLs) with an Hg velocity of about 1 m/min. These tests have now been supplemented with a series of experiments designed to examine the influence of increased flow velocity and possible cavitation conditions on compatibility. In one experiment, the standard TCL design was modified to include a reduced section in the hot leg that provided a concomitant increase in the local velocity by a factor of five. In addition, a pumped-loop experiment was operated with a flow velocity of about 1 m/s. Finally, a TCL was modified to include an ultrasonic transducer at the top of the hot leg in an attempt to generate cavitation conditions with corresponding extreme local velocity associated with collapsing bubbles. The results indicate that compatibility of type 316L/316LN stainless steel does not depend significantly on liquid metal velocity in the range of 1 m/min to 1 m/s. Benchtop cavitation experiments revealed susceptibility of 316L coupons to significant weight losses and increases in surface roughness as a result of 24 h exposure to 1.5 MPa pressure waves in Hg generated ultrasonically at 20 kHz. However, attempts to generate cavitation conditions on coupons inside the TCL with the ultrasonic transducer proved largely unsuccessful.
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## 1. Introduction

The Spallation Neutron Source (SNS) will utilize type 316L/316LN stainless steel as the primary containment material for a liquid mercury target. To investigate potential thermal gradient mass transfer in flowing Hg, previous 316L/316LN compatibility experiments for the SNS target station [1–4] utilized thermal convection loops (TCLs) with prototypic temperature gradients (about 70 °C) but relatively high peak temperatures (near 300 °C, compared to about 130 °C expected in the SNS) to encourage wetting of stainless steel surfaces by Hg. In the initial TCL experiment [1], coupon wetting and weight loss proved to be a strong function of temperature, with 316L coupons exposed to pure Hg above 250 °C exhibiting the development of a porous surface layer substantially depleted of Ni and Cr. Coupons exposed at lower temperatures revealed no interaction with the Hg (no change in weight, appearance, or microstructure). Analysis of the weight loss data as a function of temperature above 250 °C in the TCL [1] indicated that the rate-controlling step in the corrosion/dissolution process had a relatively low activation energy. In combination with the lack of a gradient of Cr and Ni in the depleted zone, this result suggested rate control by solute diffusion in the saturated liquid boundary layer adjacent to the corroding surface as opposed to solid state diffusion of solute to/across the solid/liquid boundary. This latter observation suggests that corrosion of the 316L target container could be sensitive to Hg velocity at temperatures above 250 °C. Where flow velocity was approximately 1 m/min in the previous TCL experiments, mercury velocity in

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0022-3115/03/$ - see front matter Published by Elsevier Science B.V.
doi:10.1016/S0022-3115(03)00021-7

the SNS target is expected to be in the range of 1–2 m/s near the target window, so a velocity effect on containment corrosion could be significant.

To examine this possibility, two additional experiments to manipulate flow velocity were performed. Initially, the standard TCL design was modified to include a venturi-shaped restricted section to locally increase the Hg velocity by a factor of about five compared to that achieved in the previous TCL tests. Subsequently, a pumped-loop facility was utilized to generate an Hg flow velocity of approximately 1 m/s in the sections containing test coupons.

In addition to potential effects of bulk velocity in the target, extremely high local velocities of Hg at/near the target containment surface are possible through a process termed cavitation. In the SNS, the incident proton beam interacting with the target Hg will be pulsed. The duration of each pulse is short (<1 μs) and the temperature rise of the affected volume is small (a few °C), but the rate of temperature rise during each pulse is exceptionally high (order of $10^7$ °C/s). The expansion of the affected volume with each pulse gives rise to a thermal-shock induced pressure wave that travels into the surrounding Hg. When the compression wave reaches a boundary (e.g., the containment wall), it will be reflected back with a change of phase. The resulting rarefaction wave travels back into/through the Hg, exposing the Hg to transient negative pressures. When the liquid Hg is exposed to sufficient negative pressure, microscopic bubbles are expected to form in the Hg. Previous research [5,6] indicates less than one MPa is required to generate bubbles in Hg of nominal purity at SNS temperatures. When the bubbles collapse (in principle, with each pulse cycle) at/near the containment surface, some of the energy released – typically a 'jetting' action of liquid at extreme velocity – can effectively erode the surface through a scrubbing action. This form of mechanical damage, termed cavitation erosion, potentially could be a localized wastage issue for the target container. In addition, due to the erosive action of the collapsing bubbles, cavitation may be expected to remove the air-formed passive film from the container surface, thus rendering it more susceptible to chemical wetting by Hg and, therefore, subsequent dissolution and mass transfer.

Calculations [7] for the SNS operating conditions suggest that negative pressures sufficient to induce cavitation will be routinely present in the target near the beam window. Therefore, through the process of cavitation erosion, there is a potential for wetting and interaction between the Hg and stainless steel in the SNS even though it will operate at temperatures well below those used in the prior TCL tests. In recognition of this concept, an experiment was performed to examine cavitation as a mechanism to induce wetting in TCLs operating at prototypic SNS temperatures. Although there is uncertainty as to whether the frequency and magnitude of pressure oscillations examined here will be prototypic of the dominant modes expected in the SNS, the ultrasonic energy in these tests could potentially act as a tool to encourage low temperature wetting (at temperatures relevant to SNS, compared to unrealistically high values above 250 °C required to generate wetting in the TCL tests). This is deemed important because wetting of the target containment, particularly near the beam window, may occur as a result of the combined action of irradiation damage, cavitation erosion, and mechanical strains that are not simulated in the TCL experiments. Establishment of wetting via this mechanism could permit evaluation of potential low temperature interactions with Hg that are not evident in the previous TCLs due to the absence of wetting below 250 °C.

## 2. Experimental

### 2.1. Thermal convection loop with reduced section

A schematic of the TCL design is shown in Fig. 1. The loop was fabricated of mill-annealed 316L seamless tubing (25.4 mm ID, 1.8 mm wall). The thermocouple wells, which protruded about halfway into the flow channel, were also seamless, mill-annealed 316L tubing (6.4 mm OD, 0.7 mm wall). The valves and a few other metallic accessories (connectors, transfer lines, etc) were also 316 or 316L stainless steel.

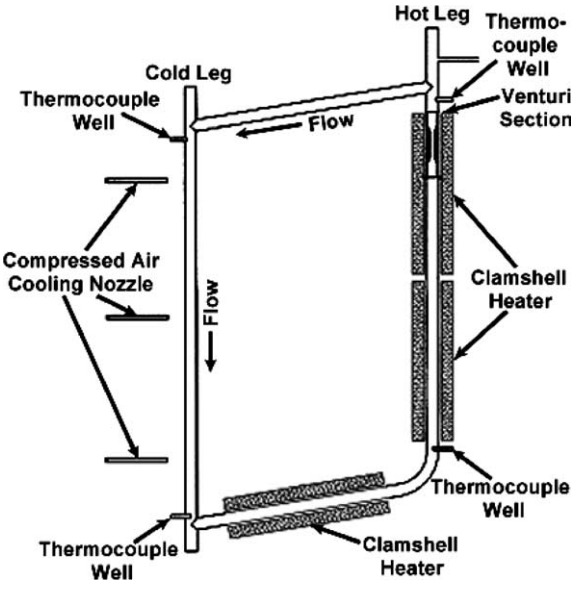


Fig. 1. Schematic of the thermal convection loop design used for these experiments. The approximately horizontal sections are about 1 m apart and the vertical sections are about 0.5 m apart.

In previous TCL experiments (details recorded in [1–3]), the loop tubing had the same diameter all the way around the loop. In the present experiment, the heated vertical leg of the TCL (typically termed the 'hot leg') was modified at the top to include a venturi-shaped flow path designed to locally increase the Hg flow velocity [3]. Several different combinations of maximum and minimum diameters, reduced section lengths, and transition profiles were tested using a glass TCL with water (with about 20% ethylene glycol) as the working fluid operating with about a 60 °C temperature gradient. Fluid velocities in the glass loop were confirmed by monitoring the progress of small suspended particles (neutral density) in the water/antifreeze mixture, and the reduced section which generated the highest local velocity without impeding overall loop flow was implemented in the stainless steel TCLs. Compared to the flow in the bulk of the loop, the average velocity increase in the reduced section for this design was almost a factor of five and included a significant amount of turbulence at both the inlet and outlet sections.

Each TCL in this experiment contained a chain of 316L specimens in both the hot leg and cold leg (the cooled vertical section). Due to the dimensions of the reduced section in the hot leg, specimens for the hot leg were significantly smaller than those used in previous tests [1–3] and were fabricated from a different heat of 316L material than that used previously. To permit easy comparison, specimens for the cold leg chain also used the smaller specimen design even though there was no reduced section. Each specimen chain consisted of 28 small coupons (19 mm × 3.2 mm × 0.76 mm thick, 0.9 mm holes near each end) and two large size coupons (25 mm × 19 mm × 1 mm thick, 1.0 mm holes in each corner) that were the same size/heat as those used previously [1–3]. The specimens were joined together with pieces of 0.4 mm diameter 316 stainless steel wire via the holes in each end of the specimens. The wire attached to the bottom specimen in each chain was welded to the bottom of the respective vertical section to keep the chains from floating to the top of the Hg and to keep the specimens positioned such that the top and bottom of the chain corresponded approximately to the thermocouple well positions in each leg. The compositions of the specimens and tubing are given in Table 1.

Most of the specimens on each chain were exposed in the standard mill-annealed/surface-ground condition. Although each heat of material may have slight variations in treatment, the mill-annealing process nominally describes a solution heat treatment at 1050 °C for 30 min followed by water quenching. The nominal surface finish of the coupons can be described by the average surface roughness (produced by grinding/milling) of 0.8 μm (32 μin.). However, in order to examine potential sensitivity to wetting, two specimens representing each of the following conditions were exposed at specific locations in each of the hot leg and cold leg:

- Gold-coated: both sides of these mill-annealed/surface-ground coupons were sputtered with $Ar^+$ in a high-vacuum chamber to remove the oxide film, then sputter coated with gold to a thickness of approximately 0.6 μm in the same chamber without readmitting air.
- Polished: one side of these otherwise mill-annealed/surface-ground specimens was polished through 1200-grit polishing paper.
- Etched: these mill-annealed/surface-ground specimens were immersed in a 40% reagent grade sulfuric acid solution at 75 °C for about 6 min; the black 'smut' that formed on the specimens was removed by ultrasonic cleaning in acetone.
- Sensitizing heat treatment: these mill-annealed/surface-ground specimens were sealed in glass tubes with a small partial pressure of helium and heated 20 h at 650 °C; subsequently, they were lightly pickled (2 min in ambient 10% nitric acid with 3% hydrofluoric acid in water) to remove the slight surface tarnish.

Table 1
Composition of tubing and specimens (weight percent from mill certification)

| Element | TCL tubing | Venturi section | Pumped loop test sections | Small 316L coupons | Large 316L coupons | Large 316LN coupons |
|---|---|---|---|---|---|---|
| C | 0.03 | 0.024 | 0.018 | 0.010 | 0.022 | 0.009 |
| Cr | 16.27 | 16.24 | 16.56 | 16.67 | 16.05 | 16.31 |
| Mn | 1.30 | 1.83 | 1.81 | 1.41 | 1.82 | 1.75 |
| Mo | 2.06 | 2.12 | 2.14 | 2.28 | 2.08 | 2.07 |
| N | | 0.04 | | 0.079 | 0.04 | 0.11 |
| Ni | 12.16 | 10.19 | 11.24 | 11.11 | 10.11 | 10.2 |
| P | 0.039 | 0.030 | 0.036 | 0.027 | 0.022 | 0.029 |
| S | 0.005 | 0.000 | 0.008 | 0.028 | 0.002 | 0.002 |
| Si | 0.46 | 0.55 | 0.49 | 0.35 | 0.48 | 0.39 |

In addition, the 316LN coupons contained 0.23% Cu and 0.16% Co. By difference, the balance of each alloy is iron.

The specimens in each chain were individually numbered, cleaned ultrasonically in acetone, and weighed (before and after each 'treatment') prior to assembly of the chain. Two nominally identical TCL tests were conducted, except that one TCL received an internal steam cleaning following final assembly and just prior to operation while the other loop experienced no steam cleaning.

After assembly and leak checking, each loop was alternately evacuated (internal pressure of a few microns of mercury) and filled with helium several times. Subsequently, the loop was evacuated and filled with mercury from the reservoir at the top (the ullage of which was also evacuated and purged with helium). Approximately one atmosphere of helium was used as a cover gas for the mercury in the loop.

Virgin mercury from the same batch as that used for previous 316L SS loops [1–3] was used for these experiments. Standard chemical analysis of representative samples indicated the Hg was quite pure, containing only about 85 ppb Ag and 100 ppb Si above detection limits. Immediately prior to use in the loops, the Hg was 'filtered' through cheesecloth to remove the small amount of residual debris (oxides) floating on the surface.

Clamshell-type heaters were placed on the vertical leg (two heaters) and on the near-horizontal lower portion of the loop (one heater) for long term operation. The vertical section of each cold leg was cooled by compressed air delivered from three roughly equi-spaced copper tubes with outlets placed close to the outer loop surface and an array of small fans providing air movement across the entire cold leg. After a day or so of heater temperature and airflow adjustments, the temperature at each thermocouple well position and the mercury flow rate became quite stable. The temperature at each location varied only by about ±2 °C over the entire 2000 h duration of the experiments. The only interruption was a 2-h loss of electrical power to each loop. Table 2 gives the temperatures at each thermocouple well for these TCLs.

The overall average flow rate of the mercury inside the TCL was determined via a localized 'temperature spike' test. In this test, a propane torch was used to heat a small area in the middle of the roughly horizontal section at the top of the loop for about 15 s. The time required for the resultant temperature 'spike' to reach each thermocouple in sequence around the loop along with the distance between thermocouples was used to estimate the velocity of the mercury. The overall mercury flow rate was found to be approximately constant at 1.2 m/min in each loop (same as for previous experiments). Because the flow rate in the similar reduced section in the glass loop (filled with water and neutral density beads) was measured to be about five times faster than the bulk velocity, the flow rate of Hg in the reduced section of the actual TCL was presumed to be about five times faster than the bulk, or approximately 5 m/min.

Table 2
Representative temperatures at each thermocouple well around the TCLs described here

| Thermowell location | Temperature (°C) |
|---|---|
| Bottom of the hot leg | 264 |
| Top of the hot leg | 304 |
| Top of the cold leg | 278 |
| Bottom of the cold leg | 234 |

For an individual TCL, the temperature at any point nominally drifts about ±2 °C over the test duration. The reduced section was located just below the thermocouple well at the top of the hot leg.

### 2.2. Pumped loop

This flow loop – a room-sized collection of tubing, tanks, and operating equipment previously used for thermal-hydraulic experiments in another portion of the SNS research program – was fabricated with 316/316L tubing and components that contact Hg. The Hg velocity was controlled with an electromagnetic pump fabricated by the University of Latvia, Institute of Physics. A series of test section elements fabricated from 25.4 mm OD tubing (with 1.6 mm wall) with machined inserts to contain the test specimens and form the small rectangular flow channel for high Hg velocity adjacent to the specimens were installed downstream of the pump.

The inserts were machined such that the mating halves would hold the test coupons in place and form the Hg flow channel – which was about 12 mm wide and 2 mm deep – along the central portion of each side of the coupon. The length of the insert was determined by the number of specimens it was to contain (either 2 or 10 specimens). The rectangular test coupons – each 25 mm × 19 mm × 1 mm thick with 1 mm holes in each corner – fit into the inserts as indicated by the sketch in Fig. 2. In this configuration, with the mating halves of the inserts pressed together and force-fitted into the test section tubing, the specimens were rigidly held in place. Approximately conical internal contours at the end of each of the inserts provided a transition from the approximately 25 mm circular flow cross-section of the bulk loop to the much smaller rectangular cross-section in the test sections. The test sections were joined to the bulk of the loop by way of nut-and-ferrule compression-type fittings.

The loop design included a bypass around the test section leg that permitted redirecting the Hg flow such that only a small fraction of the 20 l Hg inventory in the loop actually passed through the heaters and test sec-

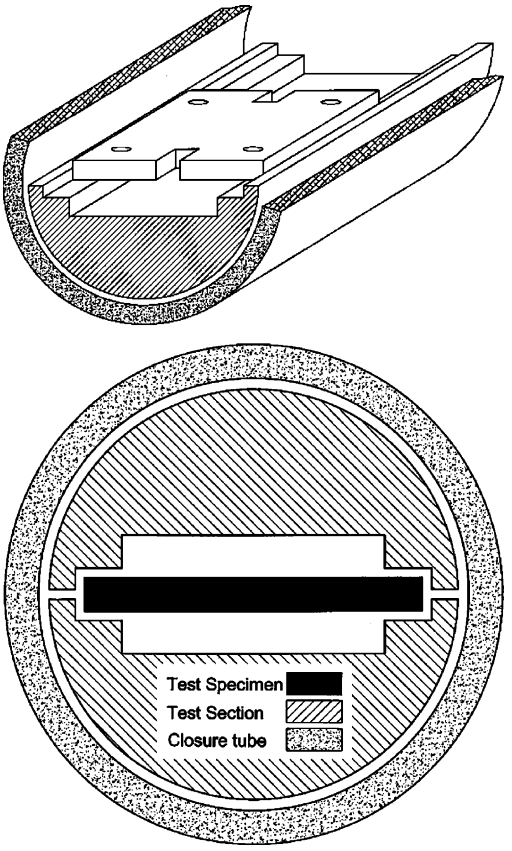


Fig. 2. Schematic of test-section arrangement in exploded view and cross-section showing coupon placement and Hg flow channel formed by the mating halves of the test sections.

tions. The control valve used to determine the flow split was set such that the volumetric flow in the bypass loop was about five times that in the test section loop. This division of flow enabled adequate heating/cooling of the test section Hg without taxing the heat exchanger capability. The heat exchanger utilized Hg on the shell side and chilled water on the tube side. The tests were conducted such that the Hg temperature in the hot leg test section was 250 °C (just downstream of the heaters) and the temperature in the 'cold leg' test section was 100 °C (just downstream of the heat exchanger and pump). Although wetting in the standard TCLs was not observed below 250 °C, this temperature was selected for the hot leg as the maximum temperature that could be reliably achieved (long term operation) and which was an extreme possibility (at hot spots) in the SNS. A schematic diagram representing the pumped loop system appears in Fig. 3.

A test section containing 10 specimens and a test section containing two specimens, along with a bypass valve arrangement, were located in both the hot leg and cold leg of the loop. The bypass valve arrangement permitted the 2-sample test section to be removed from the loop and replaced with a new 2-sample test section during operation. During the cumulative 1000-h exposure, specimens in the 10-sample test section were exposed to the entire test while specimens in the 2-sample sections were exposed to the first 500 h and the second 500 h, respectively.

The corrosion tests were conducted by setting the pump speed to provide a velocity in the test section leg of 1 m/s. Velocity was confirmed using strategically placed venturi flow meters in both the test section loop and the bypass loop.

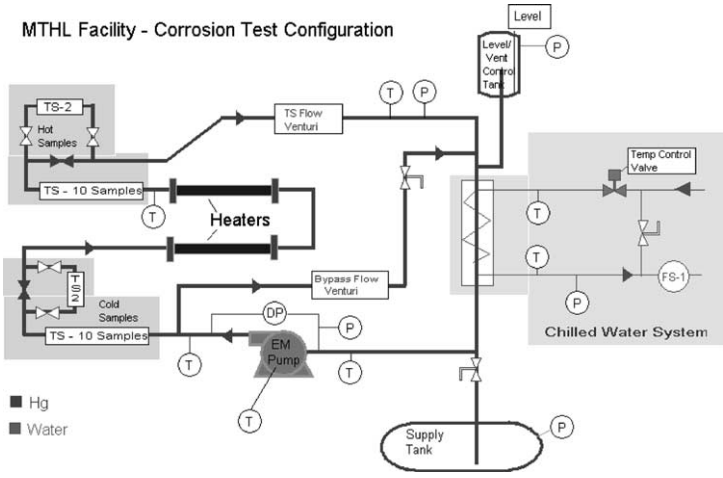


Fig. 3. Schematic diagram of the pumped loop system. Measurement points for pressure (P), temperature (T), and pressure drop (DP) indicated on the diagram. TS = test sections containing 2 or 10 specimens, as indicated.

The rectangular test specimens – identical in dimensions to coupons used previously [1–3] – were 316LN stainless steel with a composition given in Table 1. Many of the coupons were tested in the mill annealed and surface ground condition, but additional conditions were also examined:

- gold-coated as described previously,
- polished as described previously,
- welded: an electron beam welder was used to generate an autogenous weld pad in the central two-thirds of both sides of the coupon,
- oxidized: heated in air for 1 h at 900 °C and slowly cooled in the furnace.

A series of problems with the flange seals and loop heaters caused many starts/stops of the experiment while the initial 500 h of time-at-temperature was accumulated. Each interruption included a drop in the Hg temperature to ambient and the flow rate to zero. In one instance, the coupons were also briefly exposed to air because the Hg had to be drained from the loop. With only one minor perturbation, the second 500 h was accomplished without interruption. The mercury used in this experiment was taken from the same master batch of Hg used in all of these experiments.

### 2.3. Cavitation experiments

In an initial bench-top experiment, the simple arrangement shown in Fig. 4 was utilized. The apparatus consisted of a cylindrical glass container filled with pure Hg from the master batch of material used for all of the compatibility experiments to date. A rectangular coupon (25 mm × 19 mm × 1 mm) of mill-annealed and surface-ground 316L stainless steel, identical to those used in many previous experiments [1–3], was attached to the end of a support rod and positioned in the center of the glass cylinder. A cylindrical ultrasonic transducer was coupled to the outside surface of the glass container and a programmable waveform generator and amplifier were used to drive the transducer. A pill-microphone (and later a calibrated pressure transducer) was used to monitor changes in the wave shape and as an indicator of the onset of cavitation.

Initially, specimens were exposed to the Hg for 24 h at room temperature without power to the transducer as a 'control' experiment. Subsequently, with the ultrasonic transducer driven at ~20 kHz – the first resonant mode of the system – with peak-to-peak pressure wave oscillation of about 1.5 MPa, cavitation was induced on the specimen surfaces for 24 h at 20–22 °C (ambient temperature). These conditions (20 kHz, 1.5 MPa) are considered to be within the realm of possibility for SNS operation [5–7].

Following the initial proof-of-concept tests, the standard TCL design was modified to include an ultrasonic transducer near the top of the hot leg. An approximately 8-cm length of TCL tubing was cut from the region just below the thermocouple well at the top of the hot leg. In its place, a section of tubing from the same heat of material with a donut-shaped transducer affixed with high temperature epoxy was welded as shown in Fig. 5. In addition, a standard flange was welded to the top of the hot leg opening to facilitate placement of the specimens into the area receiving ultrasonic energy.

Instead of the interlocked chain of rectangular 316L coupons used in previous TCL tests [1–3], small pieces of tubing were placed in a stack on a welding wire mounted in the top of the hot leg via a flange. The tubing from which the specimens were cut was mill-annealed 316L with an OD of 3.2 mm and an ID of 2.0 mm, and each specimen was 6.0 mm in length. The welding wire on

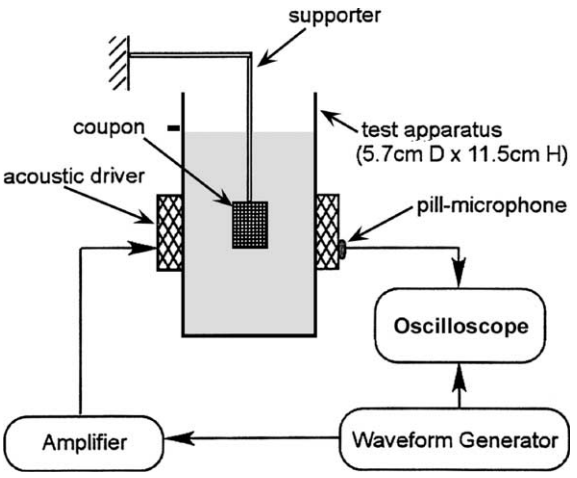


Fig. 4. Schematic arrangement of the bench-top cavitation experiments.

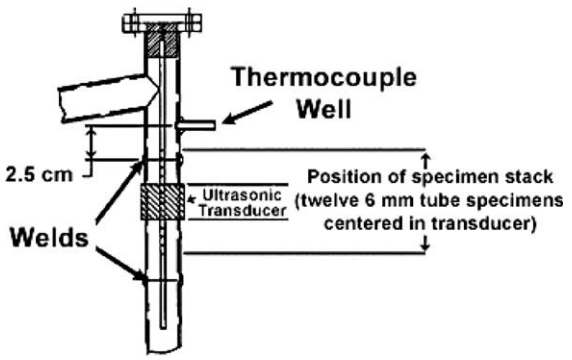


Fig. 5. Modification to TCL which includes an ultrasonic transducer at the top of the hot leg. Also note the specimens were deployed as small cylindrical coupons on a rod loaded from the top of the hot leg.

Table 3
Summary of general test conditions for each experiment

| | Thermal convection loop | Pumped loop | Cavitation exposure |
|---|---|---|---|
| Total exposure time (h) | 2000 | 1000 | 24 (coupons), 140 (loop) |
| Hot leg temperature (°C) | Up to 304 | 250 | Ambient (no gradient) |
| Cold leg temperature (°C) | Down to 234 | 100 | |
| Hg velocity (m/min) | 1.2 (bulk), ~5.0 (venturi) | 60 (test sections) | Bulk solution was stagnant |
| Nominal coupons | Annealed 316L | Annealed 316LN | Annealed 316L |
| Coupon variations | Gold-coated, polished, etched, sensitized | Gold-coated, polished, welded oxidized | |

which the cylindrical specimens were stacked was approximately 1.7 mm in diameter. The individual specimens in the stack were numbered and then cleaned ultrasonically in acetone prior to loading in the stack. The individual specimens were placed such that they were centered at the vertical location of the transducer, as shown in Fig. 5, allowing several individual specimens to be placed 2–3 cm above and below the precise position of the transducer as well as adjacent to the transducer. The remaining space on the specimen stack was filled with 20 mm long pieces of the same 3.2 mm tubing, with the bottom end held firmly in place by a tubing-to-wire spot weld. This specimen arrangement was found to hold the specimens rigidly in place during each test and to permit easy disassembly of the specimen stack for evaluation and reassembly for the next test.

A series of 48–72 h test exposures were performed to determine the optimum conditions for operating the transducer to develop appropriate – and reproducible – cavitation and interaction with the specimen surfaces in the TCL. Details of the loop operation have been recorded previously [8]. The goal was to eventually operate the TCL at temperatures relevant to SNS to determine if cavitation could generate wetting and containment dissolution.

Table 3 gives a summary of the experiments in this investigation. The summary includes exposure time, temperature, velocity, and coupon types.

## 3. Results and discussion

### 3.1. Thermal convection loop with reduced section

Following the test – independent of specific coupon location (temperature) and whether the TCL received steam cleaning or not – Hg was observed clinging to significant portions of most hot leg specimens but little adherent Hg was observed on specimens from the cold leg. It has been a consistent observation in this research that even when wetting of stainless steel has occurred – as evidenced by adherent Hg with a low contact angle, or in some cases [1,4] leaching of Ni and Cr from the interaction surface – extended exposure of the wetted surfaces to air at temperatures below about 250 °C causes the Hg film to bead (become spherical droplets with a high contact angle and little adherence). That some amount of wetting by Hg was observed several hours after exposure of the specimens to ambient lab air suggests that the coupon surfaces in the hot leg were wet to a significant degree during the test. Since this observation applies to both TCLs, it appears that the steam cleaning treatment afforded one of the loops was not significant to the wetting process.

The role of nominal (mill-annealed, surface-ground) 316L coupons on the chain was two-fold: first, to function as near-neighbor control specimens for the other coupons with a range of surface/structure variations and, second, to permit comparison with other coupons around the loop as well as previous coupon data from similar TCL experiments. In summary [3] – independent of coupon position/exposure temperature – the nominal 316L coupons revealed negligible and essentially random weight changes ($\pm$ up to 0.2 mg on 1.5 $cm^2$ for all coupons) and none of the nominal coupons revealed a change in microstructure or surface roughness ($\pm$2–3 μm) compared to virgin coupons. In particular, nominal coupons placed in the reduced section (temperature 300–305 °C, highest velocity), near the reduced section (similar temperature, lower velocity with turbulence), and remote to the reduced section (temperature and velocity lower than in reduced section) revealed indistinguishable results, indicating Hg velocity, within the parameters of this test, is not a significant factor in compatibility with 316L stainless steel. However, the leaching reaction depleting Ni and Cr from the stainless steel surface [1,4] – which was thought to be potentially velocity dependent – was not observed on any coupons in this experiment. Absence of this leaching reaction may be the result of insufficient time at temperature, but in any case the weight loss results in the present experiment are significantly less than in the previous experiments in which the leaching reaction was observed [1,4]. (Compared to the maximum weight loss detected in previous experiments of this type in which the leaching of Ni and Cr was observed [1,4], the weight loss observed here is much lower in all cases. In the former experiment, the maximum loss was about 16 mg

on 10.5 $cm^2$ in 5000 h; correcting for smaller coupon (1.5 $cm^2$) and shorter exposure time (2000 h), the greatest mass loss per area and time in the present case is a factor of 4–5 lower.)

The gold-coated specimens behaved similarly to the nominal coupons. All of the observed weight changes were small – each coupon lost the mass of gold applied plus an amount similar to nearby nominal coupons – and there was no evidence of surface roughening or microstructural changes. This result is consistent with previous tests [2] indicating gold-coating was not particularly effective at increasing the interaction between stainless steel and Hg under these conditions.

The polished coupons all exhibited weight changes similar to nearby nominal coupons and revealed surface roughness and structure indistinguishable from as-polished but unexposed specimens. This result is also consistent with previous tests [2] indicating initial surface roughness is not a significant factor in compatibility within the bounds of the exposure conditions.

Specimens receiving the sensitizing heat treatment were generally a dark golden brown and exhibited weight changes similar to nearby nominal coupons. However, two of the heat-treated coupons exhibited a somewhat larger weight change than nearby nominal coupons. Post-test metallographic analysis of all the heat-treated coupons revealed pronounced intergranular attack around the hole areas on each specimen, while the remainder of the coupon surfaces remained relatively smooth and indistinguishable from the virgin coupon condition – see Fig. 6.

Subsequent investigation of this behavior (details in [3]) revealed that the specimens were substantially carburized adjacent to the holes on each specimen and that the sensitizing heat treatment was generating heavy carbide precipitation about 5–6 grains deep around the hole. Most of the intergranular attack was attributed to the brief pickling treatment these specimens received to remove light oxidation products from the heat treatment, as heat treated specimens without the pickling treatment (but subsequent exposure to Hg at 300 °C in a separate experiment) revealed little or none of the same attack pattern around the holes.

Further study identified the source of the carburization. The specimens for this study were cut from sheet stock using two separate electro-discharge machining (EDM) processes. The outer profile of each specimen was cut with a brass wire electrode and deionized water as the cutting fluid. However, the flat strips were stacked to cut the small holes using a ram-type graphite electrode and a hydrocarbon cutting fluid. In concert with the observations that the carburization (and intergranular attack following pickling) was limited to the hole surfaces and that solution treatment could eliminate the susceptibility to attack near the holes, it appears that the graphite rod and carbonaceous cutting fluid carburized this area significantly rendering it susceptible to sensitization during the heat treatment. Because carburization and potential sensitization can adversely affect the mechanical properties and corrosion resistance (in a pickling solution or any number of environments capable of attacking Cr-depleted grain boundaries or chromium carbides) of 316L/316LN, this result indicates caution is required in selecting machining processes for critical components.

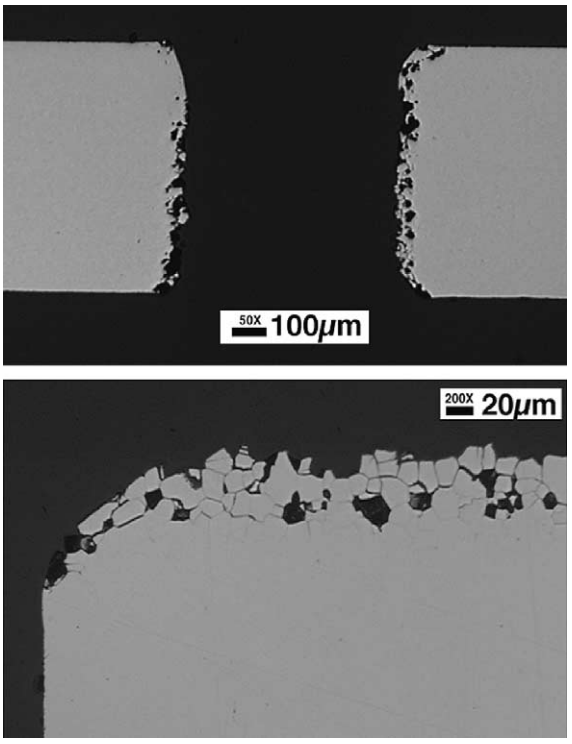


Fig. 6. Intergranular attack observed in heat-treated coupons near the specimen hole. Original magnification (prior to reproduction) is given above the scale marker of specified length.

The etched coupons in the TCLs also exhibited no attack along the bulk surfaces but there was irregular attack on the specimens adjacent to the hole areas. Based on comparison of microstructures for etched specimens with and without exposure to Hg, it is suspected that the heavily carburized material in this area was subject to attack by the etching procedure itself and the Hg exposure contributed little, if anything, to the observed attack adjacent to the holes.

After completion of the experiment, each TCL was destructively analyzed. Cross-sections of the loop were prepared for metallographic analysis from a region near each thermocouple well, and from several areas of the venturi section. In each case, the process side of the tubing revealed no signs of attack (no increase in surface roughness, porosity, or film/deposit) compared to virgin material.

### 3.2. Pumped loop

Post-test visual examination of the coupons from each test section revealed little or no sign of macroscopic wetting. Independent of exposure temperature (100 or 250 °C) and exposure time (up to 1000 h), only a few small regions of the specimens retained any adherent Hg after opening the test sections, which required some amount of jostling. Only the gold-coated and the oxidized specimens appeared discolored compared to the initial condition. In the case of the gold-coated specimens, they each lost all of the dull yellow coloration associated with the virgin coating and looked rather similar to the nominal (uncoated) coupons. (This was true for the entire coupon surface, including the small area that was not exposed directly to the flow channel but was firmly grasped by the sample slot portion of the test section. The Hg velocity in this small region is unknown but likely to be much less than that in the flow channel.) In the case of the oxidized coupons, the rather uniform dull brown color of the unexposed specimens gave way to a rather non-uniform brown, with some areas almost returned to the pre-oxidation treatment color of light silver/gray. (Again, this includes areas of the coupon not directly exposed in the flow channel.)

Weight loss data from all the coupons indicated that only the gold-coated coupons and the oxidized coupons lost weight. The oxidized coupons each lost approximately the mass increase associated with oxidation prior to exposure. Metallographic examination indicates that only small 'islands' of oxide remain on the post-test coupons, but it is not clear from this data if the Hg was able to attack the oxide directly or if the oxide removal was a result of the erosive quality of rapidly flowing Hg. Because the relative affinity of Hg for oxygen is relatively low compared to the oxide formers in stainless steel such as Fe and Cr [9], and because Hg appears to have difficulty wetting stainless steel at relatively low temperature, oxide removal due to an erosive quality of the specimen/Hg arrangement seems most likely. The gold-coated coupons each lost weight corresponding to the mass of gold applied (confirmed by the absence of the dull-yellow film) plus a small additional amount that appeared random in terms of coupon exposure position and temperature. While the gold-coated coupons experienced the greatest net weight loss (up to 2.7 mg over-and-above the weight loss associated with the original mass of applied gold on about 10 $cm^2$ exposed surface area – 7 $cm^2$ directly exposed to flow of Hg, but the other 3 $cm^2$ held tightly by the mating halves of the flow channel were equally cleaned of gold), they exhibited no detectable increase in surface roughness (<2–3 μm increase) or other evidence (e.g., leaching of Ni) of interaction with Hg.

Post-test metallographic analysis of each coupon included a cross-section from the central portion (that was fully exposed in the Hg flow path) as well as near the edge (that was shielded from the Hg by contact with the sample slot). Each coupon surface was found to exhibit isolated regions in which the surface roughness and relief was somewhat irregular, but the extent of the variability appears largely within the scatter of the surface condition on the coupons (features with ±3 μm of surface relief/roughness) and not a function of Hg exposure conditions.

An insert from each of the hot leg and cold leg test sections was also cross-sectioned in several places to search for signs of interaction between Hg and the 316L containment surfaces exposed to the high flow velocity. No indications of attack – such as discoloration, increased surface roughness, or change in near-surface microstructure – were observed on any of the cross-sections.

Because there is no significant change in weight or surface roughness for coupons exposed to Hg flowing at 1 m/s at 100 and 250 °C (and because the small changes observed are equivalent to those for much lower velocity experiments), the results of this experiment suggest that Hg velocity, in and of itself, is not a critical variable in the compatibility of Hg with 316LN stainless steel. However, since no coupons were observed to exhibit the leaching reaction detected in the original experiments [1], it is difficult to estimate the full influence of velocity.

### 3.3. Cavitation experiments

Relatively little work has been done to examine cavitation in Hg as a mechanism by which wetting of stainless steel can occur. In one study associated with SNS [10], polished specimens of stainless steel were attached to the tip of a velocity transformer (vibratory horn) and ultrasonically agitated at 20 kHz in Hg at various vibrational amplitudes to examine cavitation damage on the surfaces. The authors found apparent wetting (low contact angle, adherent Hg) and relatively significant damage on the stainless steel surfaces measured as wastage of many μm/h after up to 8 h in tests with an average Hg temperature of 42 °C. Other authors [11,12] have also examined cavitation erosion of materials in Hg and found qualitatively similar results regarding wetting and damage rates. In addition, a more detailed study of cavitation in Hg specifically in support of the SNS was also undertaken [13]. Frequency and amplitude relevance to SNS aside, however, a subtle but perhaps important difference between these experiments [10–13] and possible cavitation erosion in the SNS target containment is that the target surfaces will receive pressure pulses generated at distant locations in the Hg. In the experiments of [10–13], the surfaces themselves created pressure pulses via externally driven vibrational motion. In the present experiments, rather than injecting energy into the Hg through the test specimen, pressure

waves were created by energy deposited in the Hg from vibrations in the container wall, which then interact with the surrounding structures and specimens.

In the bench-top experiments (apparatus shown in Fig. 4), 24 h exposure at room temperature under cavitation conditions (20 kHz, 1.5 MPa) resulted in apparent wetting, as a thin film of Hg was clinging to much of the 316L coupon surface immediately following the test. Following the cavitation exposure, dewetting in room air was observed to progress over a period of several hours, as evidenced by change of the Hg film to individual beads of Hg resting on the surface with a very high contact angle. Final clean-up was accomplished with a brief ultrasonic treatment in an aqueous sulfur-containing bath that chemically binds Hg, followed by rinsing in water, ultrasonic treatment in acetone, and forced air drying. After this cleaning, the post-test coupon exhibited a significant weight loss (about 10 mg on approximately 10 $cm^2$) and a significantly roughened surface. For comparison, 5000 h at 305 °C resulted in about a 16 mg weight loss on the same size 316L coupon from a TCL in which wetting/interaction was observed [1].

To examine the extent of cavitation damage, it is useful to compare the surface profiles of 316L specimens before and after exposure to cavitation conditions in Hg. Fig. 7 shows representative backscattered electron images of the surface of an unexposed coupon. The photos show that the surface is largely smooth with occasional lapping marks or locations where an inclusion has been dragged from the surface by the machining operation. Profile analysis (details in [8]) indicated that the unexposed surfaces exhibit only ±0.5 μm of surface relief in most areas, with small regions of slightly greater relief.

In contrast, the surface of the specimen exposed to Hg under cavitation conditions exhibited much more surface relief over most of the coupon surface. Fig. 8 gives representative examples of this observation, for which profile analysis indicates typical surface relief of 14–15 μm from the surface to the bottom of the craters.

Electron microprobe chemical analysis of the surface and cross-section of the specimen exposed to room temperature cavitation in Hg did not reveal any leaching of Cr or Ni (which was observed in the case of the maximum interaction of 316L with Hg in a TCL [1]). It is possible that the initial development of a very thin layer depleted in Ni and/or Cr coincides with its mechanical removal from the surface due to cavitation-induced erosion, as any leaching reaction renders the affected material structurally weakened and more susceptible to microcracking or wastage. However, it is also likely that the low temperature ambient exposure was inadequate to leach significant Ni and Cr even if chemical wetting had been established.

Taken together, these observations suggest that the exposure of the coupon to room temperature cavitation contributes to significant mechanical damage to the surface (akin to erosion), but there is no indication of chemical interaction with Hg (for example, leaching of Ni and Cr from the attacked surfaces or residual Hg resulting from tenaciously wetting). In any case, the physical removal of material from the coupon surface as a result of cavitation under these conditions suggests a possible damage mechanism in the SNS target and a path by which relatively low temperature wetting of target surfaces could occur.

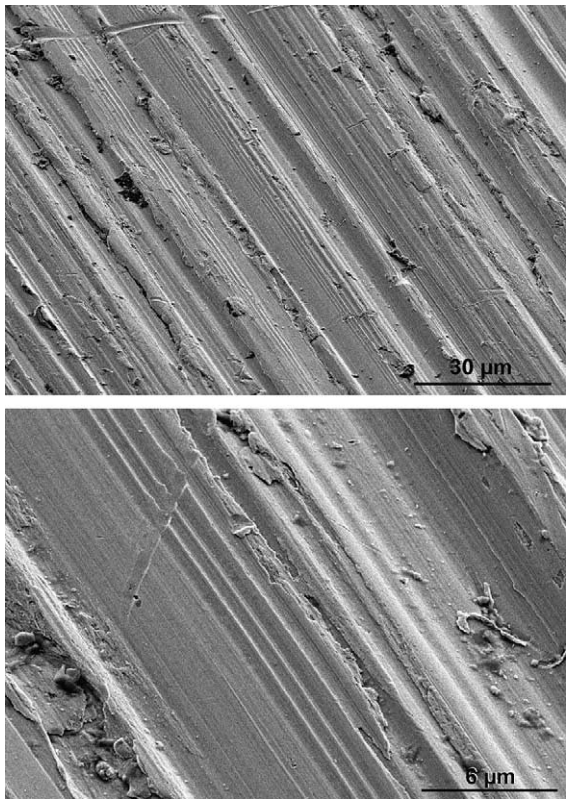


Fig. 7. SEM images of the surface of an unexposed coupon.

Based on the relative success of inducing cavitation on coupon surfaces in the bench-top experiments, a TCL was modified by addition of a donut-shaped transducer at the top of the hot leg as shown in Fig. 5. A number of experiments were performed for a range of transducer settings (frequency and peak-to-peak voltage on the waveform generator) which included the values that induced cavitation in the bench-top experiments. Precise frequency values were selected by maximizing the audible noise coming form the TCL, which was considered indicative of cavitation bubbles in the region of the transducer. However, at the end of each of these experiments, the cylindrical coupon stack was analyzed and none of the specimens revealed any change in appearance (still smooth and shiny, no adherent Hg) or weight.

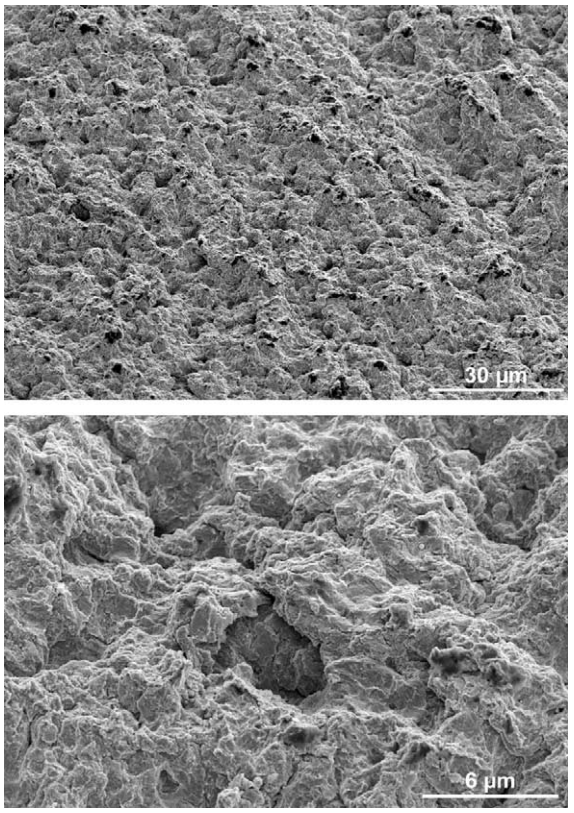


Fig. 8. SEM images of a coupon exposed to cavitation conditions in Hg.

Following a series of unsuccessful attempts to generate cavitation in the TCL, the loop was partially drained of Hg such that a borescope could be inserted into the hot leg to (and past) the position of the transducer to look for cavitation damage on surfaces other than the removable specimens. The ID surfaces of the TCL tubing were examined over a range of several cm above and below the position of the transducer and found to be bright and shiny, and very smooth – only a few scattered drawing marks from the original fabrication of the tube were observed. This result indicates that the cumulative 'damage' for the series of tests in the TCL, which included more than six days at various conditions, left no readily apparent sign of cavitation or wetting on the TCL surfaces. The only minor sign of wetting (clinging Hg) was observed on the thermocouple well at the top of the hot leg. This piece of 316L tubing – but not the TCL walls adjacent to the tubing – appeared 'silvered' by a thin film of Hg. This result suggests that, due to geometry effects, the pressure wave energy was inadvertently focused away from the specimens and that, perhaps, the thermocouple well was acting as the initiator for cavitation onset. This result could not have been predicted from the simplified bench-top experiments, in which only a short cylinder was used compared to the complex geometry of a TCL.

Inability to reproduce cavitation damage in the TCL of the type generated in the bench-top tests in glass and stainless steel tubing led to the discontinuation of these experiments prior to the planned long-term exposure. It is not clear why cavitation/wetting could not be reproduced in the TCL. In addition to the thermocouple well acting as an initiator as noted above, it is possible that the small volume in the bench-top vessel confined the acoustic energy in a way that made it more focused and destructive compared to the very large Hg volume and essentially 'infinite' test cylinder of the hot leg in the TCL. In addition, the shape of the test specimens (cylindrical in the TCL and flat rectangles in the bench-top tests) and/or the protuberances inside the TCL perhaps contributed to a geometry effect that was not anticipated from the simplified bench-top tests. In any case, the attempt to use acoustic energy to generate cavitation and aid wetting in the TCL was considered unsuccessful for practical use.

## 4. Conclusions

In a TCL with a reduced section at the top of the hot leg to increase local Hg velocity, specimens in the reduced section (Hg about 300 °C at about 5 m/min) and specimens outside the reduced section (Hg from about 230 to 300 °C at 1.2 m/min) revealed similar insignificant weight changes and little if any change in visual appearance and surface roughness. In addition, specimens in a pumped Hg loop revealed no significant effects of exposure to Hg at 100 and 250 °C flowing at 1 m/s. Taken together with previous results, the present experiments suggest compatibility of type 316L/316LN stainless steel with Hg does not depend significantly on liquid metal velocity.

In the TCL and the pumped loop, a range of surface and heat treatment conditions for type 316L/316LN was examined and all were found to be similarly compatible with Hg based on insignificant weight and surface roughness changes. However, post-test metallographic analysis of specimens receiving a sensitizing heat treatment (exposed in the TCL) served to highlight a potential fabrication concern for the EDM process. Areas adjacent to holes cut with a graphite rod and carbonaceous cutting oil were heavily carburized and thus rendered susceptible to intergranular carbide precipitation during heat treatment. As the carbides can negatively influence corrosion resistance and mechanical properties, this result suggests caution when specifying machining processes.

In the most extreme example of Hg velocity, bench-top experiments revealed susceptibility of 316L surfaces to cavitation erosion damage in Hg. Significant weight

losses and increases in surface roughness were observed on 316L specimens as a result of 24 h exposure to 1.5 MPa pressure waves in Hg generated ultrasonically at 20 kHz at room temperature. Attempts to transfer the ultrasonic energy source to a thermal convection loop as a mechanism to enhance wetting with Hg proved unsuccessful.

## Acknowledgements

The authors would like to acknowledge the contribution of many helpful individuals. C.B. Herd and L.P. Smarsh fabricated the TCLs. H.F. Longmire performed specimen metallography and L.R. Walker performed microprobe and profile analysis. M.T. McFee operated and maintained the pumped loop and S.H. Kim performed many of the cavitation experiments. R.B. Ogle and S.N. Lewis provided Industrial Hygiene services for controlling mercury exposures. J.H. DeVan (recently deceased) offered many helpful insights over the course of this work. J.R. DiStefano and L.K. Mansur also participated in helpful discussions and provided critical review of the manuscript. F.C. Stooksbury and K.A. Choudhury helped to prepare the manuscript and figures. The work was funded by the Spallation Neutron Source.

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