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**MATERIALS COMPATIBILITY  
STUDIES FOR THE  
SPALLATION NEUTRON  
SOURCE**

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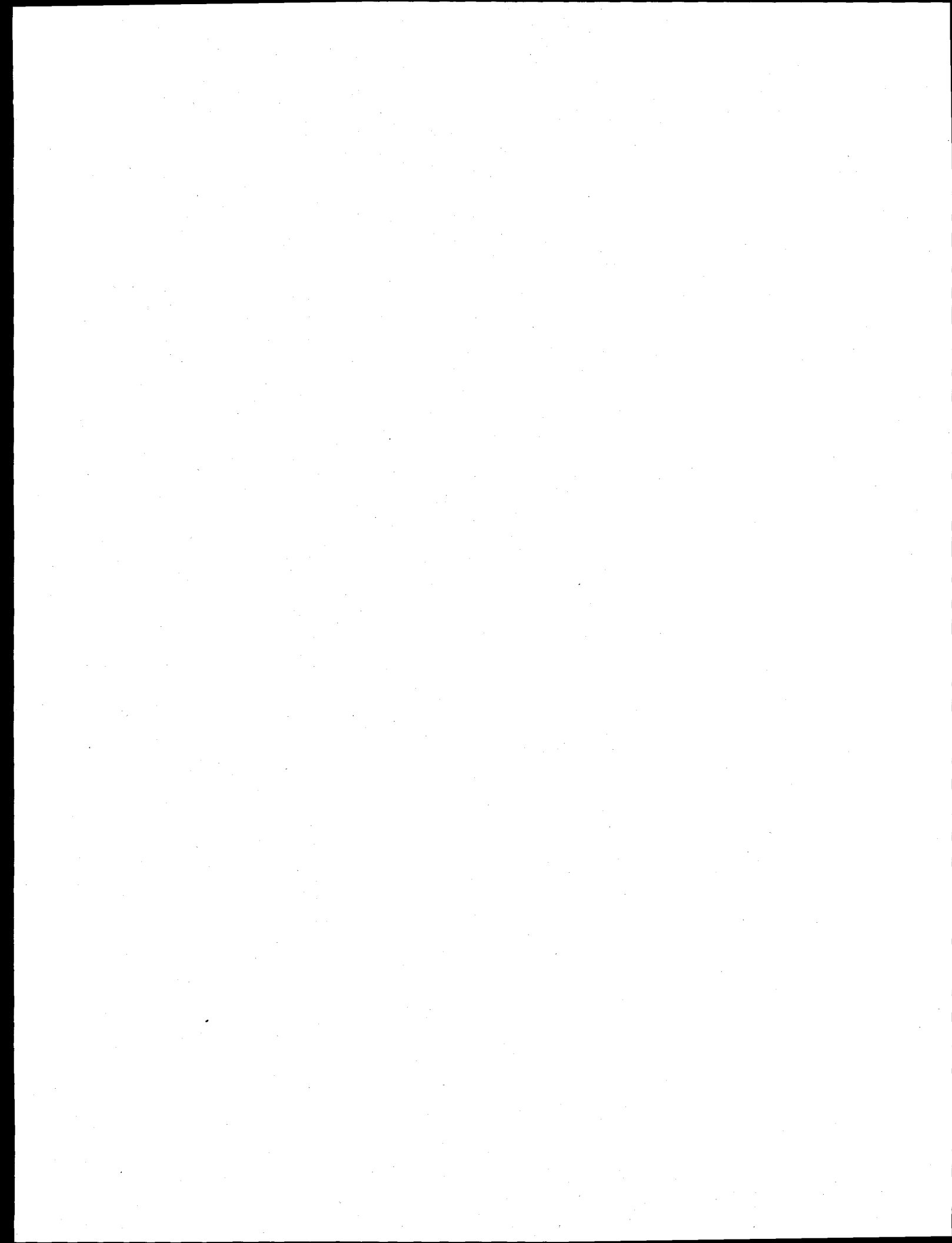
**MATERIALS COMPATIBILITY STUDIES FOR THE  
SPALLATION NEUTRON SOURCE (SNS)**

J. R. DiStefano, S. J. Pawel, and E. T. Manneschildt

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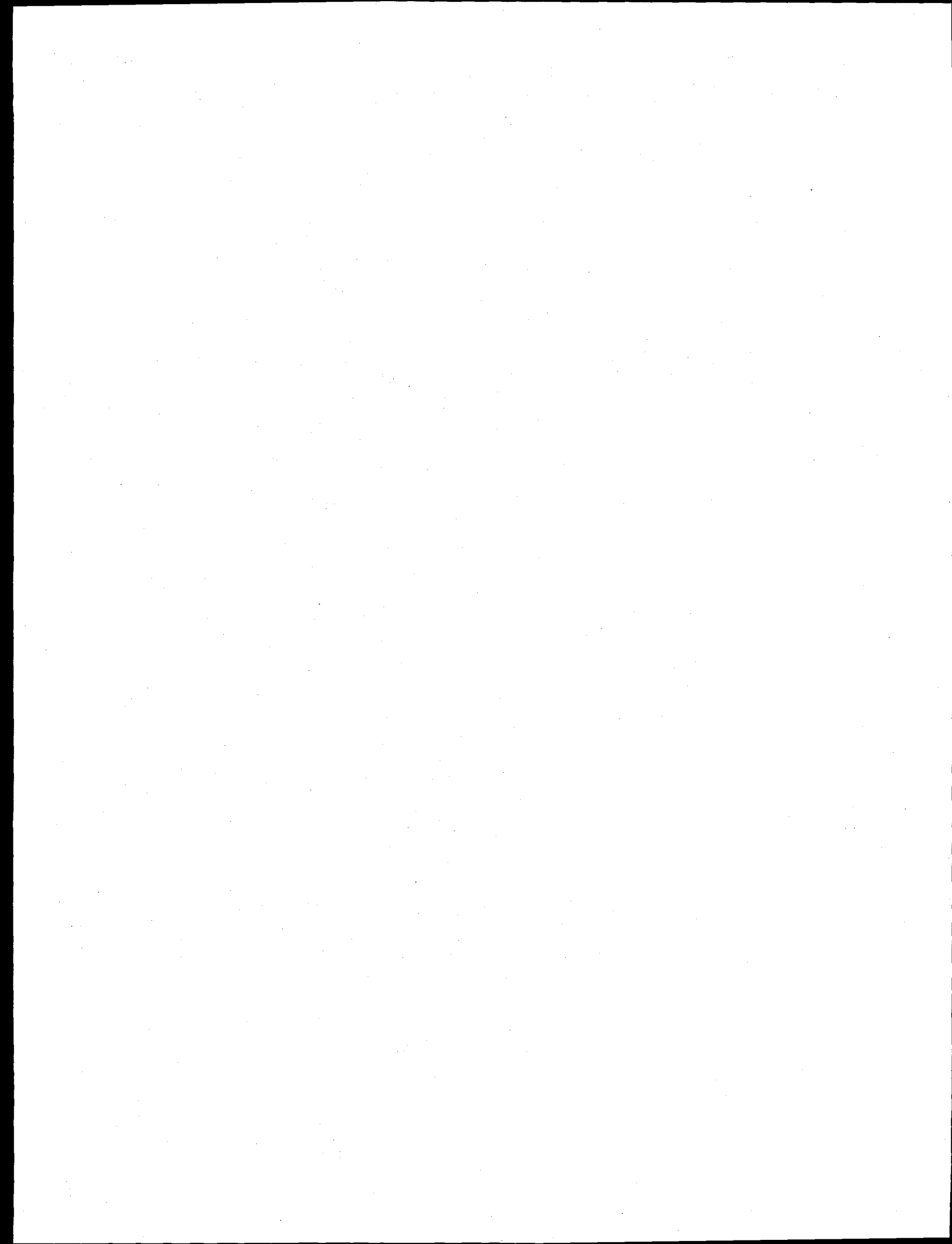
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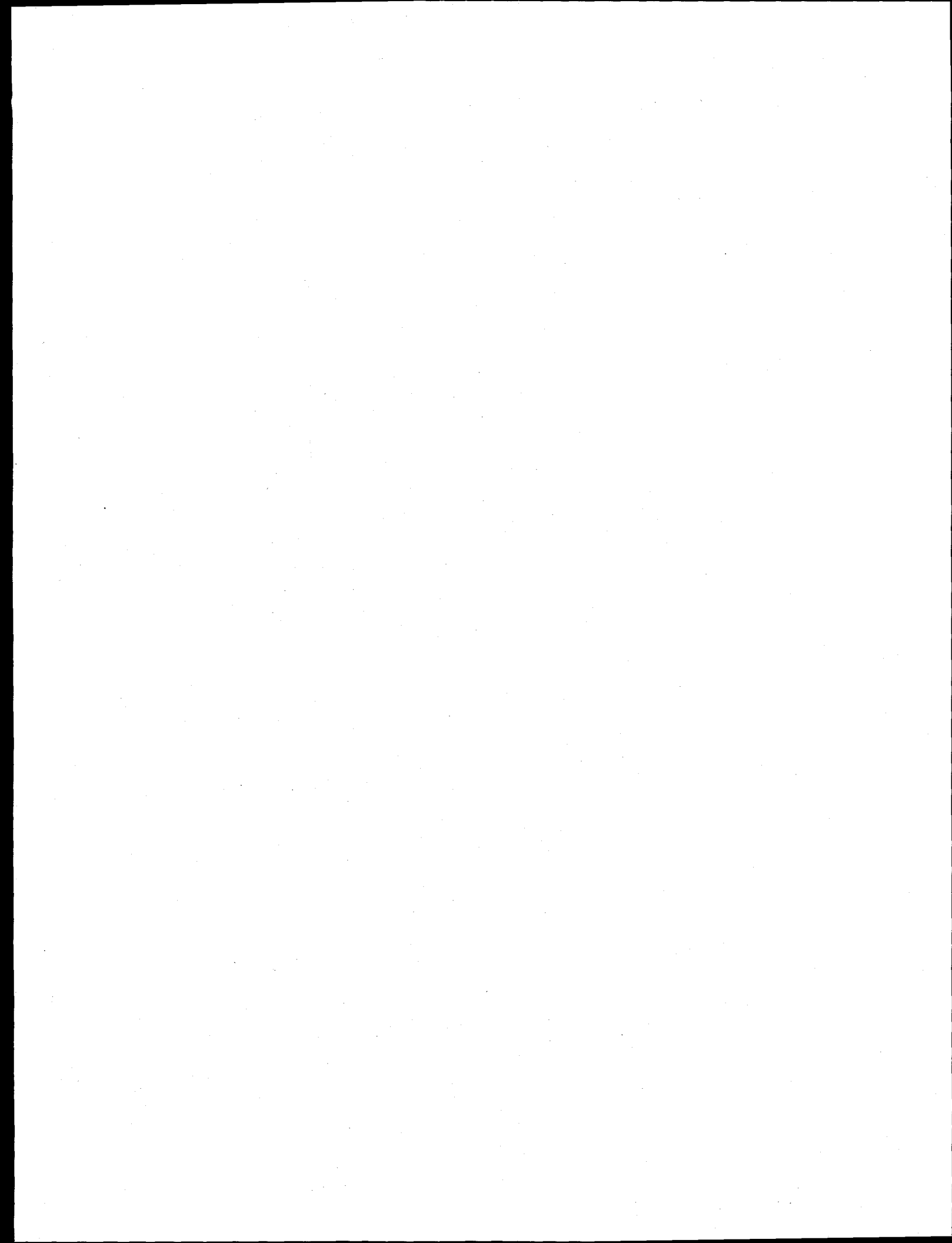
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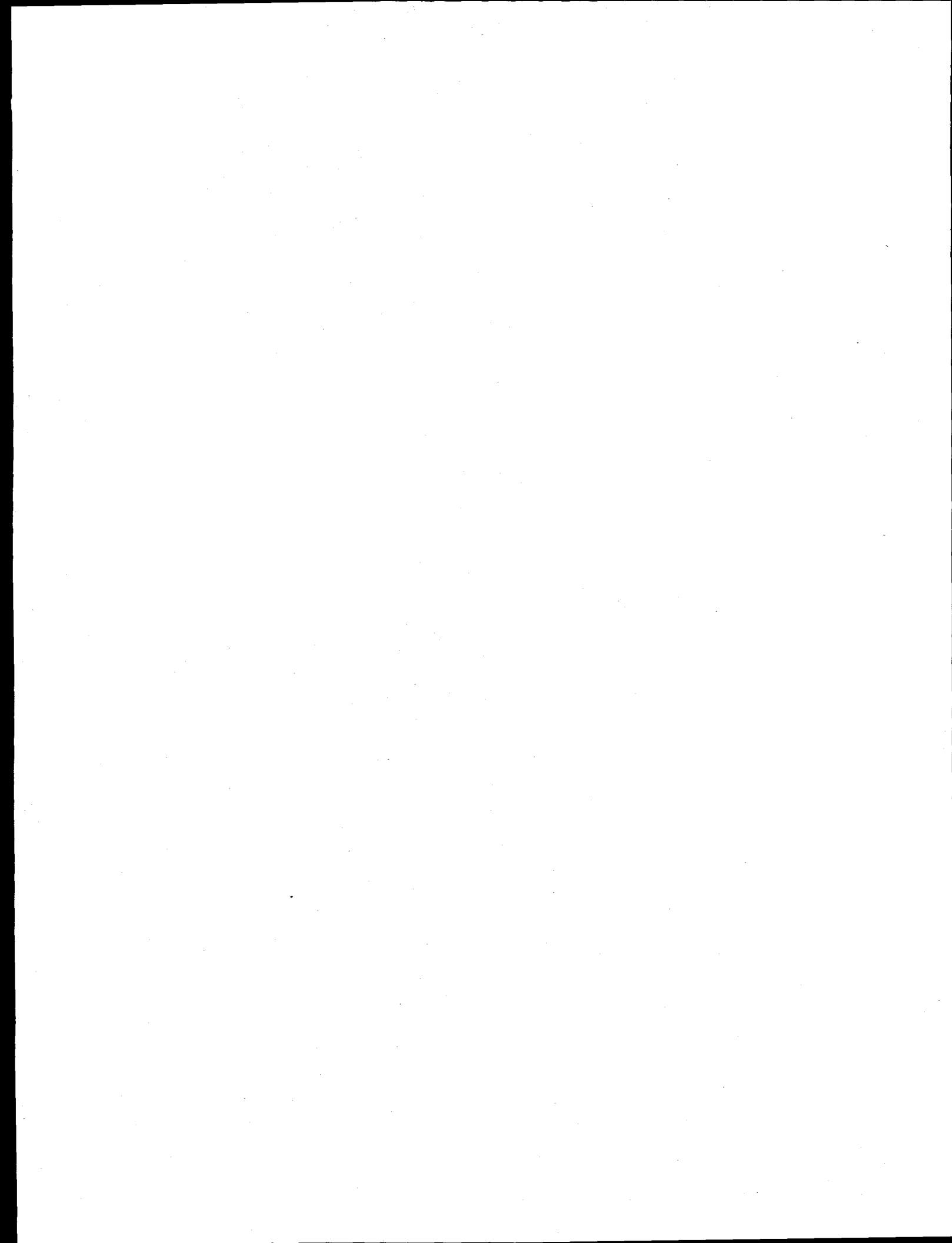
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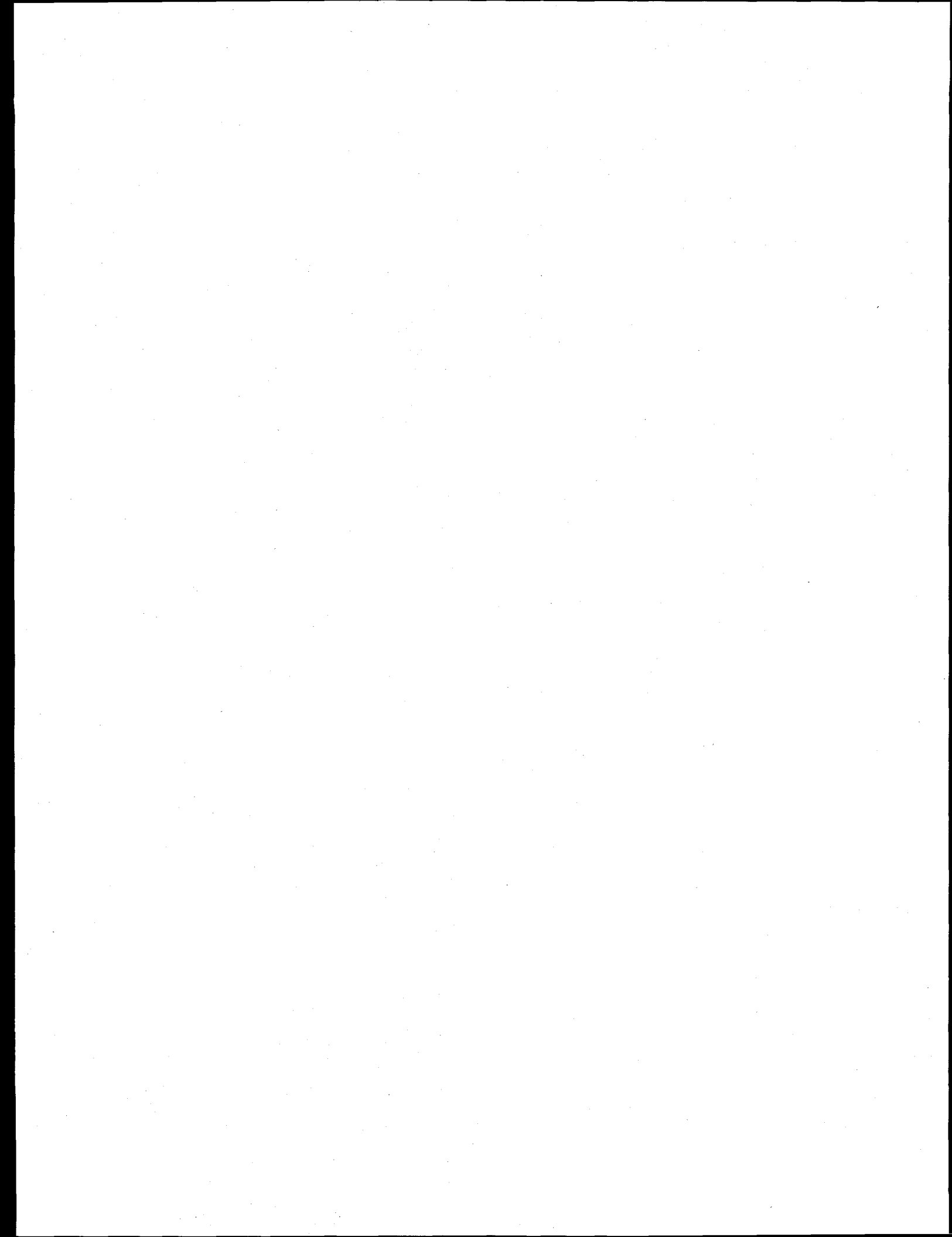
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## ACRONYMS

LME	liquid metal embrittlement
MA	mill annealed
SNS	Spallation Neutron Source



## ABSTRACT

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.

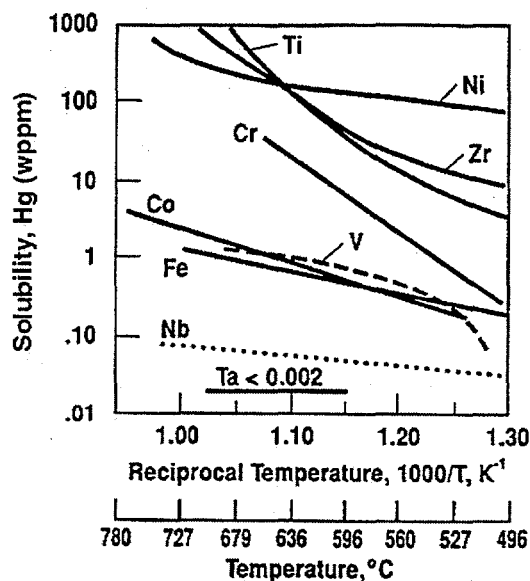
Preliminary evaluation of mass transfer of either type 304 SS, 316 SS, and Inconel 718 in mercury or mercury-gallium at 300-350°C (maximum temperature) did not reveal significant effects. Two 5000 h thermal convection loop tests of type 316 SS are in progress, with specimens in both hot and cold test regions, at 300 and 240°C, respectively.

## 1. 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.<sup>1</sup> 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.<sup>2</sup> In addition, corrosion studies include evaluation of Inconel 718 because it has been successfully used in previous water cooled spallation neutron systems as a window material.

Use of mercury as a heat-transfer medium in power systems was first attempted in the early part of the twentieth century. In a 1942 review paper,<sup>3</sup> Hackett stated that for large systems operating at 388°C "it was discovered that the heat-absorbing ability of a tube filled with mercury was not only uncertain but unpredictable to such a degree that pronounced overheating and actual damage to the boiler tubes occurred. Furthermore, mercury did cause disintegration of the steel to such a degree, by dissolving slowly great quantities of iron from the inside walls of the tubes and redepositing it in the cooler regions, that the tubes became plugged at their upper ends." Later, in 1970, Deverall<sup>4</sup> described results of heat pipe experiments for 10,000 h at 330°C in which magnesium and titanium were added to the mercury and good wetting was achieved with negligible changes in the type 347 SS wall thickness.

Experiments involving mercury have continued that indicate iron-base alloys may be suitable for mercury containment depending upon composition and temperature. Figure 1 shows a plot of the solubility of some elements in mercury as a function of temperature.<sup>5</sup> Iron has a relatively low solubility while that of nickel is much higher. Thus, as shown in Fig. 2, the ferritic stainless steels<sup>6</sup> (Cr, no Ni) have better resistance to mercury than austenitic stainless steels (Cr and Ni).



Weeks: Liquidus Curves and Corrosion of Fe, Cr, Ni, Co, V, Cb, Ta, Ti, and Zr in 500°-750 °C Mercury

Fig. 1. Solubility of some selected elements in mercury.

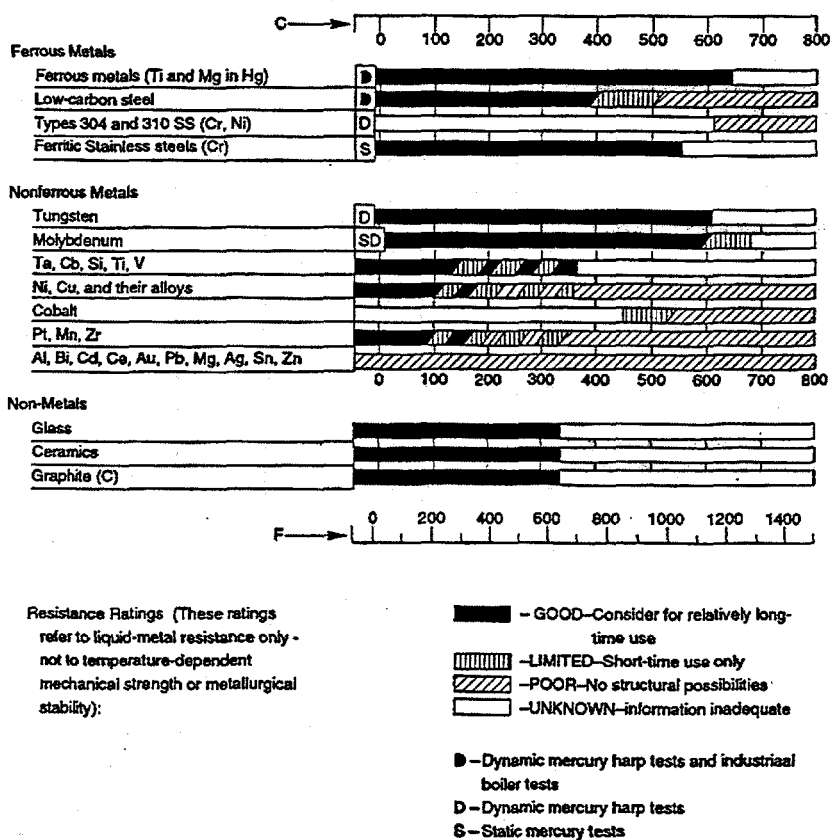
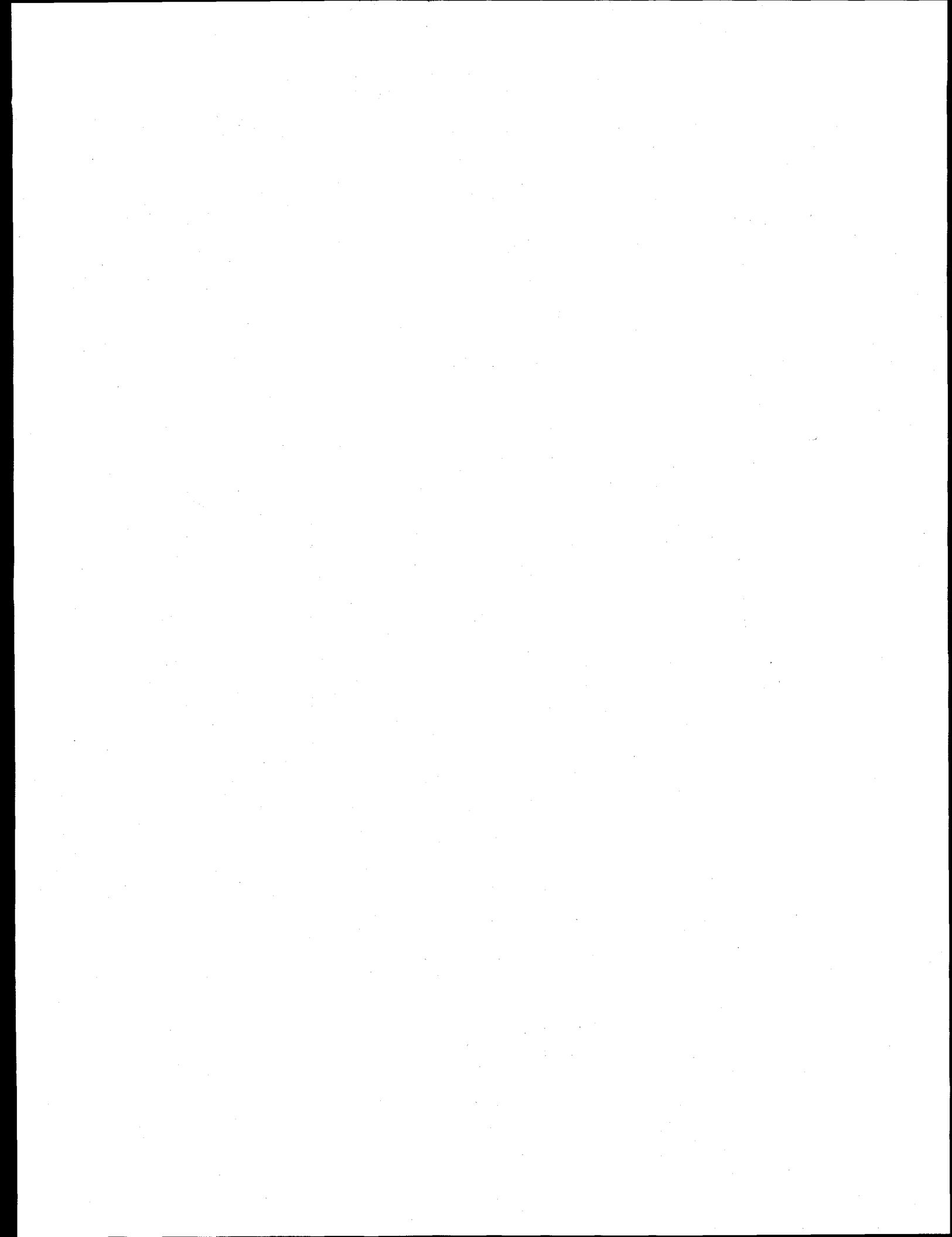


Fig. 2. Resistance ratings of selected materials to mercury. Results from both static and dynamic tests.



Note that Fig. 2 distinguishes between static and dynamic systems (dynamic involves flowing mercury and, usually, a temperature gradient). In a static system with no temperature gradient, corrosion primarily depends upon solubility or other forms of chemical interaction such as compound formation. Such reactions proceed until equilibrium is achieved. When there is an activity gradient in a flowing system, such as results from a system  $\Delta T$ , corrosion can continue indefinitely as material is dissolved at the higher temperature and deposited at the lower temperature. Factors such as  $T$ ,  $\Delta T$ , and velocity can all be important as well as solid state diffusion if there is preferential dissolution of an alloying element of the containment material. Nickel has the highest relative solubility of the elements in type 316 SS 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.

In addition to temperature gradient mass transfer, the other compatibility issue relative to 316 SS/mercury and Inconel 718/mercury being examined is liquid metal embrittlement (LME). Although the phenomenon of LME has been known for over 100 years, it is still not well understood.<sup>7</sup> 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.<sup>8</sup> Hayden and Floreen<sup>9</sup> also found that Fe-Ni was embrittled by Hg-0.1% In, and Krupowicz<sup>10</sup> 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.



## 2. 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 ( $10\text{ }\mu\text{m}$ ),
3. acid cleaned with acid subsequently displaced by mercury, and
4. specimen abraded 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, whereas other proven wetting aids such as Mg, Ti, and Zr have very low solubilities even at 200°C. When small amounts 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:

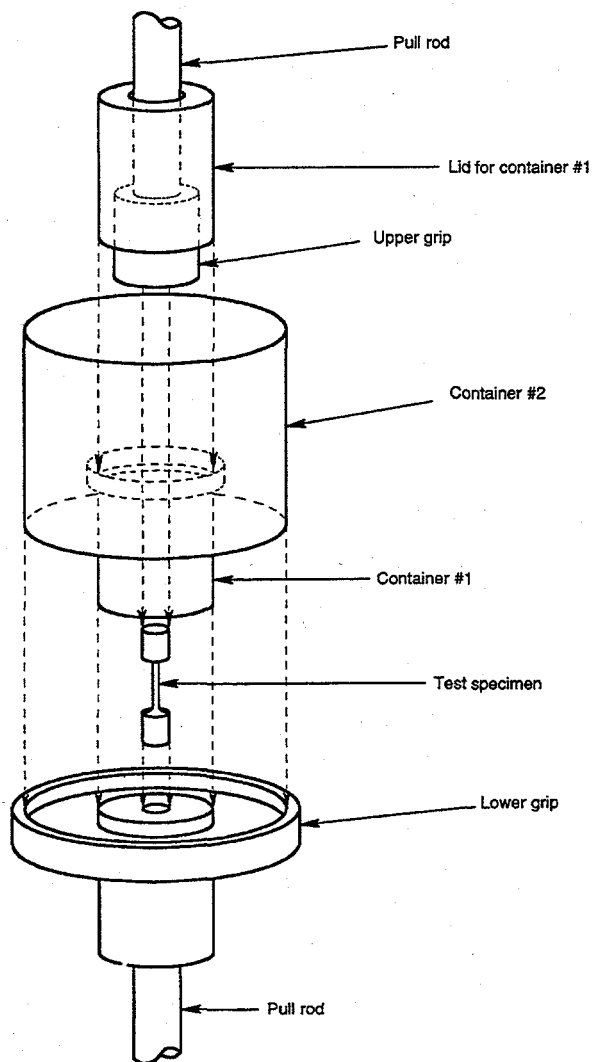
1. No scum was observed for additions of up to 0.5 wt. % gallium in mercury, even after cooling the solution to room temperature.
2. No visual evidence of wetting of Inconel 625 or type 316L SS was noted up to 230°C for mercury containing up to 2 wt. % gallium.
3. When the 2 wt. % solution was cooled 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.

Even though wetting by Hg-Ga was problematic at best, 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, specimen container and the mercury-gallium solution, was ultrasonically agitated for thirty minutes prior to testing.

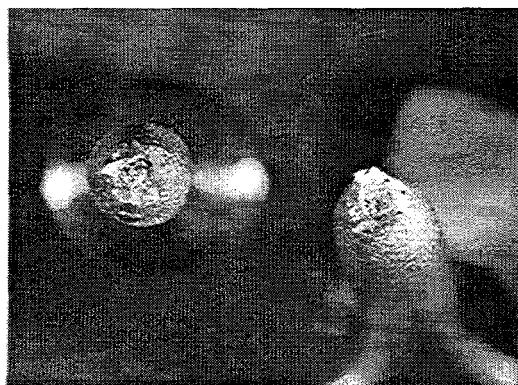
### 3. RESULTS

#### 3.1 Liquid Metal Embrittlement (LME)

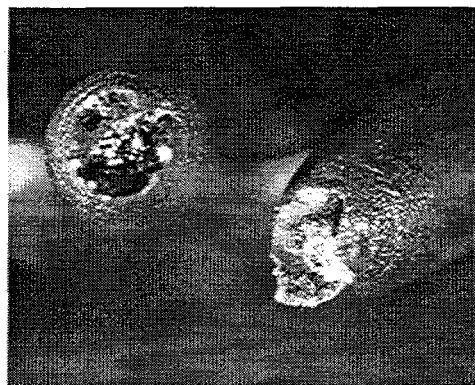
Both type 316 SS and Inconel 718 were evaluated for LME based on tensile tests at room temperature and 100°C in mercury. In order to contain the mercury during the tensile tests, the test system shown in Fig. 3 was used. Except for small areas on the fracture surface that were in mercury, no wetting 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 (Fig. 4).



**Fig. 3. Liquid metal embrittlement test apparatus assembly schematic.**



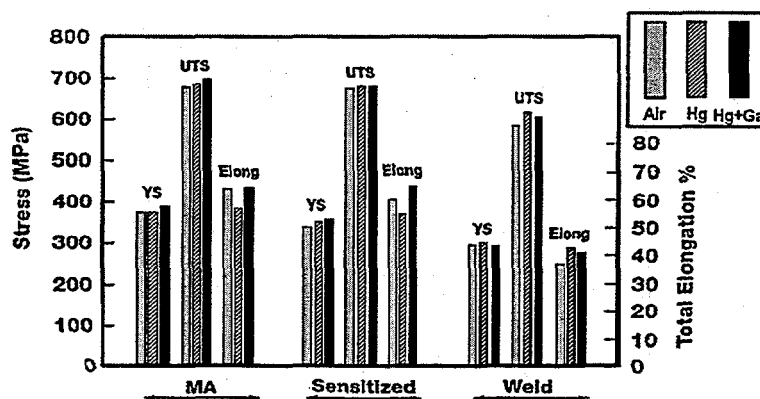
Tested in mercury



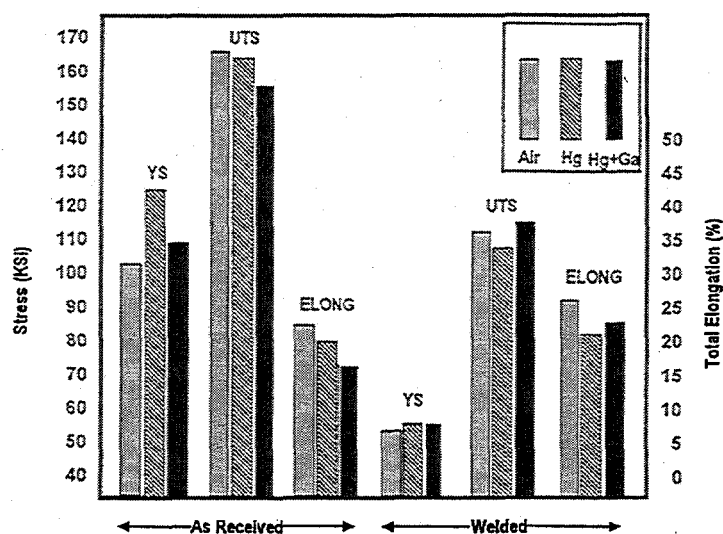
Tested in mercury with 0.5% gallium

**Fig. 4. Fracture surfaces of 316 stainless steel after exposure to mercury at 100°C.**

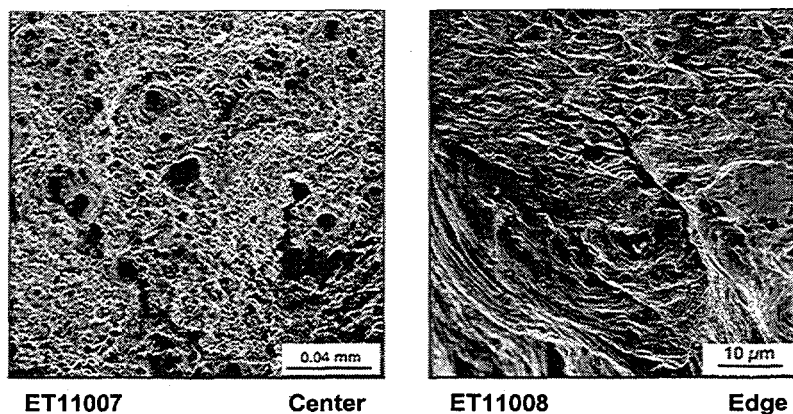
Figures 5 and 6 show the results of constant extension rate tensile tests at room temperature for type 316 SS and Inconel 718 in air, mercury, and mercury-gallium. For the type 316 SS, one set of specimens was tested in the mill annealed (MA) condition; another was heat treated to produce a microstructure 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.7). One type 316 SS sample in the MA condition was also tested in Hg-100 wppm Ga by ramping the load to ~90% of ultimate, holding in that condition for 500 h, and then increasing the load to failure. Both UTS and total elongation were once again unchanged indicating no effect of the Hg-Ga.



**Fig. 5. Mercury or mercury-gallium did not change the room temperature tensile properties of type 316 stainless steel in LME tests.**

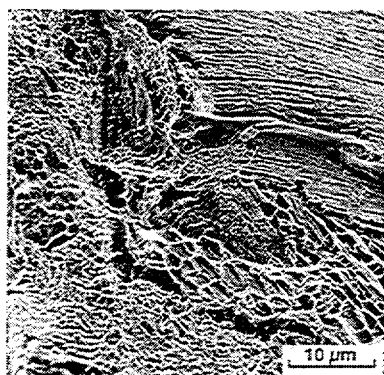


**Fig. 6. Mechanical property results from constant extension rate tests for Alloy 718.**



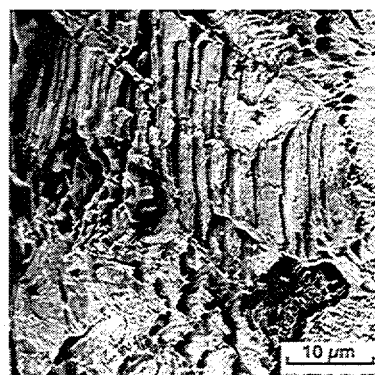
**Fig. 7. SEM micrographs showing the appearance of type 316 SS tested in Hg-Ga at room temperature.**

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. 8). For specimens tested in mercury or mercury-gallium, both ductile and brittle features were found (Figs. 9 and 10).



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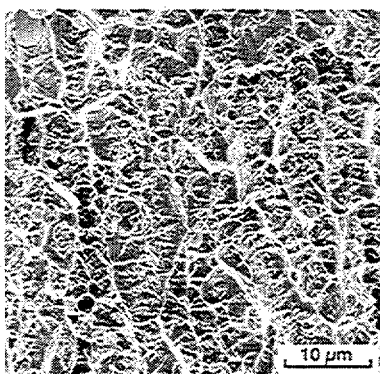
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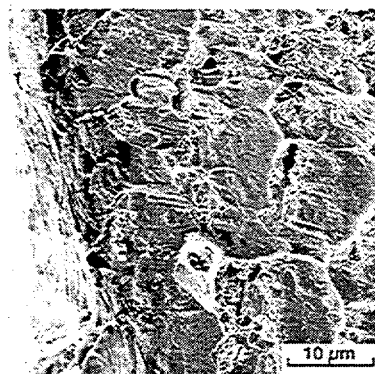
Edge

**Fig. 8. SEM photographs of the center and edge of the fracture surface of the as-received In 718 specimen tested in air.**



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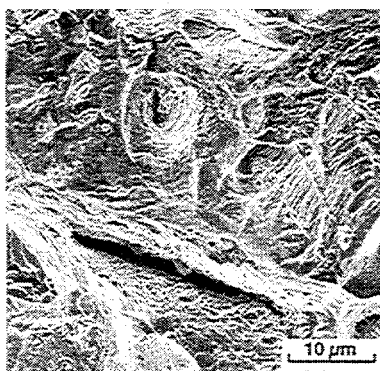
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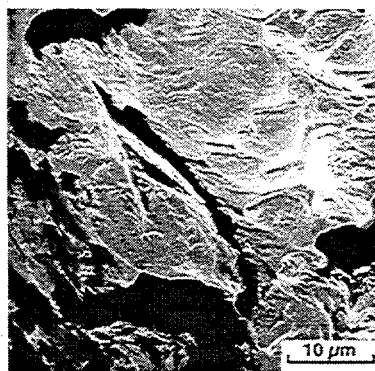
Edge

**Fig. 9. SEM photograph of the center and edge of the fracture surface of the welded In 718 specimen tested in Hg.**



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Center



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Edge

**Fig. 10. SEM photographs of the center and edge of the fracture surface of the welded In 718 specimen tested in Hg-Ga.**



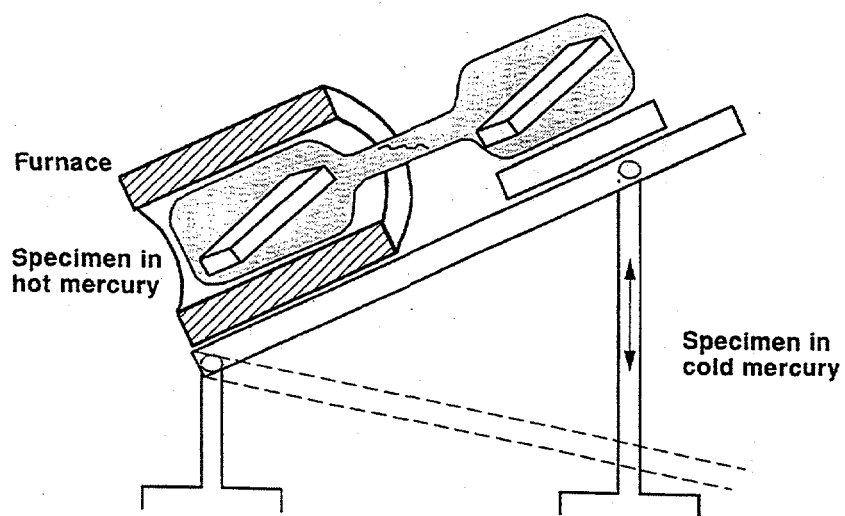
Additional tests have recently been completed to investigate LME effects at 100°C as well as to evaluate an additional method to achieve wetting.

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.

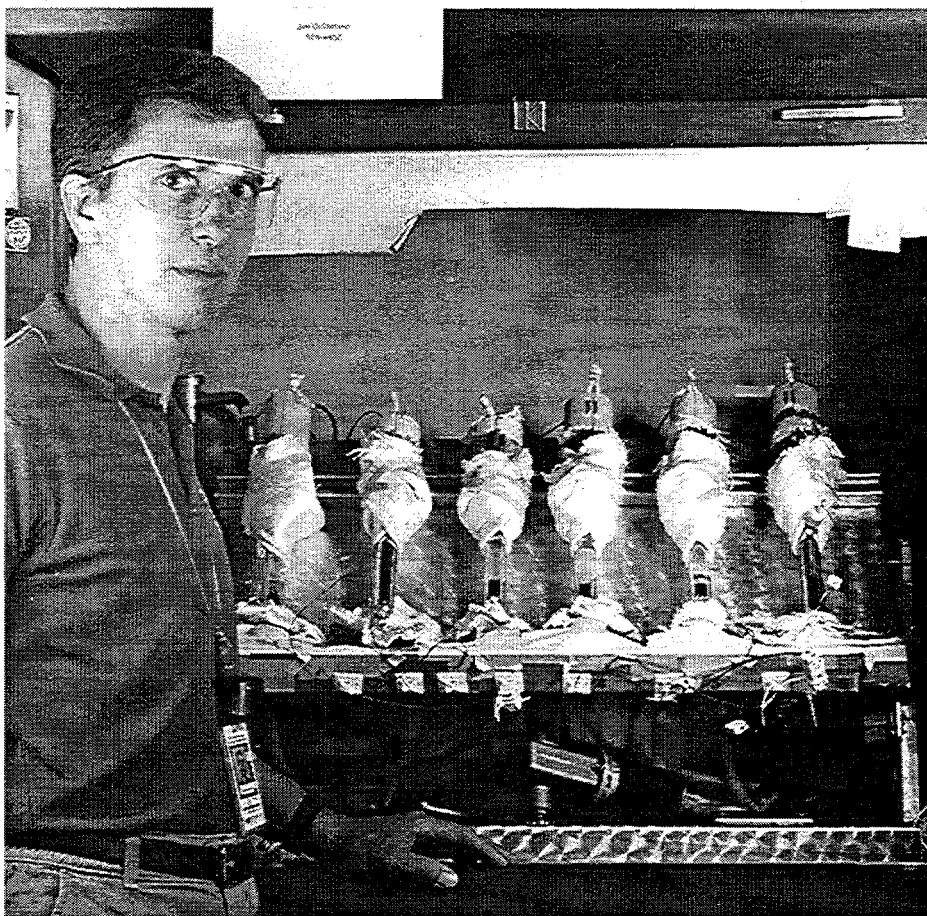
A test was also performed on a specimen coated with silver solder along its gage length. The coated specimen was first pre-exposed to mercury at room temperature to permit some dissolution of the tin/silver and then it was transferred to the test container with fresh mercury. When tested at room temperature, 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.

### **3.2 Mass Transfer**

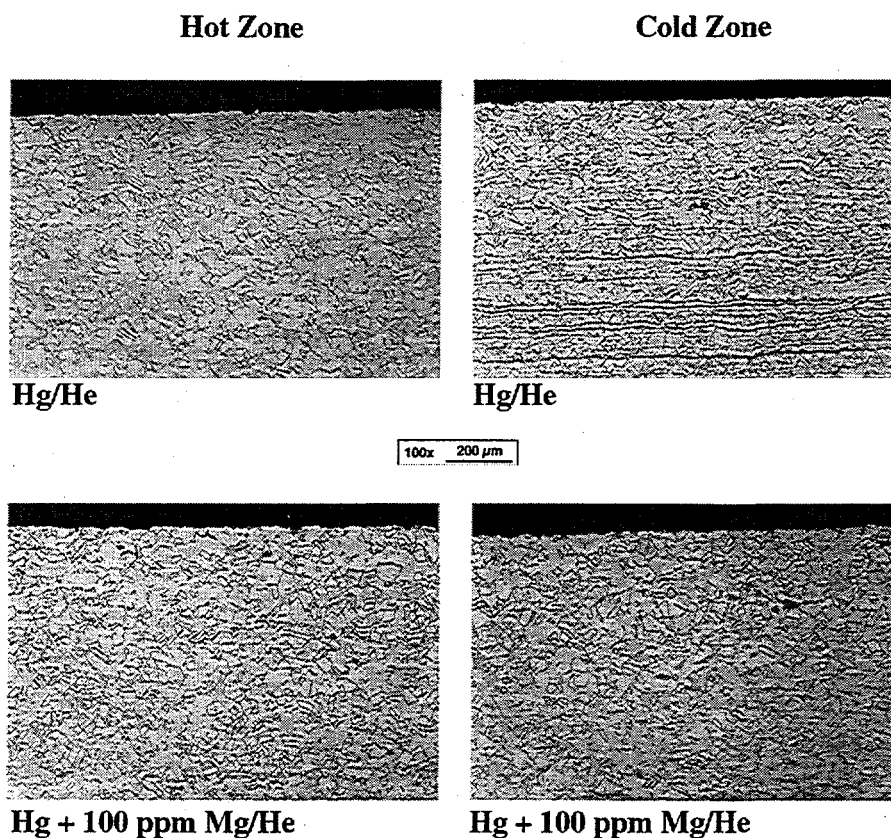
Initially, screening tests were conducted in which mercury was cycled between hot and cold regions of a sealed quartz capsule containing a type 304 SS specimen in each region (Fig. 11-12). These tests were conducted at temperatures from 150-200°C above those expected in the target system in order to accelerate the mass transfer process. Weight change results in Table 1 indicate small losses in type 304 SS specimens from both hot and cold regions, and metallographic examination indicated very limited corrosion (Fig. 13). Environments within the closed quartz capsules were either air, He, or vacuum. Although the vapor pressure of Hg was the same (as determined by the test temperature) in the tests, the vacuum environment allowed boiling to occur. Based on the results in Table 1, this seemed to promote wetting and weight loss. The addition of 100 wppm Mg to the Hg also appeared to promote wetting, but weight change results were inconsistent with respect to wetting effects.



**Fig. 11. Rocker apparatus used to screen for thermal gradient mass transfer.**



**Fig. 12. Photograph of rocker tests in operation.**



**Fig. 13. Microstructure of 304 stainless steel after exposure to mercury in rocker tests. 200 h at 300°C with 78°C ΔT.**

Table 1. Initial rocker screening tests of type 304 SS in mercury indicated only small changes						
Average maximum temperature: 300°C						
Average ΔT: 78°C						
Time: 2000 h						
Cycles (60s/18s): 92,000						
Capsule environment	Mg in Hg	Visual wetting		Weight change mg/cm <sup>2</sup>		Comments
		Hot	Cold	Hot	Cold	
Air	None	No	Yes	-.14	-.12	Hg fairly bright and shiny
He	None	No	No	-.14	-.02	Hg very bright and shiny
Vac	None	Yes	Yes	-1.53	+.004	Metallic crystals on cold leg surface
Air	100 wppm	No	Yes	-.04	-.02	Dark residue in Hg
He	100 wppm	Yes	Yes	-.92	-.03	Dark residue in Hg

Results from a second rocking test series with type 316 SS and Inconel 718 in mercury are summarized in Table 2. Losses increased when magnesium or gallium was added to the 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. 14. There was no evidence of internal corrosion of either material, only surface dissolution or deposition.

Table 2. Weight change (mg/cm <sup>2</sup> ) from rocking test of type 316 SS and Alloy 718 in mercury after 2800 h (10,000 Cycles: 15 min/2 min; He environment)				
Material	Hot zone 350°C			Hot zone 375°C
	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	

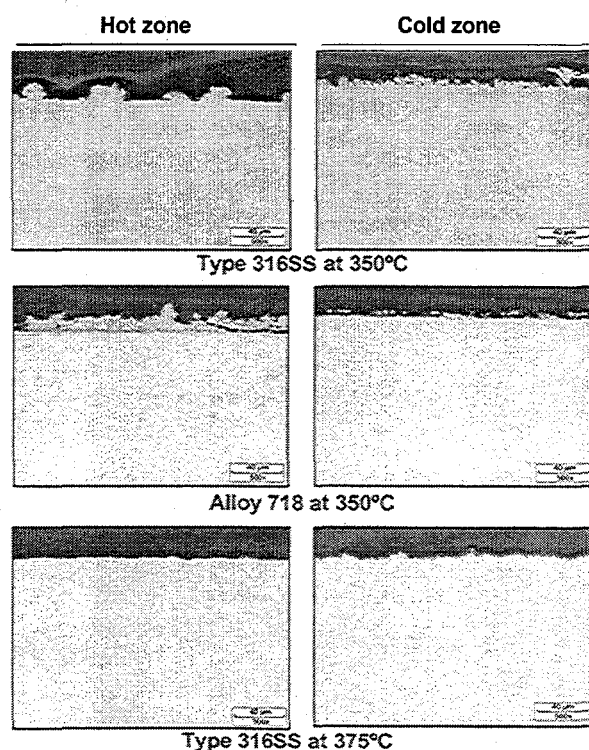
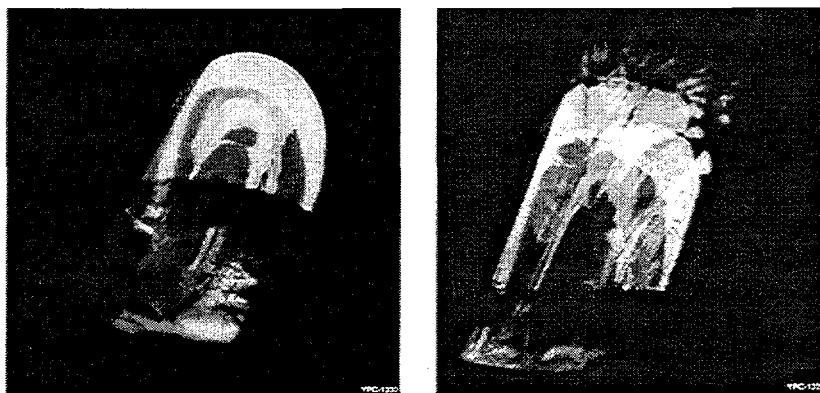


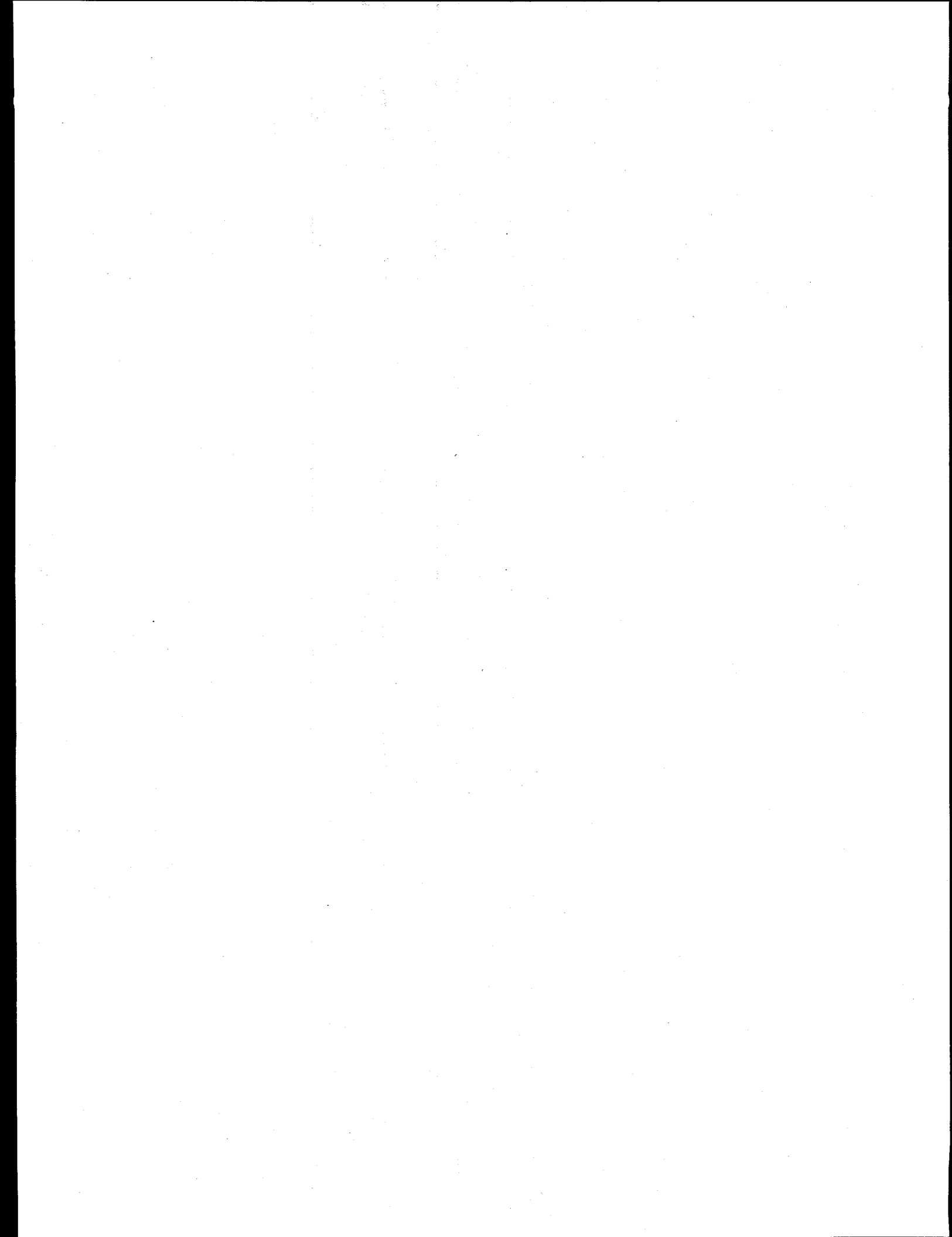
Fig. 14. Behavior of type 316 SS with Alloy 718 exposed to Hg-Ga.

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. 15. A recent publication,<sup>11</sup> 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.



**Fig. 15. Light observed in rocking tests after ~2500 h of testing.**

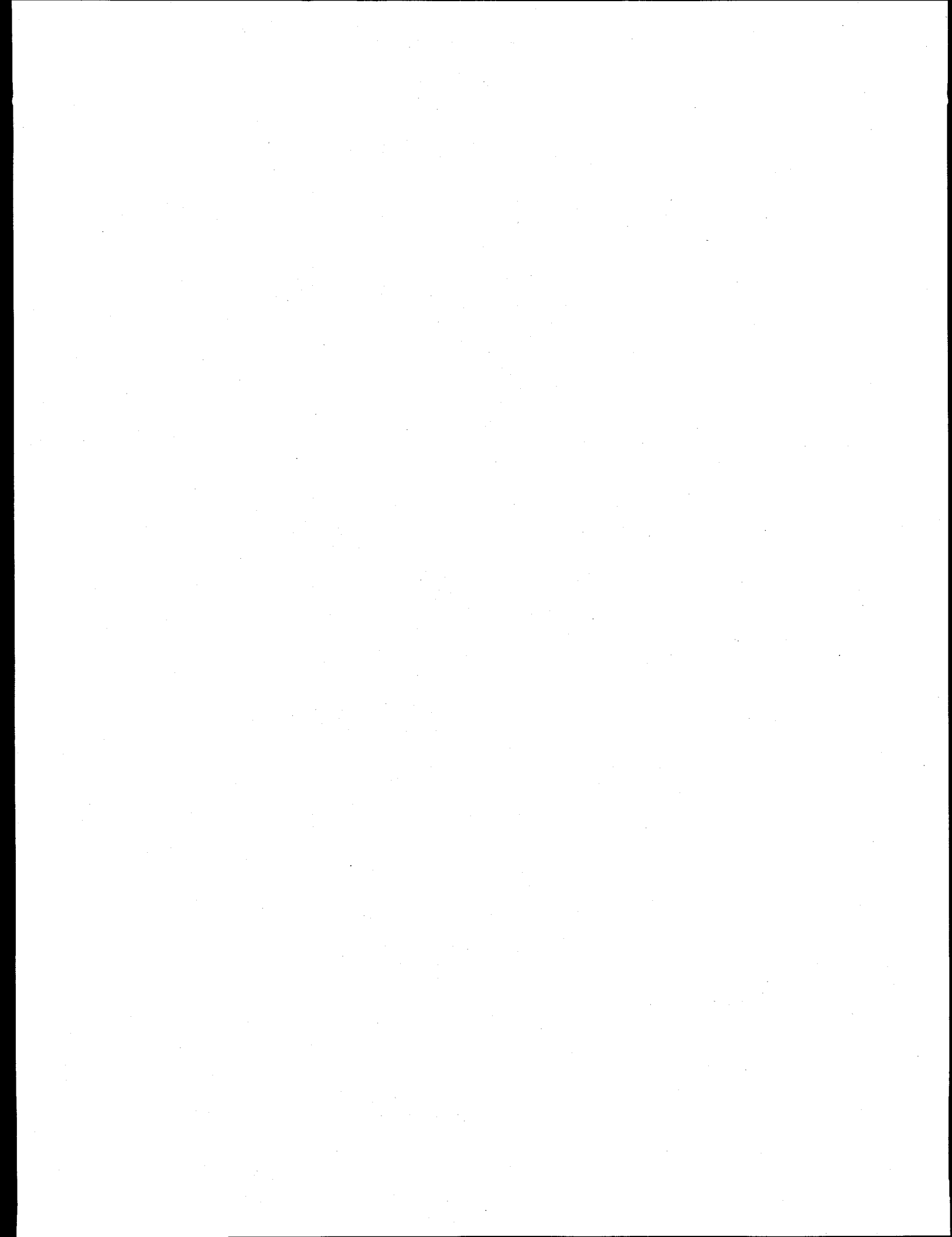
More prototypic testing for mass transfer effects is continuing in two type 316 SS thermal convection loops as shown in Fig. 10. Each loop contains specimens in both hot (300°C) and cold (240°C) vertical legs. Mercury or mercury-0.1 wt. % gallium is being circulated at approximately 1 m/min for a scheduled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



#### 4. 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 304 SS, 316 SS, and Inconel 718 in mercury or mercury-gallium at 300-350°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.

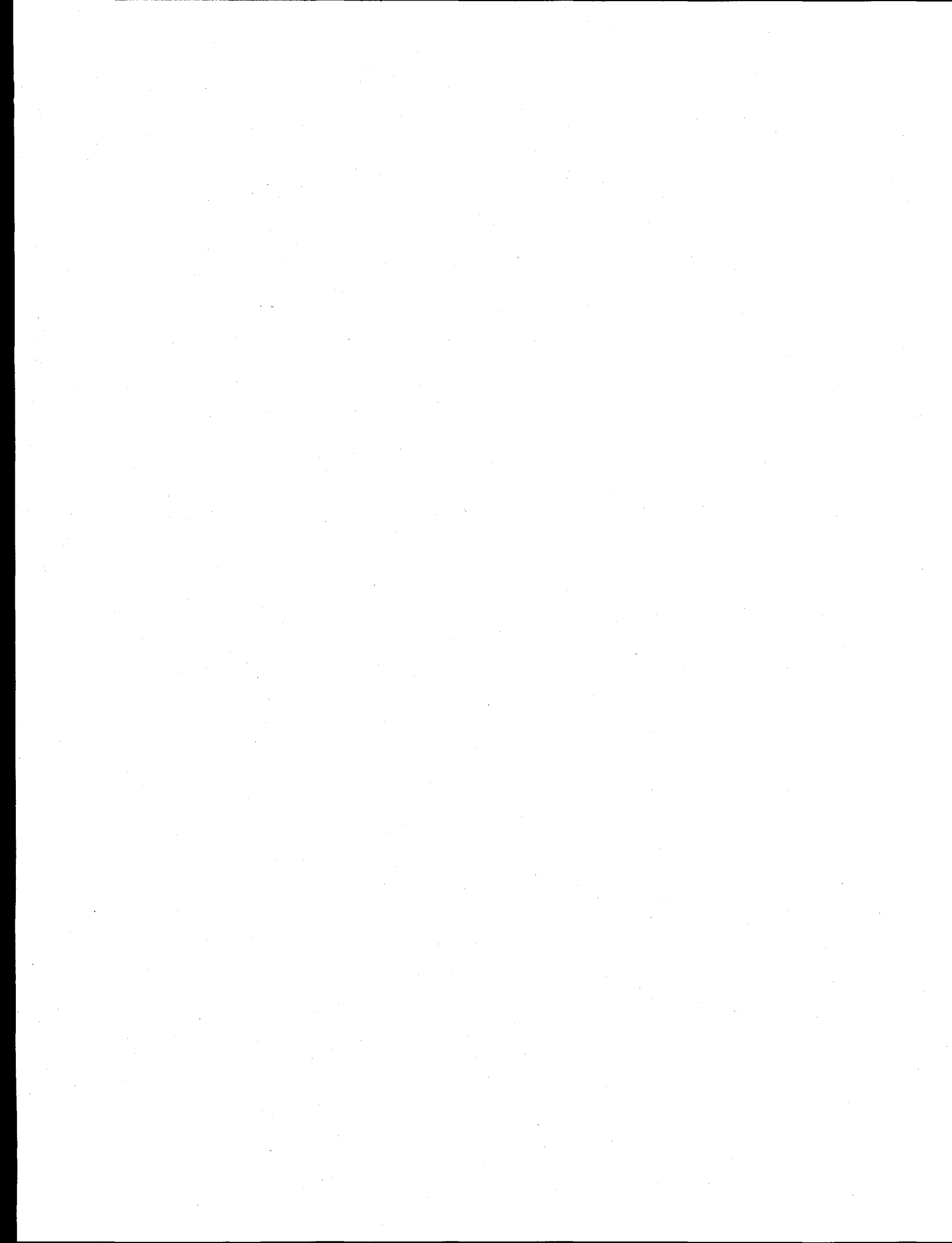




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