THE EFFECT OF COLD WORK ON THE LIQUID METAL INDUCED EMBRITTLEMENT OF BRASS BY GALLIUM

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ABSTRACT

Liquid metal induced embrittlement (LMIE) reduces the stress intensity required to cause fracture in metals. Because of the difficulty in studying this phenomenon, most tests have used simple tensile specimens, but the results of these tests can be ambiguous. This paper describes recent studies of LMIE using fracture mechanics. The brass/liquid gallium couple is used to model behaviour at higher temperatures and give some insight into the micromechanisms of LMIE and the way in which tensile specimens fail in liquid metals.

KEYWORDS

Liquid-metal embrittlement, environmental fracture, brass, liquid gallium, LMIE

INTRODUCTION

One of the methods frequently used to study liquid metal induced embrittlement (LMIE) is to tensile test the relevant metal coated with the liquid metal. Susceptibility to LMIE is determined by establishing whether LMIE fractures had occurred and over what range of test temperatures LMIE was operating. A considerable amount of data from these tests has been gathered already. However, failure of tensile specimens by LMIE occurs by an interaction between LMIE crack initiation and propagation and to date, there have been few studies which have examined each aspect separately. In the studies described in this paper, the failure of a brass by LMIE due to liquid gallium is examined using both tensile tests and fracture toughness tests.

Fracture of tensile specimens by liquid metal induced embrittlement initiates at the surface of the specimen in regions where the liquid metal wets the surface of the specimen. Therefore, the surface between the liquid gallium and the brass must be important in determining the way in which the specimen fails. In most systems, LMIE failure of the solid metal doesn't occur until the specimen has yielded, even if this yielding is only localised (Kamdar, 1983). In fact in many systems, including the brass/gallium system studied here, a considerable amount of plastic deformation is necessary to initiate LMIE failure, albeit less than the deformation required for unembrittled fracture.

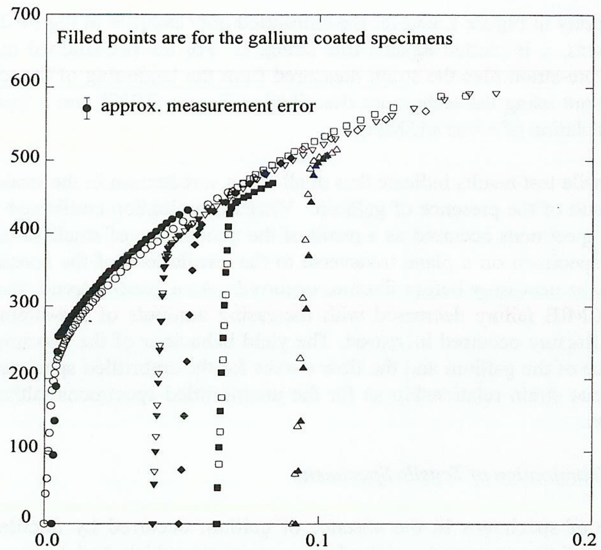

Figure 1 True stress *vs* true strain for brass tensile specimens.

It is well known that as a tensile specimen deforms plastically, the surface of the specimen deforms unevenly on a microscopic scale and a rough surface texture can develop, sometimes called an "orange peel" effect. This uneven deformation can lead to small fissures or others areas of localised stress concentration and such sites of stress concentration would provide ideal areas for the initiation of LMIE cracks. In this study, the effect of roughening the surface of tensile specimens by to plastic deformation on LMIE fracture are investigated, in order to see if LMIE crack initiation is in fact dependent upon the roughened state of the surface.

EXPERIMENTAL PROCEDURE

The brass used in these experiments was BS2874 CZ121. This was a cold rolled, leaded brass with a nominal composition of 58Cu 39Zn 3Pb. The material was supplied in strip and rod form. The strip had cross sectional dimensions of 50 × 12.5mm (2inch × 1/2inch) and the rod had a diameter of 12.5mm (1/2inch).

The tensile specimens were made from material which had been annealed at 900°C for two hours and water quenched. Three sets of fracture toughness specimens were used, each at a different degree of cold work starting from the "as-received" state. The three conditions tested were; as-received, 13% cold worked from the as received condition and 24% cold worked from the as-received condition. The specimens were cold rolled in a rolling mill using approximately 3 passes to achieve 13% reduction and 6 passes to achieve 24% reduction. In order to ensure stable crack propagation in the cold worked material, all fracture toughness specimens were stress

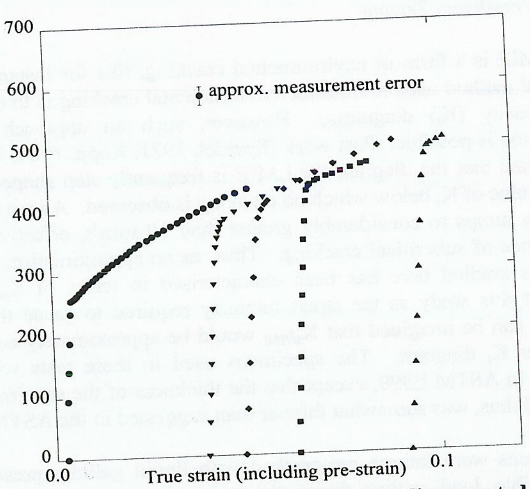

Figure 2 As for Fig. 1, showing only the gallium coated specimens.

relieved by heating to 180°C for 1 hour. It is acknowledged that such a treatment would have produced a degree of recovery, and thus affected the cold worked structure of the brass, but without stress relieving, no measurement of crack resistance would have been possible.

Tensile Testing

Ten tensile specimens were tested. The dimensions of the specimens were in accordance with ASTM E8M; a gauge length of 30 mm and a gauge diameter of 5 mm (nom.). The specimens were divided into pairs and each pair was tested at a different pre-strain. All but the first pair were pre-strained prior to the final surface preparation. The first pair were tested in the annealed (non pre-strained) state. Pre-straining was carried out in a testing machine, with the degree of pre-strain being determined using an extensometer. The specimens were then re-machined over the gauge lengths to a depth of at least 0.3mm on radius in order to remove any surface texture. The gauge lengths were then ground and polished to produce clean surfaces essentially free of the residual surface plastic deformation produced by the machining. The specimens and testing grips were pre-heated to approximately 40°C prior to testing and the gallium was applied externally to cleaned surfaces. Although no special steps were taken to ensure good wetting, the gallium appeared to have little difficulty in wetting the polished surface. Testing was carried out in an Instron 6025 fixed crosshead displacement machine using a 100kN load cell and a 12.5 mm Instron extensometer.

Fracture Toughness Testing

LMIE is a form of environmental cracking, like for instance stress corrosion cracking. The typical method used to describe environmental cracking is to develop crack speed (*da/dt*) *vs* stress intensity (K_I) diagrams. However, such an approach is time consuming and a simplification is possible. Past work (Speidel, 1971; Kapp, 1984; Wheeler and Hoagland, 1986) has suggested that the diagram for LMIE is frequently step shaped. That is, there is a distinct threshold value of K_I below which no cracking is observed. Above this threshold value, the crack speed often jumps to considerably greater than 10 mm/s, effectively fast LMIE fracture, with little evidence of subcritical cracking. Thus, as an approximation, the *da/dt vs* K_I behaviour of the systems studied here has been characterised in terms of K_{Ilme}, loosely defined for the purposes of this study as the stress intensity required to cause the LMIE crack to propagate quickly. It can be imagined that K_{Ilme} would be approximately equal to the threshold value in the da/dt *vs* K_I diagram. The specimens used in these tests were made with the standard dimensions in ASTM E399, except that the thickness of the specimens was the full thickness of the strip and thus, was somewhat thinner than suggested in the ASTM Standard.

The specimens were fatigue pre-cracked with liquid gallium present at the tip of the crack. Where possible, load cycling during the last region of crack growth was carried out with the maximum stress intensity during the cycle, K_{Imax}, being less than 60% of the critical stress intensity for fast fracture. However, this was not always possible with the gallium embrittled material, as some cracks appeared to arrest below a critical ΔK_I range and this was often greater than K_{Imax}. This and other anomalous fatigue crack behaviour prompted a series of investigations into the behaviour of fatigue cracks in the presence of liquid gallium and these are reported elsewhere (Clegg, 1993; Clegg and Jones, 1994). Fracture toughness testing was carried out in either the Schenk fatigue testing machine or in the Instron 6025 testing machine. Problems with pre-cracking of the gallium embrittled specimens meant that in some cases, specimens failed during the fatigue pre-cracking stage of testing. In these situations, an estimate of K_{Ilme} was obtained by estimating the crack length at which fast fracture occurred and the maximum load in the fatigue cycle. In this case, the crack length could only be determined to ±1.0 mm and an error analysis of this method indicated that measurement errors of ±3MPa√m could be expected for determining K_{Ilme}, whereas measurement errors of only ±0.25MPa√m would occur if testing were carried out using fatigue pre-cracked specimens in which crack length had been determined accurately.

SEM Examination

SEM examination of the specimens was carried out in a Cambridge Instruments scanning electron microscope (SEM). Examination was carried out in order to investigate the size and development of surface defects during plastic deformation.

RESULTS

Tensile Testing

The specimens were pre-strained to true strains of 4.0%, 5.0%, 6.5%, 9.5% before testing, and specimens were also tested in the annealed condition. The results of these tests can be seen

collectively in Figure 1. and for the embrittled only samples in Figure 2. In all of these diagrams, true stress, *s*, is plotted against true strain, *e*. For the pre-strained material, *e* is the degree of plastic pre-strain plus the strain measured from the beginning of the test. An error analysis was carried out using the techniques described in Taylor (1982) and a typical measurement error in the calculation of *s* was ±15MPa.

The tensile test results indicate that in all cases, a reduction in the strain to fracture was observed as a result of the presence of gallium. Visual examination confirmed that failure of the gallium coated specimens occurred as a result of the penetration of small LMIE cracks from the surface of the specimen on a plane transverse to the tensile axis of the specimen. In all cases, plastic strain was necessary before fracture occurred. As a general trend, the plastic strain required to cause LMIE failure decreased with increasing amounts of pre-strain, while the true stress at which fracture occurred increased. The yield behaviour of the specimens was unaffected by the presence of the gallium and the flow curves for the embrittled specimens followed the same true stress/true strain relationship as for the unembrittled specimens, although the strain at fracture was less.

SEM Examination of Tensile Specimens

Failure of specimens in the absence of gallium occurred by ductile fracture initiated on the outside of the specimen. All of the specimens which had been coated with gallium were embrittled and small, thumbnail shaped cracks could be seen on the fracture surfaces. The liquid metal did not always completely cover the fracture surface and areas of LMIE fracture surface apparently clear of gallium could be seen. SEM examination of the outer surfaces of the brass specimens revealed once again the presence of surface flaws. Figure 3 shows a typical surface flaw formed after approximately 11% strain.

In order to investigate the development of these flaws, a slightly different test routine was adopted for the last two pairs of specimens. These pairs of specimens had been pre-strained to 9.5 and 5% respectively. The gallium coated specimens were tested to failure as before and the strain at fracture was noted. The remaining two specimens were tested in the absence of gallium and were strained incrementally until failure occurred. After each increment of strain, typically 1/2% of nominal plastic strain, the specimen was removed from the Instron and the polished surface was examined in the SEM and the size and development of the flaws was noted.

It was found that for the specimen pre-strained to 5%, the surface flaws started to form well before the strain at which LMIE fracture occurred suggesting that LMIE crack initiation does not occur immediately the surface flaws form, but when they have developed to a certain degree. For specimens pre-strained to 9.5% it was found that initially, no evidence of surface flaws was found, while some small flaws were detected at higher strain. This suggests that surface flaws need to be less well developed to initiate LMIE for high pre-strain than for lower pre-strain.

Effect of Cold Work on Resistance to LMIE Fracture

Three degrees of cold work of the brass were tested: 0%, 13% and 24%. The results from the specimens are as shown in Table I.

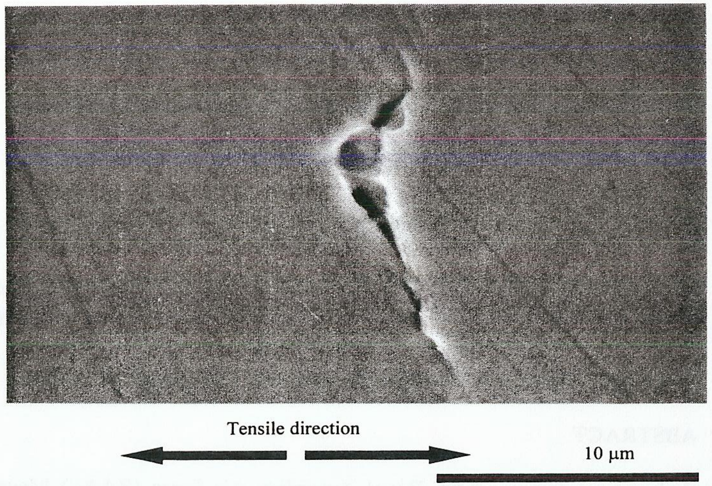

Figure 3 Surface flaw on the specimen strained to approx. 11%. Such flaws are likely sites for crack initiation.

Degree of Cold work (%)	*K_{Ilme} (MPa√m)*	*Measurement Error (MPa√m)*
0 (as-received)	23.0	2.0
13	20.6	1.2
24	20.3	0.25

Table I - Results of fracture toughness testing of brass embrittled by gallium

DISCUSSION

The most important conclusion from this series of experiments is that stress and degree of cold work are not sufficient criteria for the initiation of LMIE cracks. The development of surface texture is important to LMIE crack initiation in these specimens and if it is removed by machining, a new surface texture must develop before LMIE fracture can occur. Of all the surface features observed, the type of surface flaw shown in Figure 3 appears to be the most likely candidate for sites of crack initiation. In all cases, failure did not occur until these had appeared. Their role in crack initiation is not entirely clear, but the stress and strain at the roots of these flaws would be highly concentrated. Once these conditions reached a critical state, LMIE crack initiation would ensue.

A number of micromechanisms for the initiation of LMIE cracks on plane tensile specimens have been proposed. Gordon and An (1978) studied the time dependence of crack initiation of indium embrittled steel and concluded that crack initiation was limited by diffusion of the embrittler along grain boundaries. However, it is difficult to see that interdiffusion of gallium into the brass would be significant at room temperature. Roth *et al* (1980) proposed that crack initiation occurred when the shear stress ahead of a slip band at the point at which it intersected a grain boundary reached a critical value. That value determined by the relative surface energies of the gallium, brass and air. Lynch (1988) has proposed that liquid metals tend to enhance plasticity, which may increase the stresses ahead of the slip band increasing the likelihood of brittle crack initiation. A micromechanism such as this may indeed be operating, but it is clear that the stress and strain concentrations at the roots of the surface flaws are necessary before the critical conditions for initiation are achieved.

There appears to be little effect of cold work on the LMIE crack propagation characteristics of the brass. Table I indicates a slight reduction in K_{Ilme} with increasing amounts of cold work. This trend may not be significant when compared with the errors associated with the K_{Ilme} determination but is consistent with experience with unembrittled materials. It must be remembered that the brass used in this test was already cold rolled and so, cannot be used directly to describe the effect of cold work on the LMIE crack propagation behaviour of annealed specimens. However, it is a good guide to the effect of plastic straining during the tensile tests.

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