The effects of loading variables on fatigue crack growth in liquid metal environments

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Liquid-metal-induced embrittlement (LMIE) refers to the loss of ductility in normally ductile metals and alloys when stressed while in contact with a liquid metal. In this study, the fatigue crack growth behaviour of brass in molten gallium is examined using standard LEFM testing procedures. The effects of environment, load ratio, cyclic frequency and load waveform on the rate of crack growth are investigated. At $\Delta K > \Delta K_{th}$, crack growth rates are approximately an order of magnitude faster in liquid metal than in air. However, at low ΔK values (low fatigue crack growth rates) and/or where the crack tip is exposed to the liquid metal for long periods of time, crack tip blunting by dissolution of the solid metal into the liquid metal occurs. This markedly decreases the rate of crack growth and results in apparent ΔK_{th} values in liquid metal that are significantly higher than the unembrittled threshold values.

(Keywords: liquid-metal-induced embrittlement; fatigue crack growth; load ratio; cyclic frequency; load waveform; crack tip blunting; dissolution; brass; gallium)

Liquid-metal-induced embrittlement (LMIE) refers to the loss of ductility in normally ductile metals and alloys when stressed while in contact with a liquid metal. Although this form of environmentally assisted cracking has not received as much attention as hydrogen embrittlement and stress corrosion cracking, there have been many catastropic failures attributed to LMIE. These have recently been reviewed by Fernandes et al.[1].

LMIE has been extensively investigated since it was first reported on in 1874[2], and its characteristics have been generally well established. These can be briefly summarized as follows.

1. There must be intimate contact or 'wetting' of the solid metal by the molten metal.
2. Crack initiation occurs at the solid–liquid metal interface.
3. Fracture is by rapid, brittle crack propagation. Crack growth rates in the order of 1 m s^{-1} have been reported[3].
4. Crack growth is predominantly intergranular.
5. The presence of liquid metal at the crack tip is essential for crack propagation by LMIE. If the liquid metal is depleted during crack growth, or the crack runs ahead of the liquid metal supply,

crack growth will occur by a conventional fracture mechanism, e.g. cleavage, microvoid coalescence or shear.

The mechanisms of LMIE have also been the subject of substantial research. Significant progress has been made in this area by, amongst others, Stoloff and Johnston[4], Westwood and Kamdar[5] and Lynch[6]. These mechanisms are reviewed by Nicholas and Old[3] and Stoloff[7].

Experimental difficulties encountered in the study of LMIE (e.g. the very fast crack growth rates observed, and the necessity to use elevated testing temperatures, particularly in the case of industrially important liquid metals such as zinc, tin and lead) have led to the use of simple testing techniques. More sophisticated concepts and testing procedures such as fracture mechanics have thus not been extensively employed in this field. This is surprising given the advantages of these techniques in the study of other environmentally assisted cracking phenomena.

In this investigation, standard LEFM concepts and testing procedures are used to study the fatigue crack growth (FCG) behaviour of brass in molten gallium ($T_m = 29.8$ °C). In particular, the effects of load ratio, cyclic frequency and load waveform on the rate of crack growth are investigated.

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FATIGUE IN LIQUID METAL ENVIRONMENTS

Reports of LMIE failures due to cyclic loading have prompted a number of investigations into the fatigue behaviour of metals in liquid metal environments. Early investigations were primarily concerned with the effect of these environments on the *S–N* behaviour. In some cases it is found that the number of cycles to failure, N_f, is reduced over the entire stress range, e.g. zinc–mercury[8], steel–mercury[9] and steel–sodium[10]. In other cases, N_f is reduced only in the high-stress range and the endurance limit is not significantly altered, e.g. copper–mercury[9,10].

The use of fracture mechanics in the study of fatigue crack growth in liquid metal environments is a relatively new approach and only a few such studies have been reported to date. Kapp and co-workers[11,12] investigated the effect of cyclic frequency on the embrittlement of aluminium alloys by mercury and found that FCG rates in liquid mercury were two to three orders of magnitude higher than in air. Increased FCG rates were observed at low frequencies, in agreement with predictions made by the superposition model for corrosion fatigue[13]. Matlock and co-workers[14–16] studied the effect of temperature and cyclic frequency on the FCG behaviour of 2.25Cr–1Mo steel in molten lithium. They found that FCG rates decreased as the cyclic frequency increased up to a critical frequency, f_{cr}. Above f_{cr}, the crack growth rate increased as the cyclic frequency increased. Their results are shown schematically in *Figure 1*.

EXPERIMENTAL PROCEDURE

Compact tension (CT) specimens were prepared from 63/37 brass (CZ108) plate according to the guidelines given in the ASTM E647 standard testing procedure[17]. A specimen thickness, *B*, of 12.5 mm was used in all cases. The mechanical properties of this material are given in *Table 1*. Specimens were prepared such that crack growth occurred in the direction of rolling (*T–L* orientation). A reservoir for the liquid gallium was made by sticking tape along the notch to a distance of approximately 5 mm from the notch tip.

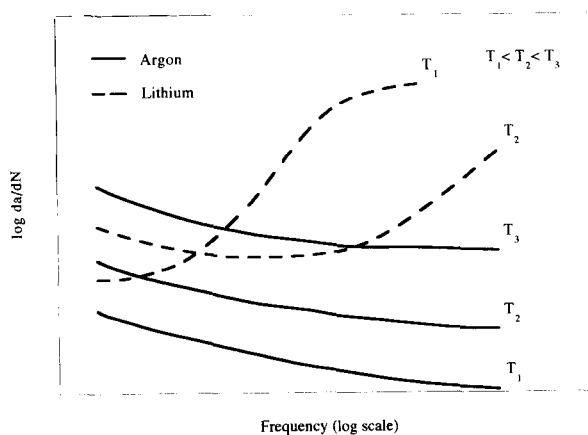Fatigue tests were carried out in accordance with

Table 1 The mechanical properties of 63/37 brass (CZ108)

0.2% proof stress (MPa)	UTS (MPa)	% elongation	% reduction in area
141	360	58	75

the ASTM E647 standard testing procedure[17] using a Schenck Hydropuls PSA hydraulic machine. Liquid gallium was introduced into the notch prior to fatigue precracking to ensure good wetting. The constant load amplitude (increasing ΔK) testing procedure was used, and special care was taken to ensure that LEFM conditions were not violated. Tests were conducted at various load ratios, cyclic frequencies and load waveforms. For the purpose of comparison, tests were also carried out under identical load conditions in laboratory air, i.e. without liquid gallium. All tests were performed at $35°$ C $\pm$ $2°$ C.

Crack length was measured optically on both sides of the specimen using a travelling microscope and a magnification of 35×. It was found that crack fronts were generally straight and no crack curvature corrections were required. FCG rates were calculated using the secant method and results presented in the form of $\log(da/dN)$–$\log(\Delta K)$ plots.

RESULTS

The effect of load ratio

FCG tests were carried out over a range of positive load ratios in both molten gallium and laboratory air. A cyclic frequency of 5 Hz and a sinusoidal load waveform were used in all these tests. These results are shown in *Figure 2*.

The rate of FCG in air at intermediate ΔK values, i.e. $\Delta K > \Delta K_{th}$, is found to be independent of load ratio. Crack growth rates are given by the Paris law relationship, where

$$\frac{da}{dN} = 1.28 \times 10^{-8}(\Delta K)^{3.5} \tag{1}$$

Figure 1 A schematic diagram of the effect of temperature and cyclic frequency on the FCG rate of 2.25Cr–1Mo steel in molten lithium[14]
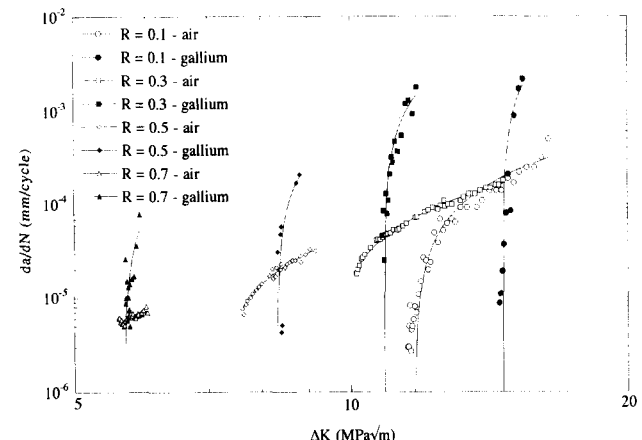

Figure 2 The effect of load ratio on the FCG of 63/37 brass in molten gallium and laboratory air (open symbols, results in air; solid symbols, results in gallium)
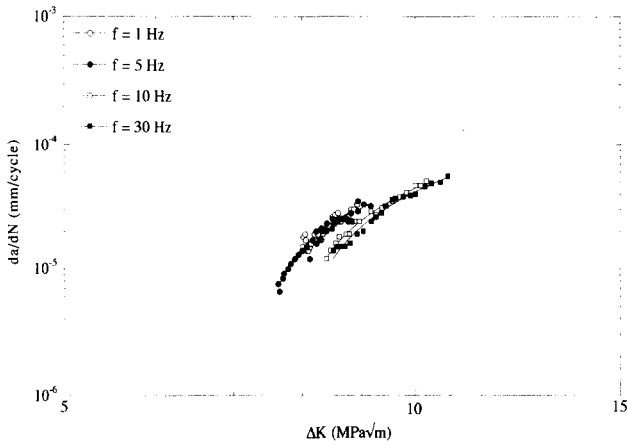
Figure 3 The effect of cyclic frequency on the FCG rate in air

da/dN is measured in mm cycle^{-1} and ΔK in MPa m$^{\frac{1}{2}}$. In the threshold regime, however, da/dN is strongly dependent on load ratio, and ΔK_{th} increases as the load ratio decreases.

For the tests carried out in liquid gallium only data from the threshold regime have been obtained as at higher ΔK values LEFM conditions would be violated. Crack growth rates at these low ΔK values increase rapidly with increasing ΔK, and are one to two orders of magnitude faster than that in air. The variation in ΔK_{th} with load ratio in molten gallium is similar to that observed in air. Thus ΔK_{th} increases as the load ratio decreases. In addition, it is interesting to note that at $\Delta K \approx \Delta K_{th}$, the rate of crack growth in gallium is actually lower than that in air.

The effect of cyclic frequency

FCG tests were carried out over a range of cyclic frequencies in air and molten gallium. In these tests, a constant load ratio of 0.5 and a sinusoidal load waveform were used. The results for FCG in air and molten gallium are given in *Figures 3* and *4* respectively.

Figure 3 shows that the effect of frequency on FCG in air is small and that the rate of crack growth increases as the cyclic frequency decreases. In molten gallium, however, the opposite effect is observed, and
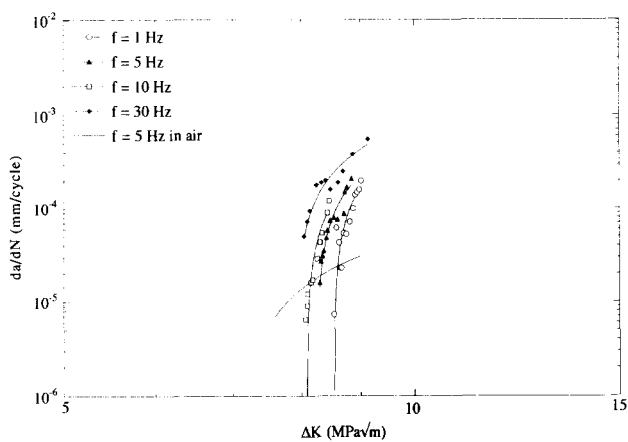
FCG rates increase as the frequency increases. Once again, at $\Delta K \approx \Delta K_{th}$, the rate of FCG is lower in gallium than in air.

The effect of load waveform

The effect of load waveform on FCG was examined using sinusoidal, triangular and square load waveforms. Tests were carried out in molten gallium at a load ratio of 0.5 and frequency of 5 Hz. These results are shown in *Figure 5*.

It is evident from these results that the load waveform does have a significant effect on FCG. Low ΔK_{th} values and high FCG rates were observed under square wave loading, suggesting that this is the most severe loading condition. Under triangular wave loading, ΔK_{th} is lower than that under sinusoidal loading, but at higher ΔK values the difference between these loading conditions is reduced. It is again interesting to note that at $\Delta K \approx \Delta K_{th}$, the rate of crack growth in molten gallium is lower than that in air for all load waveforms.

DISCUSSION

Various theories have been proposed to predict the rate of FCG as a function of ΔK. Of these, the damage accumulation models proposed by Rice[18] and Weertman[19] have proved particularly useful. In these models, the FCG rate is given by an equation of the form

$$\frac{da}{dN} = \frac{A(\Delta K)^4}{\sigma_y^2 U^*} \quad (2)$$

where A is a constant, ΔK is the stress intensity range, σ_y is the yield stress and U^* is the critical amount of absorbed hysteresis energy or plastic work required per unit crack growth.

Lynch[6] has proposed a model for LMIE in which adsorption of liquid metal atoms at the crack tip results in a localized reduction in the shear strength of the solid. This facilitates dislocation emission and movement, and leads to failure by extensive localized plasticity immediately ahead of the crack tip. Under these conditions the volume of material in which
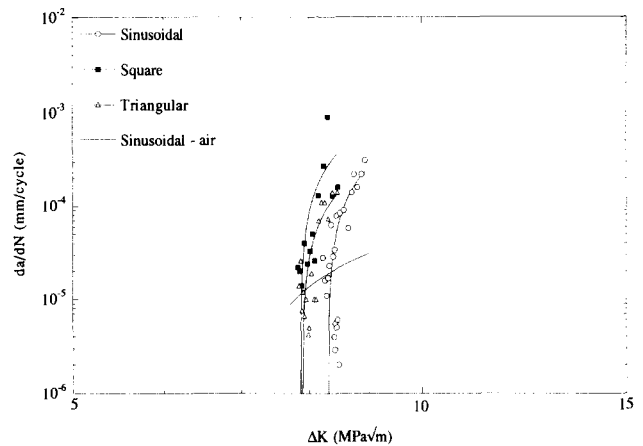

Figure 4 The effect of cyclic frequency on the FCG rate in molten gallium. The results for the tests in air at a frequency of 5 Hz are also shown

Figure 5 The effect of load waveform on the FCG rate in molten gallium. The results for tests in air under sinusoidal wave loading are also shown

the critical damage must be reached is significantly reduced[20]. This effectively reduces U^* in Equation (2). Thus, if U^* is reduced by a factor β^* such that $U^*_{env} = U^*_{inert}/\beta^*$, Equation (2) becomes

$$\left(\frac{da}{dN}\right)_{env} = \frac{\beta^* A(\Delta K)^4}{\sigma_y^2 U^*} = \beta\left(\frac{da}{dN}\right)_{inert} \tag{3}$$

Equation (3) has certain attractive features. First, in inert environments it predicts a Paris law exponent of 4. This is close to the value obtained in the present investigation. Furthermore, by making β^* a function of ΔK and time, i.e. $\beta^* = f(\Delta K, t)$, synergistic effects between environment and mechanical growth are readily accounted for. This is in contrast to the simple superposition or additive models proposed for corrosion fatigue[13]. Note that the exact form of β^* is not known at this stage.

The effect of load ratio on FCG is shown in *Figure 2*. In laboratory air load ratio effects are observed only in the threshold regime, where crack growth rates decrease rapidly as ΔK decreases. A similar load ratio effect is observed in liquid metal environments. These observations can be explained by considering crack closure effects, where crack closure results in premature contact between the fatigue surfaces during the unloading potion of the load cycle. These effects have been considered in detail elsewhere[21].

An interesting observation in the present study is that at $\Delta K \approx \Delta K_{th}$, the rate of crack growth in liquid gallium is less than that in air. This suggests that the liquid metal is actually beneficial in suppressing FCG at low ΔK values, as crack growth rates in this regime are lower than expected if the crack-driving force were purely mechanical. This behaviour can be explained in several ways. First, crack closure effects lead to a reduction in the effective stress intensity range at the crack tip, and thus reduce the crack driving force. This issue has been discussed previously[21]. Second, at low ΔK values the crack tip opening displacement is significantly reduced. This may restrict the access of liquid metal to the crack tip and therefore lead to reduced FCG rates. However, it is unlikely that the exclusion of liquid metal from the crack tip will lead to crack growth rates that are lower than that expected for purely mechanical crack growth. Third, the reduced FCG rates can be due to crack branching. This would reduce the effective stress intensity at the crack tip and therefore lead to reduced FCG rates. However, metallographic analysis of the specimens on completion of the FCG tests shows that no crack branching occurred.

Finally, the high ΔK_{th} values in gallium can be due to crack tip blunting resulting from the dissolution of solid metal into the liquid gallium. Crack blunting reduces the effective stress intensity range and therefore leads to a reduction in the crack-driving force. At low ΔK values and low FCG rates, more time is available for dissolution to occur, and crack blunting is able to suppress crack growth. At high crack growth rates, or where the crack tip is exposed to the liquid metal for very short periods of time, the extent of crack blunting is reduced and crack growth dominates. A number of factors affect the extent of crack blunting by dissolution, namely liquid and solid metal composition,

temperature, mean stress and the length of time the crack tip is exposed to the liquid metal. The latter depends on the cyclic frequency and the load waveform.

Figure 3 shows that the rate of FCG in laboratory air increases as the cyclic frequency decreases. These results are in agreement with those observed in many material–environment systems[22,23]. During crack growth in air, oxidation of the fatigue surfaces takes place. The oxide layers thus formed give rise to irreversible slip and hence increase the total FCG rate. At low frequencies more time is available for oxidation and slip irreversibility becomes more severe. The effect of frequency on FCG rates in molten gallium is shown in *Figure 4*. It is clear that in this environment, the rate of crack growth increases as the cyclic frequency increases. This result is the opposite to that observed in air, and is also contrary to that reported for the aluminium–mercury system[11,12].

The effect of cyclic frequency on the rate of crack growth can be accounted for by the crack tip dissolution mechanism proposed earlier. At low cyclic frequencies, more time is available for dissolution to occur during each load cycle, and crack blunting therefore proceeds to a greater extent. This, in turn, reduces the crack growth rate. As the cyclic frequency increases, less time is available for dissolution and the rate of crack blunting is insufficient to suppress crack growth.

It is interesting to consider the results of Matlock and co-workers[14–16] (*Figure 1*), in terms of the crack tip dissolution mechanism. In the high-frequency regime, FCG rates increase as the frequency increases, in agreement with the results of the present investigation. The effects of temperature on FCG can also be explained in terms of crack tip dissolution. At high temperatures the rate of dissolution is increased and crack blunting proceeds to a greater extent. This leads to reduced FCG rates. In the low-frequency regime, however, FCG rates increase as the temperature increases and the cyclic frequency decreases. Under these conditions, crack growth is by grain boundary penetration of the liquid metal ahead of the crack tip, and is enhanced by high temperatures and low frequencies.

The effect of load waveform on FCG in liquid metal environments (*Figure 5*) can similarly be explained in terms of the crack tip dissolution mechanism. The extent of crack blunting by dissolution depends on the length of time the crack tip is exposed to the liquid metal. For the square load wave, the load rise time, t_R, is significantly shorter than for the triangular or sinusoidal load waveforms. The amount of dissolution taking place during the loading and unloading portion of each load cycle is therefore less under square wave loading. In addition, for this load waveform it is important to consider the behaviour of the crack tip in the interval when $K = K_{max}$. If $K_{max} < K_{ILMIE}^*$, crack blunting can occur during this time interval. However, if $K_{max} > K_{ILMIE}$, static crack growth will occur. This will reduce the amount of crack blunting and increase the crack growth rates. In the present investigation, K_{max} is approximately equal to K_{ILMIE},

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* K_{ILMIE} is the threshold stress intensity for static crack growth in liquid metal environments, and is 16–18 MPa m$^{\frac{1}{2}}$ for the brass–gallium system used in the present study.

and static crack growth effects therefore contribute to the total crack growth rate.

SUMMARY

FCG tests have been carried out on 63/37 brass in laboratory air and molten gallium environments. The following observations have been made.

1. At $\Delta K > \Delta K_{th}$, FCG rates in air are independent of load ratio and can be described by the Paris law relationship.
2. The FCG rates in molten gallium at $\Delta K > \Delta K_{th}$ are one to two orders of magnitude faster than that in air. It is tentatively proposed that crack growth rates in this regime can be described by the damage accumulation models for fatigue.
3. In the threshold regime, FCG rates in both laboratory air and molten gallium are affected by crack closure effects, which result in an increase in the apparent ΔK_{th}.
4. At $\Delta K \approx \Delta K_{th}$, crack growth rates in the liquid gallium are lower than that in laboratory air.
5. In liquid gallium, dissolution of the solid metal into the liquid metal occurs, leading to crack tip blunting. This reduces the effective stress intensity at the crack tip and leads to lower crack growth rates.
6. At high cyclic frequencies, insufficient time is available for dissolution to proceed to any great extent, and crack growth processes therefore dominate. This leads to an increase in FCG rates at high cyclic frequencies.
7. The effect of load waveform on FCG is due to changes in the time available for crack tip dissolution to occur and to static crack growth effects. FCG rates are most severe for the square load waveform where load rise times are short and $K = K_{max}$ for a finite period of time.
8. The extent of dissolution depends on the liquid and solid metal composition, temperature, applied load and the time available for dissolution. These effects must be taken into consideration in any analysis of FCG in liquid metal environments.

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