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Fatigue in lithium fluoride crystals at elevated temperatures

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Summary

When lithium fluoride single crystals are cyclically strained below 400° C strain hardening occurs very rapidly and no failure occurs; above 400° C both cross slip and dislocation climb can occur quite freely. Cyclic straining under these conditions results in a steady decrease in density associated with the generation of micro-pores. This development eventually produces considerable opacity in the specimens and crack nucleation by void accumulation at deformation induced tilt and kink boundaries. Once cracks nucleate however, their growth is relatively slow. Experiments performed with silver chloride under conditions where cross slip was observable but dislocation climb was difficult suggest that climb is necessary for the production of porosity in both of the materials. The mechanism of fatigue damage described is thought to be present in metals at elevated temperatures.

Introduction

In spite of the numerous studies on the development of fatigue damage, the mechanisms by which such damage is produced in metals at room temperature are still in question, and the fatigue process at elevated temperatures is even less clearly understood. In this investigation, lithium fluoride, a member of a class of semi-brittle materials, was fatigued at elevated temperatures where metal-like ductility was observable. Examination of specimens took place at room temperature where the samples could be sectioned by cleavage without affecting the fatigue damage. The object of the study was to investigate the high temperature fatigue of LiF in order to obtain fundamental information on the fatigue process in general. Transparency, photo-elastic behavior, etchability, and stability at elevated temperatures are additional factors which contributed to the choice of LiF as a material useful for a fundamental study. Ionic crystals have been employed previously in some fatigue investigations. Forsyth [1], McEvily and Machlin [2], and McClintock and Groll [3] found that the process of crack nucleation in AgCl was very similar to that occurring in ductile metals. On the other hand, McEvily and Machlin [2], and Subramanian and Washburn [4] could not produce fatigue failure at room temperature in LiF and MgO respectively, regardless of the magnitude of the stress amplitude.

Experimental procedure

Lithium fluoride single crystals of the highest available purity were used. The yield stress of the crystals was about 100 gm/mm^2 between room temperature and 350 °C, and rose slowly to a value of 200 gm/mm^2 at 500 °C, indicating that although impurity effects were not entirely absent they were not important. The crystals were cleaved into prismatic slabs of approximately $4{\cdot}5 \times 6 \times 30$ mm dimensions. A gauge section was produced in the center by hand grinding a 12·5 mm radius on both sides – leaving a neck of $4{\cdot}5 \times 3$ mm. All specimens had cleaved cube surfaces; orientations other than (100) were not tested. Specimens were polished in fluoboric acid, rinsed in methanol and ether and were stored in polyethylene bags in a dessicator until used. No signs of significant mechanical damage in the gauge section resulting from the preparation could be found in the specimens.

Fatigue tests were performed with a Sonntag SF-2 fatigue machine in cantilever bending under fully reversed constant load amplitudes at a frequency of 1800 c/min. Specimens were located in a tubular stainless steel vacuum system enclosed by a split furnace. Motion was introduced into the vacuum system through a flexible bellows joint. All tests were performed in a dynamic vacuum of 10^{-3} Torr. The temperature was monitored by a thermocouple suspended near the specimens. At the termination of a test an initial cooling rate of 50 °C/min was obtained in the chamber by opening the furnace and directing an air blast against the exposed furnace tube. This was sufficient to minimize thermal alterations in the fatigued crystal subsequent to fatigue testing.

In the experiments conducted below 400 °C where rapid hardening in the specimens produced essentially elastic bending, the maximum bending stress on the outer surfaces of specimens was used as a parameter to characterize the fatigue process. In experiments conducted above 400 °C much of the specimen cross section was plastic, thus the total strain amplitude after saturation, calculated from the maximum radius of curvature during bending was used as the characterizing parameter. Some tests were interrupted to study the development of fatigue damage by visual or microscopic examination. In these tests some annealing of fatigue damage may have occurred upon re-heating the specimens prior to re-application of the cyclic load. This may have resulted in some extension of the fatigue life of the specimens but such effects appeared to be minor and were ignored.

The damage in the fatigued specimens was studied at room temperature by visual and internal microscopic examination. In addition specimens were sectioned by cleavage both in and transverse to the plane of bending. Cleavage surfaces were examined by both optical and scanning electron microscopy. Internal stresses were measured in some specimens

by the photoelastic method. Conventional electron microscopy by the replica method proved too cumbersome because of the difficulty in removing replicas from the rough cleavage surfaces.

Experimental results

Fatigue experiments below 400 °C confirmed the earlier observations of McEvily and Machlin [2] that cyclic loading produces rapid strain hardening in LiF and does not result in a fatigue failure. In one test at 370 °C the stress amplitude was raised in steps to a final level of 1400 gm/mm^2 during $1{\cdot}6 \times 10^6$ cycles. The specimen failed after 30,000 cycles at the highest stress. An examination of the fracture surface showed that the fracture originated from a small crushed area showing no sign of slow crack growth characteristic of fatigue but had the appearance of an impact site. The slip traces on the top and bottom surfaces at these lower temperatures were straight and shallow as shown in Fig. 1.

Above 400 °C, where Gilman [5] has observed cross slip on the (100) planes, the behavior was quite different. Fig. 2 shows the slip traces on top surface of a crystal subjected to ¼ cycle at 510 °C. Strong cross slip traces of the (221) and (111) planes can be seen in addition to the prominent traces of the two (110) planes normal to the plane of bending. Traces of the (110) planes oblique to the plane of bending, visible at the top of the figure, were found only near the side surfaces. There was no evidence of slip on these planes in the interior of the specimen, no doubt because slip on these planes would not produce the changes of curvature required by the imposed deformation conditions.

Cyclic straining at 510 °C rapidly produced cells of single slip in the specimen in which only one of the primary (110) systems was active. In spite of the fully reversed nature of the imposed cyclic load, lattice misorientations between these cells of single slip of 2-4 ° were measured in some samples. In this manner many tilt and twist boundaries became established inside the specimen separating cells of a fraction of a millimeter size .

The cyclically reversed plastic strain at 510 °C also produced very wavy and deep slip bands on the surface of the specimens as shown in Fig. 3. This rough slip topography contained prominent extrusions and intrusions as shown in the scanning electron micrograph of Fig. 4; the reversal of the direction of the extrusions across a deformation induced twist boundary can be clearly seen. The deep intrusions and the blocklike extrusions are shown in Fig. 5 by a photomicrograph taken from a longitudinal cleavage surface.

In addition to the effects discussed above, cyclic straining at 510 °C also resulted in a gradual loss of transparency which became visibly noticeable after about 15,000 cycles and increased steadily with further cycling until the highly strained regions of the crystals were opaque. This effect is evident in Fig. 6 which shows a transversally illuminated specimen which had been subjected to 620,000 cycles at a strain amplitude of 0·26%. While the neutral plane region of the specimen appears dark, the outer portions of the specimen subjected to large strain reversals scatter light into the microscope. Detailed examination showed that the light was scattered by numerous small pores in the specimen. Fig. 7 (a) and 7 (b) show longitudinal cleavage surfaces from such a fatigued specimen illuminated by reflected and transmitted light respectively. The many elongated dark patches in Fig. 7 (a) are voids intersected by the cleavage plane: the many light dots are subsurface voids reflecting light into the microscope. All voids regardless of their position relative to the cleavage surface appear as dark patches in the transmitted light view of Fig. 7 (b). The scanning electron micrograph of Fig. 8 provides a clearer picture of the voids intersected by a longitudinal cleavage surface.

The development of opacity by void formation and growth must certainly be accompanied by a decrease in overall density of specimens. The change in density with cycling, prior to the development of opacity was measured with a conventional density column using a mixture of bromoform and carbon tetrachloride. In this experiment, slivers from the top and bottom surfaces of cycled specimens were used. The difference in density between these specimens and uncycled specimens subjected to the same thermal treatment was measured. The results shown in Fig. 9 indicate that the decrease in density is gradual and begins from the very start of the cycling. Only after about 15,000 cycles does the density change produce observable traces of opacity.

With continued cycling a clustering of voids was observed along: (a) deformation induced tilt boundaries and kink walls as shown in Figs. 10 and 11, (b) deformation induced twist boundaries as shown in Fig. 12, and (c) the active slip direction in the form of beads in cells of single slip as shown in Figs. 7 (a) and 7 (b). The presence of a resolved normal stress component across the tilt and kink boundaries resulted in eventual crack formation by void clustering as shown in Figs. 10 and 11. The absence of a resolved normal stress across the twist boundaries resulted in only an intense porosity as shown in Fig. 12.

In addition to the tests at a strain amplitude of 0·26%, tests were performed at strain amplitudes of 0·36% and 0·59%. These tests showed no fundamentally different feature other than more rapid development of opacity and accumulation of damage.

Cycling at 650°, as shown in Fig. 13, produced wavier slip and rougher surface topography in comparison to cycling at 510 °C. The development of intrusions and extrusions also became more prominent as shown in Fig. 14. While much less opacity developed at 650 °C in comparison to 510 °C, damage accumulation along deformation induced boundaries was more rapid.

An annealing experiment was performed to test the stability of the porosity developed at 510 °C. An opaque specimen which had been cycled at a strain amplitude of 0·59% for 218,000 cycles was annealed at 750 °C for 4 hours. The annealing restored full transparency to the specimen except in regions which already possessed large voids. The voids in these regions assumed a very regular cubical shape.

The slip distribution in all the experiments was quite homogeneous on the surface. Although prominent intrusions did develop, etching the longitudinal cleavage surfaces revealed no special dislocation configuration at the continuation of the intrusions in the crystal. The crystals fatigued above 400 °C showed very little birefringence. Internal stresses were measured at a few spots and found to be roughly of the same magnitude as the yield stress.

Although cyclic deformation above 400 °C produced widespread porosity, intense damage accumulation along deformation induced boundaries, and crack nucleation along tilt and kink boundaries, no fatigue failure could be produced within millions of cycles. Once nucleated, cracks grew only very slowly. In the majority of cases, the vast segmentation resulting from the extensive porosity and cracks made the specimens so compliant that it was impossible to keep the fatigue machine running at the excessively large excursions of the load arm. For this reason no S-N curve can be given.

Discussion of results

Although the need for cross slip in the development of fatigue damage has been emphasized by many investigators [2, 6], the specific role of cross slip has not been satisfactorily established. It has been proposed that cross slip serves to eliminate internal stresses of strain hardening and that no fatigue damage can develop at a temperature and stress level where strain hardening cannot be saturated [7, 8]. That no fatigue damage developed below 400 °C in LiF where cross slip was absent is consistent with this view. It must be noted, however, that 400 °C is 0·59 of the absolute melting temperature of LiF where rapid dislocation climb should be possible. To determine whether climb or cross slip is the more important ingredient for the development of porosity, similar experiments were performed with AgCl in which cross slip develops at a temperature well below that for dislocation climb. These experiments showed that similar porosity and accumulation of damage around grain boundaries developed in AgCl at 150 °C [9] which is again 0·58 of the absolute melting temperature. Cycling AgCl below 150 °C developed only the persistent slip plane damage characteristic of ductile metals: no porosity and damage accumulation along grain boundaries was observed. From this we conclude that the development of widespread porosity requires climb. The presence only of cross slip produces the more familiar fatigue damage inside persistent slip bands, but when both climb and cross slip can occur, slip is dispersed by climb and the high temperature form of fatigue damage is dominant. In the absence of both climb and cross slip, rapid hardening develops without fatigue damage as is the case with LiF and MgO at room temperature.

All known instances of fatigue fit this general pattern. Ductile metals such as Cu, Ag, Au, Cd, Mg and Al in which cross slip is possible at all temperatures, show fatigue even at 4·2°K [10]. This is most likely by the persistent slip band deterioration mechanism. Metals in which cross slip is restricted at low temperatures such as Zn and Fe do not show fatigue at these low temperatures but undergo brittle fracture [10]. The same behavior of course, holds for LiF [2] and MgO [4] as already discussed above. The development of porosity observed by Wood and associates in brass [11, 12], copper [12] and iron [13], by Forsyth in aluminum and its alloys [1], and AgCl [1] are all of the low temperature persistent slip band type. The elevated temperature porosity found by Ronay, Reimann and Wood in their highest temperature experiments in alpha brass [14] and that found by Grant, Blucher and Ritter in aluminum [15] are of the type in which climb is the dominant mechanism of internal stress relief.

Although it is premature to be precise in detail, the mechanism of development of fatigue damage leading to crack nucleation can be described in its general outlines. At low temperatures, where climb is minimal, cross slip plays an essential role in the localization of slip inside persistent slip bands. The elimination of screw dislocations by cross slip and the associated shunting of slip between a large number of neighboring slip planes inside the persistent slip bands produces only islands of edge dislocation dipoles which form the boundaries of intricate and regular cells [16, 17]. The to and fro slip activity between these cell walls inside the persistent slip bands steadily produces a large number of vacancies by the intersections of primary screw dislocations and forest screw dislocations. These vacancies coalesce either by very short range diffusion inside the persistent slip bands or are swept to and fro by edge dislocations. This process should become increasingly difficult at low temperatures but should never cease because the local temperature rise due to slip [18] at very low temperatures can still provide sufficient thermal energy for very short range diffusion of vacancies. Recent measurements of large resistivity changes in fatigue at cryogenic temperatures in copper [19, 20] and aluminum [21] all show that large numbers of vacancies are continuously produced inside persistent slip bands. The isochronal annealing experiments and transmission electron microscopy observations of Eikum [21] on aluminum fatigued at cryogenic temperatures show that more than half of the measured changes of resistivity are associated with di-vacancies and single vacancies.

At temperatures where climb is rapid, the effects of strain hardening are undone over the entire specimen resulting in uniform dispersal of slip and elimination of persistent slip bands. The production of vacancies by dislocation intersections is now more uniformly spread over the entire volume. The vacancies produced can now diffuse over much longer ranges. Random vacancy condensation produces void nuclei throughout the volume resulting eventually in the opacity of the fatigued crystals. Longer range diffusion of vacancies to sinks such as deformation induced boundaries produces intense porosity and eventual crack nucleation at these sites. The measured rate of decrease in density, $7{\cdot}8\times10^{-8}$ gm/cm^3-cycle, referred to above implies a very reasonable rate of vacancy production which is quite compatible with the actually observed densities of forest dislocations and expected density of mobile dislocations.

Once cracks have nucleated at deformation induced boundaries by the vacancy condensation mechanism, they do not grow rapidly because the increased numbers of vacancies produced by slip concentration around the crack are dispersed into the surrounding volume by diffusion instead of concentrating at the crack tip. The crack grows primarily by the somewhat increased rate of arrival of vacancies at the crack front. A very rare case of localized production of porosity in the shear zone of a surface notch is shown in Fig. 15.

Similar observations have been made by Grant, Blucher, and Ritter [15] on aluminum, suggesting that the mechanism of fatigue damage observed in LiF is also present in metals at elevated temperature.

Acknowledgments

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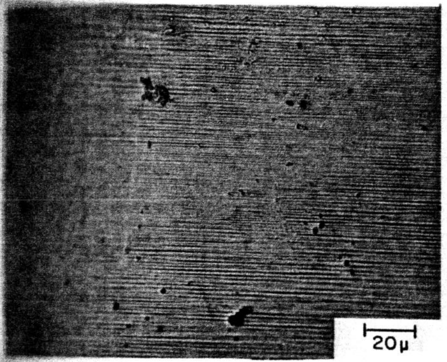

Fig. 1. Fatigue slip bands after 7·5 × [illegible] cycles at 900 g/mm² and 400° C.

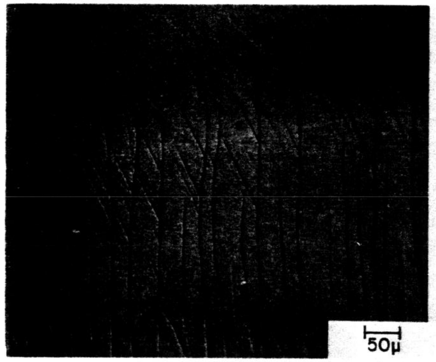

Fig. 2. Slip traces on top surface of crystal bent ¼ cycle at 510° C.

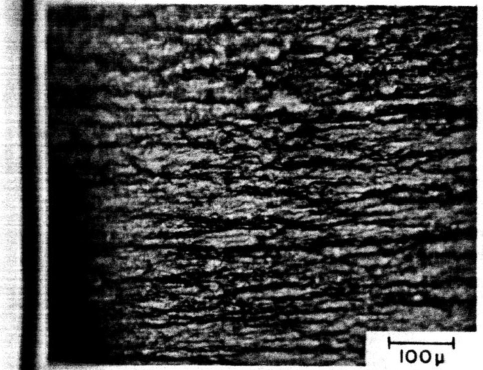

Fig. 3. Fatigue slip bands after 1·16 × 10^5 cycles at a strain amplitude of 0·26% at 510° C.

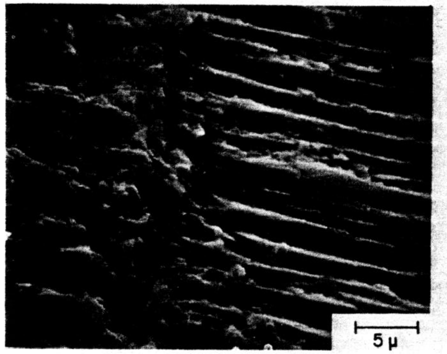

Fig. 4. Scanning electron micrograph of extrusions on top surface of crystal fatigue at 510° C. showing also deformation induced twist boundary.

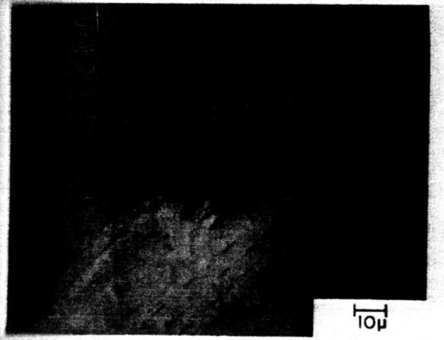

Fig. 5. Cross-section by longitudinal cleavage of surface topography in crystal cycled at 510° c at a strain amplitude of 0·26% for 0·41 × 10^5 cycles.

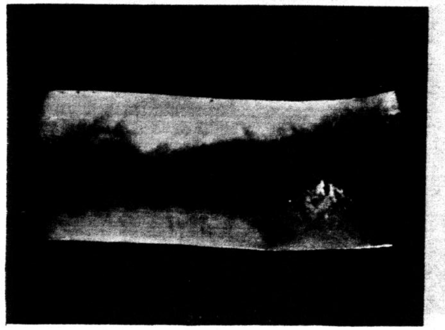

Fig. 6. Appearance in transverse illumination of crystal cycled at 510° C at a strain amplitude of 0·26% for 6·2 × 10^5 cycles.

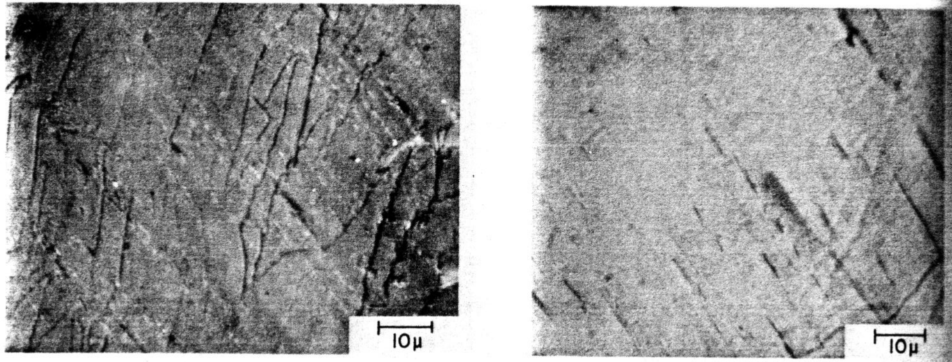

Fig. 7. (a) View in reflected light of voids in crystal cycled at 510°C at a strain amplitude of 0·59% for 3·15 × 10^5 cycles. (b) Same view as above in transmitted light.

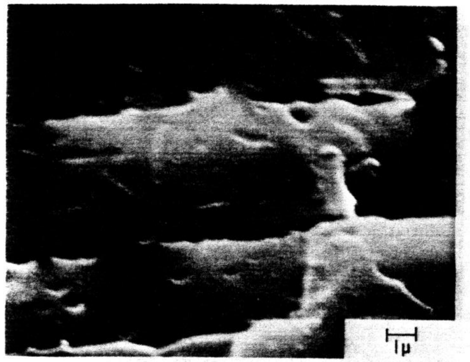

Fig. 8. Scanning electron micrograph of voids intersected by a longitudinal cleavage surface.

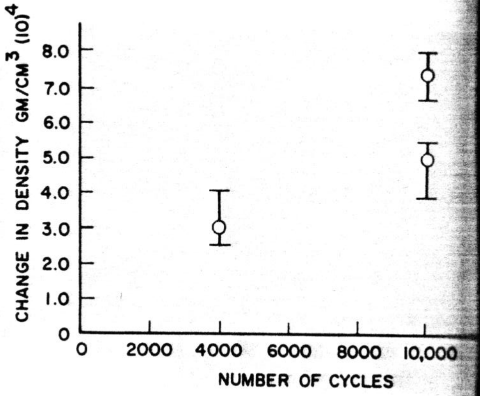

Fig. 9. Change in density of LiF fatigue specimens after testing at 510° C at a strain amplitude of 0·26%.

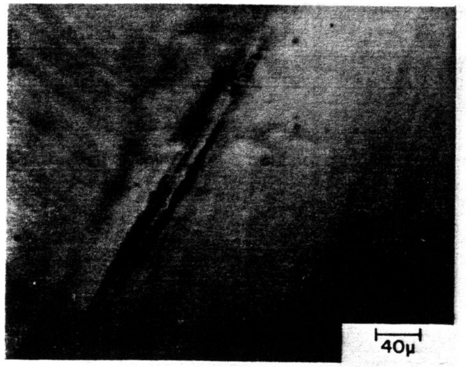

Fig. 10. Void accumulation and crack nucleation at deformation induced tilt boundary in crystal at 510° C at a strain amplitude of 0·26% after 2·25 × 10^6 cycles.

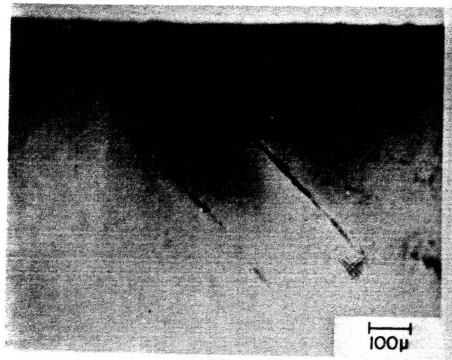

Fig. 11. Void accumulation and crack nucleation at kink boundaries in crystal at 510° C at a strain amplitude of 0·36% after 10^5 cycles.

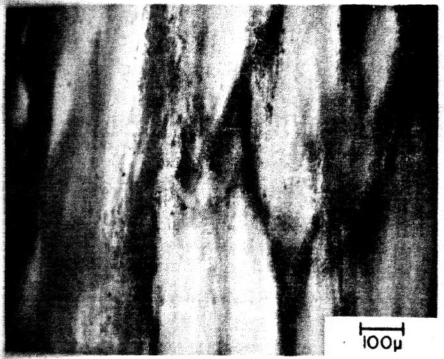

Fig. 12. Void accumulation at deformation induced twist boundaries in crystal at a strain amplitude of 0·59% after 1·18 × 10^5 cycles.

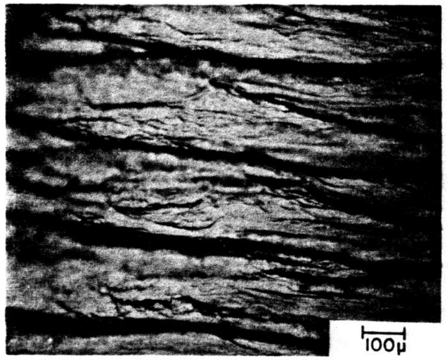

Fig. 13. Fatigue slip bands after 5 × 10^4 cycles at a strain amplitude of 0·68% and 650° C.

Fig. 14. Surface topography and damage in crystal cycled at 650° C at a strain amplitude of 0·68% for 5 × $(10)^4$ cycles.

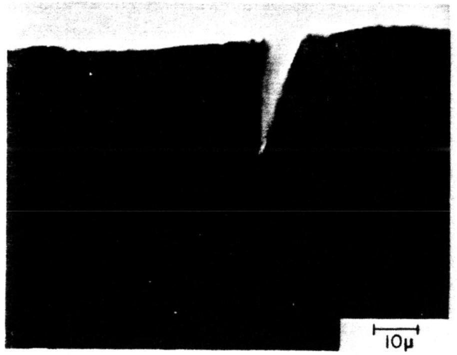

Fig. 15. Pore damage in shear zone ahead of surface crack after 7·5 × 10^4 cycles.