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TEM study of the recovery process of cyclically deformed MgO single crystals

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Single crystals of MgO were subjected to plastic strain-controlled push–pull cyclic deformation at elevated temperatures. Below 400° C the crystals were very brittle and failed with a few fatigue cycles. At 470° C a large number of cycles could be obtained before failure, and the cyclic stress–strain response showed a period of rapid hardening followed by a period of decreasing hardening rate. TEM investigations of the lower temperature samples show structures of isolated dislocation dipoles, multipoles and debris. At 470° C dense bundles of dislocations were observed aligned perpendicular to the Burgers vector direction. The regions between the bundles were relatively dislocation free, but they contained a high density of debris. Bowed out screw dislocations are observed between the edge dislocation bundles, suggesting that screw dislocations were largely mobile. Comparisons are made with the cyclic deformation and structure of f c c metals and other NaCl structure single crystals.

1. Introduction

The dislocation structures following fatigue of some metal single crystals have been well characterized by various investigators. During the early cycles the structure consists of dislocation debris, which gradually gives rise to well-defined dislocation configurations in the saturation regime of cyclic deformation. In the case of f c c single crystals such as copper, oriented for single slip, deformation is ultimately confined to persistent slip bands (PSB) consisting of ladder-like dislocation structures [1–7]. The walls or rungs of the ladder are oriented perpendicular to the [1 2 1] direction, and are mainly composed of immobile edge dislocations. The regions in between the ladders are relatively dislocation and debris free, suggesting that a large fraction of debris formed during the early stage of fatigue is swept up by the motion of dislocations between the ladders. Similar structures have been observed in h c p materials such as magnesium [8], as well as in b c c materials at sufficient impurity concentrations and low strain rates [3].

In comparison to metals, there have been only limited investigations on the cyclic deformation behaviour of rock salt type crystals, and an early review on the subject is given in [9]. An extensive work on the push–pull cyclic deformation behaviour of LiF single crystals [10–12] indicated that at temperatures above 300° C, the cyclic behaviour and dislocation configuration had similarities with that of f c c single crystals. Thus at such elevated temperatures and in the saturation regime of fatigue, typical ladder-like PSB structures were observed, though variations in the dislocation configuration were found in various parts of the crystals. On the other hand, during the early stages of fatigue and at low homologous temperatures, dense arrays of dislocation etch pits were observed along slip bands. During this period hardening was almost linear per cycle, and the crystals could sustain as much as ten times the initial flow stress. At very low amplitudes the samples withstood a very large number of cycles before fracture, and a decrease in cyclic hardening rate could be observed. Similar recovery behaviour was also noticed during push–pull fatigue of MgO single crystals at room temperature [13]. Etch pit studies on LiF [10] cycled at room temperature, indicated some banding of dislocations during the recovery period. However, since dislocation densities were extremely high it became difficult to resolve the dislocations by etch pit techniques, even using TEM replicas. Also the Burgers vector of the dislocations were not unambiguously determined. Attempts to conduct transmission electron microscopy on LiF samples were unsuccessful, since the crystals degraded under the electron beam.

The work presented in this article, on the fatigue of MgO single crystals, gives direct evidence of the dislocation behaviour during the initial cyclic recovery stage. Essentially it is intended to augment the work on LiF crystals cycled at temperatures below 300° C. Since the melting point of MgO (3037 K) is much larger compared to LiF (1115 K), so elevated temperatures had to be chosen for the investigation in order to achieve homologous temperature comparable to that of LiF at room temperature. Note that while LiF single crystals are not amenable to TEM study, MgO single crystals are excellent for the purposes of electron microscopy.

Previous investigations on the dislocation structure in monotonically deformed MgO single crystals indicated a dislocation debris structure [14] and elongated edge dislocation pairs. The stability of the pairs depends on the testing temperature, with narrow pairs

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breaking up into small prismatic loops at 750° C and above. TEM investigations on MgO crystals deformed under reversed bending at room temperature [9, 15] showed essentially a debris structure, and dislocations entangled in dipole and multiple configurations. Etch pit studies indicated slip bands increasing in width with the number of cycles, but the density of etch pits remained approximately constant within a slip band. TEM observations showed that the debris structure was essentially constant after a certain number of cycles, and there was no evidence of any form of recovery. In the present work MgO single crystals have been subjected to push–pull strain-controlled fatigue loading at 370 and 470° C. Push–pull loading ensures a uniform stress state throughout the sample, and in addition allows any dynamic recovery to be easily noticed from the plot of stress amplitude against number of cycles. As already mentioned, elevated temperatures are necessary for the homologous temperature of deformation to be comparable to that of LiF at room temperature. The latter crystals, it may be remembered, showed recovery at low strain amplitudes, and therefore a similar behaviour is expected in the case of MgO. It is believed that the elimination of edge dislocation dipoles during dynamic recovery occurs by a sweep-up operation, possibly by the mechanism proposed in [16]. Sufficient stress is required for the process, and that is why recovery can only occur after a significant amount of hardening. Easier propensity of cross slip also helps in the clean-up operation, and therefore higher temperatures aid the process. The MgO crystals, fatigued under axial loading, have finally been subjected to TEM observations for determination of dislocation structure.

2. Experimental procedures

2.1. Mechanical testing

Single crystals of MgO were purchased from the Norton Research Corporation (Canada) Ltd. The exact impurity concentration is, however, unknown. Cleaved and polished specimens of undeformed crystals showed very low dislocation density, when observed in the electron microscope. The crystals were cleaved into sizes approximately 1.9 mm × 1.9 mm × 30 mm, and the gauge length region was reduced in cross-section by using emery paper. After a final polish with 600 grit paper, the crystals with {1 0 0} faces were chemically polished in stirred orthophosphoric acid at about 60° C. About 200 μm was removed from the surface by this method, and the edges of the crystals became rounded. The crystals were then mounted in cement-filled cups, and tested on a servohydraulic testing system, fitted with a proportionally controlled furnace. The final exposed dimensions of the samples were around 1.7 mm × 1.4 mm × 4 mm. The loading direction was [1 0 0]. An LVDT assembly, similar to the one used in [11] was used to monitor and control the strain. A specially built plastic strain amplitude controller was used in order to control the plastic strain, rather than the total strain amplitude.

The strain rate was fixed at $10^{-5}\,\mathrm{sec}^{-1}$ for all the tests. Higher strain rates usually led to premature failure. Testing was conducted at two temperatures: 370 and 470° C. The plastic strain amplitude was 0.15% for all the tests. Higher strain amplitudes could not be used as the crystals were too brittle even at these temperatures to withstand larger plastic strains.

2.2. Electron microscopy

Failure of the crystals always occurred at the grips, indicating the presence of large stress concentration there. After failure, the crystals were cleaved in half, parallel to the loading axis. These were then coated with an enamel paint, except for a small region where perforation was desired. The crystals were then chemically thinned from both sides, in warm orthophosporic acid. After sufficient thinning the paint was removed by dipping the specimens first in ethyl alcohol and then in acetone. the entire crystal was then chemically polished in warm H_3PO_4, until perforation was observed. The polished specimens were washed and rinsed in reagent grade acetone, and a thin coating of carbon was evaporated on both sides. The purpose of this step was to prevent charging up of the specimens by the electron beam. The specimens were then placed between two copper grids, and the dislocation structure examined on a Jeol-100B electron microscope, operating at 100 kV.

3. Results

3.1. Mechanical testing

It must be mentioned at the very outset that the crystals are extremely brittle, and all the samples fractured before any apparent saturation in the peak stress was reached. At 370° C, $T/T_{\mathrm{m.p.}} = 0.21$, the crystals failed within 20 cycles. However, hardening was rapid, and the flow stress doubled within these few cycles. At 470° C, $T/T_{\mathrm{m.p.}} = 0.24$, approximately 250 cycles could be withstood at 0.15% plastic strain amplitude, before the crystals fractured. Such brittleness is not unusual when we compare the behaviour with that of LiF single crystals fatigued at room temperature [10], where, $T/T_{\mathrm{m.p.}} = 0.27$.

The cyclic hardening curve of MgO at 470° C is shown in Fig. 1, where the average normal peak stress is plotted against the cumulative plastic strain $\epsilon_{\mathrm{cum}} = 4\,\Sigma\,\Delta\epsilon_{\mathrm{p}}/2$. The hardening curve shows a period of rapid hardening followed by a decreasing rate. The

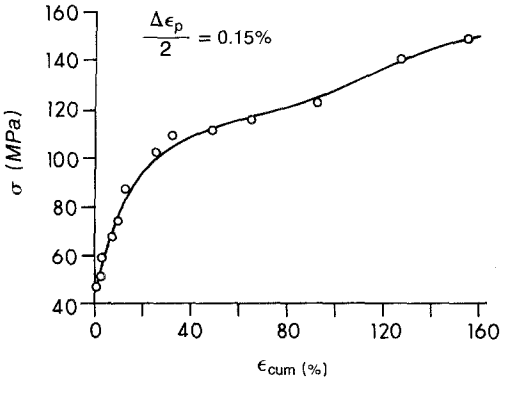

Figure 1 Fatigue hardening curve for MgO single crystal fatigued at 470° C. σ is the normal peak stress, and ϵ_{cum} is the cumulative plastic strain.

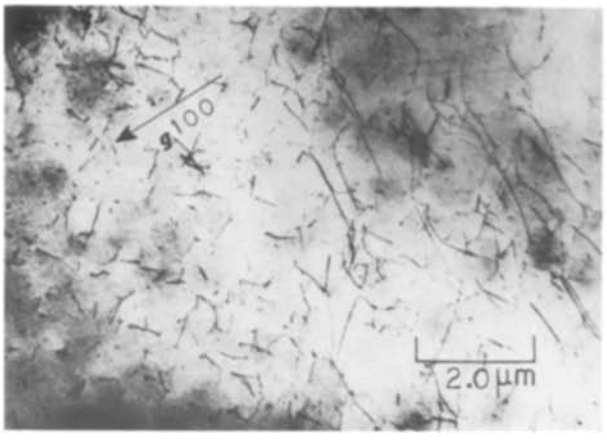

Figure 2 Transmission electron micrograph of an MgO single crystal, cycled at 370° C.

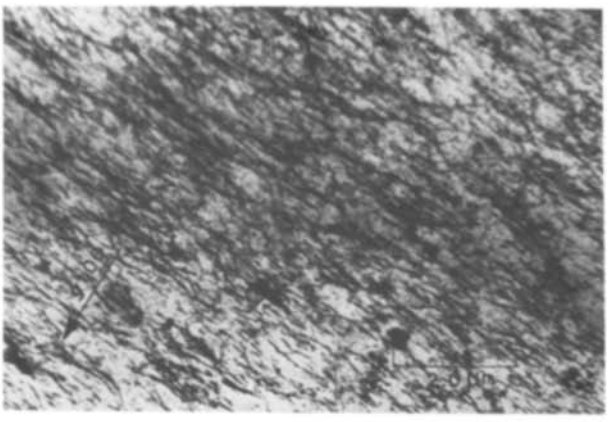

Figure 4 TEM of MgO single crystal at 470° C. The plastic strain amplitude was 0.15% for 250 cycles.

flow stress rises by as much as three and a half times the initial yield stress. A decrease in hardening rate during the later cycles probably implies that some dynamic recovery takes place. Etching of the crystals in warm H_3PO_4 after fracture, showed that in a given region slip took place on predominantly one slip system. Since the crystals were rounded, good etch pit micrographs could not be obtained.

3.2. Transmission electron microscopy

3.2.1. Fatigue at 370° C

Fig. 2 shows the dislocation structure of a crystal fatigued at 370° C for 17 cycles. Fig. 3 shows a similar region viewed at higher magnification. The structure consists of long and short dislocation dipoles and jogs randomly arranged. Interconnected dipoles are also observed, as at A (Fig. 3). The star-shaped arrangement (B, Fig. 3) is probably formed by dislocation bowing between pinning points and then cross slipping. In addition, pinching of loops are observed (C, Fig. 3), and such debris formation may occur by the mechanism first suggested by Johnston and Gilman [17]. Some multipoles are also observed but these are extremely few in number. In short, the structure essentially consists of dislocation debris and dipoles, and is similar to the observations [9, 15] made on MgO single crystals, fatigued in bending at room temperature.

3.2.2. Fatigue at 470° C

The dislocation structure of a crystal cycled at 0.15% plastic strain amplitude for 250 cycles, is shown in Fig. 4. Fig. 5 is a larger magnification micrograph of a similar region. The dislocation density is much higher when compared to the structure observed at 370° C, and bundles of dislocations are observed, aligned perpendicular to the [1 0 0] direction. Burgers vector analysis indicate that the Burgers vector of the dislocations is [1 0 1]. The arrangement of the dislocations is best explained by referring to Fig. 6. The TEM specimen is polished parallel to the plane BCGF, and the electron beam is almost normal to this plane. Invisibility of the dislocations is observed under $\boldsymbol{g} = [0\,1\,0]$. Thus the dislocations lie on the (1 0 1) plane with Burgers vectors of the type $\langle \bar{1}\,0\,1 \rangle$. In addition to the edge dislocation bundles, primarily screw segments can be seen lying between the bundles (A, Fig. 5). The screw segments are fewer in number and many of them retained their bowed out configuration even without external stresses, probably due to internal stresses and high friction stress of the material. Such curvature of the dislocations imply that the screw segments are largely mobile. This, and the fact that the dislocation density is extremely high within the edge bundles, suggest that most of the plastic strain is accommodated by screw and mixed screw segments. The edge segments probably become rapidly immobilized by dipole structures.

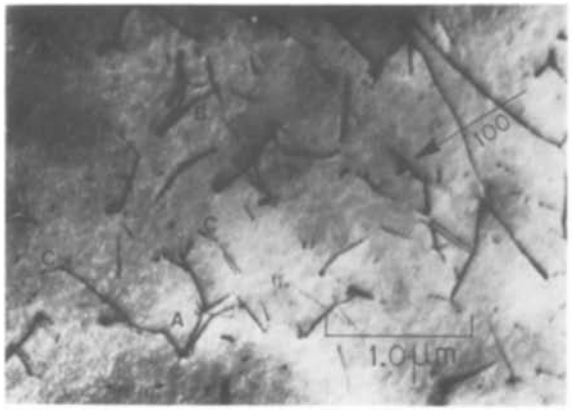

Figure 3 TEM of MgO single crystal cycled at 370° C. Higher magnification micrograph of Fig. 2.

Figure 5 TEM of MgO single crystal cycled at 470° C. Higher magnification micrograph of Fig. 4.

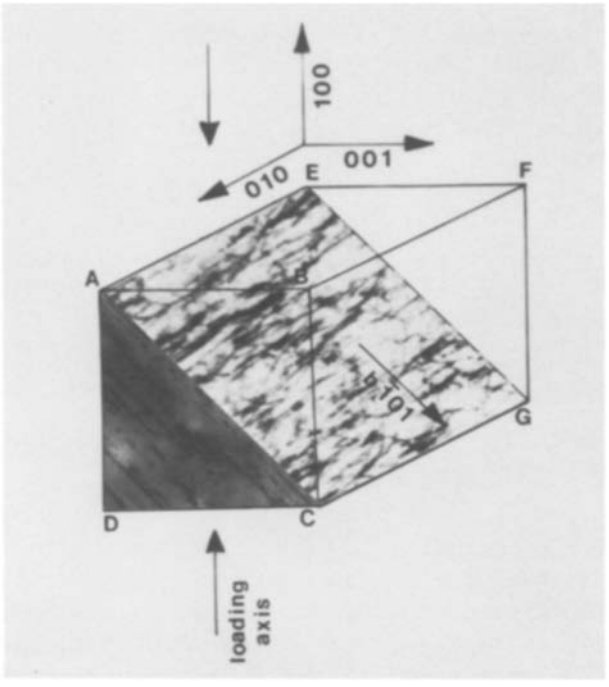

Figure 6 Three-dimensional montage of dislocation structures of MgO single crystals, fatigued at 470° C.

In some regions of the crystal, the density within the bundles is even higher and such a high dislocation density make it difficult to obtain clear micrographs of the dislocation structure. Essentially the structure is similar to that in Figs 4 and 5, except for higher dislocation densities. Fig. 7 is a weak beam micrograph of a bundle at extremely large magnification. Most of the dislocations are edge in character, and are arranged in dipole configurations. A screw segment (AB, Fig. 7) can also be seen bowing between the bundles. Besides the dislocations, a large number of dots can be seen in the micrograph. These are probably small loops and point defect clusters left in the wake of moving dislocations.

Since the crystals are small, it is not possible to obtain TEM specimens parallel to both planes BCGF and ADCB (Fig. 6), from the same sample. Fig. 8 is a TEM micrograph of a crystal fatigued for 100 cycles at 0.15% plastic strain amplitude. The TEM specimen has been sectioned parallel to plane ADCB (Fig. 6). As opposed to Fig. 4, the dislocations here are observed to lie nearly along crystallographic $\langle 1\,0\,1 \rangle$ direction. Burgers vector analysis show that the Burgers vector of the dislocations is of type $[\bar{1}\,0\,1]$, so that the dislocations in Fig. 8 are of screw character. A closer look shows that the long straight dislocations (AB, Fig. 8) lie approximately 5° away from the [1 0 1] direction. This apparent deviation can be accounted for by the orientation of the crystal with respect to the electron beam [18]. Special features may be observed in Fig. 8. For example, the region such as C clearly show where the dislocations have cross slipped. A large number of small loops are also observed (D, Fig. 8); and high magnification micrographs indicate that many of these loops (A, Fig. 9) do not lie on the (1 0 1) plane. A segment of Fig. 9 shows a sufficiently long dislocation found by joining up of dislocations on neighbouring (1 0 1) planes. A kink and interconnected loops can be seen at B and C, respectively. In some regions of the crystal the dislocations are much more fragmented than in Fig. 8. Such fragmentation is probably due to annihilation by cross slip and pinching off due to climb.

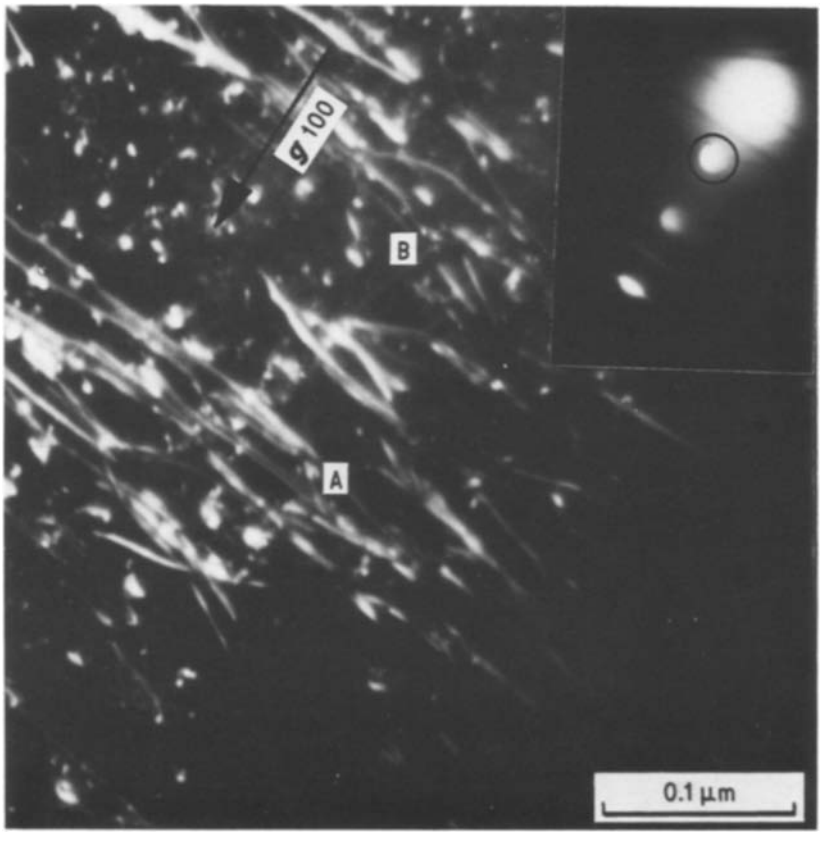

Figure 7 Weak beam micrograph of dislocation bundles in an MgO single crystal, cycled at 470° C. A bowed out screw dislocation segment can be seen along AB.

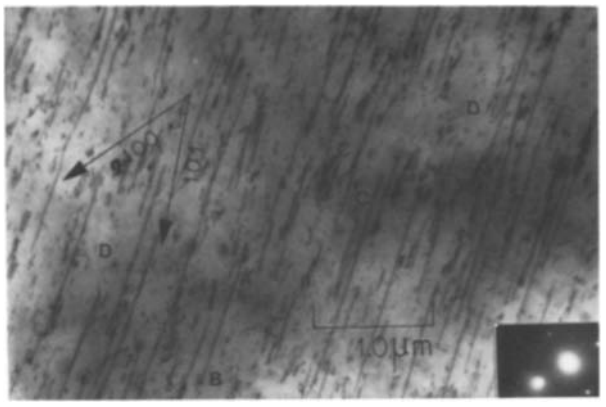

Figure 8 TEM of MgO single crystal fatigued at 470° C for 100 cycles. Section cut parallel to plane ADCB in Fig. 6.

4. Discussion

At 370° C and low cumulative strains, the dislocation structure in fatigued MgO single crystals consists of a random arrangement of dislocation dipoles, jogs and loops. This type of debris structure is similar to that observed earlier on MgO crystals, fatigued in reverse bending at room temperature [9, 15]. Multiple cross glide and trailing of edge dislocations behind jogged screw dislocations [17] are likely mechanisms for such a structure formation. Pinching off of dipoles are also observed. This could occur mechanically by cross slip, if the dislocations are not in pure screw orientation. Alternatively, even though the homologous temperature, 0.21, is quite low, pinching off could occur by climb of edge dislocations, aided by narrow dipole widths and by pipe diffusion.

Debris structure is also a characteristic feature during the initial stages of fatigue of many metal single crystals, cycled at or below room temperature [19–21]. However, there is one important difference. While the present results at 370° C indicate that the dislocations are randomly arranged, the structure in aluminium fatigued at 78 K [21] shows that the debris consists mainly of loops, aligned parallel to the ⟨1 1 2⟩ direction.

At 470° C some dynamic recovery occurs, as observed from a decrease in the cyclic hardening rate of the cyclic hardening curve. Although four {1 1 0} slip systems are equally stressed, slip is confined to only one slip system. This is concluded from contrast experiments, which show that all the dislocations have the same Burgers vector. Dense bundles of dislocations are observed, aligned perpendicular to the Burgers vector direction. The orientation of the bundles suggest that they are probably formed by cooperative cross glide of a number of screw dislocations. A significant number of dipoles and loops exist between the bundles, and these too are aligned perpendicular to the Burgers vector direction. Such an orientation is similar to the ⟨1 1 2⟩ type debris arrangement in aluminium, fatigued at low strain amplitudes at 78 K [21]. An important difference can, however, be noted between the bundle structure in MgO, and the bundles formed during the initial stages of fatigue of fcc metal single crystals, oriented for single slip [19–21]. A high concentration of debris exists between the bundles in MgO, while in metal single crystals the region between the bundles is relatively free of dislocations. The immobility of jogs (lying on (1 0 0) planes) and narrow dipoles is the primary cause for such a difference in behaviour, since they cannot be easily swept into the bundles by other dislocations [16, 22]. It would require edge dislocations lying on planes closer to the arms of the dipole than the dipole width, to bow out from the bundles and thus annihilate the dipoles or pull them to the next bundle. Such a process would probably need an even higher dislocation density within the bundles and consequently a larger applied stress. Nevertheless

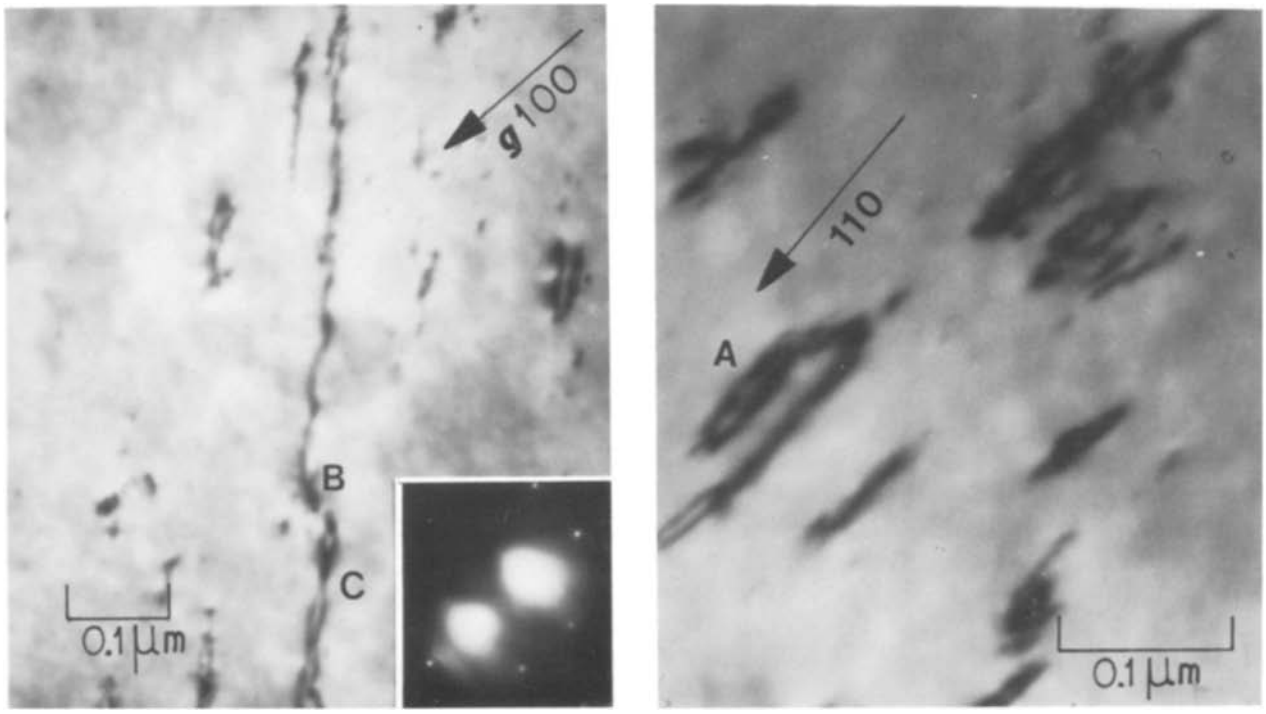

Figure 9 Higher magnification micrographs of samples sectioned parallel to plane ADCB in Fig. 6.

some clearer regions in the TEM specimens suggest that recovery must be occurring at the temperature and stresses of this investigation, and it is surmised that dynamic recovery is directly related to the sweeping up of dipoles and loops lying between the bundles. Only then would there be an easier passage for mixed screw dislocations to accommodate the plastic strain. The clearing of regions between bundles is also aided by disintegration of dipoles by climb. Thus, though the homologous temperature is only 0.24, the small loops in the weak beam micrograph presented earlier indicate that local diffusion does play an important role. In the case of metals fatigued at very low temperatures, climb could occur by pipe diffusion. The importance of thermal activation in fatigue cannot, therefore, be discounted.

The bowed out configuration of screw dislocations between bundles suggest that they are largely mobile. Heating up by the electron beam also indicate that they are relatively easier to move than edge dislocations within the bundles. Hence, as mentioned earlier, it is concluded that most of the plastic strain is accommodated by screw and mixed screw segments. The edge components are rapidly immobilized, and even if they do move their mean free path is extremely small due to debris existing between the bundles.

Observation on planes perpendicular to the primary slip plane show that screw components are mostly wavy and jogged. Small loops are found which do not lie on the primary slip plane, and must be formed by cross-slip. In some places quite a few cross-slipped dislocations are grouped together. Since the jogs are immobile, these must be sites where edge dislocation bundles are formed. Many of the observed kinks on long screw segments could be formed by climb of edge components of dislocation loops on nearby slip planes. Such a climb mechanism could also be a significant factor in bundle formation.

Before concluding it may be mentioned that PSB structure, typical of fcc metal single crystals oriented for single slip, have not been observed. However, these crystals fractured before any saturation in the peak stress was reached. It remains speculative if structures resembling PSB structures would have occurred had saturation been reached. The results of LiF single crystals [11, 12] suggest that testing at higher temperature would show a ladder-like PSB structure. Further careful experiments are needed to verify this.

5. Conclusion

The dislocation structures in MgO single crystals, fatigued for 250 cycles at 470° C, show some resemblance to the structure observed during the early stages of fatigue of fcc metal single crystals. The main difference lies in the fact that while in metals the region between the bundles is relatively free of dislocations, in MgO a significant concentration of debris exists between them. Immobility of jogs and dipoles in MgO is thought to be the primary reason for such a difference in behaviour. It is believed that dynamic recovery is directly related to the cleaning up of the debris between the bundles. Accordingly higher fatigue strength can be achieved in metals by making the dislocation debris more immobile. Lower self diffusivity would have a similar effect, since it would prevent dipole disintegration by climb.

Bowed out screw dislocations between bundles suggest that plastic strain is mainly accomodated by screw and mixed screw segments. This is similar to the situation that occurs in the PSBs of fcc metals at saturation. The implication is that the mobility of screw dislocations in fcc metals is probably a major factor in fatigue, even before saturation is reached.

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