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THE EFFECT OF OXYGEN ADSORPTION ON THE FATIGUE BEHAVIOUR OF LEAD SINGLE CRYSTALS + POLYCRYSTALS

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ABSTRACT

A study has been made of the fatigue properties of single crystals of lead in air at pressures from 760 to 2 x 10^{-6} torr. The fatigue lives of crystals at a plastic shear strain amplitude of 0.2% were significantly greater at air pressures below 1 torr. However, the air pressure dependence of the fatigue life of the single crystals was not as marked as that of polycrystalline material.

In both the high and low air pressureranges, the fatigue experiments showed that, at the same resolved shear strain amplitude, crystals with "hard" orientations had lives which were up to five times greater than those of crystals with "soft" orientations.

The results are discussed in terms of a previously proposed model for atmospheric corrosion fatigue, the effect of orientation on the cross-slip process, and the blocking of the growth of fatigue striations and cracks by dislocation tangles and loops produced by the active secondary slip systems.

1. Introduction

Previous studies of the fatigue properties of polycrystalline lead have shown that the fatigue life and mode of failure depend to a marked extent on the partial pressure of oxygen in the surrounding test atmosphere[1]. A reduction in the partial pressure of oxygen below 3 x 10^{-2} torr results in a sudden and striking increase in fatigue life and a marked reduction in the amount of inter-crystalline cracking. A change in cycle frequency or in the time of the experiment does not affect the form of the air pressure dependence of the fatigue life, nor the pressure at which the life suddenly increases[2]. The present work was undertaken to determine the effects of atmosphere on the fatigue properties of lead single crystals and to examine the mechanism for atmospheric corrosion fatigue in the absence of grain boundaries.

Although there have been numerous investigations of the fatigue behaviour of metal single crystals, there have been few in which attention was given to the effects of atmosphere on fatigue behaviour$^{\phi}$.

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ϕ One case which can be cited is an investigation by Broom and Summerton[3] who noted that a few experiments on Zn single crystals which were coated with butyl rubber indicated that the coating increased the life by about 30%.

Studies of the influence of orientation on the fatigue life of single crystals in air at atmospheric pressure have also been made in relatively few cases. Lipsitt and Horne[4], and Hempel[5] found no marked effect of orientation on the fatigue life of α - iron single crystals. Similarly Broome and Ham[6], and Ebner and Backofen[7] found no clear evidence for an orientation dependence of the fatigue life of Cu single crystals. However, Ebner and Backofen noted that with two crystal orientations, the rotational orientation about the principal axis had an effect which indicated that the life was longest when the slip direction was parallel with the specimen surface.

Roberts and Honeycombe[8] on the other hand obtained a marked orientation dependence of the fatigue life of Al single crystals at the same resolved shear stress or initial plastic strain amplitude such that crystals with "hard" orientations (i.e. principal crystal axis near the boundaries or the (001) and (111) corners of the standard triangle) had a high fatigue resistance compared with the fatigue resistance of "soft" crystals (i.e. principal axis near the centre of the standard triangle).

In contrast to the work of Roberts and Honeycombe, measurements by McEvily and Boettner[9] of the rate of crack propagation at constant resolved shear stress amplitude in Al single crystals which contained a small stress raiser indicated that the rate of crack propagation was highest near the (001) - (111) boundary (i.e. "hard" orientations). This suggests that such crystals would have low fatigue resistance.

In view of the above conflicting results on orientation effects, it was decided to examine the influence of orientation on the fatigue behaviour of lead single crystals both in the presence and absence of atmospheric oxygen.

2. Experimental Methods

Single crystals were grown under vacuum (approx. 10^{-5} torr) in a split mould made from high purity graphite using the Bridgeman technique. The lead was from the same batch of material as that used in an earlier study of polycrystalline lead (99.999% Pb.)[1]. The specimen shape and dimensions were the same as those used previously[11]. Crystals were annealed at 100°C for 24 hr., chemically polished and fatigued in reverse plane bending at 500 cycles/min. at 22° ± 2°C using the methods described elsewhere[1,12].

Preliminary fatigue tests on two crystals showed that recrystallization was absent except for that which occurred outside the gauge length in the region of the edges of the gripped section. In these tests in air at atmospheric pressure, the presence of a recrystallized region lead to intercrystalline cracking outside the gauge section and premature failure of the crystal. The tendency for recrystallization was overcome by covering the faces of the grips with lead foil of similar purity to that of the specimen. After failure, crystals were etched and carefully examined to ensure that recrystallization had not occurred during the test.

The reverse plane bending machine used in the present tests subjected a specimen to a constant total strain amplitude, i.e. the specimen deflection was constant throughout the test. The elastic strain component is liable to

vary with orientation because of the orientation dependence of fatigue hardening (see Ebner & Backofen[7]) and because the elastic constants for lead are anisotropic. In order to estimate the magnitude of the elastic strain, the width of the undeformed region near the neutral axis on the sides of specimens was measured for several polycrystalline and single crystal specimens. By assuming that the strain was proportional to the distance from the neutral axis, the elastic resolved shear strain component was estimated to be approximately from 0.01% to 0.02% which is small compared with the magnitude of the total imposed resolved shear strain, 0.2%. Examination of the polycrystal data given in Fig. 2 for the low pressure region shows that at γ = 0.002, a change of strain of 0.0002 changed the life by about 40%. This estimate indicates that the present tests can be considered to be made at approximately constant plastic strain amplitude. A similar conclusion was reached by Ebner and Backofen[7] from estimates of the elastic strain in Cu crystals based on bending moment measurements at similar total strains.

The orientations of crystals were determined by the Laue back-reflection technique. The results are summarised in Table 1 which gives: the angle between the slip direction and the tensile axis, ϕ; the angle between the tensile axis and the normal to the slip plane, λ ; the angle between the plane defined by the tensile axis and the slip direction and the plane defined by the tensile axis and the normal to the broad surface of the specimen, δ ; and the number of cycles to failure, N. In defining δ , the same definition has been used as that given by Ebner and Backofen[7]. It may be noted that the longer lives listed in Table 1 represent continuous test times of 2 to 4 weeks duration.

Stereographic plots of the location of the principal axis of crystals are given in Figs. 1 and 2. The areas outlined by dashed lines in the triangles correspond approximately to the three areas used by Paterson[13] to discuss the effects of orientation on the cyclic work hardening curves of Cu single crystals.

3. Experimental Results

3. 1. Effect of Atmosphere on the Fatigue Life of Lead Single Crystals:

The air pressure dependence of the fatigue life of the lead single crystals at the constant strain amplitude of ϵ = 0.092% is shown in Fig. 1 together with results for polycrystalline specimens tested at the same strain amplitude.

Although it was not possible to determine the air pressure dependence of the fatigue life of crystals with identical orientation, the lives of crystals which had similar orientations, i.e. near the centre of the triangle, (E, H, S, T, U and W in Fig. 1) suggest that the air pressure dependence of the fatigue life is similar in form to that exhibited by the polycrystals shown as the full curve in Fig. 1.

No marked orientation dependence of the fatigue life of the single crystals was evident in the data shown in Fig. 1.

3. 2. The orientation Dependence of the Fatigue Life in the High and Low Pressure Ranges:

An orientation dependence of the fatigue life became evident when the data were plotted on the basis of the resolved shear strain amplitude, Fig. 2. The resolved shear strain amplitude is given by $\gamma = \epsilon/\mu$ where ϵ is the principal bending strain amplitude and μ is the orientation factor $\mu = \cos\phi . \cos\lambda$. The results for polycrystals have been included in Fig. 2 by following the procedure used by Kemsley and Paterson[14] in which the principal bending strain is divided by a factor 0.435*.

In Fig. 2, the single crystal data have been divided into two groups according to whether the crystals were tested in the high or low air pressure ranges; the boundaries of the two groups are indicated by dashed lines in the lower part of the figure. Examination of the distribution of the lives of the single crystals with regard to shear strain amplitude and pressure region revealed a trend for crystals with "hard" orientations, i.e. near the (001) - (111) boundary and toward the (001) and (111) corners, to have longer lives than "soft" crystals, i.e. near the centre of the triangle, in both the high and low pressure ranges. For instance, in the high pressure range, crystals A, C, Q and P form a group with long lives whereas crystals E, H, S and T form a group with relatively short lives. Crystals I, K and R form an intermediate group. Similarly, in the low pressure range, crystals G and O have long lives, crystals M and J. have intermediate lives, and crystals U and W have short lives. Reference to the values of $|\delta|$ given in Table 1 suggests there was no marked rotational orientation effect superimposed on the influence of axial orientation indicated in Fig. 2.

Fig. 2 shows further that on a basis of the same resolved shear strain amplitude and similar orientation, the influence of atmosphere is not as great on the fatigue resistance of the single crystals as on the fatigue resistance of the polycrystals, e.g. at an approximate shear strain amplitude of 0.20% the average life of crystals U and W was about 4 times the average life of crystals E, H, S and T. Similarly the life of crystal O is about 5 times that of crystal P. These figures may be compared with the overall ratio for the single crystal data at about γ = 0.20% of 4.7 (i.e. comparing the average life of crystals, J, O, U and W with the average life of C, E, H, I, P, Q, R and S) and with the corresponding ratio for the polycrystals of about 30. It is evident from Fig. 2 that the difference between the ratios for the single crystals and the polycrystals can be attributed to the lives of the single crystals in the high pressure region which are longer than the corresponding lives of the polycrystals in the same pressure region.

3. 3. Observations on Crack Nucleation and Propagation:

Fatigue striations were evident on the chemically polished surfaces of crystals in the first 10^2 to 10^3 cycles. The striations formed parallel to the most favoured slip systems and gradually spread over the gauge section.

* This is equivalent to using the average ratios of principal bending strain to resolved shear strain for all orientations in the stereographic triangle. As Kemsley and Paterson note, this method of plotting the polycrystal data introduces some uncertainty into the comparison with the single crystal data, but it is better than a direct plot of the polycrystal data according to principal bending strains.

By 10^4 to 10^5 cycles, some striations had intensified as part of the general disturbance of the surface, Fig. 3. This figure shows an area of the side surface near the edge, and an area on the broad face of crystal O after 8 x 10^5 cycles. Fig. 3a shows the topography of the slip movements forming the hills and valleys in the top surface. The development of a valley about 20 microns deep is indicated at A. In some areas, tongues of metal, which were parallel with traces of the primary system, protruded in the slip direction above the surface of the crystal, e.g. extrusions of this type occur at the positions marked B in Fig. 3a; a similar observation has been made by Thompson[15] on a Cu single crystal. Figs. 3a and b show that slip on the primary system predominated.

At later stages of the life certain striations had developed into recognizable crevices or grooves which were approximately parallel with the traces of the active slip systems. These were similar in appearance to the grooves in the maximum shear directions observed on polycrystals fatigued in the low pressure region[1, 16]. Fig. 4a shows a crack propagating at a small angle to traces of the critical system which was one of the most favoured slip systems in crystal C. This area and other areas of crystal C exhibited slip on the critical, primary and to a lesser extent on the cross systems. It was noted that crystals oriented near the (001) - (111) boundary and in the region of the (112) showed variations in the extent to which the primary and conjugate systems operated. Similar effects have been reported by Paterson[13] and others[14 17]. Close examinations of the tip of the crack shown in Fig. 4a revealed that an area of intense slip occurred in the immediate region of the tip. Fig. 4b shows the same area as in Fig. 4a after a light polish and etch, and confirms that the intense disturbance in Fig. 4a was in fact a crack. (The contrast around the crack in Fig. 4b is due to staining effects produced by the etchant which tends to become trapped in the crack). These polishing and etching techniques did not reveal any sign of sub-boundary formation ahead of propagating cracks.

In general, crystals fatigued in the high pressure region showed a similar development of deformation to crystals fatigued in the low pressure region; however, the degree of deformation was greater in the latter crystals, as might be expected from the greater number of cycles to failure of this group.

4. Discussion

The present work shows clearly that lead single crystals were significantly less prone to atmospheric corrosion fatigue than polycrystals. The results indicate that at an equivalent resolved shear strain amplitude of about $\gamma = \pm 0.20\%$, polycrystalline lead was about 6 times more susceptible to atmospheric corrosion fatigue than lead single crystals. This difference is attributed to the inter-crystalline cracking that occurs in polycrystalline lead at high oxygen partial pressures[16].

The fatigue life of the single crystals of similar orientation (near the centre of the standard triangle) varied with air pressure in a similar manner to that shown by the polycrystals except that the height of the "step" in the curve was not as marked. The similarity in the form of the air pressure dependence of the fatigue life of the single crystals and polycrystals suggests that the mechanism of atmospheric corrosion fatigue is likely to be the same as that proposed on the basis of changes in cycle frequency and time of test for polycrystals[2]. In this mechanism which is illustrated in Fig. 5, the supply of oxygen to the tip of the crack is controlled by the equilibrium population density of the physically absorbed oxygen molecules at the external surface. Oxygen reaching the crack tip reacts with the freshly exposed metal and lowers the cohesion of the metal for the next cycle. In the single crystal case, however, the oxygen attack takes place at the tip of a crack originating from a slip striation.

The fatigue life of the single crystals exhibited the same orientation dependence when fatigued either in the high or low pressure regions (i.e. air pressures greater or less than 10^{-1} torr). This and the similar sequence of fatigue deformation shown by the single crystals in both pressure regions suggest that the presence of oxygen in the test atmosphere simply speeds up the rate of crack propagation.

The life tended to increase as the orientation moved from the centre of the stereographic triangle towards the (001) - (111) boundary and the (001) and (111) corners. This dependence is in agreement with that shown by Al single crystals found by Roberts and Honeycombe[8] and is in contrast to the dependence shown by Cu single crystals observed by Ebner and Backofen[7]. In view of the proposed formation of fatigue cracks by the cross-slip mechanism, the orientation dependence of the fatigue life should be consistent with the tendency for cross-slip to occur, i.e. crystals with orientations toward (001) and (111) should exhibit the lowest fatigue resistance whereas orientations along the (011) - (112) great circle should show the highest fatigue resistance. The experimental data, however, show no evidence for the fatigue resistance to vary in such a manner. In fact a trend in the opposite direction was observed. An absence of a correlation between fatigue life Al single crystals and the resolved shear stress on the cross-slip system was also noted by McEvily and Boettner[9].

The observed orientation dependence of fatigue life does correlate with the activity of the secondary slip systems. This suggests that growth of slip striations or cracks may be determined by obstacles produced by dislocation activity on the secondary slip systems. In particular, mobile dislocations associated with a spreading striation or with the observed plastic region ahead of fatigue cracks may be blocked by the dislocation tangles and dislocation dipoles lying in their path[18]. On this basis dislocations associated with a spreading striation or crack are expected to "see" more forest dislocations the greater the secondary slip activity. Furthermore, Orowan[19] has pointed out that obstacles to crack propagation tend to divert cracks out of their plane and that these deviations can produce an additional limitation to crack propagation.

The present observations on crack propagation in lead, which has a low stacking fault energy ($\sim$25 erg/cm^2 [20]) showed no evidence for sub-boundary formation ahead of cracks similar to that observed in aluminium[21] which has a high stacking fault energy ($\sim$ 238 erg/cm^2 [20]). This is consistent with the explanation given by Grosskreutz[22] that sub-boundary formation ahead of fatigue cracks occurs more readily the higher the stacking fault energy.

5. Conclusions

The conclusions which are derived from this paper are :-

(i) The fatigue life of lead single crystals increases by a factor of about five at oxygen partial pressures less than 2 x 10^{-2} torr.

(ii) At comparable strain amplitudes to those used for the single crystal tests, the fatigue life of polycrystalline lead increased by a factor of 30 at oxygen partial pressures below 2 x 10^{-2} torr.

(iii) An orientation dependence of the fatigue life was found in which lead crystals with "hard" orientations had lives which were about five times those of crystals with "soft" orientations.

(iv) The orientation dependence of the fatigue life was independent of oxygen partial pressure.

(v) The form of the air pressure dependence of the fatigue life of the single crystals was in accord with a model for oxygen corrosion fatigue of polycrystals in which the supply of oxygen was controlled by an equilibrium number of physically absorbed oxygen molecules.

(vi) No evidence was found for the fatigue life to decrease with increasing values of the Schmid factor for the cross-slip system.

(vii) No evidence was found for sub-boundary formation ahead of fatigue cracks in lead single crystals. This is consistent with the low stacking fault energy of lead.

(viii) The orientation dependence of the fatigue life can be accounted for in a qualitative manner by interactions between fatigue cracks and the obstacles produced by secondary slip activity, and the effect of crack deviations on the rate of crack propagation.

Acknowledgments

This investigation forms part of the research program of The Broken Hill Associated Smelters Pty. Ltd. and the author would like to thank Mr. R.J. Hopkins for his continued interest and encouragement. The work was carried out under the general direction of Professor J.N. Greenwood and the author would like to express his thanks to Professor Greenwood and Professor Worner for the facilities made available.

REFERENCES

(1) K.U. Snowden, Acta Met., 12, 295 (1964)
(2) K.U. Snowden, Phil. Mag., 10, 435 (1964)
(3) T. Broom and J.M. Summerton, Phil. Mag., 8, 1847 (1963)
(4) H.A. Lipsitt and G.T. Horne, Proc. International Conference on Fatigue of Metals, London, 513 (1957)
(5) M.R. Hempel, Basic Mechanisms of Fatigue, A.S.T.M. Special Publication No. 237, Philadelphia, 52 (1959)

(6) T. Broom and R.K. Ham, Proc. Roy Soc., A, 251, 186 (1959)
(7) M.L. Ebner and W.A. Backofen, Trans. Met. Soc. A.I.M.E. 215, 510 (1959)
(8) E. Roberts and R.W.K. Honeycombe, J. Inst. of Metals, 91, 134 (1962-63)
(9) A.J. McEvily and R.C. Boettner, Acta Met., 11, 725 (1963)
(11) K.U. Snowden, Phil. Mag., 6, 321 (1961)
(12) K.U. Snowden, J. Sci. Instrum., 41, 470 (1964)
(13) M.S. Paterson, Acta Met., 3, 491 (1955)
(14) D.S. Kemsley and M.S. Paterson, Acta Met., 8, 453 (1960)
(15) N. Thompson, Fracture, Ed. B.L. Averback, John Wiley and Sons, New York, 354 (1959)
(16) K.U. Snowden and J.N. Greenwood, Trans. Met. Soc. A.I.M.E. 212, 626 (1958)
(17) K.U. Snowden, Acta Met., 11, 675 (1963)
(18) K.U. Snowden, Electron Microscopy Society of America, Denver, U.S.A. (1963)
(19) E. Orowan, Fatigue in Aircraft Structures, Ed. A.M. Freudenthal Academic Press, New York, 121 (1956)
(20) G.F. Bolling, L.E. Hayes and H.W. Wiedersich, Acta Met., 10, 185 (1962)
(21) J.C. Grosskreutz, J. Appld. Phys., 33, 1787 (1962)

(22) J.C. Grosskreutz, Fatigue - An Interdisciplinary Approach, Syracuse University Press, New York, 27 (1964)

Table 1: Orientation Parameters & Fatigue Life of Lead Single Crystals

Crystal	ϕ	λ	$\lvert\delta\rvert$	Air Press Torr	N (Cycles)	Crystal	ϕ	λ	$\lvert\delta\rvert$	Air Press Torr	N (Cycles)
A	32.5°	63°	23°	760	2.60×10^6						
C	39°	53°	37°	"	3.85×10^6						
H	48°	43°	22°	"	2.52×10^6						
P	44°	52°	12°	"	3.98×10^6						
Q	40°	55°	2°	"	2.82×10^6						
S	53°	40°	2°	"	2.56×10^6	M ø	36°	54°	44°	6×10^{-4}	2.98×10^6
T	43°	48°	4°	"	0.956×10^6	W	43°	48°	46°	"	10.71×10^6
E	48°	45°	73°	1.0	1.69×10^6						
I	34°	56°	30°	"	2.53×10^6						
K	32°	63°	78°	"	1.15×10^6	G	36°	68°	85°	2×10^{-6}	5.50×10^6
R	33°	58°	70°	"	2.04×10^6	J	37°	54°	40°	"	10.60×10^6
						O	46°	49°	14°	"	20.90×10^6
						U	50°	41°	62°	"	6.30×10^6

ø Crystal M was tested at a total strain amplitude of 0.140%, all other crystals were tested at a total strain amplitude of 0.092%.

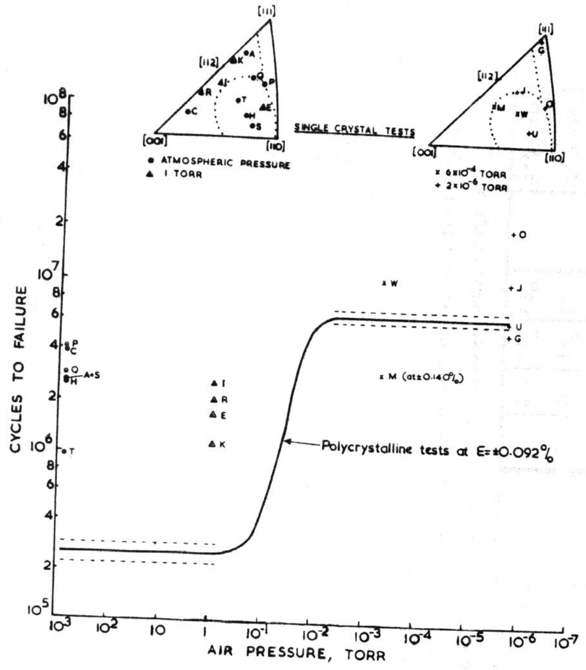


Fig. 1.

The influence of air pressure on the fatigue life of lead single crystals at a total strain amplitude of ±0.092%. Crystal M was tested at a total strain amplitude of 0.140%. The polycrystal results are indicated by the full curve; the dashed lines indicate ± the standard deviation.

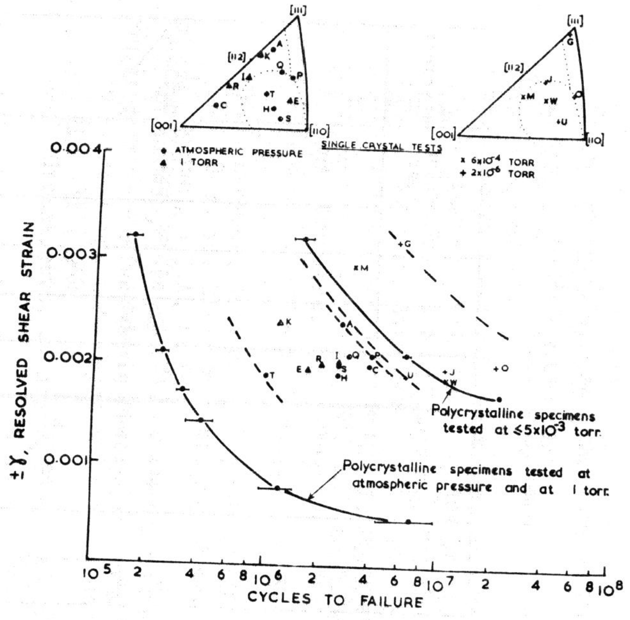
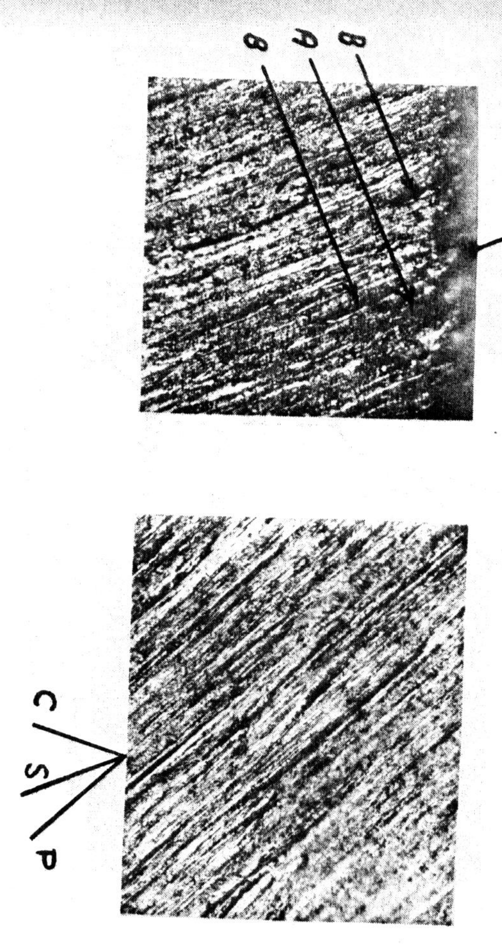


Fig. 2.

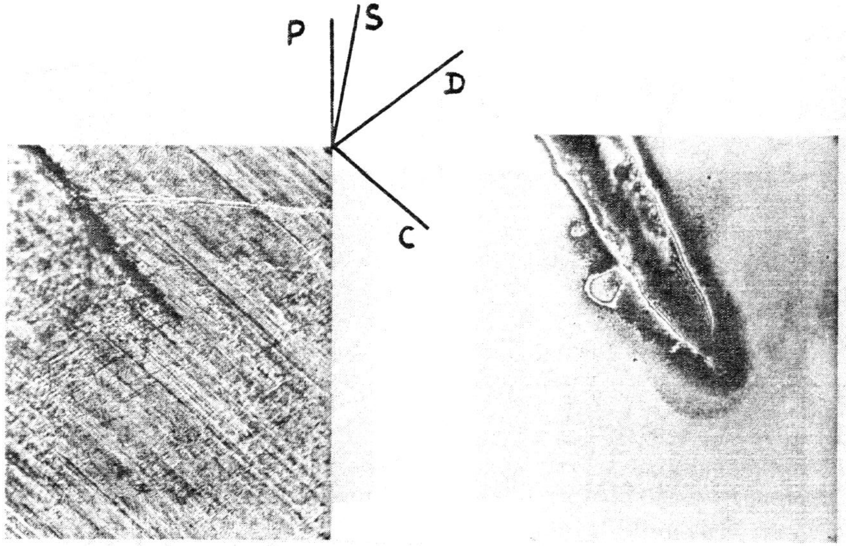
The effect of resolved shear strain amplitude on the fatigue life of lead single crystals in the high and low air pressure ranges. The full curves are for polycrystals tested at an equivalent shear strain amplitude.



(a) (b)

Fig 3.

Side and top surfaces of crystal O after 8 x 10^5 cycles. Specimen axis horizontal. (x 200)
P, S and C are the directions of the primary, conjugate and critical slip traces, respectively.



(a) *(b)*

Fig. 4.

The left hand side shows a propagating fatigue crack in crystal C at 3.6 x 10^6 cycles. Slip on the critical and cross systems in evidence (x 200). The right hand side shows the same area after a light polish and etch. (x 200)

C, D, P and S are the directions of the critical, cross, primary and conjugate slip traces, respectively.

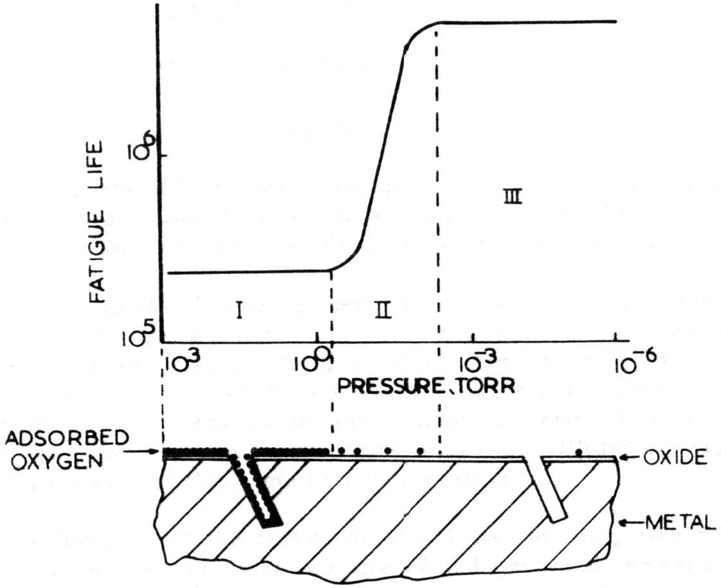


Fig. 5.

Diagram illustrates the variation in physically adsorbed oxygen with pressure.