

ON THE MECHANISM OF LOW TEMPERATURE INTERGRANULAR FRACTURE

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INTRODUCTION

Previous attempts to explain low temperature intergranular fracture have concentrated on cracking at grain boundary precipitates [1], or on a postulated decrease in the work of fracture proportional to the energy, γ , required to form free surfaces from grain boundaries [2 - 7]. The quantity γ is defined by $\gamma = 2\gamma_s - \gamma_b = (\partial F/\partial A)_{T,V}$; the subscripts s and b indicate surface and boundary respectively, F is the Helmholtz free energy, A the interfacial area, T the temperature and V the volume. This paper concerns the cases where no precipitation occurs and the segregant is present at grain boundaries as the equivalent of less than one monolayer. The grain boundary capacity for solute under equilibrium segregation conditions has been observed [8] to be at least several monolayers, whereas concentrations of the order 100 ppm of solute are necessary to produce an equivalent monolayer in a typical polycrystalline metal. Embrittlement is known to occur at much lower concentrations than this e.g. 5 ppm can cause brittleness in Cu-Bi e.g. [9]. The adsorption of solute is therefore dilute in at least some embrittled alloys so that the Gibbs adsorption isotherm applies. Under these conditions γ has been shown [10, 11] to be independent of solute segregation, and at higher levels up to 2/3 monolayers less than a 1% change in γ is found according to B.E.T. [12] and McLean isotherms [13]. It therefore follows that models of boundary embrittlement based on a presumed relation between γ and the actual energy dissipated in fracture, in a Charpy test for example, cannot be valid for low concentrations of impurities.

The purpose of this paper is to describe a new model in which the reduced energy of fracture and intergranular mode of fracture in low temperature embrittlement are related to the constraint of plastic flow at a notch or crack lying near a grain boundary. It is suggested that the major constraint to plastic flow ahead of the flaw is the propagation of slip across a grain boundary. Propagation mechanisms in pure materials are considered, an additional impedance introduced by impurity segregation is discussed subsequently.

PROPAGATION OF SLIP ACROSS GRAIN BOUNDARIES

The yielding of a polycrystal is envisaged as beginning in a favourably oriented grain. Slip is then propagated into neighbouring grains (a) by forcing dislocations through grain boundaries or (b) by operating lattice dislocation sources at grain boundaries or in neighbouring grains. Optical microscopy of dilute Fe-Si alloys [14] and transmission electron microscopy of nickel [15] support these views of the yielding process.

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The stress at the head of a pile up of dislocations leads to further yielding in a ductile metal and may lead to fracture in a material embrittled by segregation.

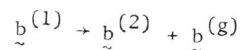
The transmission of dislocations through grain boundaries is considered according to the complexity of the geometry which is imposed by the crystallography of slip and the conservation of Burgers vector [16]. There are three distinct situations.

(a) Misorientation axis parallel to a crystal Burgers vector

In fcc metals for example dislocations with Burgers vectors parallel to a $\langle 110 \rangle$ axis of rotation are lattice vectors in both grains and can therefore move across a grain boundary without any dislocation reaction at the boundary. The simplest case is a $\langle 110 \rangle$ tilt boundary where transmission can occur by a process analogous to cross slip, Figure 1a. When the rotation axis and Burgers vector are not parallel to the boundary plane the dislocation line must rotate in the boundary plane into an orientation parallel to a glide plane of the second grain, Figure 1b; in general this process requires climb. Figure 2a shows some examples of the first type of process for a coherent twin boundary in a thin foil of high purity aluminum. Slip was caused by the stresses resulting from heating by the electron beam in the transmission electron microscope. Figure 2b shows similar observations by scanning electron microscopy of a lightly deformed Cu 6% Al alloy. No examples of the second type of transmission process have been observed so far possibly because crystal dislocations dissociate into grain boundary dislocations during any reorientation process and this prohibits transmission in the way envisaged above.

(b) Glide planes continuous across grain boundaries

For fcc metals there is always a continuous glide plane for crystal dislocations when there is a $\langle 111 \rangle$ rotation axis. However whilst no reorientation of a dislocation line is required a reaction of the following type must occur at the boundary.



where the superscripts (1), (2) and (g) refer to crystal (1), crystal (2) and the grain boundary respectively. For coincidence related boundaries $\underline{b}^{(g)}$ would be a DSC vector [17]. So far no clear observations of this process have been made. Figure 3 is an electron micrograph of a coincidence related boundary in high purity aluminum where a continuous glide plane exists (misorientation is $[111]/47^\circ$). The contrast behaviour of the features in the grain boundary was consistent with the DSC Burgers vector $a/38$ [10, 13, 15]. These boundary dislocations lie along the intersection of the continuous glide planes with the boundary and are close to edge character. These are the kind of dislocations which would be left in a boundary as a result of the transmission process envisaged above. The dislocations in the pile up shown in grain (1) are in the process of being incorporated into the boundary, but were not observed to be transmitted. The Burgers vector of these dislocations is $a/2 [110]$ and they are gliding in the (111) plane of crystal (1) which is not the continuous plane.

(c) No continuous glide planes or common Burgers vector

In general, dislocations which approach a grain boundary can be transmitted only by a combination of the reorientation and dislocation reaction mechanisms discussed above.

In fcc metals, except for the $\Sigma = 3 \{111\}$ coherent twin, a maximum of one $a/2 \langle 110 \rangle$ slip dislocation may be transmitted across a grain boundary without the need to generate a residual grain boundary dislocation. In general therefore the transmission of dislocations through grain boundaries requires an increase in the elastic energy of the system and/or climb. An additional, probably small, impediment to the transmission of slip dislocations can arise as a consequence of any relative translation of two neighbouring grains [18]. It is anticipated that the transmission process becomes more difficult as the density of dislocations or the solute concentration in the boundary increases.

FRACTURE

As discussed above, transmission of slip across grain boundaries generally requires production and motion (glide and/or climb) of grain boundary dislocations (gbds). Any impediment to gbd motion is therefore expected to inhibit plastic flow near grain boundaries. As a result of this restraint, we envisage a reduction in the extent of plastic deformation associated with an advancing intergranular crack, as depicted schematically in Figure 4, and a corresponding reduction in fracture toughness. By restricting the strain directly ahead of a crack in this way, less plastic work is involved in crack propagation and less crack tip blunting occurs. Since the exact plastic solution for such a case has not yet been obtained, we restrict our remarks to qualitative effects.

At low solute concentrations, most isotherms predict that $\Gamma_g \propto C$, where Γ_g and C are the interfacial and overall solute concentrations respectively. If we assume that impurity segregation to boundaries causes local solid solution hardening (i.e. impeding gbd motion) similarly to that in the grain interior, we expect the flow stress in the boundary region, σ_g , to be related to Γ_g by $\sigma_g \propto \Gamma_g^p$, where p takes values near $1/2$ (for grain interior solid solution hardening $1/2 \leq p \leq 1$). Since grain boundary distribution coefficients are very large generally, almost all of the solute in a dilute solution will reside on grain boundaries, and Γ will depend not only on C but on the grain diameter, d , $\Gamma_g \propto C d$. The boundary flow stress will then be given by $\sigma_g \propto \Gamma_g^{1/2} \propto (C d)^{1/2}$.

If we assume that the plastic work done during fracture, E_f , is inversely related to σ_g we obtain, $E_f \propto (C d)^{-1/2}$. This is in qualitative agreement with experimental results which show fracture toughness to be approximately proportional to $d^{-1/2}$ [19 - 21]. In addition, σ_g is expected to depend on the elastic misfit of impurity atoms analogously to lattice solid solution hardening. We therefore anticipate large reductions of E_f when atoms of large misfit are segregated to grain boundaries, in agreement with the discussion of Seah [11].

According to our model, E_f will decrease as the temperature falls for two reasons: 1) Γ_g increases, and 2) the diffusion rate of both solute and solvent atoms is decreased, thereby further slowing the gbd climb necessary for transmission of slip.

The occurrence of brittle intergranular fracture may accordingly be related to the capacity of the grain boundary region to relax stress concentrations. Large reductions in fracture toughness in equilibrium segregated alloys are expected to occur only in cases where sufficient impediment to gbd motion occurs so as to dictate an intergranular fracture path or to make crack nucleation at grain boundaries much more likely. Some of the parameters expected to be important in this model are as follows:

- 1) the degree of equilibrium segregation to grain boundaries which depends on distribution coefficients, impurity content, temperature and grain size,
- 2) the degree of elastic misfit of solute at the grain boundary and, therefore, the degree of elastic dislocation - solute interaction,
- 3) impediment to grain boundary dislocation climb, related to the diffusion rate of solutes and solvent in the grain boundary,
- 4) details of grain boundary crystallography which determine the dislocation reactions required for propagation of slip.

A model of low temperature embrittlement based on a position-dependent K_{IC} has been introduced. Sharp cracks are envisaged to nucleate and grow near grain boundaries with a high solute concentration at external stresses lower than those required for nucleation and growth of cracks within grains.

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REFERENCES

1. SMITH, E., Proc. Conference Physical Basis of Yield and Fracture, 36, Inst. Phys., Phys. Soc., Oxford, 1966.
2. MORTIMER, D. A., Fourth Bolton Landing Conference on Grain Boundaries in Engineering Materials, 1974.
3. RAMASUBRAMANIAN, P. V. and STEIN, D. F., Met. Trans. 2, 1971, 809.
4. JOSHI, A. and STEIN, D. F., ASTM-STP 499, 1972, 59.
5. CAPUS, J. M., ASTM-STP 407, 1968, 3.
6. LOW, J. R., Trans. TMS-AIME 245, 1969, 2481.
7. HONDROS, E. D. and McLEAN, D., Phil. Mag. 29, 1974, 771.
8. SEAH, M. P. and HONDROS, E. D., Proc. Roy. Soc. (London) A335, 1973, 191.
9. JOSHI, A. and STEIN, D. F., J. Inst. Metals, 99, 1971, 178.
10. SEAH, M. P., Surf. Sci., 53, 1975, 168.
11. SEAH, M. P., Proc. Roy. Soc. (London) A349, 1976, 535.
12. BRUNAUER, S., EMMETT, P. H. and TELLER, E., J. Am. Chem. Soc., 62, 1940, 1723.
13. McLEAN, D., Grain Boundaries in Metals, Oxford University Press, London, 1957.
14. WORTHINGTON, P. J. and SMITH, E., Acta Metall., 12, 1964, 1277.
15. MALIS, T., LLOYD, D. J. and TANGRI, K., Phys. Stat. Sol.(a) 11, 1972, 275.
16. CHRISTIAN, J. W., J. de Phys. 35, 1974, C7-65.
17. BOLLMANN, W., "Crystal Defects and Crystalline Interfaces", Springer, Berlin, 1970.
18. POND, R. C. and SMITH, D. A., Can. Met. Quart., 13, 1974, 39.

19. LOW, J. R., Symposium on Relation of Properties to Microstructure, 163, ASM, 1954.
20. HORNBOKEN, E., Sonderdruck aus Zeitschrift Metallkunde, 66, 1975, 511.
21. BERNSTEIN, I. M. and RATH, B. B., Met. Trans., 4, 1973, 1545.

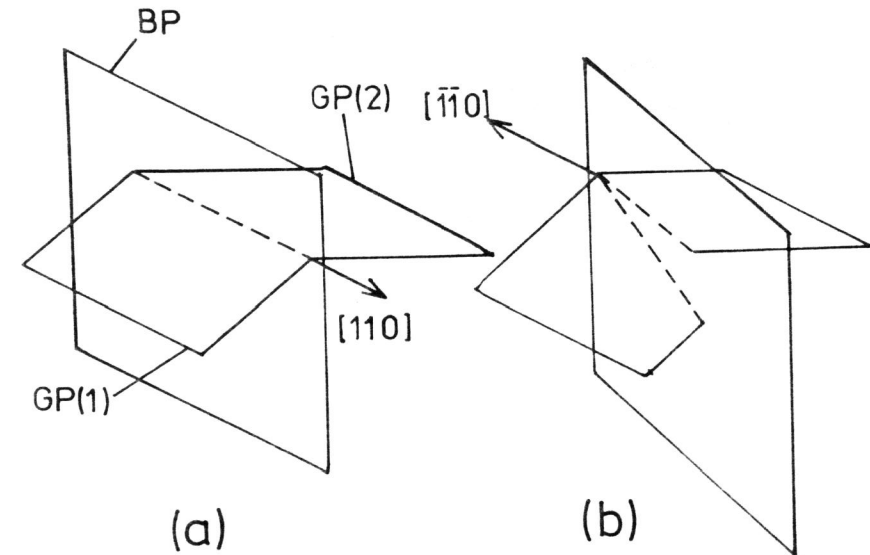
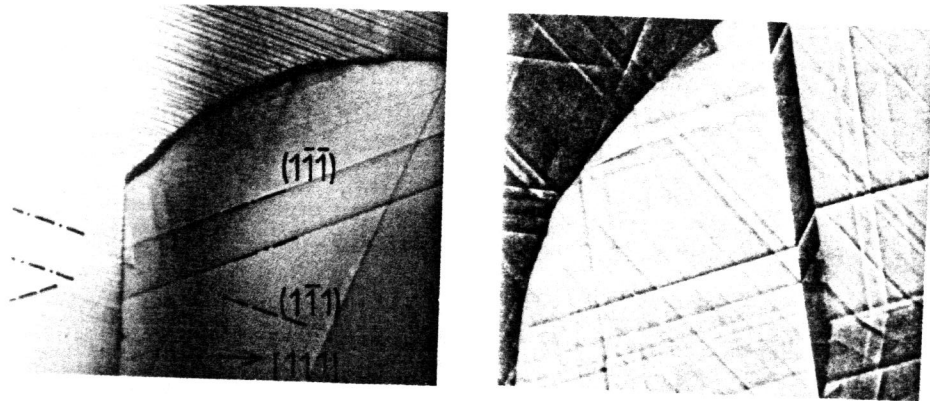


Figure 1 (a) Schematic Representation of Dislocation Transmission Through a Grain Boundary by 'Cross Slip' from a Glide Plane in One Crystal, GP (1), to a Glide Plane in the Other, GP (2). In this Case the Misorientation Axis is Contained in the Boundary Plane (BP) and has the Form $\langle 100 \rangle$ in f.c.c. Material
(b) As for (a) but when the Misorientation Axis Does Not Lie in the Boundary Plane



(a)

(b)

Figure 2 (a) Electron Micrograph of Slip Traces of Dislocations which have Penetrated a Coherent Twin Boundary, (111) , in Pure Aluminum. The Electron Beam Direction was $[\bar{1}01]$ so that the (111) Boundary and $(\bar{1}\bar{1}1)$ Glide Plane Appear Edge On
 (b) Transmission of Slip Through a Coherent Twin Boundary in Cu 6% Al. (Courtesy JEOL and T. Yamamoto)

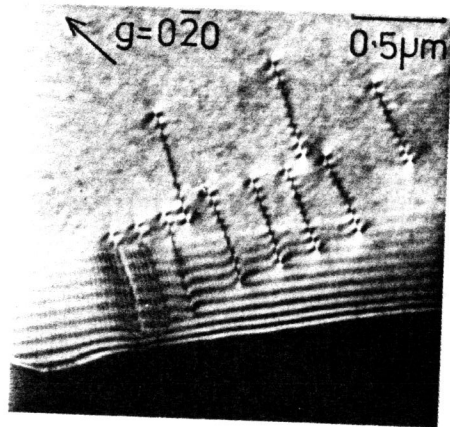
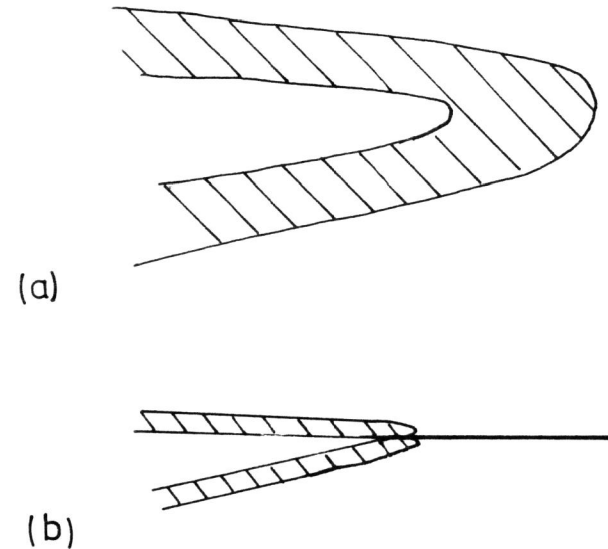


Figure 3 Electron Micrograph of a Pile-Up of Lattice Dislocations Entering a Boundary in Aluminum. The Two Grains are Misoriented About a $[111]$ Axis



(a)

(b)

Figure 4 (a) Schematic Diagram of Plastically Deformed Region (Shown Hatched) Developed During Propagation of a Blunted Crack
 (b) Plastically Deformed Region Developed During Propagation of a Crack with Limited Strain at the Crack Tip