

Mechanisms of Stress Relief in Polycrystalline Films

Abstract: The stress required to operate dislocation sources within a grain, at a grain boundary, and at surfaces is found to be larger than the intrinsic stresses observed in polycrystalline films. It is therefore unlikely that a dislocation flow mechanism can relieve stresses in films. Grain boundary sliding and diffusional creep can, however, relieve stresses in films and equations describing the kinetics of stress relaxation are derived. It is suggested that stress relief occurs primarily by a diffusion-creep mechanism. Growth of hillocks during annealing of a film is briefly discussed in terms of the diffusion-creep mechanism.

Introduction

It is observed that thin films prepared by evaporation onto a substrate contain stresses.^{1,2} The stresses are considered to originate from defects generated during growth of a film (growth stresses), from mismatch at the interface between substrate and film, and from differential thermal expansion.² This paper considers some mechanisms of plastic flow by which the elastic strains can be relieved. It confines itself primarily to films in which the grain size is very small and the thickness of the film is much greater than the grain size. For the purpose of discussion we divide this paper into four parts: analysis of stress in film and substrate; dislocation mechanisms of flow; grain boundary sliding; and diffusional creep.

Stress analysis

In the following it is assumed that: (a) the elastic constants of film and substrate are the same; (b) isotropic elasticity theory is valid; and (c) the film and substrate are bonded at their common interface. The film and substrate are shown schematically in Fig. 1. The film lies in the x_1, x_2 plane. The origin is chosen at the surface of the film and the thickness of the film measured in the x_3 direction is $2C$. The thickness of the film plus that of the substrate is $2D$.

The stresses in the film and substrate can be obtained by replacing the discrete positions of defects by a continuous distribution of infinitesimal defects. The stress distribution can then be calculated in a manner analogous to

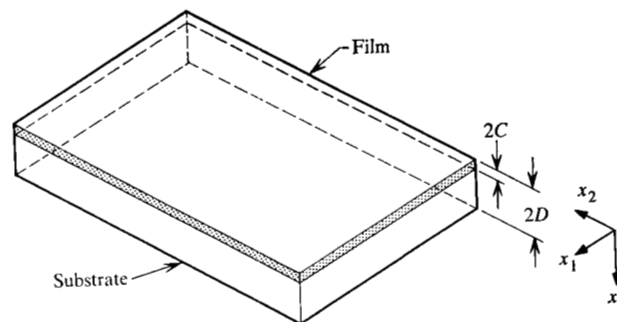


Figure 1 The geometry of a film and substrate.

that for thermal stresses.³ This procedure gives the stress distribution in a film as

$$\sigma_{11} = \sigma_{22} = \frac{E}{1-\nu} \left[\epsilon_{11}(x_3) - \frac{1}{2D} \int_0^{2C} \epsilon_{11}(x_3) dx_3 + \frac{3(x_3 - D)}{2D^3} \int_0^{2C} (x_3 - D) \epsilon_{11}(x_3) dx_3 \right], \quad (1a)$$

and the stress distribution in the substrate is

$$\sigma_{11} = \sigma_{22} = -\frac{E}{2D(1-\nu)} \left[\int_0^{2C} \epsilon_{11}(x_3) dx_3 - \frac{3(x_3 - D)}{D^2} \int_0^{2C} (x_3 - D) \epsilon_{11}(x_3) dx_3 \right], \quad (1b)$$

where $\epsilon_{11}(=\epsilon_{22})$ is the normal elastic strain associated with the defects (or differential thermal expansion) in a film; $\epsilon_{11}(x_3)$ is a function that describes the variation of

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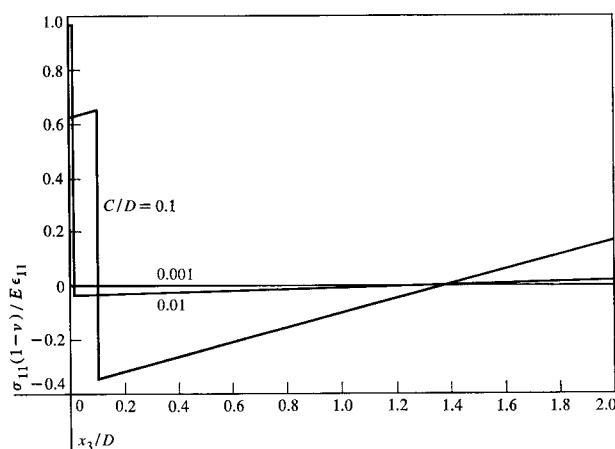


Figure 2 Stress distribution in film and substrate for several values of the ratio of film thickness to substrate thickness.

strain components ϵ_{11} or ϵ_{22} as a function of x_3 ; $\sigma_{11}(=\sigma_{22})$ is the normal stress; $\sigma_{33} = 0$; ν is the Poisson's ratio and E the Young's modulus. Equation (1) accurately describes the stress distribution far away from the ends of the film and substrate edges.

In the simple case for which the strain is uniform and independent of x_3 , $\epsilon_{11}(x_3) = \epsilon_{11}$ and Eq. (1a) gives the stress in a film as

$$\sigma_{11} = \sigma_{22} = \frac{\epsilon_{11}E}{1-\nu} \left\{ 1 - \frac{C}{D} \times \left[1 + \frac{3(C-D)(x_3-D)}{D^2} \right] \right\}, \quad (2a)$$

and Eq. (1b) the stress in the substrate as

$$\sigma_{11} = \sigma_{22} = -\frac{\epsilon_{11}EC}{(1-\nu)D} \left[1 + \frac{3(C-D)(x_3-D)}{D^2} \right]. \quad (2b)$$

The variation of σ_{11} (or σ_{22}) with x_3 for several values of the ratio C/D is shown in Fig. 2. For $C \ll D$ the components of the stress tensor in the film corresponding to a planar stress have the following values:

$$\begin{aligned} \sigma_{11} &= \sigma_{22} \cong \frac{\epsilon_{11}E}{1-\nu} \\ \sigma_{33} &\cong 0, \\ \sigma_{12} &= \sigma_{13} = \sigma_{23} = 0. \end{aligned}$$

The values of the normal stresses in a film where $C \ll D$ obtained in the present analysis differ from those quoted frequently in the literature by the factor $(1-\nu)$ in the denominator. This factor arises from the presence of a biaxial system of stress in the film, rather than from the uniaxial stress usually assumed.

In the following development the driving force for plastic flow is provided by the shear component of stress. We shall, however, present our final results in terms of normal components of stress to facilitate comparison with experimental information. Similarly, the components of strain contained in the equations are transformed from shear to normal strains. The transformations are carried out by the second-rank-tensor transformation law; for example, the shear stress σ on a plane inclined at an angle α to the normal of the plane of the film is given by $\sigma = \sigma_{11} \sin \alpha \cos \alpha$. It is also convenient in the present analysis to relate stresses to strains in terms of the shear modulus μ rather than Young's modulus E using the relation $E = 2\mu(1+\nu)$.

Dislocations

The value of σ_{11} determined experimentally in films is found to depend on several factors such as the temperature of deposition, rate of deposition, and the physical properties of the films. Values of σ_{11}/μ as high as 10^{-2} have been observed.² This value is to be compared with the critically resolved shear stress of most bulk single crystals which lies in the range of $(10^{-5} \text{ to } 10^{-4})\mu$. Therefore the question we wish to address in this section is: Why do films retain these high elastic strains in view of the relatively low value of the critically resolved shear stress in bulk materials?

Elastic strains in films can be relieved by flow either in the substrate or in the film. For the case of a uniform and constant strain in a film the stress in a substrate is given by Eq. (2b) and is shown in Fig. 2 for several values of the ratio C/D . We note that in most cases of deposition the condition $C \ll D$ exists, so that the stress in a substrate is a very small fraction of the stress in a film. For a film thickness of one micrometer and a substrate thickness of one millimeter, the maximum value of stress in the substrate is less than one-half percent of the stress in the film. A stress as large as $10^{-2}\mu$ in a film means that the maximum value of stress in the substrate is barely in the range of the critically resolved shear stress of soft, single-crystal substrates. For films where $C/D \leq 10^{-4}$ we may therefore neglect plastic flow by a dislocation mechanism in substrates. When the ratio C/D is larger than 10^{-4} , deformation of the substrate can occur. In these cases the plastic flow properties of the particular substrate have to be considered to determine the extent to which dislocations in it can relieve the elastic strains in film and substrate.

The inability of dislocations in films to relieve high elastic strains can be due to difficulties in generating dislocations or to the immobility of dislocations.⁴ In what follows it is shown that high elastic strains in films can be sustained (from the point of view of dislocation theory) because of the difficulty in operating dislocation sources. This difficulty is attributed to the fine grain size in films.

There are three places where dislocation sources can be located. These are: (a) within the volume of a grain; (b) at the grain boundary; and (c) at free surfaces. A dislocation source located within a grain generates a dislocation loop, the size of which is determined by the dimensions of the grain. Consider a cylindrically shaped grain of diameter d and height h (Fig. 3a) in which a dislocation loop has been generated. The plane of the loop is inclined at an angle α with respect to the free surface.

The theoretical normal stress $\hat{\sigma}$ required to generate a dislocation loop of the type shown in Fig. 3a can be calculated and compared with the experimentally determined value of σ_{11} . The theoretical value is obtained by equating the energy of a loop to the work done by the stress in generating the loop. The energy of an elliptical loop is given by⁵

$$E = \frac{\mu b^2 \varepsilon d}{2\pi(1-\nu) \cos \alpha} \left\{ \left[1 - \frac{\nu(1-\rho \cos^2 \alpha)}{\sin^2 \alpha} \right] \times \ln \left(\frac{4d}{eb \sqrt{\cos \alpha}} \right) + \frac{(1-\rho)(1-2\nu) \cot^2 \alpha}{2(1-\nu)} - \frac{\rho(1-\nu + \cos^2 \alpha)}{2} \right\}, \quad (3)$$

where $e = 2.71828 \dots$, $\rho = \mathcal{K}/\varepsilon$, and ε and $\mathcal{K}$ are complete elliptic integrals of the first and second kind, respectively, of modulus $\sin \alpha$, and b is the magnitude of the Burgers vector. The work done by the stress in generating the loop is

$$W = \frac{\pi}{4} \hat{\sigma} b d^2 \sin \alpha. \quad (4)$$

The value of $\hat{\sigma}$ is obtained from the condition $[\partial(E-W)/\partial d]_{\alpha} = 0$. The value of $\hat{\sigma}$ is shown in Fig. 4a as a function of d keeping α constant and in Fig. 4b as a function of α keeping d constant. The value of $\hat{\sigma}$ decreases with increasing grain size and, as shown in Fig. 4a, it is approximately proportional to the inverse of the grain diameter d . The relative minimum in the curve of $\hat{\sigma}$ as a function of α keeping d constant is attributed to the dependence of the shear stress in the slip-plane on the angle α and the dependence of the energy of an elliptical loop on its modulus.

Provided that $\sigma_{11} < \hat{\sigma}$, dislocation loops cannot be generated in the film because the work done by σ_{11} during plastic flow (expansion of the loop) is less than the energy expended in generating the loop. Experimentally one finds that values of σ_{11} as high as $10^{-2} \mu$ are associated with growth stresses in films that have a grain size of several hundred angstroms. For these values of grain diameter $\sigma_{11} < \hat{\sigma}$, so that dislocation sources within the volume of a grain do not play a significant role in relieving the observed elastic strains.

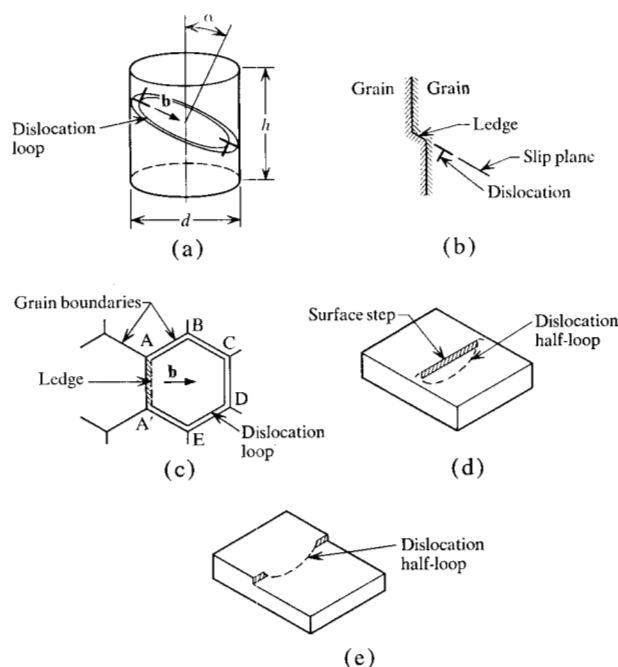


Figure 3 Dislocation sources in polycrystalline materials. (a) Source within a grain. (b) Grain boundary source associated with a ledge. (c) Dislocation loop within a grain operated from a grain boundary source. (d) Dislocation source at an atomically flat free surface. Dislocation half-loop moves into film-generating step (shaded area) on surface. (e) Dislocation source at a step on a free surface. Dislocation half-loop moves into film and removes step; remaining area (shaded) left on surface.

Dislocation sources at grain boundaries are present at ledges.⁶ A dislocation loop generated from a ledge is shown schematically in Figs. 3b, c. The energy E' required to generate a dislocation loop from a ledge is equal to the energy of an elliptical loop given by Eq. (3) minus a correction term. The latter is associated with the absence of the segment AA' and the change in grain boundary energy when a loop is generated. The self-energy of the segment AA' and its interaction with the rest of the loop can be calculated using Blin's formula⁷ for the interaction energy E_{ij} between two loops c_i and c_j :

$$E_{ij} = -\frac{\mu}{2\pi} \oint_{c_i} \oint_{c_j} \frac{(\mathbf{b}_i \times \mathbf{b}_j) \cdot (\mathbf{dl}_i \times \mathbf{dl}_j)}{R} + \frac{\mu}{4\pi} \oint_{c_i} \oint_{c_j} \frac{(\mathbf{b}_i \cdot \mathbf{dl}_j)(\mathbf{b}_j \cdot \mathbf{dl}_i)}{R} + \frac{\mu}{4\pi(1-\nu)} \oint_{c_i} \oint_{c_j} (\mathbf{b}_i \times \mathbf{dl}_i) \cdot \mathbf{T} \cdot (\mathbf{b}_j \times \mathbf{dl}_j), \quad (5)$$

where $\mathbf{b}_i$ and $\mathbf{dl}_i$ are the Burgers vector and an infinitesimal

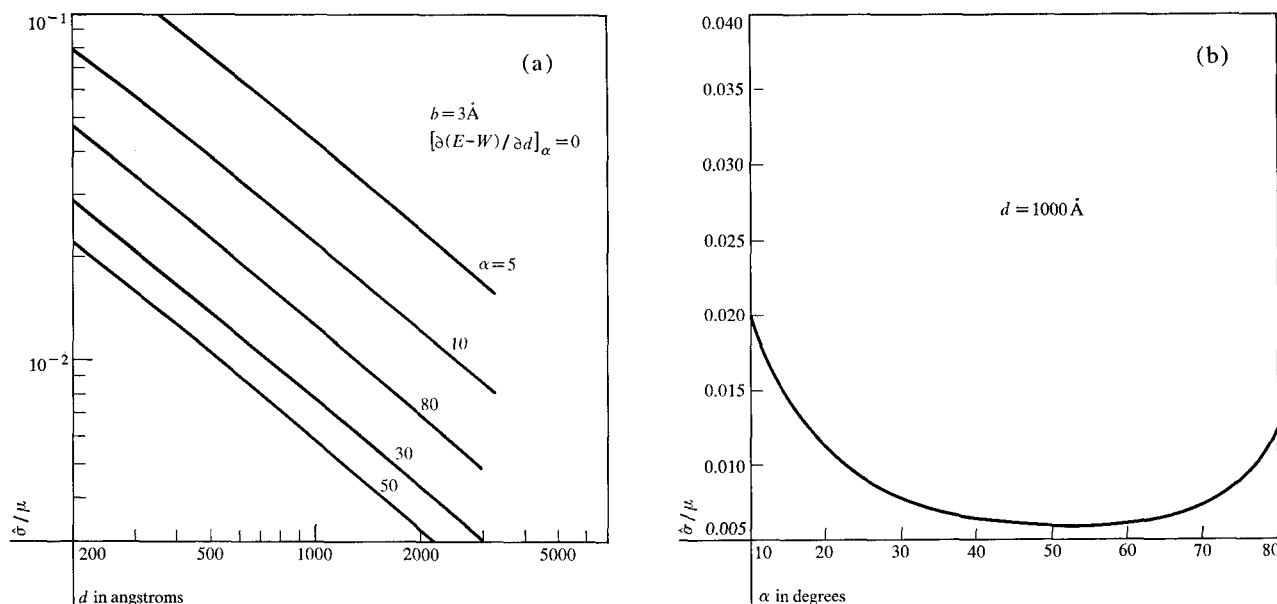


Figure 4 Stress divided by shear modulus required to operate dislocation source in grain. (a) Stress divided by shear modulus as a function of grain diameter keeping α constant. (b) Stress divided by shear modulus as a function of α keeping grain diameter constant.

line element of the loop c_i , respectively. In Eq. (5) T is a tensor with components

$$T_{\alpha\beta} = \delta^2 R \mid \delta X_{\alpha} \delta X_{\beta}; \quad (6)$$

here

$$R^2 = X_1^2 + X_2^2 + X_3^2, \quad (7)$$

$$X_1 = x_i - x_j, \quad X_2 = y_i - y_j, \quad X_3 = z_i - z_j,$$

and x_i, y_i, z_i are the Cartesian coordinates of the line element dl_i . The energy of the dislocation segment AA' according to Blin's formula is⁸

$$E_{ii} = \frac{\mu b^2 L}{(1-\nu)} \ln \left(\frac{4L}{eb} \right), \quad (8)$$

where L is the length of the segment. The interaction energy of the segment AA' with the rest of the loop can be calculated by making the simplification that the energy of a smooth loop is equal to that of a piecewise loop when their areas are equal.⁹ With this simplification the integrals involved in Blin's formula [Eq. (5)] can be evaluated in a straightforward manner. For our case the interactions between the segments AA' and AB , AA' and BC , AA' and CD , AA' and DE , and AA' and EA' were determined. If we write this interaction energy as E_{ij} , the energy of a loop generated from a ledge can be calculated using the following equation:

$$E' = E - (E_{ii} + E_{ij} + \gamma Lb), \quad (9)$$

where γ is the grain boundary ledge energy per unit area.

Using Eqs. (4) and (9), we calculated the value of the theoretical normal stress σ' required to operate a source for several values of γ as a function of d and α . This is shown in Figs. 5a, b. We note that, to a first approximation, σ' also varies inversely with the diameter of the grain and has a minimum as a function of α . The value of σ' is smaller than σ for a given value of d and α . Thus grain boundary sources can be operated at lower values of stress than those required for volume sources. A comparison of σ_{11} with σ' reveals, however, that the experimentally found values of growth stress cannot operate grain boundary sources in films because of their fine grain size.

We now consider the third possibility, which is surface sources. Dislocation sources at a good surface, that is, an atomically flat surface, require a value of stress to operate that is comparable to those for sources within a grain (volume sources). This is due to image terms and the extra energy required to create a step on the surface. The operation is shown schematically in Fig. 3d. For the special case of $\alpha = 90^\circ$ the energy of a half-loop generated at a surface is equal to that of a full loop within a grain.¹⁰ With the energy needed to generate a step on the surface, the theoretical stress required to operate a surface source is larger than the stress needed to generate a similar loop within a grain. For loops lying on planes with normals at an angle α less than 90° , the image terms lead to somewhat more complex expressions for the energy. An approximate analysis shows that the energy in these cases is still comparable to that for volume sources. A lower energy is required to operate a surface source where a

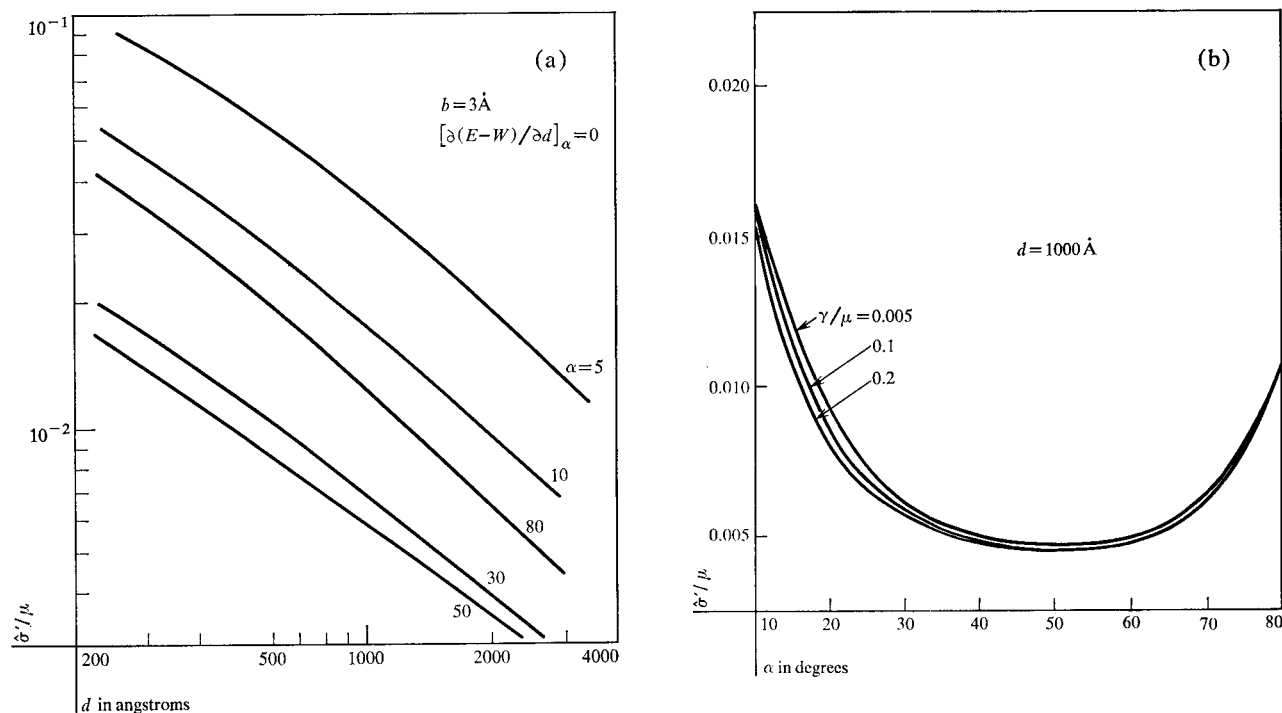


Figure 5 Stress divided by shear modulus required to operate dislocations at a grain boundary ledge. (a) Stress divided by shear modulus as a function of grain diameter keeping α constant with $\gamma/\mu = 0.1$. (b) Stress divided by shear modulus as a function of α keeping γ/μ constant with $d = 1000\text{Å}$.

step is already present. This is due to the reduction in free-surface areas and, through that, a reduction in surface energy when a dislocation half-loop is nucleated (Fig. 3e). Hirth¹⁰ has calculated the stress required to nucleate a dislocation half-loop at various types of surfaces and concludes that the stress required to do so is about one-third the value of the theoretical stress at room temperature. This value is lowered somewhat by the stress concentration at a step that acts like a notch.¹¹ Even if a loop could be nucleated at a surface, the size of such a loop is limited by the grain size. In polycrystalline films where the grain diameter d is much smaller than the film thickness, sources located at the surface can at most relieve stresses confined to a thin layer of the dimensions of a grain. We therefore conclude that the observed levels of stress can be maintained in films with a fine grain size because of the large values of stress required to operate dislocation sources.

Our calculations so far have not included the contribution from thermal fluctuations. Inclusion of thermal energy shows that for temperature T , where $T > 0$, the value of σ' is lowered. However, this decrease is not sufficient to change the conclusions reached earlier. Inclusion of temperature effects in the calculation of σ' shows that σ' is a function not only of temperature, but also of the rate of deposition. The limiting value of the observed stress at

$T = 0$ or at very high rates of deposition is σ' , provided dislocation mobility is high and other physical characteristics of the film, such as grain size, remain constant.¹² Although the effect of temperature is to reduce the critical stress for generation of a source, the presence of obstacles to the motion of dislocations tends to increase the critical stress. In most thin films, point defects in excess of the equilibrium concentration are probably present so that the critical stress to generate a dislocation may be larger than those shown in Figs. 4 and 5.

The present calculations show that the critical stress to operate dislocation sources is larger than the observed values. They do not show what the maximum value of elastic strain in a film may have been. For, clearly, had the stress in a film been larger than the critical stress to operate a source, dislocation motion would have led to plastic flow and subsequent reduction in elastic strain until the stresses were nearly equal to those for generation of sources. Any dislocation source mechanism that requires a critical stress to operate cannot relax the stresses in a film beyond approximately the critical stress. It follows then that dislocation mechanisms such as those considered here cannot entirely relieve the elastic strains in films. We therefore consider next two mechanisms of plastic flow that do not require dislocation motion and may relax the strains in a film completely.

Grain boundary sliding

The elastic strains in films which give rise to stresses are less than one percent. It is therefore conceivable that grain boundary sliding without appreciable flow of bulk material could relieve the elastic strain. The rate of deformation or, alternatively, the relaxation of the elastic strains can be derived from the grain boundary sliding formulation considered by Nabarro.¹³ We have

$$\frac{d\epsilon_{11}^p}{dt} = \frac{D_b \sigma_{11} b^3}{dkT} \sin \alpha \cos^2 \alpha$$

when $\sigma_{11} b^3 \sin \alpha \cos \alpha < kT$, (10)

where $D_b = D_{0b} \exp(-U_b/kT)$ is the boundary diffusion coefficient, U_b the activation energy for grain boundary diffusion, k the Boltzmann constant, and T the temperature. The value of $\sigma_{11} \sin \alpha \cos \alpha$ is expected to be small because most of the grain boundaries in a film are nearly perpendicular to the free surface of the film. If all grain boundaries were exactly perpendicular to the free surface, the plastic strain contribution from grain boundary sliding would be zero because, under biaxial tension or compression, no shear stress acts on these boundaries.

The rate of annealing is given by

$$\frac{d\sigma_{11}}{dt} = -\frac{D_b E \sigma_{11} b^2}{(1-\nu) dkT} \sin \alpha \cos^2 \alpha \quad (11a)$$

or

$$\sigma_{11} = \sigma_{11}^0 \exp \left[-\frac{D_b E b t \sin \alpha \cos^2 \alpha}{(1-\nu) dkT} \right], \quad (11b)$$

where σ_{11}^0 is the initial value of the elastic stress and t the time. We note that the annealing rate is a function of the projection of the grain diameter on the film surface and is independent of the height of the grain.

Diffusional creep

Diffusional creep or Nabarro-Herring creep occurs by the movement of point defects (usually vacancies) under a concentration gradient generated by the applied stress.¹³⁻¹⁶

In thin films under biaxial stress this concentration gradient is present between grain boundaries parallel and perpendicular to the free surface. The analysis of diffusional creep in thin films under uniaxial stress has been carried out¹⁷; we therefore consider only the annealing kinetics and make comparison with grain boundary sliding. The plastic strain rate is given by

$$\frac{d\epsilon_{11}^p}{dt} = \frac{B D_v}{dh} \left[\exp \left(\frac{\sigma_{11} b^3}{kT} \right) - 1 \right] \quad (12a)$$

or

$$\frac{d\epsilon_{11}^p}{dt} = \frac{B' D_b b}{dh^2} \left[\exp \left(\frac{\sigma_{11} b^3}{kT} \right) - 1 \right], \quad (12b)$$

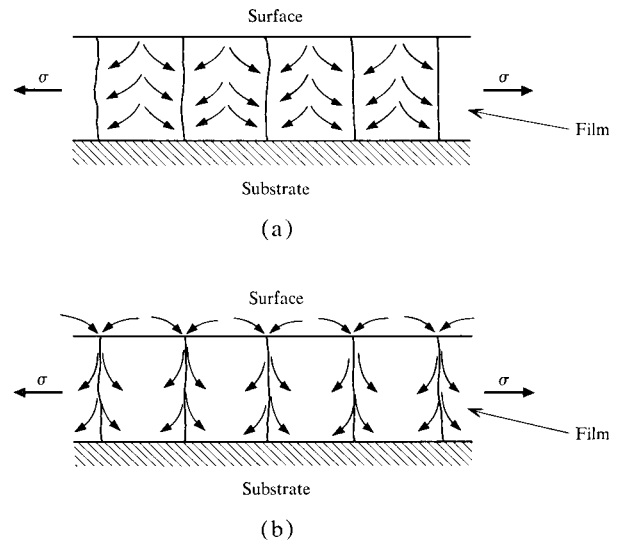


Figure 6 Schematic representation of diffusion currents in a film. (a) Volume diffusion is rate-controlling. (b) Grain boundary diffusion is rate-controlling.

where D_v is the volume diffusion coefficient, B and B' are constants which can be determined,¹⁷ and h is the height of a grain. The annealing rate is

$$\frac{d\sigma_{11}}{dt} = -\frac{E B D_v}{(1-\nu) dh} \left[\exp \left(\frac{\sigma_{11} b^3}{kT} \right) - 1 \right] \quad (13a)$$

if volume diffusion predominates, and is

$$\frac{d\sigma_{11}}{dt} = -\frac{E B' D_b b}{(1-\nu) dh^2} \left[\exp \left(\frac{\sigma_{11} b^3}{kT} \right) - 1 \right] \quad (13b)$$

if grain boundary diffusion is dominant. The stress as a function of time for the case of volume diffusion is given by

$$\begin{aligned} & \left[1 - \exp \left(-\frac{\sigma_{11} b^3}{kT} \right) \right] \exp \left[\frac{b^3 (\sigma_{11}^0 - \sigma_{11})}{kT} \right] \\ &= \left[1 - \exp \left(-\frac{\sigma_{11}^0 b^3}{kT} \right) \right] \exp \left[-\frac{E B b^3 D_v t}{(1-\nu) dh kT} \right] \end{aligned} \quad (14a)$$

and for grain boundary diffusion is

$$\begin{aligned} & \left[1 - \exp \left(-\frac{\sigma_{11} b^3}{kT} \right) \right] \exp \left[\frac{b^3 (\sigma_{11}^0 - \sigma_{11})}{kT} \right] \\ &= \left[1 - \exp \left(-\frac{\sigma_{11}^0 b^3}{kT} \right) \right] \exp \left[-\frac{E B' b^4 D_b t}{(1-\nu) dh^2 kT} \right]. \end{aligned} \quad (14b)$$

When $\sigma_{11}^0 b^3 kT \ll 1$, Eqs. (14a) and (14b) reduce to the form (11b). Compared with Eq. (11b), the difference in the annealing kinetics between grain boundary sliding and diffusional creep by grain boundary diffusion is the de-

pendence of the latter on the height of a grain. The two forms of diffusional creep can be distinguished not only by their dependence on grain height, but also by their temperature dependence through the activation energies associated with the diffusion coefficients.

We have outlined above the kinetics of annealing by a diffusion-creep mechanism in a polycrystalline film containing elastic strains. In doing this it was implicitly assumed that, on the average, all areas of the free surface of a film are equally efficient sources and sinks of vacancies. If this property of the film is eliminated, then a diffusion-creep mechanism can lead to the formation of hillocks (or growths) on a film surface.

Figures 6a and 6b show schematically the diffusion currents that occur by a diffusion-creep mechanism when volume diffusion and grain boundary diffusion are dominant, respectively. In both cases we note that material transfer occurs between the free surface and the interior of the film. For a tensile stress in a film, material is transferred from the free surface into the interior and for a compressive stress it is transferred out of the interior of a film onto its surface. Both Figs. 6a and 6b correspond to the case in which all areas of the free surface are, on the average, equally efficient sources and sinks. However, unusual behavior results from removing this condition and allowing selected areas to be more efficient than the remaining areas. Physically the selected areas may correspond to holes or cracks in an oxide layer (or film with low diffusion rates) on a film in which diffusional creep is occurring. Another possibility is selected diffusion paths (where diffusion rates are high compared with the rest of the film) combined with low surface diffusion or high surface energy that is strongly orientation dependent.

Consider the case of one film covered with a second film that has a hole or crack in it (Fig. 7a). Under biaxial stress, caused by growth defects or by differential thermal expansion, a film relieves its elastic strains by flow of matter between surface and interior of the film. In the case of compressive stress in a film, matter is transferred onto the free surface and the area of film exposed at the crack can accommodate this "extruded" material. The remaining area covered by the second film (see Fig. 7b) cannot do this at a rate comparable with that in the exposed area because of slower diffusion. Material extruded at the crack or hole manifests itself as a "hillock" (or growth). It is interesting to note that for a tensile stress the present model predicts removal of matter from the surface into the interior of the film and therefore the formation of a depression at the exposed area (Fig. 7c). Although hillocks have been observed in films^{18,19} the presence of depressions has not been reported.

The idea that compressive stresses in films may lead to growth on film surfaces has been used by Pennebaker¹⁹ to explain his observations on gold. A major difference

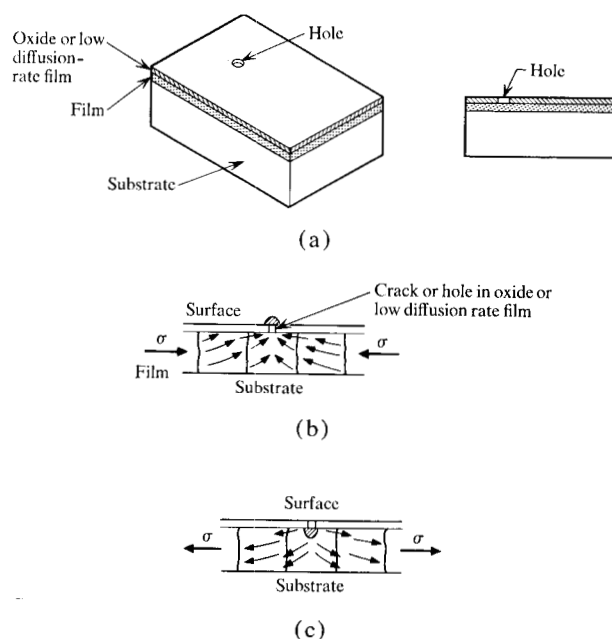


Figure 7 Film on a substrate covered by another film that has lower diffusion rates. (a) Hole or crack in a local spot in the covering film. (b) Mass flows out of the film under a compressive stress and is deposited on the surface of covering film. (c) Mass flows into the film under a tensile stress leaving a crater or depression near the hole or crack.

between the present model and that proposed by Pennebaker¹⁹ lies in the manner in which the driving force provided by the stress leads to mass flow.

Our analysis so far has assumed that the stresses in a film are uniform. A non-uniform stress distribution affects the annealing kinetics outlined above. We consider diffusion by a vacancy mechanism as an example. The elastic strain field of a vacancy, approximated by a center of relaxation, interacts with the hydrostatic component of stress. If the interaction energy varies with the location, the vacancy experiences a drift velocity given by the Einstein relation $v = -(D/kT) \text{grad } E_{kl}$, where E_{kl} is the interaction energy. In thin films on a substrate, a gradient of the interaction energy is present even though the strain caused by the defect may be uniform [see Eq. (1a) or (2a)] and therefore a drift force on vacancies exists in the x_3 direction. This drift force affects the expressions for annealing kinetics outlined in this section. It also provides for a mechanism of very limited hillock growth on a single-crystal film. The vacancy migration results not from a concentration difference, as in Nabarro-Herring creep, but from the stress gradient.

Summary

Calculations of the stress required to operate sources of dislocations within a grain, at grain boundaries, and at a free surface show that in fine-grained material the stress

to operate a source is high. This stress is approximately proportional to the inverse of the grain diameter. Films that contain high growth stresses are associated with very fine grain size and in these films the observed growth stresses are less than the stress to operate a dislocation source. Dislocation mechanisms therefore cannot be expected to relieve the observed growth stresses. At temperatures where diffusion rates are appreciable, two other mechanisms may be able to relieve the stresses in films. One of these is grain boundary sliding. This mechanism, while possible, is probably slower than the second, a diffusion-creep mechanism. It is suggested that diffusional creep in fine-grained films provides stress relief. The growth of hillocks on the free surface of a film is also discussed in terms of a diffusion-creep mechanism.

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References

1. R. W. Hoffman, *Thin Films*, H. G. F. Wilsdorf, Ed., American Society for Metals, Cleveland, p. 99 (1964); *Physics of Thin Films*, Volume 3, G. Hass and R. E. Thun, Eds., Academic Press, New York, p. 211 (1965).
2. E. Klokholm and B. S. Berry, "Intrinsic Stress in Evaporated Metal Films," *Research Report RC 1969*, IBM Watson Research Center, Yorktown Heights, N.Y., 1968.
3. S. Timoshenko and J. M. Goodier, *Theory of Elasticity*, McGraw-Hill Book Co., Inc., New York, p. 399 (1951).
4. J. W. Menter and D. W. Pashley, *Structure and Properties of Thin Films*, C. A. Neugebauer, J. B. Newkirk, and D. A. Vermilyea, Eds., John Wiley and Sons, Inc., New York, p. 111 (1959).
5. J. C. M. Li and G. C. T. Liu, "Energy of Elliptical Dislocation Loops," *Phil. Mag.* **14**, 413 (1966).
6. J. C. M. Li, "Petch Relation and Grain Boundary Sources," *Trans. AIME* **227**, 239 (1963).
7. J. Blin, "Energie Mutuelle de Deux Dislocations," *Acta Met.* **3**, 199 (1955).
8. T. Jøssang, J. Lothe, and K. Skylstad, "Explicit Expressions for the Energy of Dislocation Configuration Made Up of Piecewise Straight Segments," *Acta Met.* **13**, 271 (1965).
9. J. P. Hirth and T. Jøssang, "Dislocation Energies and the Concept of Line Tension," *J. Appl. Phys.* **37**, 110 (1966).
10. J. P. Hirth, *Relation between Structure and Strength in Metals and Alloys*, H. M. Stationery Office, London, p. 218 (1963).
11. J. Friedel, *Electron Microscopy and Strength of Crystals*, G. Thomas and J. Washburn, Eds., Interscience Publishers, New York, p. 605 (1963).
12. P. Chaudhari, to be published.
13. F. R. N. Nabarro, *Proceedings of the Conference on the Strengths of Solids*, Physical Society (London), p. 75 (1948).
14. C. Herring, "Diffusional Viscosity of a Polycrystalline Solid," *J. Appl. Phys.* **21**, 437 (1950).
15. R. L. Coble, "A Model for Boundary Diffusion Controlled Creep in Polycrystalline Materials," *J. Appl. Phys.* **34**, 1679 (1963).
16. I. M. Lifshitz, "On the Theory of Diffusion-Viscous Flow of Polycrystalline Bodies," *Soviet Phys.—JETP* **17**, 909 (1963).
17. G. B. Gibbs, "Diffusion Creep of a Thin Foil," *Phil. Mag.* **13**, 589 (1966).
18. F. d'Heurle, L. Berenbaum, and R. Rosenberg, "On the Structure of Aluminum Films," *Trans. AIME* **242**, 502 (1968).
19. W. B. Pennebaker, "Recrystallization, Hillock Growth, and Stress Relief in Sputtered Gold Films," *Research Report RC 2105*, IBM Watson Research Center, Yorktown Heights, N.Y., 1968.

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