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Paul E. Murray

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Effect of an eigenstrain on slow viscous flow of compressible fluid films

Paul E. Murray

Idaho National Engineering Laboratory, EG&G Idaho Inc., Idaho Falls, Idaho 83415-2414

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We present a general formulation of the mechanics of slow viscous flow of slightly compressible fluid films in the presence of an eigenstrain. An eigenstrain represents a constrained volume change due to temperature, concentration of a dissolved species, or a chemical transformation. A silicon dioxide film grown on a silicon surface is an example of a viscous fluid film that is affected by a constrained volume change. We obtain a general expression for pressure in a fluid film produced by a surface chemical reaction accompanied by a volume change. This result is used to study the effect of an eigenstrain on viscous stress relaxation in fluid films.

Viscous stress in fluid films is affected by the presence of an eigenstrain. A strain associated with a constrained volume change in a material that is free from external forces is called an eigenstrain.¹ For example, thermal stress occurs as a result of a temperature gradient in the fluid. In this case, the eigenstrain is αT , where α is the coefficient of thermal expansion and T is the temperature. Another eigenstrain is βc , where β is the activation volume and c is the concentration of a dissolved chemical species. Moreover, an eigenstrain may be used to represent a constrained volume change that accompanies a chemical reaction or phase transformation.

An amorphous oxide film is a slightly compressible, viscous film that is affected by an eigenstrain.² For example, a residual compressive stress occurs in a silicon dioxide film grown on a silicon substrate. Although residual thermal stress occurs during cooling of the film, an initial residual stress occurs at the growth temperature. This initial residual stress is a result of the volume change that accompanies the oxidation reaction. The oxide film flows to accommodate the volume change. But because the oxide film is constrained by adhesion to the substrate, a residual stress occurs.

The eigenstrain that occurs in oxide films during growth is not known. As discussed previously, residual stress had been attributed to the volume change that accompanies the oxidation reaction. The volume change is defined as $\gamma = 1 - v_s/v_f$, where v_s is the specific volume of the solid substrate and v_f is the specific volume of the fluid film. In the case of silicon oxidation, $\gamma = 5/9$. But this is obviously not the eigenstrain! In fact, the residual stress at the growth temperature has been measured, and it is approximately equal to 400 MPa which is associated with a strain of approximately 0.01.³

Measurements of residual strain in oxide films show the following trend. At a low growth temperature ($T < 1000^\circ\text{C}$), the residual strain is high. At a high growth temperature ($T > 1000^\circ\text{C}$), the residual strain is low. In order to explain the dependence of residual strain on the growth temperature, several studies had suggested that residual strain relaxes by viscous flow of oxide.^{4,5} However, the process of viscous stress relaxation in the presence of an eigenstrain has not been examined.

In this letter, we present a general formulation of the mechanics of slow viscous flow of slightly compressible

fluid films. The formulation includes an eigenstrain to represent a constrained volume change due to temperature, concentration of a dissolved species, or a chemical transformation. We consider a viscous fluid film produced by a chemical reaction at a solid surface and accompanied by a constrained volume change. The equations of motion are solved to obtain a general expression for the pressure in the fluid film. The results are used to study the effect of an eigenstrain on viscous stress relaxation in fluid films.

The equations of motion for a compressible, viscous, Newtonian fluid are given in tensor notation by

$$\rho \frac{\partial u_j}{\partial t} + \rho u_k \frac{\partial u_j}{\partial x_k} = -\frac{\partial p}{\partial x_j} + \frac{\partial}{\partial x_i} \mu \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right) + \frac{\partial}{\partial x_j} \lambda \frac{\partial u_k}{\partial x_k} + f_j, \quad (1)$$

$$\frac{\partial \rho}{\partial t} + \frac{\partial}{\partial x_k} (\rho u_k) = 0, \quad (2)$$

where x_i ($i=1,2,3$) are the coordinate directions, t is time, u_i ($i=1,2,3$) are the velocity components, ρ is the density, p is the pressure, f_i ($i=1,2,3$) are the body force components, and μ and λ are the first and second coefficients of viscosity.⁶ The equations of motion are supplemented by an equation of state $\rho = \rho(p, T, c)$ where T is temperature and c is concentration of a dissolved chemical species.

We apply the equations of motion to study the mechanics of slow viscous flow of a slightly compressible fluid film on a solid surface. The geometry and coordinate system used for this problem are shown in Fig. 1. We assume that fluid is produced by a surface reaction accompanied by a volume change. The fluid flows to accommodate the

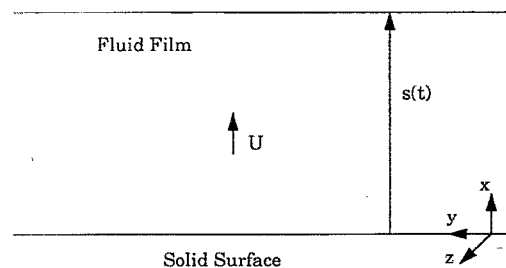


FIG. 1. Geometry for a viscous fluid film grown on a solid substrate.

volume change and the fluid velocity components are $u_x = U$, $u_y = 0$, $u_z = 0$. The time dependent film thickness is $s(t)$ and $U = ds/dt$ is the film growth rate.

We make the following assumptions: (a) the time scale of fluid flow is much larger than the time scale of reaction, and therefore the flow process is quasistatic; (b) the fluid is very viscous, and therefore inertia forces are negligible; and (c) Stoke's hypothesis, $\lambda = -(2/3)\mu$. Using these assumptions, the equations of motion reduce to

$$\frac{\partial \sigma_x}{\partial x} = 0, \quad (3)$$

$$\frac{\partial}{\partial x} (\rho u_x) = 0, \quad (4)$$

for $0 < x < s(t)$, where σ_x is the x component of normal stress given by

$$\sigma_x = -p + \frac{4}{3}\mu \frac{\partial u_x}{\partial x}. \quad (5)$$

Furthermore, we assume that the fluid is slightly compressible and introduce the eigenstrain ϵ^* into the equation of state

$$\rho = \rho_0 / (1 - \kappa p + \epsilon^*), \quad (6)$$

where κ is the isothermal compressibility and $\rho_0 = \rho(p=0, \epsilon^*=0)$.

Integrating Eq. (4) and applying the boundary conditions $u_x = U$ and $p = p_0$ at $x=0$, we obtain

$$u_x = U(1 - \kappa p + \epsilon^*). \quad (7)$$

Integrating Eq. (3) and applying the boundary condition $\sigma_x = 0$ at $x = s(t)$, we obtain

$$-p + \frac{4}{3}\mu \frac{\partial u_x}{\partial x} = 0. \quad (8)$$

Using Eq. (7) in Eq. (8), we obtain an equation for the pressure

$$p + \tau U \frac{\partial p}{\partial x} = \frac{\tau U}{\kappa} \frac{\partial}{\partial x} \epsilon^*, \quad (9)$$

where $\tau = (4/3)\mu\kappa$ is the time scale of compressibility.

We integrate Eq. (9) and apply the boundary condition $p = -\epsilon^*/\kappa$ at $x=0$ to obtain a general expression for pressure in the fluid film

$$p(x) = \frac{1}{\kappa} \left[\epsilon^*(x) - 2\epsilon^*(0)e^{-x/\tau U} - \frac{1}{\tau U} e^{-x/\tau U} \int_0^x \epsilon^*(\xi) e^{\xi/\tau U} d\xi \right]. \quad (10)$$

We use Eq. (10) to study the pressure in fluid films in the presence of an eigenstrain. As a special case, consider the eigenstrain $\epsilon^* = \epsilon_0^*[1 - (x/s)]$ for $0 \leq x \leq s$, where $\epsilon_0^* = \epsilon^*(0)$. For example, this eigenstrain may represent a linear temperature distribution in the film, or a constrained volume change due to a chemical reaction. Using this eigenstrain, Eq. (10) reduces to

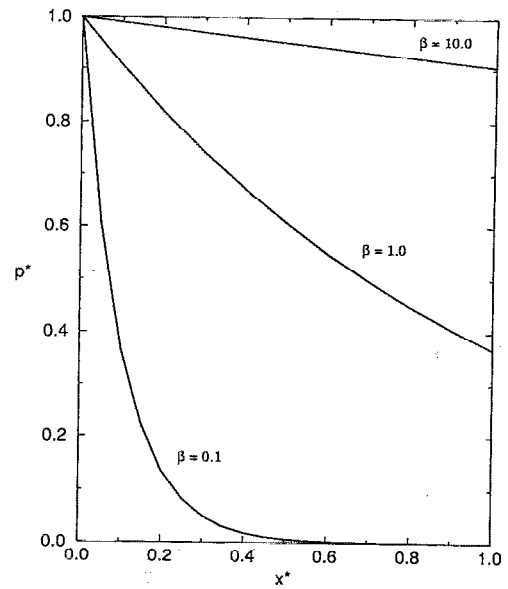


FIG. 2. Pressure in a viscous fluid film.

$$p(x) = -\frac{\epsilon_0^*}{\kappa} \left[e^{-x/\tau U} + \frac{\tau U}{s} (1 - e^{-x/\tau U}) \right]. \quad (11)$$

The presence of an eigenstrain affects viscous stress relaxation in a fluid film. In order to examine this effect, we express Eq. (11) in dimensionless form.

$$p^*(x) = e^{-x^*/\beta} + \beta(1 - e^{-x^*/\beta}), \quad (12)$$

where $x^* = x/s$, $p^* = -(\kappa p)/\epsilon_0^*$ is the ratio of the compressive strain in the film to the eigenstrain, and $\beta = (\tau U)/s$ is the ratio of the stress relaxation length to the film thickness.

The pressure obtained from Eq. (12) is shown graphically in Fig. 2, using the values $\beta = 10.0, 1.0, 0.1$. A rapid decrease in pressure for small values of β illustrates viscous stress relaxation. For silicon dioxide films, a high growth temperature is associated with a small value of β . Therefore, Eq. (12) predicts viscous stress relaxation in oxide films grown at high temperature. Experimental observations of stress relaxation in high temperature oxide films confirm this prediction.⁷ In general, for a given eigenstrain, Eq. (10) is solved to determine the extent of viscous stress relaxation in a fluid film.

To determine the temperature at which stress relaxation occurs in oxide films, we integrate Eq. (12) to obtain the average pressure in a fluid film,

$$\bar{p}^* = \frac{2}{3}\beta [1 + (1 - \beta)(1 - e^{-1/\beta})]. \quad (13)$$

If the stress relaxation length is much smaller than the film thickness, $\beta \rightarrow 0$ and $\bar{p}^* \rightarrow 0$. If the stress relaxation length is much larger than the film thickness, $\beta \rightarrow \infty$ and $\bar{p}^* \rightarrow 1$.

The pressure obtained from Eq. (13) is shown graphically in Fig. 3. We use experimental data for the growth rate of oxide films in wet and dry oxidation environments⁸ and experimental data for the viscosity of fused silicon dioxide⁹ to determine β for temperatures between 920 and

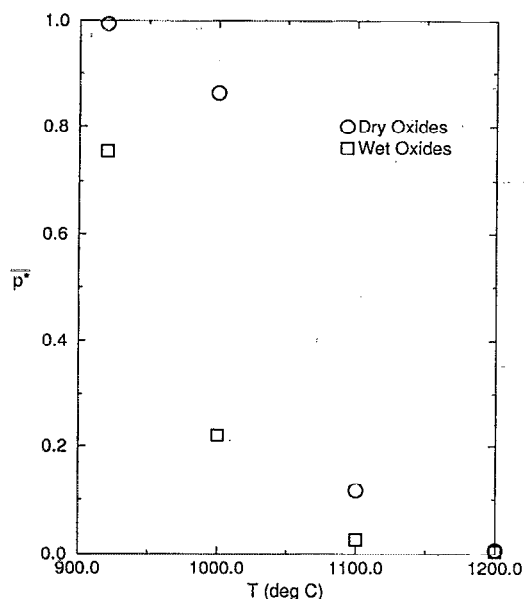


FIG. 3. Stress relaxation in an oxide film.

1200 °C. For wet oxide films, the values of \bar{p}^* predict stress relaxation in the entire temperature range. For dry oxide films, the values of \bar{p}^* predict stress relaxation at temperatures greater than about 950 °C. Experimental observations of stress relaxation in wet oxide films and the onset of viscous flow in dry oxide films confirm these predictions.⁴

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¹T. Mura, *Micromechanics of Defects in Solids* (Martinus Nijhoff, The Hague, Netherlands, 1987).

²P. E. Murray, *Appl. Phys. Lett.* **58**, 2488 (1991).

³E. Kobeda and E. A. Irene, *J. Vac. Sci. Technol. B* **5**, 15 (1987).

⁴E. P. EerNisse, *Appl. Phys. Lett.* **30**, 290 (1977).

⁵R. H. Doremus, *J. Electrochem. Soc.* **134**, 2001 (1987).

⁶I. G. Currie, *Fundamental Mechanics of Fluids* (McGraw-Hill, New York, 1974).

⁷E. Kobeda and E. A. Irene, *J. Vac. Sci. Technol. B* **6**, 574 (1988).

⁸B. E. Deal and A. S. Grove, *J. Appl. Phys.* **36**, 3770 (1965).

⁹G. Hetherington, K. H. Jack, and J. C. Kennedy, *Phys. Chem. Glasses* **5**, 130 (1964).