# MORPHOLOGICAL STABILITY OF MULTILAYER SYSTEMS

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**ABSTRACT**

With microstructural scale models, we examine the morphological stability and the kinetics of structural evolution for a multilayer system taking into account the different thermodynamic driving forces including chemical, elastic and interfacial energies. The description embodies the required conservation laws, the grain-size, physical and chemical properties of the individual layers, and flaw information to model the temporal evolution. We identify stability maps and estimate the system lifetime given the microstructural length scales and physical properties of the constituent layers and operating conditions including high temperature creep and thermal cycling.

In this paper, the thermal stability and the failure kinetics for a strained free-standing columnar grained polycrystalline thin film is examined. We track the shape evolution of the film when mass flow is controlled by grain boundary and surface diffusion phenomena and determine the critical time for pinch-off of the grains when the film is subjected to an imposed strain rate, simulating tensile creep conditions. The coupled time scales associated with surface diffusion, grain boundary diffusion and the applied loading rate are considered in the formulation and numerical results are derived in the limit where grain boundary diffusion is infinitely fast. The analysis shows that the pinch-off lifetime depends on only two normalized parameters: the aspect ratio of the grains and the applied strain rate.

## 1.0 INTRODUCTION

Multilayered film systems exhibit many interesting structural and mechanical properties. For instance, metallic multi-layers, with bilayer periods in the order of a few nanometers, possess yield strengths that are within a factor of two to three of the theoretical strength limit of ≈ E/30 where E is Young's modulus [1-4]. A potential application for such systems is in the area of high-temperature applications, whether in a direct load bearing application or as a protective coating. Due to the large interface density in these systems, morphological stability and creep are important considerations at high homologous temperatures. The thermodynamic driving forces that destroy layering includes chemical energy (that leads to solutionizing, intermixing, etc), elastic strain energy (residual stresses in the layers, and applied stresses), and interfacial free energies. The drive to reduce the sum total of these energies leads to instabilities in the layered structure, either through mixing of the layers, atomic rearrangement, or breakdown of the layering through capillary forces. In this paper, we examine thermal stability and the failure kinetics for a strained free-standing columnar grained polycrystalline film. We track the shape evolution driven by grain boundary and surface diffusion and determine the critical time for pinch-off when the film is subjected to an imposed strain rate, simulating tensile creep conditions. We limit our attention to the simpler problem of a single free-standing film case and deal with the multilayer case in future work. For the problem analyzed in this paper, there are three coupled time scales to consider - that associated with surface diffusion, grain boundary diffusion and the applied loading rate. We focus on the limit where grain boundary diffusion is infinitely fast but the remaining two time scales are comparable and compute the evolution kinetics by solving the coupled problem.

## 2.0 MODEL

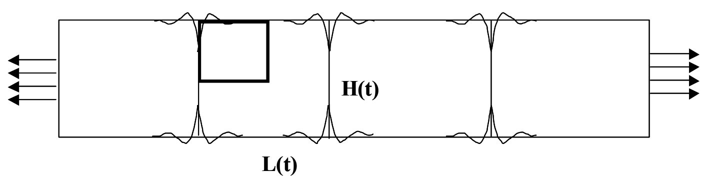


Fig 1: Schematic illustration of a thin film loaded along the axis

Consider the geometry shown in figure 1. The thin film consists of columnar two-dimensional grains of length L(t) and with grain boundaries of height H(t). The fundamental unit cell analyzed is shown in figure 1 (the grain boundary extends from $0 \le z \le H(t)/2$ and the free surface extends from $0 \le x \le L(t)/2$). Material may diffuse along the free surfaces, and also along the grain boundaries. The diffusion is driven by a variation in chemical potential, which causes atom migration from regions of higher to lower potential. We assume that the rate of mass transport is proportional to the chemical potential gradient. The governing equations for grain boundary $(0 \le z \le H(t)/2)$ stress evolution and for free surface $(0 \le x \le L(t)/2)$ shape evolution can be written [6-8] as:

$$\frac{\partial u_g(z,t)}{\partial t} = -\theta_g \frac{\partial^2 \sigma_g(z,t)}{\partial z^2} \tag{2.1}$$

$$\frac{\partial \hat{y}(x,t)}{\partial t} = -B_f \frac{\partial^4 \hat{y}(x,t)}{\partial x^4} \tag{2.2}$$

where constants $\theta_g = \delta_g D_g \Omega / 2kT$ and $B_f = \gamma_f \delta_f D_f \Omega / kT$ and where $u_g$ is the opening displacement $\delta_g D_g$ ($\delta_f D_f$) is the temperature dependent diffusion coefficient of the grain boundary (free surface), $\Omega$ is the atomic volume, $T$ is the absolute temperature, and k is the Boltzmann's constant. We examine the limiting case of infinitely fast grain boundary diffusion first. Also, we consider the thin film being subjected to a constant longitudinal external strain rate (*s*) such that $L(t) = L_0 \mathrm{Exp}(st)$. We choose the dimensionless parameters, $\hat{y} = (H_0(Y+1) - H(t))/2$ , $x = L(t)X/2$ , $\eta = H(t)/H_0$, $\tau = B_f\, t / L_0^{\,4}$, $M = (L_0/H_0)m$, and $\Delta = s L_0^4 / B_f$. Note that the temporal evolution of the surface profile ($Y(X,\tau)$) and the normalized film thickness at the groove ($\eta(\tau)$) depend on only the aspect ratio parameter $M$ and the dimensionless strain rate $\Delta$.

## 3.0 RESULTS

For the general non-zero strain rate case, the coupled partial differential equations presented in equation 2 are numerically solved and the results presented below.

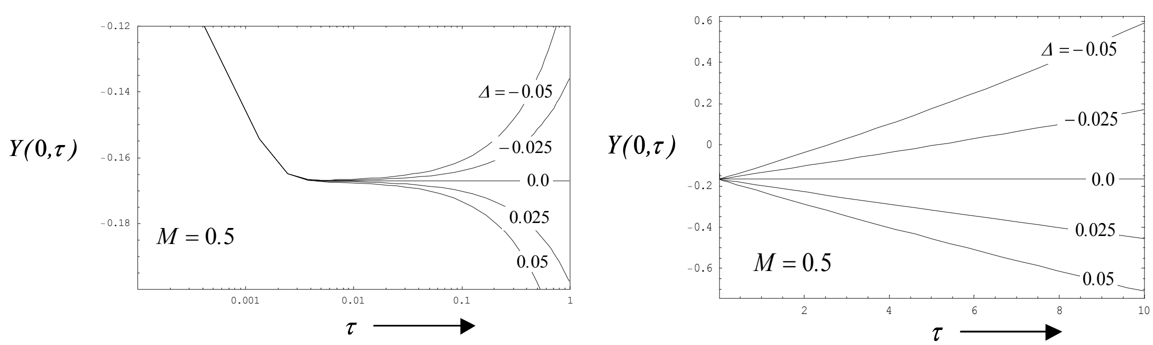


Figure 2 Temporal evolution of the surface profile for different strain rates (setting M = 0.5)

The temporal evolution of the surface profile for different strain rates is summarized in Figure 2. Material flows from the surface into the grain boundary and grooving is observed. With the choice of parameters used for rescaling the spatial dimensions, the surface profile is invariant for different strain rates at small times. However at large times the re-normalized surface profile evolution grows faster for larger strain rates and the scale invariance breaks down.

At the early stages of evolution, material depletion occurs at the groove depth irrespective of the whether the strain rate is tensile or compressive, since the dihedral angle (parameter M) dictates the material evolution at the junction. The groove depth should therefore be invariant for different strain rates. However, on marching forward in time, the surface profile at the groove root is no longer scale invariant and the profiles are no longer self-similar. The junction shows grooving under tensile strain rate conditions and hillock formation under compressive strain rate loading. Furthermore, the rate of grooving (hillock formation) is faster for more positive (negative) values of the strain rate parameter ( $\Delta$ ).

Figure 3 shows evolution of the normalized film thickness ( $\eta$ ) at the grain boundary as a function of the normalized time and for different tensile strain rates. As the strain rate is increased, the $\eta$ decreases faster. At some finite time, the thickness reaches zero and film pinch-off occurs. For the same grain aspect ratio film, pinch-off occurs earlier at larger strain rates. Also, as the grain aspect ratio parameter is increased, $\eta$ decreases leading to earlier pinch-off .

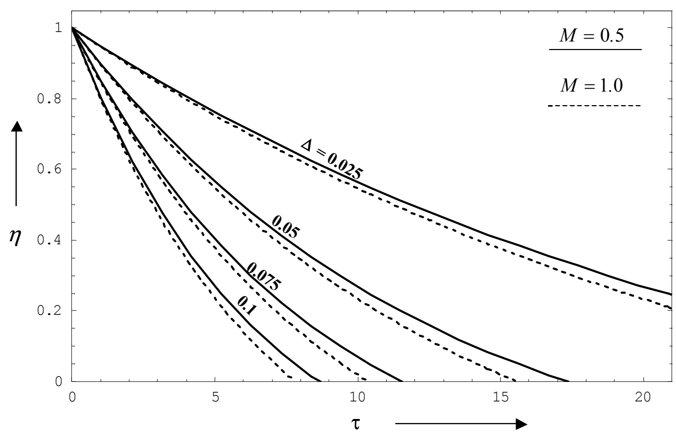


Figure 3: Evolution of the film thickness ( $\eta$ ) at the grain boundary as a function of the normalized time for different tensile strain rates and for two different aspect ratio parameters.

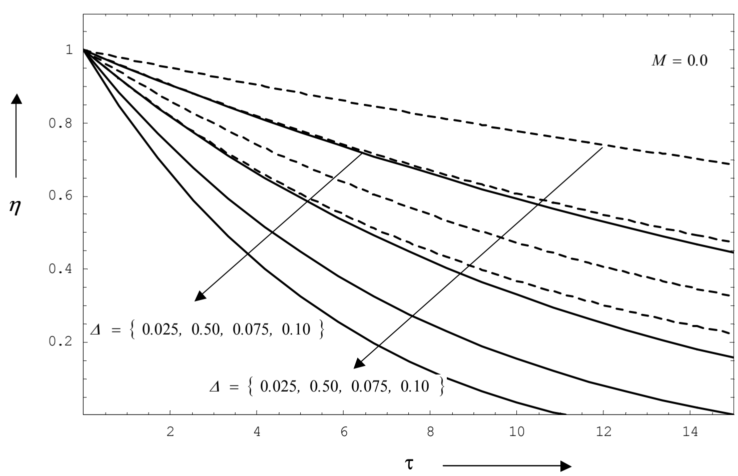


Figure 4:Evolution of the film thickness ( $\eta$ ) at the grain boundary as a function of time for different tensile strain rates (solid lines) and comparison with $\eta(\tau) = \eta_0 e^{-\varepsilon\tau}$ (dashed lines).

Figure 4 shows evolution of the film thickness ( $\eta$ ) at the grain boundary as a function of time for different tensile strain rates (solid lines) and comparison with $\eta(\tau) = \eta_0 \mathrm{Exp}(-\Delta\tau)$ (dashed lines). The dashed lines correspond to the thickness decrease of the film for the case of creep completely being accommodated by deformation of the bulk. The results suggest that the rate at which thickness decreases locally at the grain boundary is much faster than the decrease decrease for the case of bulk deformation. This is because in the case of creep by bulk deformation limit, the layer thins uniformly. On the other hand, in the creep driven by grain boundary and surface diffusion, material at the grain boundary locally thins faster.

As the tensile strain rate increases, material flows faster from the grain boundaries to the free surface in order to accommodate the deformation. However, for a finite thickness film, the available flux is finite and the film will eventually pinch-off. The time required for pinch-off is set by $\eta(\tau_c)$ =0 and $\tau_c$ can be determined Figure 5 shows the pinch-off time as a function of the strain rate and for two different grain aspect ratios. As the aspect ratio increases, the net available flux diminishes and hence pinch-off occurs earlier.

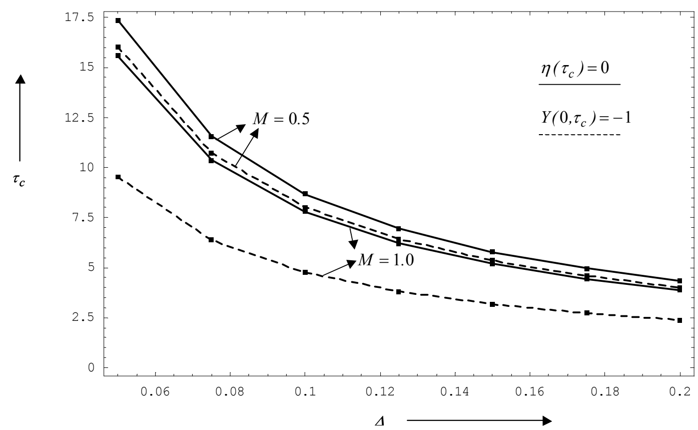


Figure 4: Time required for pinch-off as function of strain rate using two different failure criteria.

Also, shown in figure 5 is time for pinch-off time using the criteria $Y(0,\tau_c)$ =0. Since the decrease of the film thickness is first dictated by local balance of surface forces followed by thickness reduction to accommodate the imposed tensile strain, it is possible that pinch-off could occur for very thin films due to lack of insufficient material to satisfy the dihedral angle condition. As is evident in the figure, the time predicted with the $\eta(\tau_c)$ =0 criteria is expected to vastly overestimate the pinch-off time especially for very thin films.

## 4.0 SUMMARY

In this paper, the thermal stability and the pinch-off failure kinetics for a free-standing polycrystalline film under a constant applied loading rate was studied. A mathematical formulation was presented for the coupled problem where the time scale for surface diffusion is comparable to the time scale associated with the loading rate. The solution describes how the surface profile and the nominal film thickness evolve as a function of time. The pinch-off failure time under a tensile applied load is predicted as a function of the strain rate and the grain size.

## 5.0 REFERENCES


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