SELF-ORGANIZED CRACK PATTERNS IN THIN FILMS

Wei Lu and Hui-Chia Yu
Department of Mechanical Engineering, University of Michigan, Ann Arbor, MI 48109 USA

ABSTRACT

Self-organized crack patterns have drawn intensive attention recently. For instance, regular micro-cracks appear on certain polymer materials spread on substrates when drying. These cracks are generally conceived originating from the competition of two effects, i.e. cohesion force between molecules and adhesion forces between molecules and substrates. This paper develops a phase field model to study the formation and growth of crack patterns of a thin film attached to an elastic substrate. We treat the thin film as a superficial object, and specify the excess surface energy for the object. The variation of the surface energy density with the variation of the strain defines surface stress. When cracks form in the film, the surface stress is nonuniform, deforms the substrate, and reduces the total energy. This energy reduction constitutes a driving force for crack nucleation and growth. The model consists of two fields: a concentration field C, and a displacement field $\mathbf{u}$. The former phase-field parameter describes the coverage of substrate surface with film molecules, and has values between zero and one. The latter represents the elastic field in the substrate. A phase boundary in our model is represented by a concentration gradient, an approach analogous to the work of Cahn and Hilliard on spinodal decomposition. With this approach we avoid solving a moving boundary value problem which would require tracking the boundary numerically. Our model is a dynamic model and the material system can generate whatever crack patterns it favors. A nonlinear diffusion equation couples the concentration field in the film and the stress field in the substrate. An efficient spectral method is developed to solve the problem efficiently. The simulations have revealed remarkably rich dynamics in the crack pattern formation process, and produced results consistent with experiments.

1 INTRODUCTION

Self-organized crack patterns have drawn intensive attention recently. Consider a thin solid film elastically attached to a substrate. When the film contracts, elastic stresses emerges. When the stress is high enough, cracks will generate and from a pattern. A typical example is the formation of regular micro-cracks of certain polymer thin films spread on substrates when drying [1,2]. The drying causes shrinkage of the film and thus generates stresses. Similar phenomenon has also been reported in stressed metal thin film on a silicon substrate [3]. These cracks are generally conceived originating from the competition of two effects, i.e. cohesion force between molecules and adhesion forces between molecules and substrates.

Crack propagation has been the main focus of fracture mechanics in the engineering world [4]. Finite element methods have been developed to compute the process, e.g. [5]. However, the traditional linear elastic fracture mechanics does not account for microscopic process of crack propagation. In micro scales, vacancies or defects are known to drift toward and accumulate at grain boundaries in multi-granular materials or agglomerate into voids in single-granular materials, and subsequently form micro cracks. These cracks then evolve in shape and size by surface or bulk diffusions in non-rapid fracture cases. Molecular dynamic study [6,7] has strengthened this idea. With such consideration, crack pattern formation can therefore be considered as a phase transformation process [8,9].

This paper considers the crack patterns in a thin film, which is different from a thick film in several aspects. Firstly, for a very thin film with a few monolayers thickness, it cannot be treated as a bulk material. Instead, the concepts of surface energy and surface stress are required

[10]. Secondly, a crack will have two effects: a) creating the crack surface and b) exposing the substrate surface where the crack appears. The effect b can be neglected for a thick film since the crack opening is much smaller comparing with the film thickness. However, for a very thin film, the exposition of the substrate will cause energy changes comparable to that of the creation of crack surfaces. We propose a model in the following.

2 THE MODEL

We treat the thin film as an infinitely large surface and the substrate as a semi-infinite elastic body. The substrate occupies the half space $x_3 < 0$ and is bounded by the x_1-x_2 plane. The energy of the system comprises the surface energy of the film and the elastic energy in the substrate. In this paper we assume the substrate is elastically isotropic. The elastic energy per unit volume in the bulk is a quadratic function of strain with Young's modulus E and Possion's ratio ν as material constants.

The surface energy per unit area, Γ, takes an unusual form in the model. Define a phase field parameter, the concentration C, which represents the coverage of substrate surface with film molecules. It has values between zero (for crack) and one (for film material). Regard the concentration as a spatially-continuous and time-dependent function, $C(x_1, x_2, t)$. Generally speaking, Γ is a function of the concentration, C, the concentration gradient, $\partial C / \partial x_\alpha$, and the strain in the film, $\varepsilon_{\alpha\beta}$ (A Greek subscript runs from 1 to 2). Expanding the function $\Gamma(C, \partial C / \partial x_\alpha, \varepsilon_{\alpha\beta})$ in the leading order terms of the concentration gradient $\partial C / \partial x_\alpha$ and the strain $\varepsilon_{\alpha\beta}$, we have

$$\Gamma = g + h\left(\left(\frac{\partial C}{\partial x_1}\right)^2 + \left(\frac{\partial C}{\partial x_2}\right)^2\right) + f\left(\varepsilon_{11} + \varepsilon_{22}\right), \tag{1}$$

where g, h, and f are all functions of the concentration C and coordinates. We have assumed that h and f are isotropic in the plane of the surface. The leading-order term in the concentration gradient is quadratic because, by symmetry, the term linear in the concentration gradient does not affect the surface energy.

The term $g(C)$ represents the surface energy per unit area of a uniform film on a substrate. To describe phase separation, we may prescribe $g(C)$ as any function with double wells. In numerical simulations, to be definite, we assume that it takes the form of

$$g(C) = \Lambda k_b T\left[C \ln C + (1-C)\ln(1-C) + \Omega C(1-C)\right], \tag{2}$$

where Λ is the number of atoms per unit area on the substrate, k_b is Boltzmann's constant, and T is the absolute temperature. The dimensionless number Ω controls the shape of the curve. When $\Omega < 2$, the function g is convex. When $\Omega > 2$, the function g has double wells, and drives phase separation. We assume that $h = h_0$ is a positive constant. Any nonuniformity in the concentration field by itself increases Γ. In the phase field model, the h-term in Eq. (1) represents the phase boundary energy. It drives phase coarsening. The quantity, f, known as surface stress, is the surface energy change associated with the elastic strain [10]. This concentration-dependent surface stress reflects the interaction of the film and the substrate. We assume that surface stress is a linear function of the concentration, i.e. $f = \psi + \phi C$, where ψ and ϕ are material constants [10].

Atoms diffuse within the thin film to reduce the combined surface energy and the elastic energy in the substrate. The diffusion equation is obtained by energy variation, which is

$$\frac{\partial C}{\partial t} = \frac{M}{\Lambda^2}\nabla^2\left(\frac{\partial g}{\partial C} - 2h_0\nabla^2 C - \frac{(1-v^2)\phi^2}{\pi E}\iint\frac{(x_1-\xi_1)\frac{\partial C}{\partial\xi_1} + (x_2-\xi_2)\frac{\partial C}{\partial\xi_2}}{\left[(x_1-\xi_1)^2 + (x_2-\xi_2)^2\right]^{3/2}}d\xi_1 d\xi_2\right) \tag{3}$$

where $\nabla^2 = \frac{\partial^2}{\partial x_1^2} + \frac{\partial^2}{\partial x_2^2}$, and M is the mobility of atoms in the epilayer. The integration extends over the substrate surface. The strain field expressed by the double integration term is obtained by the superposition of the point force solution.

A comparison of the first two terms in the parentheses of Eq. (3) defines a length $b = \sqrt{h_0/\Lambda k_b T}$. In the Cahn-Hilliard model this length scales the phase boundary thickness. The magnitude of h_0 is on the order of energy per atom at a phase boundary. Using magnitudes $h_0 \sim 10^{-19}\,\mathrm{J}$, $\Lambda \sim 5\times10^{19}\,\mathrm{m}^{-2}$ and $k_b T \sim 5\times10^{-21}\,\mathrm{J}$ (corresponding to $T = 400$ K), we have $b \sim$ 0.6 nm. Another length, $l = Eh_0/\left[\phi^2(1-v^2)\right]$, is defined by comparing the last two terms in the parenthesis. This length reflects the competition of surface stress and phase boundary. Young's modulus of a bulk solid is about $E \sim 10^{11}\,\mathrm{N/m}^2$. A representative value for ϕ is $\sim 4\mathrm{N/m}$ [10]. The equilibrium phase size is on the order $\sim 4\pi l$, according to theoretical analysis and simulation. These magnitudes, together with $h_0 \sim 10^{-19}\,\mathrm{J}$, give $4\pi l \sim 8\,\mathrm{nm}$, broadly agrees with observed phase sizes in experiments. From Eq. (3), disregarding a dimensionless factor, we note that the diffusivity scales as $D \sim Mk_b T/\Lambda$. To resolve events occurring over the length scale of b, the time scale is $\tau = b^2/D$, namely $\tau = h_0/\left[M(k_b T)^2\right]$.

The integral makes it inefficient to solve Eq. (3) in real space. An efficient method is to solve the equation in reciprocal space. The Fourier transform converts the integral-differential equation into a regular partial differential equation. The integration operation, as well as the differentiation over space, is removed and the evolution equation is dramatically simplified. Let k_1 and k_2 be the coordinates in reciprocal space. Denote the Fourier transform of $C(x_1,x_2,t)$ by $\hat{C}(k_1,k_2,t)$, namely, $\hat{C}(k_1,k_2,t) = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty}C(x_1,x_2,t)e^{-i(k_1 x_1 + k_2 x_2)}dx_1 dx_2$. Normalizing Eq. (3) by the length b and the time τ, and applying the Fourier transform on both sides, we obtain the evolution equation in reciprocal space

$$\frac{\partial\hat{C}}{\partial t} = k^2\hat{P} - 2(k^4 - k^3 Q)\hat{C}, \tag{4}$$

where $k = \sqrt{k_1^2 + k_2^2}$, $Q = b/l$ and $\hat{P}(k_1,k_2,t)$ is the Fourier transform of $P(x_1,x_2,t) = \ln(C/1-C) + \Omega(1-2C)$.

Figure 1 shows the emergence of cracks from a void. The calculation cell size is $256b\times256b$. Material parameters are $\Omega = 2.2$, $v = 0.3$, $Q = 1$. At a given time the concentration fields are visualized by gray scale graphs. The brighter region corresponds to higher concentration (film) and the darker region corresponds to lower concentration (crack). The calculations start from

random initial conditions. The boundary condition conditions are periodic. Figure 1b shows a crack pattern from a smaller initial void. The preliminary results have revealed exciting features. Additional simulations are being carried out to under the crack pattern formation process, crack/void interactions, and effects of material anisotropy.
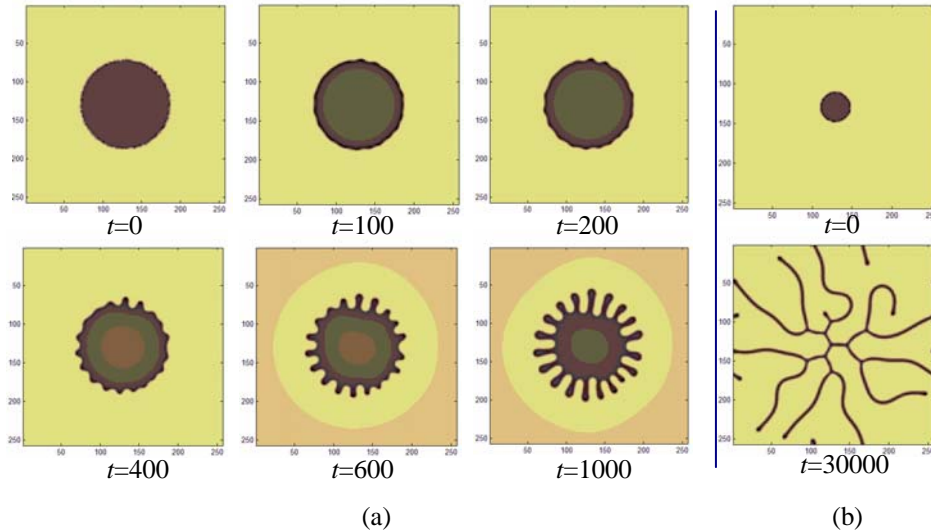

Figure 1 (a) An evolution sequence of self-assembled crack patterns. (b) A crack pattern from a smaller initial void.

3 REFERENCES

[1] Chen, X., Chen, Z., Yang, B., Zhang, G., and Shen J., "Regular patterns generated by self-organisation of ammonium-modified polymer nanospheres," J. Colloid. Interf. Sci. Vol. 269, pp79~83, 2004.

[2] Annarelli, C.C., Fornazero, J., Bert, J., and Colombani, J., "Crack patterns in drying protein solution drops," Eur. Phys. J. E, Vol. 5, pp599~603, 2001.

[3] Meyer, D.C., Leisegang, T., Levin, A.A., Paufler, P., and Volinsky, A.A., "Tensile crack patterns in Mo/Si multilayers on Si substrates under high-temperature bending," Appl. Phys. A, Vol. 78, pp303~305, 2004.

[4] Liang, J, Huang, R., Prevost, J.H., and Suo, Z., "Evolving crack patterns in thin films with the extended finite element method," Int. J. Solids. Struct., Vol. 40, pp2343~2354, 2003.

[5] Buehler, M.J., Abraham, F.F., and Gao, H., "Hyperelasticity governs dynamic fracture at a critical length scale," Nature, Vol. 426, pp. 141-146, 2003.

[6] Ashurst, W and Hoover, W, "Microscopic fracture studies in the two-dimensional triangular lattice," Phys. Rev. B, Vol. 14, pp1465~1473, 1976.

[7] Hornig, T., Sokolov, I.M., and Blumen, A., "Patterns and scaling in surface fragmentation processes," Phys. Rev. E, Vol. 54, pp4293~4298, 1996.

[8] Eastgate, L.O., Sethan, J.P., Rauscher, M., Cretegny, T., Chen, C.S., and Myers, C.R., "Fracture in mode I using a conserved phase-field model ," Phys. Rev. E, Vol. 65, Art. No. 036117, 2002.

[9] Hu, S.Y. and Chen, L.Q., "Phase-field model for evolving microstructures with strong elastic inhomogeneity,"Acta Mater., Vol. 49, pp1879~1890, 2001.

[10] Ibach, H., "The role of surface stress in reconstruction, epitaxial growth and stabilization of mesoscopic structures," Surf. Sci. Rep., Vol. 29, pp193~263, 1997.