Toughness Distribution Considerations in a Thin Film System

L. D. BROWN* and H. L. MARCUS

Center for Materials Science and Engineering, Department of Mechanical Engineering, The University of Texas, Austin, TX 78712, USA

**Currently affilated with Atlantic Richfield Oil & Gas, Central Research, 2300 W. Plano Pkwy., Plano, TX 75075, USA*

ABSTRACT

The spontaneous mechanical failure of thin films and occasionally substrates, due to the thin film deposition, are relatively common occurrences. Large internal stresses (e.g. 10 MPa to 100 MPa) are an inherent part of the thin film deposition process. In most cases where fracture mechanics has been applied to thin film systems, the failure path is confined a-priori to the substrate-thin film interface or in the thin film perpendicular to that interface. This paper considers the contributions which determine the failure path and presents a qualitative model for identifying the failure path in a thin film system taking into account the presence of residual stresses. The most important aspect of this model is that it offers an explanation of the variation in the locus of failure that does not strictly depend on the weakest point of the material (i.e. the least work of fracture) being the first to fail. It does not, inherent to its construction, restrict failure to the interface a-priori. The fracture mechanics criterion of the strain energy release rate equaling the material's resistance to crack growth leads to crack propagation at a specific location and defines the selection of the fracture path.

KEYWORDS

Thin film failure; Fracture mechanics; Strain energy release rate; Toughness distribution; Locus of failure; Residual stresses; Composites; Al/Graphite MMC.

INTRODUCTION

The spontaneous mechanical failure of thin films and occasionally substrates, due to the thin film deposition, are relatively common occurrences. Large internal stresses (e.g 10 MPa to 100 MPa) are an inherent part of the thin film deposition process. These residual stresses have been attributed to induced plasticity, surface oxide layers, defects, interfacial constraint, wetting, capillarity, etc.. Discussions on the residual stresses in thin films have been extensively reviewed (Hoffman,1966; Chopra, 1966).

Mechanical failure in thin film systems can be described in terms of linear elastic fracture mechanics. In this treatment, the stored elastic strain energy resulting from the residual stresses of the thin film deposition is evaluated relative to the crack propagation resistance of the substrate, the thin film, and their interface (Argon *et.al.*, 1988, Klokholm, 1987). Fracture

mechanics provides a definition for the onset of failure by crack growth. In most cases where fracture mechanics has been applied to thin film systems, the failure path is confined a-priori to the substrate-thin film interface or in the thin film perpendicular to that interface. This paper considers the contributions which determine the failure path and presents a qualitative model for identifying the failure path in a thin film system taking into account the presence of residual stresses.

TOUGHNESS DISTRUBUTION

The most probable failure path in a system defines the locus of failure and is determined by the toughness distribution in the system. The toughness of a material can be described in terms of the its resistance to crack propagation, its elastic properties, the stress distribution, the defect distribution, and the defect's relative stress intensity factor. A thin film system's toughness distribution can be assessed by considering the strain energy release rate, G, relative to the distribution of the system's resistance to crack propagation, R. A schematic representation of a G-R curve for an ideal homogeneous brittle solid is shown in Fig. 1. The critical value of the strain energy release rate, G_c, is determined by the stress state at which the G curve becomes tangential to the material resistance curve, R (Srawley *et.al.*, 1964). This represents the stress state at which the fraction of the elastically stored strain energy that is released by crack growth equals the energy required to create the new crack surfaces. Spontaneous failure will occur for this and greater stresses. The locus of failure in a thin film system can be identified by determining the failure path that releases the necessary strain energy relative to the local ability of the system to resist crack propagation on that failure path. In some respects, this treatment is analogous to the minimum strain energy density criteria (Sih, 1981).

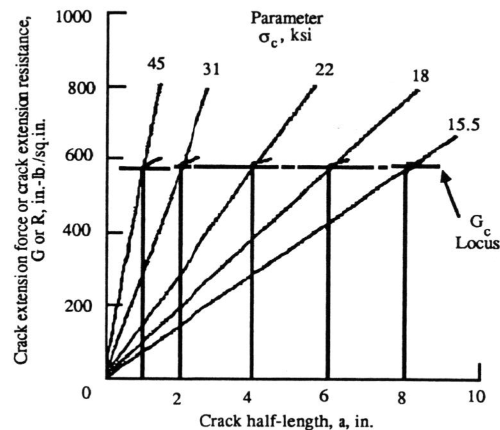

Fig. 1. A schematic representation of a G-R curve for a brittle material with a range of defect sizes. The critical stress where the G curve is tangential to the R curve is used a parameter. [From Srawley *et.al.*, 1964]

The above treatment may be applied to a thin film system by first determining the distribution of the resistance to crack growth. For an ideal brittle material, the material's resistance curve, R, is a step function equivalent to the work of fracture, W_f. This was shown in Fig. 1. The work of fracture for a brittle two phase material may be stated as

$$W_f = \gamma_A + \gamma_B - \gamma_{AB} \tag{1}$$

where γ_A , γ_B = the surface energy per unit area of each of the newly created surfaces

γ_{AB} = the interface energy per unit area if fracture occurs at a previously existing interface.

The work of fracture for a single phase of brittle material is equivalent to twice the surface energy of the solid since in this case the initial interface energy is zero. The interface energy, γ_{AB}, is determined by both chemical bonding and strain at the interface (Fine, 1964). A strong chemical bond and small strains at the interface result in a small interface energy. For the purposes of the present discussion, these surface and interface energies are considered to incorporate all mechanisms for the dissipation of energy associated with crack propagation including such effects as plasticity.

For convienence we assume equivalent structural defects, in the form of edge cracks, are distributed in the system. This is schematically represented in Fig. 2.

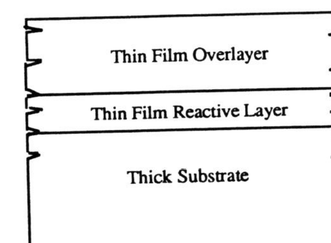

Fig. 2. A schematic of the distribution of equivalent edge cracks, exagerated, in a laminate system to analyze the distribution of strain energy release rate.

For each defect a strain energy release rate with respect to unstable crack growth can be evaluated. The strain energy release rate, G, is defined as

$$G = \frac{dU}{da} \tag{2}$$

It represents the change in the stored strain energy in a system,U, with incremental crack growth. If the increment of crack growth, da, is taken to be equivalent for each of the above

defects and since equivalent defects are used, the change in strain energy, dU, and thereby a relative G_i for each defect location may be represented as

$$G_i = U_o(a) - U_i(a + da) \tag{3}$$

Here U_o is the intial strain energy in the system with a crack length of "a" and U_i is the strain energy in the system resulting from a crack growth of da at the ith defect location. The initial strain energy is the same for all defect locations whereas U_i will vary with defect location.

The strain energy release rate is a function of the stress state, σ, the crack length, a, and the elastic properties of the components, E (i.e. Young's moduli). However, as (a + da) is considered here to be equivalent for all defects and the elastic properties remain unchanged by crack growth, the strain energy release rate distribution may be considered to be a function of the stress state. The stress distribution considered here is the elastic response of the system to the residual stresses in the deposited thin films. The residual stress distribution will adjust so as to minimize the overall strain energy, assuming no a-priori failure, consistent with the elastic properties of each component and the boundary conditions at the interfaces.

These considerations can be simplified by a one-dimensional analysis in the z-direction for a two film-substrate system where each thin film has a uniform biaxial stress, σ_{kt}. This is schematically shown in Fig. 3. In this fracture response formulation, only principle stresses, σ_x, σ_y, and σ_z, are considered and a state of uniform biaxial stress, $\sigma_x = \sigma_y = \sigma_t$, is assumed for each component (i.e. substrate and thin films) in the x-y plane. The x and y coordinates are considered parallel to the system's interfaces and the z coordinate is perpendicular to these interfaces. The uniform biaxial stress state in the substrate is considered to decay exponentially

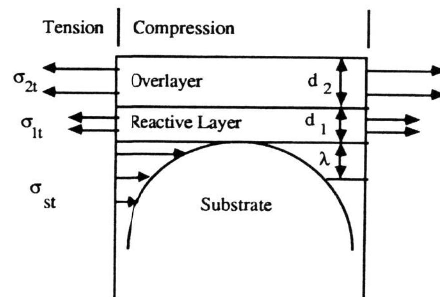

Fig. 3. A schematic representation of the biaxial stress state viewed in one dimension of two thin films on an thick substrate. Tensile biaxial stresses are shown for the thin films.

from the substrate surface with a characteristic decay length of λ (Van der Merwe, 1963). If the interfaces are considered to be strong so that continuity exists, the force balance

$$\sum_{k=1}^{3} F_k = \sum_{k=1}^{3} \int \sigma_{kt}\, dz = 0 \tag{4}$$

can be applied. It is the result of the minimization of strain energy in the system with respect to strain and simply states that the sum total of the forces associated with the thin film layers and the substrate due to the residual biaxial stress state must balance. Here the thickness of the kth thin film is d_k. The characteristic decay depth, λ, of the substrate is its apparent thickness, d_s. Solutions of this sort are commonly found for a thin film on an infinite substrate (Hoffman, 1974). The thick substrate is considered to go to infinity at one limit with the stress reducing to zero at its free surface.

From the force balance, it is seen that a stress is induced in the substrate which will oppose and balance the stresses in the thin films on its surface. These stresses can be significant since, as was mentioned previously, the residual stresses in thin films can be in the range of 10 MPa to more than 100 MPa. The above considerations indicate that a thin film system's stress distribution is a function of the film stress, film thicknesses, and the characteristic decay length of stress in the substrate.

LOCUS OF FAILURE

The locus of failure can be identified by determining the location in the system at which a defect's strain energy release rate equals the local work of fracture. In principle, this determines both the locus of failure and the critical stress state for the thin film interface model. The critical strain energy release rate, G_c, is determined by the first point of coincidence between the strain energy release rate and the work of fracture distributions. This G_c will determine the locus of failure and the failure stress for spontaneous failure when external stresses are applied.

The actual crack growth will, in part, depend on the relative defect size and the stress concentration associated with the defect with respect to all other defects in the system (Kelly *et.al.*, 1986). These considerations are combined in the stress intensity factor. A greater stress intensity factor will reduce the stress and/or the defect size required for instability by increasing the strain energy release rate of the associated defect. The locus of failure will be biased to the location with the greatest stress intensity factor and the least work of fracture. In addition, crack propagation in thin film systems corresponds to mixed modes (i.e. tensile, shear, and torsion). However, since the strain energy release rate adds linearly and independently for each of the modes, each propagation mode can be considered seperately.

This qualitative model for evaluating the toughness distribution has been applied to a thin film system of a highly oriented pyrolitic graphite substrate with thin films of a aluminum/aluminum oxide and aluminum (Brown, 1988). The graphite substrate has a basal plane oriented surface (e.g. (0001) surface/interface). The aluminum/aluminum oxide layer is deposited first by reactive evaporation to a thickness of 50 nm then the aluminum thin film is deposited by evaporation in high vacuum (e.g. 10^{-7} Torr) to a thickness of 150 nm. The reactive layer can be varied from a non-crystalline oxide to a microcystalline aluminum. The thin films were considered to have biaxial residual stresses which vary from compressive for a non-crystalline oxide film to tensile for a microcystalline aluminum thin film. The aluminum overlayer was considered to always have tensile residual stresses.

The work of fracture distribution in the thin film system represents the locus of possible points of tangency with the G curve. Values for the work of fracture for the graphite substrate and an aluminum oxide layer are 480 mJ/m^2 and 1200 mJ/m^2 respectively. The oxide's work of fracture is derived from the surface tension of aluminum oxide at its melting point and is believed to be consistent with the non-crystalline nature of the oxide thin film. The aluminum overlayer's work of fracture is much greater than the aluminum's surface tension at aluminum's melting point, 2 mJ/m^2, due to the inherent plasticity of aluminum.

The Young's moduli of the graphite parallel to the basal plane, an aluminum oxide film, and the aluminum film are 1000 GPa, 100 GPa, and 70 GPa respectively. An aluminum oxide film's modulus is the value measured for sputtered, non-crystalline thin films of aluminum oxide.

A qualitative G-R curve for this system is schematically drawn in Fig. 4 for each of the three cases of reactive layer morphologies: 1) a non-crystalline oxide layer; 2) a cermet layer; and 3) a microcrystalline aluminum layer. The cross hatched areas represent regions of modification to the work of fracture due to interfacial reactions. If the non-crystalline oxide layer is considered to be in compression and the alumium overlayer is considered to be in tension, application of the force balance indicates that stress state reversal occurs at the interfaces, G_1. Failure is predicted in the vicinity of the graphite/oxide interface. The cermet layer system has little net residual stresses resulting in a constant, G_2. Failure is predicted in the graphite substrate when external stresses are applied. By application of the force balance to the

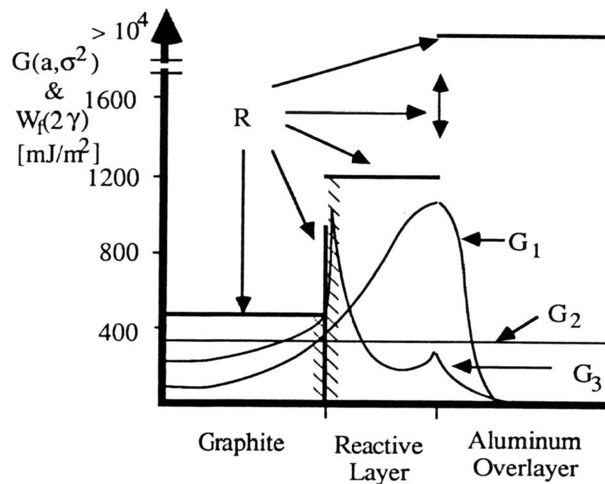

Fig. 4. A qualitative schematic of the superposition of the strain energy release rate superimposed on the work of fracture distribution in the thin film composite interface model for the three cases G_1, G_2, and G_3. The crack length, a, is fixed at an arbitrary value. The cross-hatched regions represent areas of modification to R due to substrate-film interactions.

microcrystalline aluminum layer system where the tensile residual stresses of both films must be balanced by compressive stresses in the substrate, G_3, it is observed that failure is favored in the region of the graphite/aluminum interface due to the stress reversal which occurs there.

Experimentally it was found, by failing specimens of this type in the ultra high vacuum of an auger spectroscopy system, that the failure path varied with reactive layer morphology. This is shown in Fig. 5. For a non-crystalline oxide system, the failure path was within 0.3 nm to 0.6 nm into the graphite substrate from the graphite-oxide interface. For a cermet reactive layer morphology where no net residual stress was believed to exist, the failure path was greater than 4.0 nm into the graphite substrate. As the aluminum crystallite size in the reactive layer increased, the failure path moved closer to the graphite/aluminum interface. The residual stress state in the reactive layer was believed to be increasingly tensile. At aluminum crystallite sizes of 50 nm, the failure path was essentially at the graphite/aluminum interface.

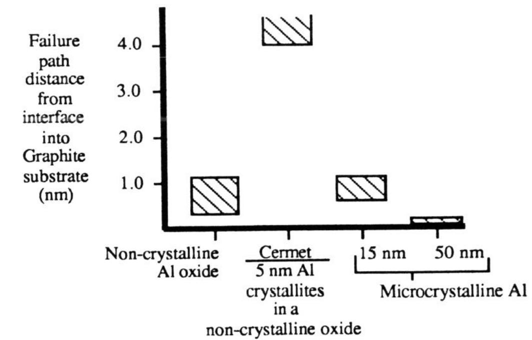

Fig. 5. The experimentally evaluated change in failure path in the graphite/reactive layer thin film system as a function of the reactive layer morphology. The failure path distance from the interface is the calculated thickness of graphite, as determined from auger spectra, remaining on the reactive layer after in-situ fracture.

CONCLUSION

The most important aspect of this model is that it offers an explanation of a variation in the locus of failure by considering both the strain energy release rate as modified by residual stresses and the work of fracture distributions in identifying the failure point of the material. It does not, inherent to its construction, restrict failure to the interface a-priori. The fracture mechanics criterion of the strain energy release rate locally equaling the material's resistance to crack growth leads to crack propagation at a specific location and defines the selection of the fracture path.

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