

MICROMECHANICS OF MACROELECTRONICS

Zhigang Suo^{1,*}, Joost Vlassak¹ and Sigurd Wagner²

¹*Division of Engineering and Applied Sciences, Harvard University, Cambridge, MA 02138, USA*

²*Department of Electrical Engineering and Princeton Institute for the Science and Technology of Materials, Princeton University, Princeton, NJ 08544, USA*

**Author to whom correspondence should be addressed. E-mail: suo@deas.harvard.edu*

Abstract The advent of flat-panel displays has opened the era of macroelectronics. Enthusiasm is gathering to develop macroelectronics as a platform for many technologies, ranging from paper-like displays to thin-film solar cells, technologies that aim to address the essential societal needs for easily accessible information, renewable energy, and sustainable environment. The widespread use of these large structures will depend on their ruggedness, portability and low cost, attributes that will come from new material choices and new manufacturing processes. For example, thin-film devices on thin polymer substrates lend themselves to roll-to-roll fabrication, and impart flexibility to the products. These large structures will have diverse architectures, hybrid materials, and small features; their mechanical behavior during manufacturing and use poses significant challenges to the creation of the new technologies. This paper describes ongoing work in the emerging field of research – the mechanics of macroelectronics, with emphasis on the mechanical behavior at the scale of individual features, and over a long time.

Keywords macroelectronics, flexible electronics, ductility, cracking, adhesion, thin films

1. Introduction

For half a century, the technology of integrated circuits has been advancing by miniaturization, squeezing more and more transistors onto each chip. While the trend to miniaturize features will continue in the field of microelectronics (Semiconductor Industry Association, 2004), a new trend to enlarge systems is gaining momentum in the nascent field known as macroelectronics or large-area electronics (Nathan & Chalamala, 2005). At present, the most visible application of macroelectronics is flat-panel displays. They entered the consumer market in the 1990s, and are now replacing cathode-ray tubes as monitors for televisions and computers. In such an application, transistors need not to be smaller than, say 10 μm , but the total surface area must be large. The flat-panel displays have enabled applications unimaginable for cathode-ray tubes. For example, the Dolphins Stadium will soon have the world's largest video display, about 15 m high and 42 m wide, comprising more than 4.6 million light-emitting diodes (<http://www.daktronics.com>).

In addition to large area, other desirable attributes for macroelectronic systems include ruggedness, portability and low cost. To realize these attributes, a growing trend is to fabricate macroelectronic products directly on flexible substrates (Nathan & Chalamala, 2005; Crawford, 2005). Current flat-panel displays are made on glass substrates and are fragile. A case in recent news is the cracking of the screens of the iPod Nano, a music player that Apple expects to be its best-selling portable device (Wingfield, 2005). By contrast, displays made on thin polymer substrates are rugged. Flexible displays of large areas will be lightweight and can be rolled up – they will be portable. Furthermore, flexible substrates will enable large areas of electronic surfaces to be manufactured by roll-to-roll printing, a process that will drastically lower the manufacturing cost (Bock, 2005). For these reasons, the field of macroelectronics significantly overlaps with that of flexible

electronics.

Macroelectronics will be a platform for many technologies. Consider, for example, all reading materials – books, newspapers, magazines, and mail-order catalogs. Given the staggering number of trees cut and amount of labor involved in printing and delivering them, it takes little imagination to see the merits of paper-like displays, provided they can be made lightweight and durable. The paper-like displays can store all the books we care to read, serve as newspapers that can be updated throughout the day, or switched to our favorite sceneries when reality is tiresome. They can be comfortably read in bed and carried around everywhere. Today's textbooks on complex and dynamic subjects, such as molecular biology of the cell, are weighty and are outdated while still in press. Their static, page-by-page format is unsuitable for learning such subjects. When paper-like displays become prevalent, printed books will become historic artifacts, and will be used for occasions of romance and nostalgia, as candles are today.

As another example, consider the global need for energy, as well as its environmental and geopolitical implications (Tester et al., 2005). Sunlight is an abundant source of clean energy, depositing 120,000 TW on the surface of the Earth, far exceeding the average power consumption of the world (13 TW). One way to use this energy resource is through solar cells (Department of Energy, 2005). Yet today solar cells are mostly made with expensive silicon-based panels and provide only about 1 GW power (Goetzberger et al., 2003; Surek, 2005). Sunlight is a distributed energy resource; harvesting solar electricity on a large scale will require solar cells to cover very large surfaces at low cost. This brings solar cells into the field of macroelectronics. Thin-film amorphous silicon solar cells are already manufactured in a roll-to-roll process on steel web (<http://www.ovonic.com>). Research on the fabrication of thin-film solar cells on polymer foils has

been conducted for many years (Okaniwa et al., 1982; Brabec, 2004; Shaheen et al., 2005). For these thin-film solar cells to penetrate the market, their cost needs be reduced, efficiency increased, and durability proven.

Macroelectronic systems are large structures of diverse architectures, hybrid materials, and small features. These structures will be subject to mechanical loads during manufacturing and use. During manufacturing, temperature change and mechanical handling will induce strains in materials. In use, paper-like displays will be bent, twisted and stretched repeatedly, and solar cells will be subject to daily temperature changes. Many consequences of hybrid materials and small features are familiar to researchers who have worked with composite materials and microelectronic devices. The following two observations are familiar but essential:

- A hybrid structure behaves unusually when its constituents are materials of extremely different kinds.
- The constituents behave differently from their bulk counterparts when features of the structure are small.

It is a common misconception that the mechanics of materials and structures is so well understood that the engineer can design a durable structure on a computer. This misconception is far from the reality. In fact, how to design complex structures of hybrid materials and small features is a grand challenge in the field of mechanical behavior of structures, a challenge painfully felt in many technologies. Recent examples include thermal barrier coatings for gas turbines (Evans et al., 2001) and interconnect structures on silicon chips (Suo, 2003; Lloyd et al., 2004; Hussein & He, 2005). The procedures to ensure durability in these and other technologies are largely empirical. The procedures are costly and time-consuming.

In technologies of relatively long standing, such as those of gas turbines and silicon chips, extensive empirical observations have led to partial understanding of likely failure modes. Such partial understanding is embodied in design rules and qualification tests. By contrast, macroelectronics is a nascent technology. Many of its challenges stem from the mechanical response of specific structures of hybrid materials and small features, but experience is too limited to formulate a set of design rules or qualification tests.

This paper describes ongoing research in the emerging field of the mechanics of macroelectronics. Section 2 explains why many macroelectronic systems will be organic/inorganic hybrid structures, and how they can be made flexible. Section 3 describes a way to realize stretchable electronics by using compliant thin-film patterns of stiff materials. Section 4 describes how to achieve high ductility of thin metal films on polymer substrates and fatigue of metal films subject to cyclic loads. Section 5 discusses cracking in brittle materials such as oxides, nitrides and amorphous silicon on polymer substrates. Section 6 outlines issues of interfacial debonding. Previous reviews of thin film mechanics (e.g., Nix, 1989; Hutchinson & Suo, 1992; Evans & Hutchinson, 1995, Vinci &

Vlassak, 1996; Volinsky et al., 2002; Freund & Suresh, 2003, Suo, 2003), largely motivated by issues concerning microelectronics, have focused on films on stiff substrates. Here we will focus on films on polymer substrates, the substrates of choice for many anticipated macroelectronic technologies.

2. Flexible Electronics with Hybrid Organic/Inorganic Materials

Enthusiasm for organic electronics aside, it is unlikely that many macroelectronic systems will be made entirely of organic materials, because organic materials cannot serve all functions of inorganic materials. Rather, most macroelectronic systems will be made as organic/inorganic hybrids. As an illustration, organic light-emitting devices (OLEDs) survive only a few hours when exposed to atmospheric oxygen and moisture. To attain a long lifetime, an OLED must be hermetically sealed. Organic materials are permeable to gases, so that a gas barrier must contain an inorganic material. In such an application, the gas barrier must also be optically transparent. A recently demonstrated gas barrier involves alternating thin films of aluminum oxide and polyacrylate (Chwang et al., 2003). The oxide films act as a barrier to gases, while the polymer films decouple defects in the oxide films. Similar hermetic seals will be required for most organic electronic devices, including organic solar cells.

As another reason to use inorganic materials in macroelectronics, electrical conductivity of organic materials is often inadequate, so that metals and optically transparent tin-doped indium oxide (ITO) are used as electrodes, and amorphous silicon is used in thin-film transistors.

The inorganic materials can be made flexible by taking advantage of two facts. First, the substrates are thin, so that bending induces small strains. According to elementary beam theory, when a substrate of thickness t is bent to a radius of curvature r , the strain on the surface of the substrate is:

$$\varepsilon = \frac{t}{2r}. \quad (1)$$

For example, $\varepsilon = 0.5\%$ for $t = 0.1$ mm and $r = 10$ mm. We have considered complications in this estimate of strain, such as the very different elastic moduli between the substrates and the films, and the residual stresses in the films (Suo et al., 1999; Gleskova et al., 1999), but the conclusion remains true that strains in flexible electronics can be made quite small. To reduce strains in brittle materials, one may even place these materials along with active devices on the neutral plane by laminating the substrate with mechanically matched encapsulation.

Second, the inorganic materials are used as thin films, whose small flaw size enables them to sustain higher strains than their bulk counterparts. According to fracture mechanics, for a brittle material the critical strain to cause fracture is approximately (Lawn, 1993)

$$\varepsilon_c \approx \sqrt{\frac{\Gamma}{Ea}} \quad (2)$$

here the fracture energy Γ and Young's modulus E are material properties independent of the feature size; the flaw size a is typically on the order of film thickness h . Taking representative values, $\Gamma = 10 \text{ N}\cdot\text{m}^{-1}$, $E = 10^{11} \text{ N}\cdot\text{m}^{-2}$, and $a = 10^{-6} \text{ m}$, we estimate that $\varepsilon_c = 1\%$. For example, a recent experiment shows that a 7 nm thick silica film can sustain strains beyond 5% (Rochat et al., 2005).

The above estimates, however, should not lead to complacency that a large margin exists between the strains needed for making systems flexible and those to cause fracture. Fracture will limit the function of flexible systems for many reasons, such as thick inorganic films, thick polymer substrates and small radii of curvature, as well as inelastic deformation under cyclic loading. In addition, flexible systems may be stretched, as well as bent; the strains caused by stretching can be much larger than those caused by bending, and are not mitigated by placing the brittle materials on the neutral plane. We next outline some of these considerations.

3. Stretchable Electronics of Patterned Stiff Materials on Compliant Substrates

Of various modes of deformation (e.g., bending, twisting and stretching), typically stretching is the most demanding, easily inducing a large tensile strain. While an elastomer substrate can recover from a large strain, inorganic materials fracture at small strains (less than about one percent, as discussed above). How to use these materials to make stretchable electronic circuits remains a challenge.

A helical spring can elongate substantially, even though the material that makes the spring can only recover from a small strain. One could fabricate electronic circuits on a helical platform, but this approach would require micro-fabrication in three dimensions, a technology that requires substantial development itself. To be compatible with planar microfabrication technology, the platform must be planar. It has been shown that serpentine metal interconnects on elastomeric substrates can sustain more than 200 cycles of elongation by 25% (Gray et al., 2004). It has also been shown that a metal film with initial microcracks can sustain large elongation of the substrate (Lacour et al., 2003, 2005).

These examples illustrate a principle: when a film of a stiff material is suitably patterned on a compliant substrate, a large elongation of the substrate induces small strains in the film, and the film accommodates the large elongation by twisting out of the plane (Li et al., 2005b). We expect that such a patterned film can serve as a platform, on which entire electronic circuits can be fabricated using the planar microfabrication technology. These circuits will function without appreciable fatigue when the substrate is repeatedly bent, twisted, and stretched.

4. Ductility of Thin Metal Films on Polymer Substrates

Thin metal films are used in macroelectronics as electrodes and interconnects. Already widely used are passive flexible circuits, mostly made of copper films on polyimide substrates (<http://www.flexdude.com>). It has been often reported that thin metal films, free-standing or polymer-supported, rupture at small strains (e.g., Alaca et al., 2002; Chiu et al., 1994). On the other hand, it has also been reported that some polymer-supported metal films can sustain strains up to 20% (e.g., Macionczyk & Bruckner, 1999; Gruber et al., 2004). The cause for the difference has been uncertain, but our recent work may have shed light on the subject.

Subject to a tensile strain, a freestanding thin metal film can deform plastically. When the tensile strain is large enough to break native oxides or other passivation layers on the surface of the metal film, dislocations in the metal film can readily escape. On further straining, the film does not harden as much as its bulk counterpart. Consequently, once the film thins preferentially at a local spot by forming a neck, further deformation is localized in the neck, leading to rupture (Fig. 1). By volume conservation, local thinning results in a local elongation of length comparable to the film thickness. The local elongation contributes little to the overall rupture strain, because the film has a small thickness-to-length ratio.

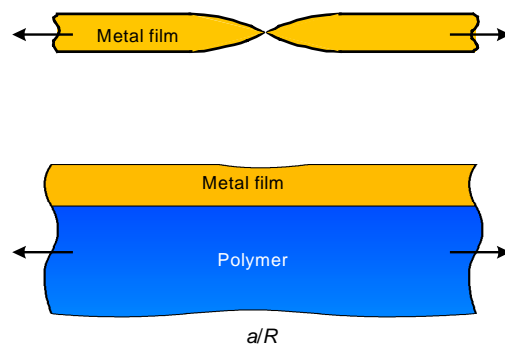


Fig. 1 A schematic to contrast a freestanding and a substrate supported metal film. A freestanding film ruptures by strain localization. A substrate may retard localization in the film, significantly increasing the strain to rupture.

While the local elongation is accommodated for the freestanding metal film by the ruptured halves moving apart, it cannot be so accommodated for a metal film bonded to a substrate. This constraint of the substrate retards strain localization, so that the metal film deforms uniformly to a large strain. The above arguments are phrased in terms of necking, but similar arguments apply to localization by shear band formation. Indeed, for such a metal film on a polymer substrate, our recent finite element simulation and linear bifurcation analysis have shown that the substrate can retard strain localization (Li et al., 2005a).

Our recent experiments with Cu films on polymer sub-

strates show that the rupture strains of the metal films are sensitive to their adhesion to the substrates (Xiang et al., 2005). Poorly bonded Cu films form channel cracks at strains about 2%, while well-bonded Cu films can sustain strains up to 10% without appreciable cracks. The well-bonded Cu films do form zig-zag cracks at strains of 30%.

Our existing theory shows, however, that a Cu film well adherent to a polyimide substrate should sustain strains in excess of 80%. This discrepancy between the experiment and theory may be caused by effects of small grain size and inadequate interfacial adhesion, as discussed below.

The effect of grain size on ductility of thin metal films on polymer substrates

One possible cause for the relatively low ductility observed for these films is that the grain size of the film is so small that dislocation motion in the grains is constrained. This may lead to excessive stress, which then causes intergranular fracture. It is also possible that trace amounts of impurities such as Bi, Sb or O may segregate to the Cu grain boundaries, embrittling the boundaries and causing intergranular fracture.

The effect of interfacial adhesion on plastic deformation of metal films

A second possible cause for the lower-than-theoretical ductility observed for these films is the loss of adhesion between the films and the substrates. If the metal film debonds from the substrate, the film becomes freestanding and is free to form a neck. When the intact laminate is subject to a modest tensile strain, we expect strain localization and debond to co-evolve. Without debond, the polymer substrate suppresses strain localization in the metal. Without localization, no traction is exerted on the interface to cause debonding. One can use cohesive zone models to study the co-evolution of necking and debonding.

The effect of a free surface or a strong interface with a hard and stiff material on plasticity in metallic films is well understood in terms of dislocation mechanisms: free surfaces allow dislocations to exit the material and reduce the work hardening rate, while strong interfaces with hard and stiff materials repel dislocations and increase hardening. This is well described by both discrete dislocation models (Nicola et al., 2005) and strain-gradient plasticity theories (Xiang et al., 2006). It is not clear, however, what happens for weak interfaces or for interfaces with soft materials, as is often the case for flexible electronics. We conjecture that when a dislocation reaches such an interface, the interface may debond or even slide allowing the dislocation to form a step. This is an important issue because it has an immediate impact on dislocation pile-up formation at interfaces and hence on the mechanical response of the metal film. Furthermore, slip steps at the interface and incipient debonds may act as nucleation sites for interfacial cracks that result in delamination of the

films and strain localization in the metal. It is currently difficult to model the effect of a weak interface on plastic deformation of thin films using either strain-gradient plasticity or dislocation dynamics methods, because the precise boundary conditions at the interface are not known.

Fatigue of thin metal films on polymer substrates under cyclic loading conditions

Above we discussed the ductility of metal films on polymer substrates under monotonic loading conditions. In some applications or during manufacturing, flexible devices may be subjected to appreciable strains for only a few cycles. In these cases, ductility under monotonic loading is important. In other cases, such as in roll-up displays, small strains of many cycles are anticipated. It is well established that most bulk materials fatigue under cyclic loads (Suresh, 1998). Fatigue is a likely failure mode in macroelectronics. For example, a recent cyclic loading experiment of an OLED identified fatigue fracture of metal electrodes as the failure mechanism (Chwang et al., 2003). The fundamental processes of fatigue of thin metal films on polymer substrates are explored in a few recent papers (e.g., Kraft et al., 2001; Zhang et al., 2005). Study along this line will lead research on fatigue, a phenomenon of lasting importance, in a new direction. In particular, one would like to determine how ductility is reduced by cyclic loading and establish the effects of interfacial adhesion and grain size.

5. Cracking in Brittle Inorganic Films on Polymer Substrates

As mentioned above, brittle inorganic materials will be used in flexible electronic systems for many essential functions – e.g., ITO as an optically transparent electrode, aluminum oxide or silicon nitride as a gas barrier, and amorphous or polycrystalline silicon as a semiconductor. Cracking in these films is a major failure mode of flexible electronics (Gleskova et al., 1999; Cairns et al., 2000; Leterrier, 2003).

Initiation vs. steady propagation of a channel crack

A brittle material fractures by breaking atomic bonds along the front of a crack. The crack driving force G is defined as the reduction of the elastic energy in the structure associated with the crack extending a unit area, when the external mechanical load is rigidly held and does no work (Lawn, 1993). As illustrated in Fig. 2, when a brittle thin film on a polymer substrate is subject to a tensile load, a flaw (A) in the film may grow into a channel crack (B). The crack driving force G is sketched as a function of the length of the crack. The trend is understood as follows.

When the crack just begins to grow from the flaw, the length of the crack, a , is typically smaller than, or comparable to, the thickness of the film, h , and the driving force

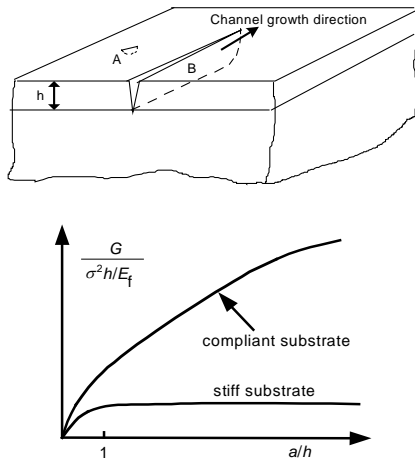


Fig. 2 Under a tensile stress in the film, a flaw (A) may grow into a channel crack (B). The crack driving force is sketched as a function of the crack length.

is given by:

$$G_{\text{initial}} = Y\sigma^2 a / E_f, \quad (3)$$

here σ is the tensile stress in the film, E_f is Young's modulus of the film, and Y is a dimensionless number of order unity. When $a/h < 1$, the crack driving force is linear in the crack length, and is insensitive to the presence of the substrate.

If the substrate film were free-standing (Fig. 3), G would continue to increase with a linearly as the crack grows. In the presence of an elastic substrate, however, the substrate constrains the opening of the crack, so that G increases with a , but no longer linearly. When the crack length exceeds some multiple of the film thickness, the driving force attains a steady state, given by

$$G_{\text{steady}} = Z\sigma^2 h / E_f. \quad (4)$$

That is, the driving force becomes independent of the crack length. The dimensionless number Z is a function of the elastic modulus ratio of the film and the substrate, and has been calculated (Hutchinson & Suo, 1992; Beuth, 1992; Huang et al., 2003; Vlassak, 2003).

In traditional microelectronic structures, the films and the substrates are both inorganic and have comparable elastic moduli, so that the crack driving force nearly attains the steady state when the crack length just exceeds the film thickness, $a/h \approx 1$ (Ho & Suo, 1993). One can simply use G_{steady} to determine the critical design condition, a practice that greatly simplifies the problem, because G_{steady} is independent of the size and the nature of the initial flaw, and because G_{steady} can be calculated by analyzing plane strain boundary value problems.

By contrast, in flexible electronics, the polymer substrate is very compliant compared to the inorganic films, so that the substrate provides less constraint on the crack opening. The crack driving force attains the steady state

only when $a/h \gg 1$ (Ambrico & Begley, 2002), and the value of Z is much larger than unity. Consequently, $G_{\text{initial}} \ll G_{\text{steady}}$. If initial flaws in a thin film are not much longer than the film thickness, as is usually the case, it will be over-conservative to design according to G_{steady} . In a realistic film, cracks are likely to initiate from a defect such as a pinhole, or a corner in a patterned film. These situations must be studied if one needs to formulate more realistic design rules.

Effect of inelastic deformation of the substrate on channel cracks in the film

Figure 3 illustrates the effect of viscoelastic deformation of the substrate on channel cracks in the film. As discussed above, the presence of an elastic substrate constrains the opening of the crack in the film, and reduces the crack driving force compared to that of a crack in the free-standing film. For a film on a viscous substrate, however, this constraint is gradually lost over time, so that the crack driving force increases; for a sufficient long time, the film is effectively free-standing. This effect will be of particular importance for slow cracks, when the substrate has sufficient time to undergo viscoelastic deformation. Recall that slow cracks are important in practice for long term reliability. Among other consequences, this viscoelastic stress can cause delayed fracture in the film under constant load. Although this viscoelastic effect can be understood qualitatively, there is limited theoretical study (Suo, 2003), and almost no experimental information on the subject.

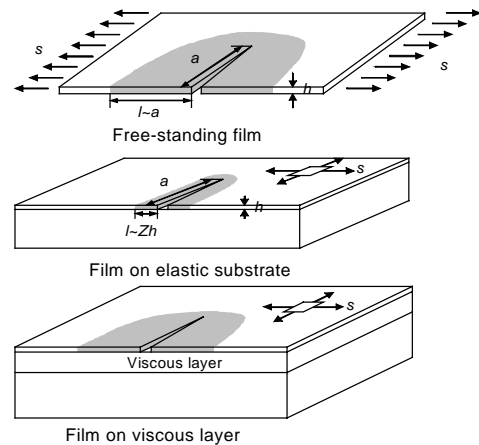


Fig. 3 Effect of constraint of a substrate on a channel crack in a film.

As another example of the possible effect of inelastic deformation of the substrate on cracking in a film, a recent experiment (Cairns & Crawford, 2005) has shown that cracks grow gradually in an ITO film on a polymer substrate under cyclic tensile loading. This observation is puzzling, as a brittle material like ITO is not particularly susceptible to fatigue under cyclic loads. The original paper described some experimental data and observations,

but offered no mechanistic explanations. Our hypothesis is that such cracking in a brittle film is due to the plastic ratcheting deformation of the polymer substrate. The phenomenon is reminiscent of crack growth in a silicon nitride film on an aluminum layer subject to cyclic temperature change (Huang et al., 2000, 2002; Begley & Evans, 2001). Further experiments and calculations are needed to evaluate this hypothesis.

Moisture-assisted cracking

Water (and other reactive molecules) in the environment may participate in the process of breaking atomic bonds along the crack front. Because the reactive molecules need to reach the crack front and to break atomic bonds there, the crack often extends at a velocity much below the sound speed in the material. The crack velocity V is an increasing function of the crack driving force G . In recent years, such V - G functions have been measured for various brittle films (Ma, 1997; Cook & Liniger, 1999; Guyer & Dauskardt, 2004; Lin et al., 2003; Lane et al., 2004).

The V - G function is specific to a given material and its environment. Once determined, the same V - G function applies when this material is integrated with other materials in a structure provided the reactive environmental molecules readily reach the crack front. In such an integrated structure, an observed crack velocity, together with the known V - G function, provides a reading of the crack driving force (He et al., 2004; Tsui et al., 2005). The principle is analogous to that of a mercury thermometer. Mercury expands when the temperature rises; the temperature-volume function can be calibrated. When an amount of mercury equilibrates with a patient, the volume of mercury provides a reading of the temperature of the patient. Furthermore, because the crack driving force depends on properties of materials adjacent to the crack, an observed crack velocity can also be used to determine the properties of materials.

6. Interfacial Debonding

Interfacial debonding has long been a leading failure mode of microelectronic devices (Hutchinson & Suo, 1992; Evans & Hutchinson, 1995; Dauskardt et al., 1998; Volinsky et al., 2002; Suo, 2003; Lin et al., 2003; Vlassak et al., 2005). Adhesion sensitively depends on processing conditions, on materials deliberately added to the interface, and on impurities segregated to the interface. The measurement of the adhesion energy provides a tool for process control and material selection. We expect that many of the same interfacial issues of microelectronics will reappear in macroelectronics. For a macroelectronic system containing both organic and inorganic materials and subject to significant mechanical loads, the following known phenomena deserve special attention. First, an island of an inorganic material on a polymer substrate will develop stress concentration at edges (Hsu et al., 2004)

and may lead to debonding (Bhattacharya et al., 2006). Second, if such an inorganic island is bonded to an organic film of an active device, the large mismatch in their properties may cause damage of the organic layer. Third, a brittle film can survive significant compressive strain in its plane if it is well bonded to the substrate (Gleskova et al., 1999), but will buckle if it is poorly bonded (Cotterell & Chen, 2000; Yu & Hutchinson, 2002).

7. Concluding Remarks

Low-cost, rugged macroelectronics will be a platform for many technologies, such as paper-like displays, medical imaging systems, and thin-film solar cells. These technologies aim to address the essential societal needs for easily accessible information, renewable energy, and a sustainable environment. Macroelectronic systems are large structures of diverse architectures, hybrid materials, and small features. The responses of such structures to mechanical loads pose significant challenges to the fundamental mechanics of structures and materials, as well as to the creation of the technologies. Research in the field of micromechanics of macroelectronics will address concerns raised in these emerging technologies, and shed insights into the working of new architectures of hybrid materials.

Acknowledgements

The work was supported at Harvard University by the National Science Foundation through the MRSEC, and at Princeton University by a DARPA-funded and AFRL-managed Macroelectronics Program (Contract FA8650-04-C-7101). The authors are grateful for many stimulating discussions with members of their research groups. Visits of ZS to the Institute of Process Engineering of Chinese Academy of Sciences have been supported by a project entitled Research Collaboration on Multi-Scale Science of Complex Systems.

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