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Relationship between mechanical damage and electrical degradation in polymer-supported metal films subjected to cyclic loading

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ABSTRACT

A detailed investigation of the correlation between mechanical damage and the change in electrical resistance induced by cyclic tensile loading is performed for polymer-supported gold, copper, and printed silver films. Four distinct types of resistance response to mechanical loading are described and linked to the mechanical properties and microstructure of thin films. It is shown that significant topographical changes induced by cyclic loading can result in minor changes in the overall electrical resistance. Presented results show that an underestimation of mechanical damage can be made when only the electrical failure criterion is used for reliability characterization of polymer-supported metal films.

1. Introduction

Mechanical testing of metals is usually performed until the failure of the sample is observed. For bulk samples or free-standing films failure is a clearly detectable event which is indicated by total fracture or catastrophic plastic deformation of the sample. The situation is different if mechanical reliability of a thin metal film which is deposited on a polymer substrate is investigated. Since polymers are much more elastic than metals, total fracture of a film-substrate system does not always occur and fracture of the film does not lead to a clear drop of applied load due to the substrate. An additional problem is that the polymer substrate prohibits strain localization of the metal film which leads to more complicated mechanisms of damage formation than in free-standing films or bulk samples [1–4]. Clear definition of the failure is especially important for cyclic (fatigue) experiments since it defines the lifetime of the material system.

Failure criteria for cyclically loaded polymer-supported metal films thus far proposed in the literature can be divided in two groups. The first group contains failure detection based on mechanical considerations and topography changes [5–8]. Several different mechanical criteria have been proposed as a failure threshold, such as a saturation of cracks or extrusion density [5], a drop of measured energy loss per cycle [6], an increase of optical scattering by the surface [7], or a point where the metal film does not further contribute to the stiffness of the sample [8]. It is evident that these failure criteria are based on fundamentally different physical processes which also results in large deviation of the lifetime estimation.

The second group provides failure criteria based on electrical resistance measurements of the metal film, typically performed in-situ during the experiment. Using electrical failure criterion a sample is treated as failed if a given threshold value of resistance growth, e.g. 25%, is overcome [9,10]. Another modification of electrical failure detection was proposed in [11,12] where the lifetime is defined by the intersection of two linear parts of in-situ resistance vs. cycle number curve. Electrical failure criterion has important advantages with respect to mechanical criteria because it is more clearly defined and can be applied to both brittle and ductile conductive films. Moreover, it reflects the degradation of electrical conductivity which is the main functional property of polymer metallizations for flexible electronics applications.

Considering electrical failure criterion, it is usually implied that the growth of resistance means the appearance of cracks in the film and if the resistance does not change with the cycle number then the mechanical damage induced in the film is insignificant [9–15]. Such implications can be well justified for brittle films but up to now there are no systematic investigations of the relationship between the electrical degradation and mechanical damage of thin films under cyclic loading.

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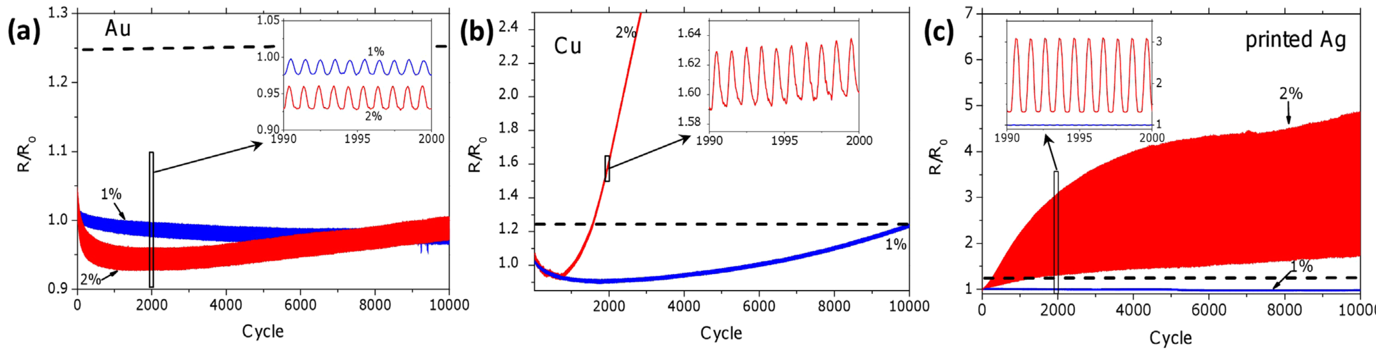

Fig. 1. Dependence of the normalized resistance on the cycle number for (a) 500 nm thick evaporated gold film on polyimide, (b) 500 nm thick evaporated copper film on polyimide and (c) 700 nm printed silver film on PEN. Two curves on each figure correspond to peak strains of 1% (blue) and 2% (red). The insets depict enlarged portions of the corresponding curves between 1990th and 2000th cycles. The dashed horizontal lines show 25% resistance growth which can be considered as the electrical failure.

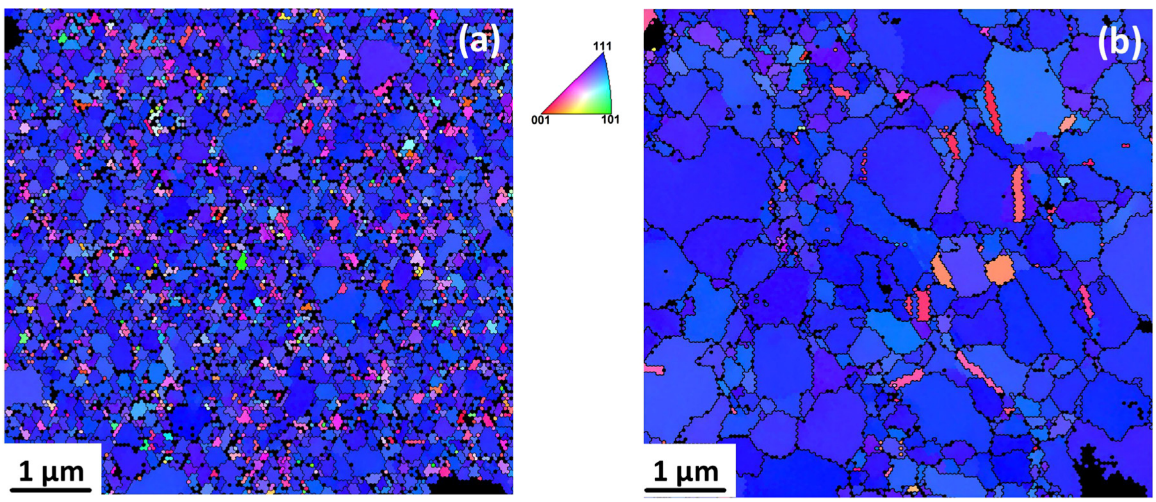

Fig. 2. EBSD crystal orientation maps of 500 nm gold films before straining (a) and after 500 cycles with 2% peak strain (b).

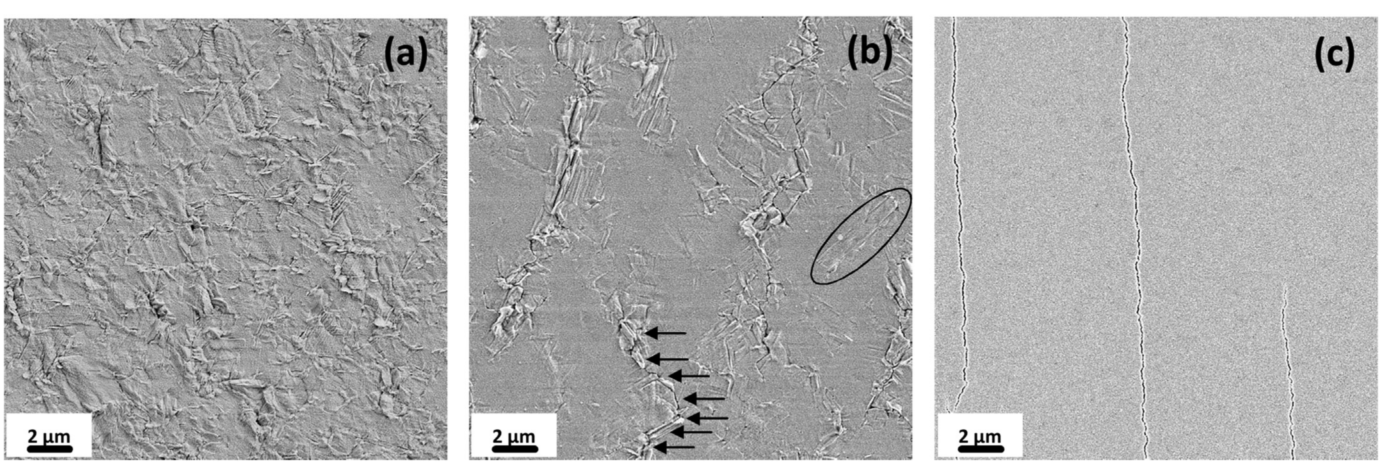

Fig. 3. SEM micrographs of the surface of (a) gold film after 10,000 cycles, (b) copper film after 4000 cycles and (c) printed silver film after 10,000 cycles with 2% peak strain. The arrows and the oval in (b) mark an example of a crack and localized slip band, respectively.

In this paper the correlation between mechanical damage and electrical degradation is analyzed in detail by comparing the cyclic behavior of gold, copper, and printed silver films on flexible substrates. Depending on the film material and initial microstructure substantial mechanical damage can result in minor electrical degradation. Thus, the amount of mechanical damage can be significantly underestimated if only an electrical failure criterion, such as a 25% increase of resistance, is used to determine the number of cycles to failure.

2. Materials and methods

Three different thin film types on polymer substrates are considered. Gold and copper films with a thickness of 500 nm were evaporated onto cleaned Upilex polyimide (PI) substrates (50 µm thick). The initial grain size of the gold films measured by electron backscatter diffraction (EBSD) is (210 ± 60) nm. The copper films exhibit a bi-modal grain size distribution where approximately 50% of the surface is covered by large grains with the average size

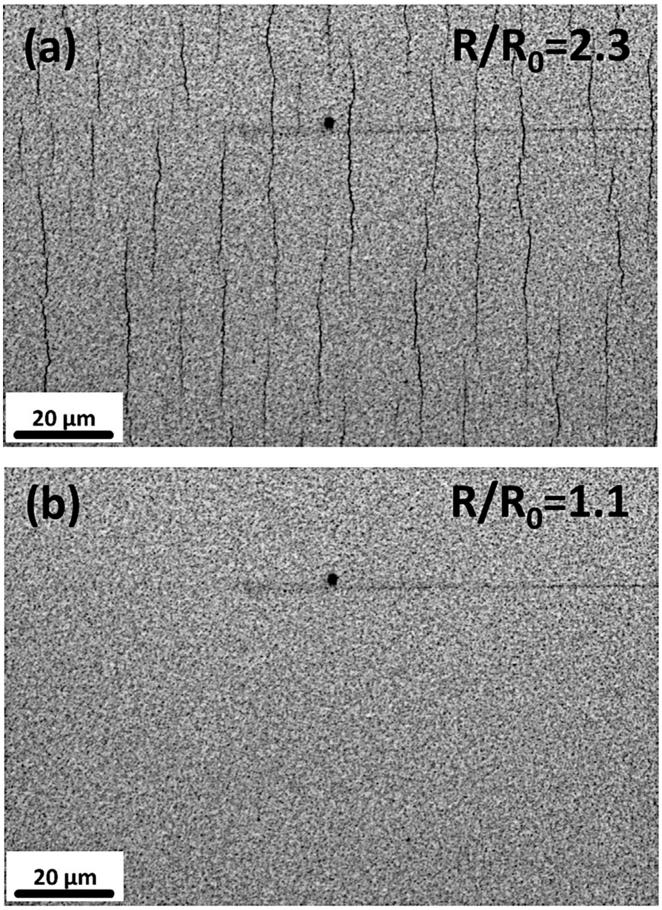

Fig. 4. CLSM images of the printed silver film taken in-situ during cyclic straining at (a) the peak strain of 2% and (b) at zero strain.

of about 1.5 μm and the remaining surface by ultrafine grains with the sizes below 250 nm. Printed silver films with the thickness of 700 nm were inkjet printed using CCI-300 Ag-nanoparticle based ink (particle size 30–100 nm) formulation from Cabot Corporation on a polyethylene naphthalate (PEN) Melinex® substrate (125 μm thick). Cyclic mechanical straining tests were performed using an MTS Tytron 250 tensile testing machine under strain-control. The loading profile was a sine function oscillating between zero strain and a defined peak strain with a frequency of 0.1 Hz. Two different peak strain values of 1% and 2% were applied up to 10,000 cycles. The resistance was measured during straining using a four-point probe geometry and Keithley 2000 multimeter with the electrical contacts incorporated into the sample grips similar to [16]. A set of in-situ straining experiments using an Anton Paar TS600 tensile stage under an Olympus OLS4000 confocal laser scanning microscope (CLSM) were performed on the printed Ag system with holding times of approximately 3 min at maximum strain for the acquisition of CLSM images of the surface.

3. Results

The dependencies of the normalized resistance of the gold, copper, and printed silver films on the cycle number are presented in Fig. 1. Due to the low straining rate each loading cycle is clearly distinguishable in the recorded resistance signal. The insets show enlarged portions of the resistance curves where each maximum corresponds to the defined peak strain and each minimum to zero strain. The three investigated film systems exhibit significant differences in the resistance behavior. Gold films show an initial resistance decrease followed by a small resistance increase for 2% peak strain and a steady resistance decrease for 1% peak strain after 10,000 cycles. Copper films also demonstrate an initial resistance decrease followed by a rapid growth of resistance after 1000 cycles for 2% peak strain and moderate increase of resistance after 2000 cycles with 1% peak strain. For the printed silver and 2% peak strain the resistance starts to grow from the first cycle and at the same time the amplitude of the resistance signal, or the resistance difference between peak and zero strain, becomes larger. For the 1% peak strain, the resistance of printed silver does not significantly change with cycle number even after 10,000 cycles. From these electrical observations, only the copper and printed silver film cycled with 2% would be considered as failed using the 25% increase in resistance criterion of Sim et al. [9,10]. The second proposed electrical failure detection method [11,12] cannot be applied to any of the shown in Fig. 1 resistance curves since they do not exhibit two clear linear parts.

4. Discussion

Four different types of electrical resistance response can be clearly distinguished in Fig. 1. First, the *decrease* of resistance with the cycle number is observed in the curves in Fig. 1a and b. For the gold films and 1% peak strain the resistance remains below the initial value after 10,000 cycles. Second, the growth of the mean value of the resistance without the change of the amplitude is observed (both curves in Fig. 1b, 2% peak strain curve in Fig. 1a). Third, simultaneous growth of the mean value and amplitude is shown in Fig. 1c for 2% peak strain. Finally, no change in both mean value and amplitude after 10,000 is observed in Fig. 1c for 1% peak strain.

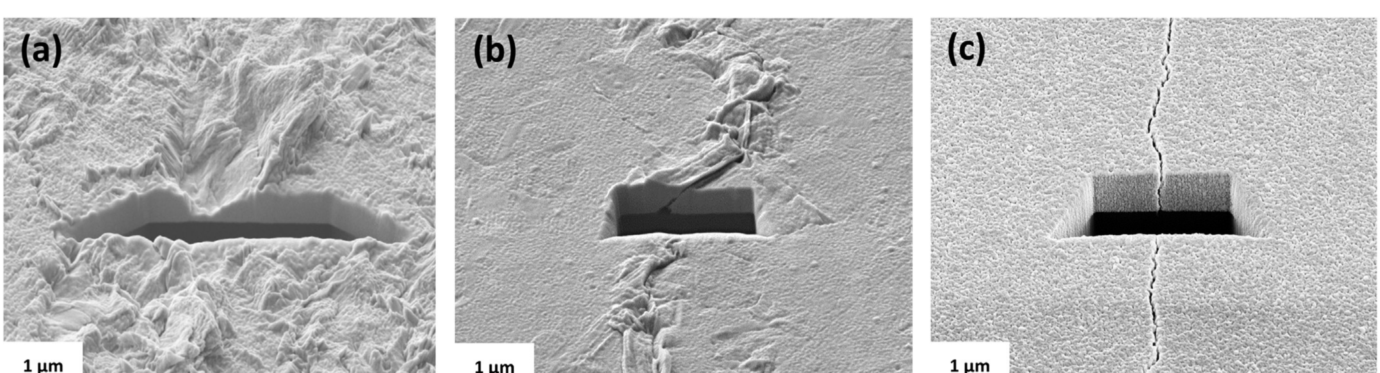

Fig. 5. FIB cross-sections of the (a) gold film after 10,000 cycles with 2% peaks strain, (b) copper film after 4000 cycles with 2% peak strain, (c) and printed silver films after 10,000 cycles with 2% peak strain.

The initial *decrease* in resistance during cyclic loading is generally an unexpected behavior and has not been much discussed in the literature. However, it was recently shown that room temperature strain-induced grain coarsening is responsible for the resistance reduction during the first 1000–2000 cycles in copper films on polyimide [17]. Even stronger grain coarsening is observed in gold films where the average grain size increases from approximately 200 nm to 800 nm after 500 cycles with 2% peak strain (see Fig. 2). It is known that the contribution of grain boundary scattering to overall resistivity becomes significant for the grain sizes below 200 nm but decays to zero if the grains are larger than 500 nm [18–20]. Observed in Fig. 1a and b reduction of the resistance is explained by the elimination of the grain boundary scattering contribution to resistivity as the grain size increases.

The increase of resistance during cyclic tensile loading is usually described by the accumulation of mechanical damage. The scanning electron microscope (SEM) micrographs of the three investigated films after mechanical loading are shown in Fig. 3. The gold film (Fig. 3a) exhibits strong roughening of the whole surface with numerous extrusion-like features. The damage induced in the gold film is delocalized and the entire film surface is plastically deformed, but no clear through thickness cracks are visible. A profound increase of optical scattering from this gold surface can be clearly seen even by the naked eye and thus an optical failure detection method [7] would indicate sample failure, even though the resistance stays well below the 25% threshold. For the copper film (Fig. 3b) interconnected networks of extrusions and cracks (an example is shown by arrows) as well as slip bands (an example is marked by an oval) are observed. In this case, the strain amplitude dictates the lifetime of the film. The larger strain amplitude induces more damage and failure after approximately 1700 cycles compared to the lower amplitude (lifetime of over 10,000 cycles). The printed silver films (Fig. 3c) exhibit long and straight cracks running perpendicular to the straining direction. Except for these cracks no evidence of plastic deformation or other surface modifications are observed which is typical for brittle fracture. The 25% resistance growth threshold is reached after 200 cycles if the resistance at the peak strain is considered and after 1500 cycles for the resistance at zero strain.

The growth of the amplitude of resistance response shown in Fig. 1c for the printed silver films can be explained by the repeated closure and re-opening of cracks during straining. For each loading segment of each cycle the crack flanks are pulled apart from each other which increases the effective current path and, consequently, the measured resistance. During the unloading segments of each cycle the crack flanks connect and the conductivity is partially restored. As evidence for this mechanism a series of in-situ straining experiments using CLSM were performed. The CLSM image in Fig. 4a of a printed silver film at the peak strain (2%) of cycle number 114 is shown. Numerous cracks are clearly visible and the measured relative resistance is around $R/R_0=2.3$. Fig. 4b shows the same area at zero strain, end of cycle 114. No cracks are visible and the in-situ resistance has decreased to $R/R_0=1.1$, which is well below the proposed 25% electrical failure criterion.

In order to better understand the correlation between the electrical degradation and mechanical damage focused ion beam (FIB) cross-sections of the accumulated damage of the three investigated thin film systems are shown in Fig. 5. One can see that despite the strong surface roughening and local thinning of the gold film (Fig. 5a) no through thickness cracks are observed. The absence of through thickness cracks in the gold films can be justified by the high ductility and strain-induced grain coarsening observed during cycling. For the copper film there is a visible crack inside the extrusion (Fig. 5b) indicating that the growth and coalescence of these cracks is responsible for rapid resistance increase in copper films. Printed silver films also exhibit clear through thickness cracks (Fig. 5c) which are responsible for the growth of the mean resistance.

5. Conclusions

A detailed investigation of the relationship between mechanical damage and electrical degradation due to cyclic tensile loading of thin polymer-supported metal films has been presented. Application of cyclic strain with the amplitude of 2% induces strong surface roughening of the gold films but an insignificant growth in resistance. Minor electrical degradation is explained by the grain coarsening and the absence of through thickness cracks. Copper films show interconnected networks of extrusions and cracks which results in rapid resistance increase with the cycle number. Printed silver films demonstrate typical brittle fracture behavior in the form of through thickness cracks. The resistance exhibits a significant increase at the peak strains of each cycle and moderate growth at zero strains of each cycle. This growth of the amplitude of the resistance response was shown to be due to crack closure at zero strain of each cycle. If an electrical failure criteria (resistance increase of 25%) were to be applied to these films, the copper and printed silver, both with 2% strain range, would be considered failed. Despite severe surface roughening, the resistance of the gold films stays well below the 25% resistance increase criteria. What the present analysis illustrates is that electrical failure criterion is mainly sensitive to the mechanical damage which induces clear topological discontinuities within the film which are oriented perpendicular to the current flow, such as through thickness cracks. If only strong surface roughening, as in the case of the gold film, is observed or if the cracks can re-connect upon unloading, such as with the printed silver, then electrical degradation does not adequately reflect the amount of mechanical damage. Therefore, failure criteria for metal-polymer systems should be a combination of electrical degradation and mechanical damage density, but also be cognizant of microstructural phenomena as well as possible crack closure which can increase the measured lifetime.

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