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Effects of stretching and cycling on the fatigue behavior of polymer-supported Ag thin films

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A B S T R A C T

The fatigue behavior of silver films on polyethylene-terephthalate substrates is studied for various levels of film thickness, pre-stretch, sample width, and applied strain range. Films with large pre-stretch have a shorter fatigue life, with failure caused by strain localization (films thicker than 100 nm) or intergranular crack formation (100 nm film). There is a significant effect of film thickness on how the strain range affects the fatigue life – we observe ‘smaller is better’ behavior for films subjected to a total strain range of $\Delta\varepsilon_t = 1.0\%$, while the opposite is true when the total strain range increases to 2.0%. We attribute this difference to a shift in failure mechanism with strain amplitude from typical fatigue failure to a more ductile-type failure. Our experimental results are well described by the Coffin–Manson relationship and a failure mechanism map is drawn based on the experimental results. Considering stretchability and long-term reliability, design suggestions are made to optimize the fatigue life of coatings subjected to uniaxial stretch and fatigue.

1. Introduction

Technological demands for low weight, low cost, and large-area integration have driven the development of stretchable, flexible electronic devices. These devices commonly use polymer materials as substrates, which have the advantages of low density and enhanced stretchability compared to silicon (Si) substrates. The use of polymer substrates has resulted in new applications including flexible displays [1,2], rollable solar cells [3], electrical sensors [4], and polymer MEMS [5]. Flexible electronic devices are often hybrid devices that consist of inorganic films of semiconductors, metals, or ceramics deposited on a polymer substrate [6–8]. While polymer substrates may survive during large and repetitive elongation, the inorganic films fail much more easily. If a metallic layer connecting functional ceramic islands on a polymer substrate fails during operation, loss of electrical connectivity occurs, resulting in device failure. In this context, large stretchability and long-term reliability of metallic films on polymer substrates are key requirements for flexible electronics.

Recent studies have focused on characterizing the mechanical behavior of thin metal films and have observed that metals in thin-film form can support much higher stresses than their bulk counterparts [9–14], a phenomenon often referred to as the ‘size effect’. This strengthening has generally been attributed to dimensional and microstructural constraints on dislocation activity in thin films [10]. Furthermore, theoretical and experimental studies of the mechanical behavior of ductile metal films on compliant substrates have demonstrated that the stretchability of thin metal films can be enhanced to strains as large as 70% without failure of the film [15–21].

The fatigue behavior of freestanding ductile metal wires, sheets and thin films has been investigated with special attention paid to the difference between these materials and their bulk counterparts [22–26]. The difficulty of testing submicron freestanding metal thin films, and the increasing interest in flexible electronics have led to studies of polymer-supported metal thin films [27–32]. These studies have observed differences in fatigue damage morphology between thin films and bulk materials, and have demonstrated effects of microstructural length scales on fatigue strength. Few studies have investigated the effect of the substrate constraint on the fatigue behavior of thin films. Given that the substrate constraint strongly enhances the stretchability of ductile metal thin films deformed in uniaxial tension experiments [18–21], one might also expect an effect on fatigue life. Previous work on polymer supported Ag thin films [33] demonstrated an increase in fatigue life for coatings with improved adhesion to the substrate. Two types of failure mode were reported: typical fatigue failure with extrusion–intrusion pairs (type I) and ductile fatigue failure with local necking (type II). These observations have motivated further work on the fatigue behavior of metal thin films on compliant substrates.

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In this paper, we present a comprehensive experimental study of the effects of strain range, film thickness, pre-stretch, and sample width on the fatigue life of thin Ag films supported by PET (polyethylene-terephthalate) substrates. The effect of pre-stretch is discussed in terms of ductility and substrate constraint, considering both uniaxial stretching and cyclic loading. We observe 'smaller is better' for films subjected to small strain amplitudes, while the opposite behavior is observed for large strain amplitudes. Design suggestions to enable films with large stretchability and long fatigue life are made.

2. Experimental

The polymer substrates used in this study were 12 μm thick PET foils with an acrylic primer (Skyrol® SH21 by SKC) grafted on one side for improved adhesion. Our previous research [21] demonstrated that PET substrates with acrylic primer layer have better adhesion. These PET substrates were patterned into rectangular shapes with a gauge length of 16 mm and a width of 300 or 1000 μm using a cutting plotter (Graphtec FC8000). Ag films with thicknesses of 100, 200, 400, and 800 nm were deposited on the PET samples at a rate of 4 Å/s by means of electron-beam evaporation (KVET-C500200). Grain sizes of the as-deposited films were measured using the linear intercept method with twins counted as separate grains. The grain sizes were determined to be 96 ± 26 nm for the 100 nm films, 99 ± 13 nm for the 200 nm films, 123 ± 10 nm for the 600 nm films, and 120 ± 10 nm for 800 nm films.

Fatigue tests were carried out using a custom-built mechanical fatigue tester [33] with in situ resistance monitoring. The fatigue tester consists of an electro-dynamic, vertical axial-loading machine with a laser displacement sensor (Keyence LK-G30). A Keithley 2700 multimeter in a four-wire measurement setup was used for in situ measurements of the electrical resistance of the sample during cyclic loading. All tested samples showed an initial resistivity of $(2.51 \pm 0.28) \times 10^{-8}\ \Omega$ m. Resistance measurements were generally extremely reproducible.

Before cyclic loading, samples were subjected to tensile strains of 2, 10, and 15% in order to evaluate the effect of pre-stretch on their fatigue behavior. These values were chosen to avoid buckling of the sample during cyclic loading and to evaluate the effect of severe plastic deformation prior to fatigue testing. Cross-sectional SEM images in a previous study [33] and in this study showed that initiation of cracks during pre-stretch increased with decreasing film thickness and increasing applied strain [20,21].

Fatigue cycling was carried out under total strain range control at a frequency of 1 Hz. The strain range ($\Delta\varepsilon_t = 1.0$, 1.25, 1.5, 1.75, 2.0, and 2.25%) was varied to establish the relationship between strain range ($\Delta\varepsilon_t$) and fatigue life (N_f). To confirm repeatability, each experiment was repeated two to five times depending on the duration of the experiment. Fatigue results were very reproducible and figures in this paper show representative data for each test. After testing, samples were attached to a rectangular (3 mm × 14 mm) Si block prior to unloading to prevent any cracks from closing up and to make it easier to observe the failure morphology. Surfaces and cross-sections of the deformed Ag films were observed using an FEI scanning electron microscope (SEM) and an FEI dual-beam focused ion beam/scanning electron microscope (FIB/SEM). Platinum was deposited on the surface of the Ag films to protect them during ion milling.

3. Experimental results

3.1. Resistance increase induced by film fracture

Fig. 1(a) shows typical resistance-cycle curves for polymer-supported Ag thin films. In order to determine the fatigue life of each film, we adopt a failure criterion that relates the electrical resistance to the structure of the metal film [33]. Observations of the film surface and film cross-sections have shown that the resistance increase is closely associated with crack formation and growth. In this context, the increase in the resistance of the film can be used as a fatigue failure criterion. Based on prior observations [33], a resistance ratio $R/R_0 = 1.25$ was selected as the fatigue failure criterion, because at this point cracks start to propagate. Fig. 1(b) depicts a typical SEM image of the surface topography at $R/R_0 = 1.25$, showing extrusion chains and sites of local necking.

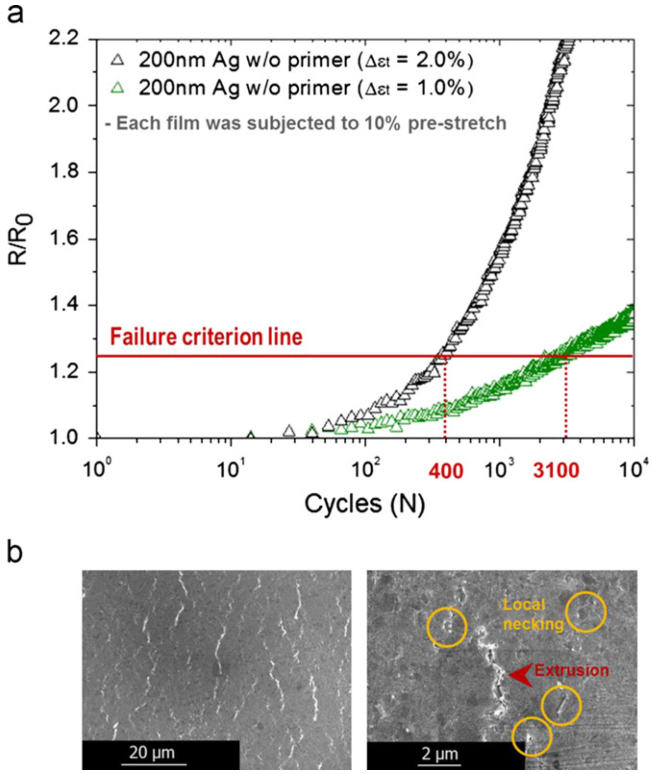

Fig. 1. (a) Normalized resistance R/R_0,where R_0 designates the initial resistance of the film before cycling, as a function of the number of fatigue cycles for two different strain ranges. (b) SEM image of the surface microstructure at $R/R_0 = 1.25$. Right image shows a magnified view.

3.2. Effect of pre-stretch

In a first series of experiments, 400 nm Ag films on bare PET were tested using a strain range $\Delta\varepsilon_t$ of 1.0%. Before cyclic loading, these films were subjected to a pre-stretch of 2, 10, or 15%. The normalized resistance R/R_0, as a function of the number of fatigue cycles, and the resultant fatigue life, are shown in Fig. 2. These results show that the pre-stretch has a significant adverse effect on the fatigue life of the coatings.

3.3. Effect of film thickness

In a second set of experiments, Ag films with various film thicknesses (100, 200, 400, 800 nm) deposited on PET substrates with primer were tested. Prior to cyclic loading, all films were stretched by 2%. Fig. 3(a) shows the normalized resistance curve and fatigue life of these films subjected to a total strain range $\Delta\varepsilon_t = 1.0\%$. The figure shows a trend of increased fatigue life with decreasing film thickness (except for the 100 nm films), consistent with results obtained by other researchers [27–31,34]. The decrease in the fatigue life of the 100 nm films is related to a