

Thickness dependent critical strain in Cu films adherent to polymer substrate

R. M. Niu, G. Liu, C. Wang, G. Zhang, X. D. Ding, J. Sun^{*}

State Key Laboratory for Mechanical Behavior of Materials and Multi-disciplinary

Materials Research Center, Xi'an Jiaotong University, Xi'an, 710049, China

Abstract — For the polymer-supported metal thin films that are finding increasing applications, the critical strain to nucleate microcracks (ε_C) should be more meaningful than the generally measured rupture strain. In this paper, we develop both electrical resistance method and microcrack analyzing method to determine ε_C of polymer-supported Cu films simply but precisely. Significant thickness dependence has been clearly revealed for ε_C of the polymer-supported Cu films, i.e., thinner is the film lower is ε_C . This dependence is suggested to cause by the constraint effect of refining grain size on the dislocation movability.

* Corresponding author: junsun@mail.xjtu.edu.cn

Metal thin films are extensively used in interconnects in multilevel metallization of ultralarge scale integrations. As the dimension of integrated circuits continues to shrink, it is becoming ever more crucial to understand the mechanical properties of metal thin films with size in submicron scale [1], in order to ensure the reliability of interconnects and favor the design and fabrication of integrated circuits. The strength of metal films, as well known, has been attracted numerous investigations and some measurement methods, such as tensile testing, indentation, microbeam bending, and bulge testing, have been developed [2-5] to determine the yield strength of films. Recently, the method of tensile testing the metal thin films on compliant substrates [6,7] has been also suggested to determine the yield strength of metal films, where the force of highly elastic compliant substrates could be subsequently subtracted from the total measured force of the compound to yield the real one of films. This provides a more precise measurement on yield strength for thin film used especially as flexible electronics that are being widely used in many applications, including paper-like display [8] and sensitive skins [9].

Besides the yield strength, ductility is another important mechanical property of the metal films. Measurement on freestanding films by using simple tensile test could provide result more closer to intrinsic ductility of the metal films, although this method involves too much complicated sample preparation and handling and is also not applicable for the films with thickness less than about 3 μm . When the freestanding metal films subject to a tensile strain, the films can deform plastically but can not harden as their bulk counterparts because the dislocations in metal films are ready to escape due to the limited thickness constraint. This means that a neck that is induced preferentially at a local spot could cause further

intense localized deformation, resulting in fast rupture. In other words, the rupture strain of freestanding films is close to the strain needed to nucleate microcrack or neck, due to the low hardening capability and small thickness-to-length ratio of the films. This is responsible for the previous reports [10,11] that the rupture strain of freestanding films is much lower than the corresponding bulk metals, such as only 0.6 % for 5.77 μm -thick Au film and only 1.3 % for 4.28 μm -thick Cu film [10].

On the other hand, when the metal film is deposited on a polymer substrate, the strain localization in film could be suppressed by the substrate due to geometrical constraint. Both theoretical simulations [12] and experimental investigations [13-15] have clearly shown that the polymer-supported metal, if well bonded to the substrate, could deform uniformly to a large strain. Two methods have been used to estimate the deformation capability of the films. One is to observe the microcracks in the films with applied strain. For example, it was found [13] that the Cu films well bonded on polymer substrate could sustain strain up to 10 % without appreciable cracks and up to 30 % with only discontinuous microcracks, far larger than its freestanding counterpart. The other one is to make real-time examinations on the change in electrical resistance of films because the damage or failure in microstructure could be sensitively revealed by the macroscale property of electrical resistance. For example, Cr thin films, deposited on polymer substrates with granular In layer as buffer layer, were found [15] to sustain strain up to 38 % with electrical resistance changed by a factor of less than 2. These crude measurements with no comparability between each other, however, were qualitatively used to present how the rupture strain of the films could be achieved. Actually in the polymer-supported metal films, the rupture strain is much larger than the critical strain

(ε_C) for nucleating microcrack or neck, and ε_C should be more meaningful for applications because this parameter indicates the begin of damage in microstructure and weakening in serve properties. Besides, the determination of ε_C could make the different measurements comparative. These make the systematic works on ε_C urgently needed especially for the applications in electronic fields, where the lifetime prediction of components is very important in order to avoid sudden failure. In this paper, we will develop two methods, i.e., electrical resistance method and microcrack analyzing method, to determine ε_C of polymer-supported Cu films with different thicknesses. The good agreement between the results measured by the two methods indicates that the intrinsic ductility of the polymer-supported metal films could be precisely determined using present measurements.

The Cu films used in present work, with thickness range from 700 nm down to 60 nm determined by Rutherford backscattering, were deposited on dogbone-shaped polyimide substrate (4 mm \times 20 mm) by magnetron sputtering. The sputter current was 0.45 A and the bias voltage - 80 V, which resulted in a deposition rate of about 5.9 nm/min. This low-energy slow deposition rate was experimentally determined to have almost no effect on the mechanical properties of the 125- μ m-thick polyimide due to its good temperature-endurance. Prior to deposition, the polyimide was firstly cleaned by Ar ion bombardment. The as-deposited films were in-situ annealed at 100°C for two hours to eliminate the residual stress. X-ray diffraction (XRD) measurements revealed that all the Cu films were polycrystalline and no obvious texture was found even in the thinnest 60-nm film. Based on quantitative microscopy, the average grain size of the films was determined using

transmission electron microscope (TEM).

Uniaxial tensile testing was performed using a MTS microtensile tester with a 50-N force transducer at a constant strain rate of $1 \times 10^{-4} \text{ s}^{-1}$. The force and displacement/strain were automatically recorded by machine and a high-resolution laser detecting system, respectively. In testing the Cu film/polyimide systems, the measured total force on the systems is a sum of forces on both Cu film and polyimide substrate. In order to obtain the intrinsic strain-stress curve of Cu films, the force on the substrate should be subtracted from the total one [6]. Therefore, pure polyimide with the same width and length as those of the Cu film/polyimide system was also tested and the strain-force data were recorded for subtracting. When the intrinsic strain-stress curve of Cu films is obtained after subtraction, the yield strength is determined as the 0.2 % offset.

In order to determine the critical strain (ε_c) for nucleating microcrack or neck, both electrical resistance method (ERM) and microcrack analyzing method (MAM) were employed for comparison. During tensile testing, the real-time electrical resistance (R) of the films was measured and the relative change in resistance ($\Delta R / R_0$, where $\Delta R = R - R_0$ and $R_0 = R(\varepsilon = 0)$) was subsequently depicted with respect to strain (ε). Two distinct regions could be clearly seen in the $\Delta R / R_0$ vs ε curves. In the first region, the films will elastically deform with conserved volume. However, the change in form (lengthening, decrease of cross-section) leads to a change in $\Delta R / R_0$ with strain. Because no damage exist, $\Delta R / R_0$ increases linearly with ε in this region. With the further increase in strain, damage will be induced and the existence of microcracks and voids will increase the electrical resistance significantly, causing the $\Delta R / R_0$ vs ε curve deviate from the linear

stage and go into the second nonlinear stage. The critical strain, where the $\Delta R/R_0$ vs ε curves transform from linear first stage to the nonlinear second stage, is defined ε_c in the electrical resistance method. On the other hand, in the microcrack analyzing method, the density of microcracks (total length of the microcracks per unit area, denoted as S) in the films was ex situ determined after different strain testing. Extrapolation treatment was subsequently employed to put the S vs ε curves back to the point of $S = 0$, where the strain was regarded as ε_c . The physical meaning of ε_c in the microcrack analyzing method is that, at this critical strain, microcracks begin to form. Fig. 1 typically shows the illustrations of the two methods for determining ε_c .

The measured thickness dependence of grain size, d , and yield strength, σ_y , of Cu films are shown in Fig. 2. The grain size is found to decrease with decreasing film thickness, t , while the yield strength enhances with decreasing t . These trends are consistent well with previous results [16-18]. Moreover, the values of yield strength we determined for Cu films are found to be close to those determined either from freestanding Cu films or from Cu film/polyimide systems [6]. The strong dependence of yield strength on Cu film thickness of submicron sizes has been suggested [6] as a result of thickness effect that could be quantitatively described by strain gradient plasticity theory [19]. Here in our experiments, the yield strength was found to be more dependent on the grain size, which follows the expression of $\sigma_y = \sigma_0 + k(d)^{-0.5}$, where σ_0 is the lattice friction stress (116 MPa for Cu), and k is a constant of $362 \text{ MPa } \mu\text{m}^{0.5}$.

Fig. 3 (a) typically shows some $\Delta R/R_0$ vs ε curves for Cu films with different thicknesses. One can find that the thicker films should undergo somewhat longer linear first

stage than the thinner ones, which means that the thicker films should have larger ε_C . In Fig. 3 (b), the ERM-determined ε_C is depicted with respect to film thickness as square dots, where ε_C exhibits a significant thickness dependence. This dependence, i.e., ε_C decreasing with film thickness, is just contrary to that of yield strength (Fig. 2), similar to the general law that strength and ductility are mutual exclusive. The constrain effect of fine grains on the dislocation movability may be responsible for the present thickness dependence of both σ_y and ε_C .

Also shown in Fig. 3 (b) is the MAM-determined ε_C with respect to film thickness (circle dots). The MAM-determined ε_C is found to be very close to the ERM-determined ε_C , which could be reasonable explained because the variation in macroscopic property of electric resistance is essentially induced by the change in microstructure and so the ERM-determined ε_C should be essentially equal to the MAM-determined ε_C . This indicates that the two methods, complementing each other, are both applicable and feasible to determine the critical strain of supported metal films. This should have significant effects on industrial applications.

From Fig. (a), it could be further revealed that thinner films exhibit much more increase in $\Delta R/R_0$ than the thicker films, especially in the second nonlinear stage. This is also related to the microstructural evolution. As seen in Fig. 4 where the respective microstructural evolution of 60 nm-, 340 nm -, and 700 nm -thick Cu films are typically presented, although all the three films show increasing microcracks with strain, more dense microcracks or large crack density are observed in the thinner films at any strain. Besides, the microcracks in the 60 nm-thick films are apt to traverse the entire gauge width of the

specimens, while in the thicker films such as the 700 nm-thick films, the microcracks are not only much less but also much shorter even at the strain up to 20 %. The lower hardening capability of the thinner films, caused by the less dislocation accumulation, should be responsible for the easy propagation of the microcracks.

In thicker films that bear certain hardening capability, the microcracks will propagate along regularly oriented direction lines with angles of about $\pm 45^\circ$ toward the loading direction. In tension, the lines are shaped like furrows. A microcrack is generally generated at a point of intersection of two deformation lines and the microcrack then propagate along a line until it connects another microcrack. This propagation process results in a zigzag form of the cracks, as commonly seen in the thicker films at somewhat low strain (e.g., in Fig. 4 (b) at strain of 20 %).

Finally, it should be necessary to address that, in present work, the thicker Cu films also have excellent rupture strain. For example, the electric resistance of 700 nm-thick film increases by less than 20 %, even as the strain up to 20 %. This indicates that the present Cu films are well bonded to the polymer substrates.

In summary, we deposited Cu films with thickness range of 60 – 700 nm on compliant polyimide and investigated the thickness dependent critical strain by using both electrical resistance method and microcrack analyzing method. The two methods yield similar results that the critical strain decreases with thickness, contrary to the dependence of yield strength on film thickness. The two methods are applicable and feasible to determine the critical strain of supported metal films.

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Figure Captions

Fig.1 Illustrations for determining the critical strain, ε_C , by using both electrical resistance method and microcrack analyzing method.

Fig. 2 Thickness dependence of grain size and yield strength for the Cu films.

Fig. 3 (a) Dependence of electrical resistance change ($\Delta R / R_0 = (R - R_0) / R_0$) on strain for different thick films; (b) Thickness dependent critical strain determined using both electrical resistance method and microcrack analyzing method.

Fig. 4 Microstructural evolution with strains in (a) 60 nm-thick, (b) 340 nm-thick, and (c) 700 nm-thick Cu films.

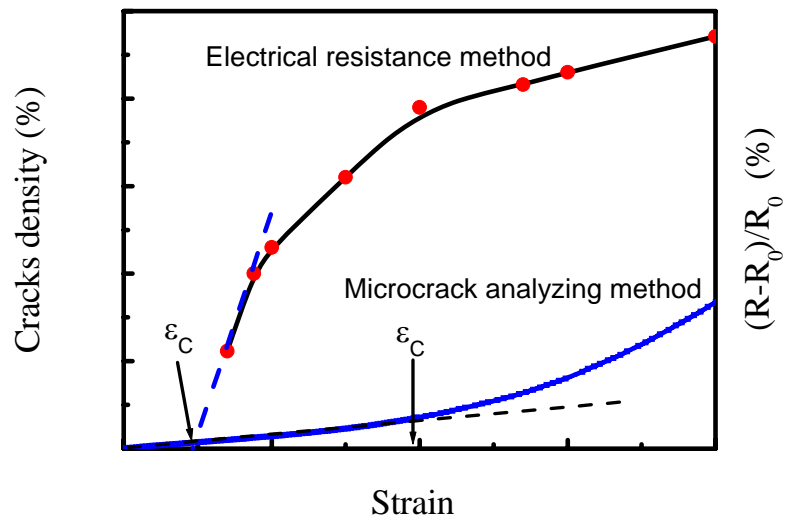


Fig. 1 Niu et al.

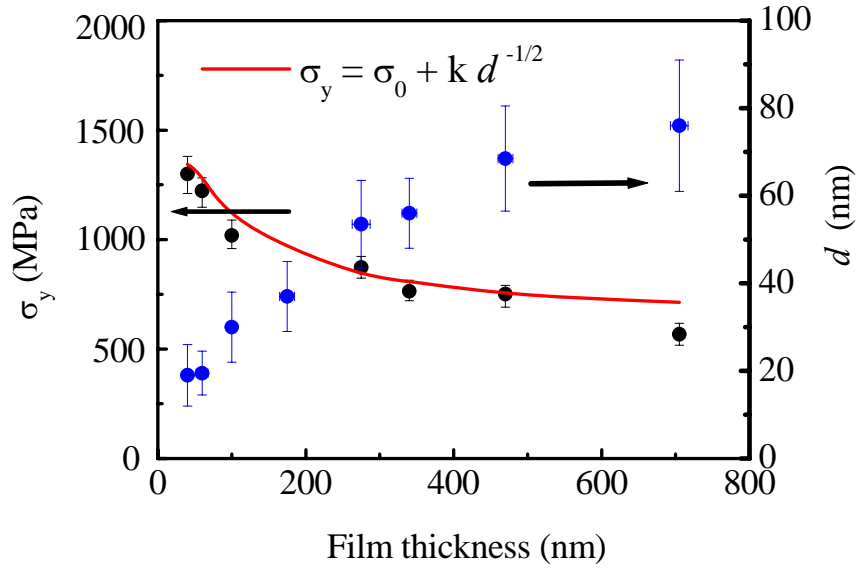


Fig. 2 Niu et al.

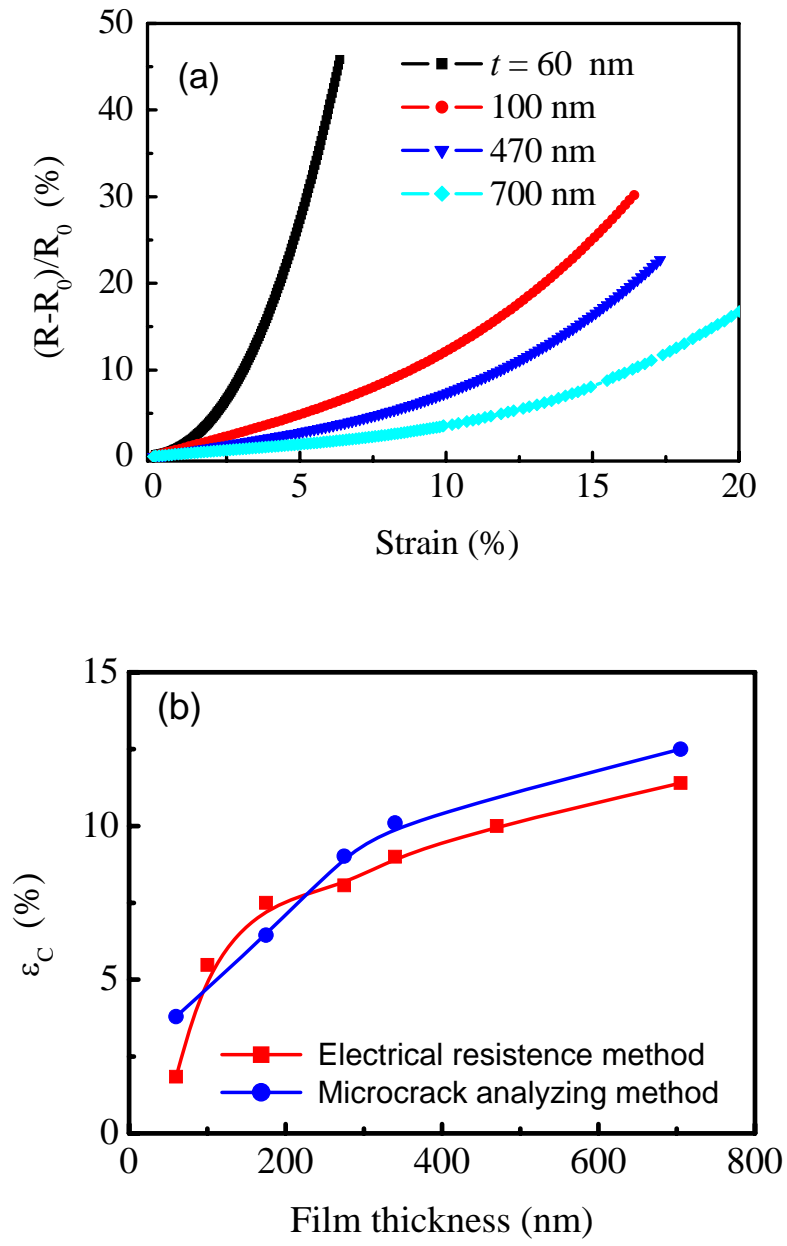


Fig. 3 Niu et al.

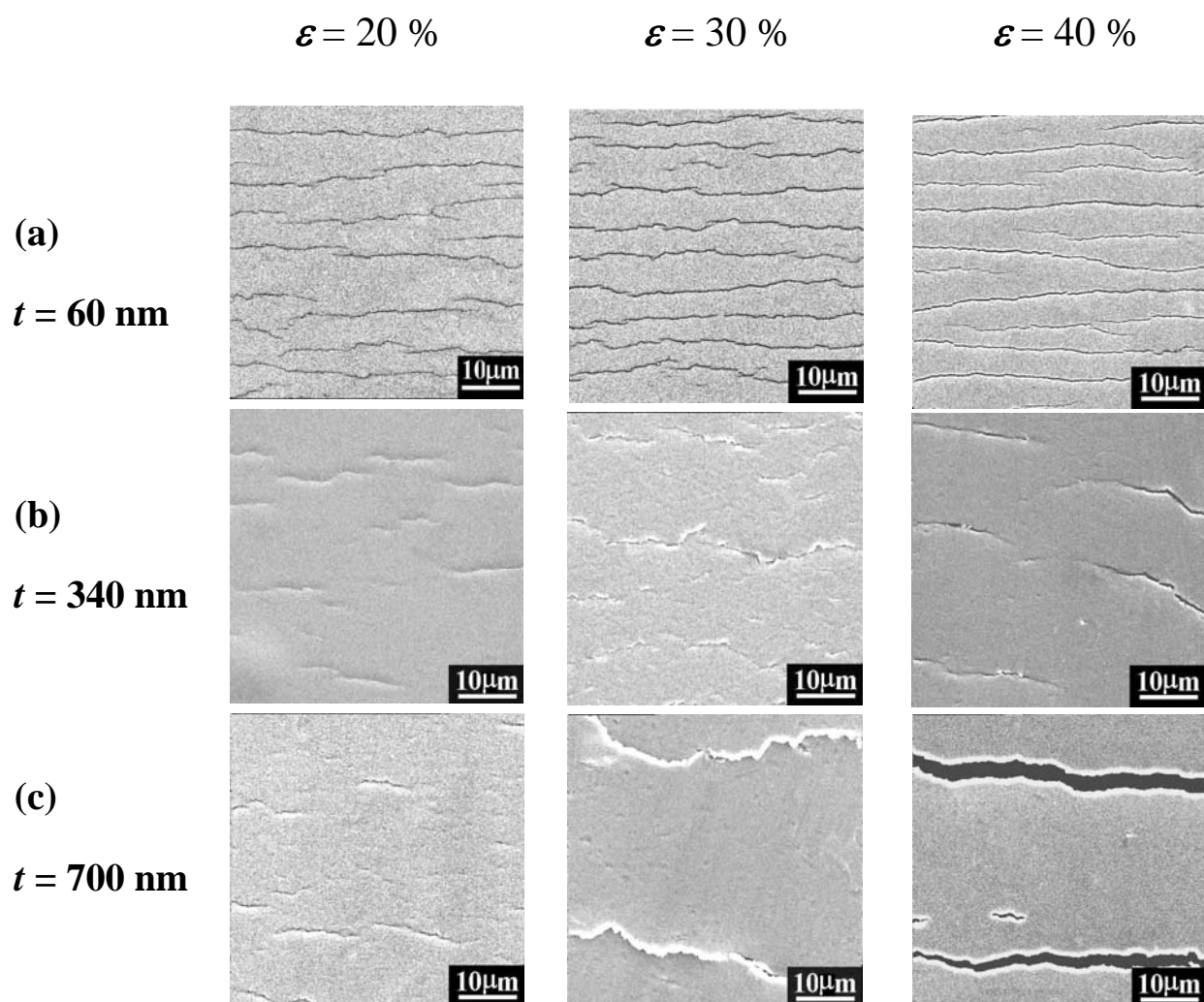


Fig. 4 Niu et al.