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Scaling of the fracture toughness of freestanding metallic thin films with the yield strength

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ABSTRACT

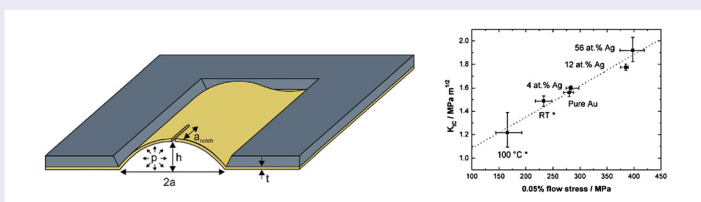
The influence of the yield strength on the fracture toughness of freestanding metallic films with a thickness of ~ 150 nm was investigated by bulge testing. For this purpose, gold films prepared by thermal evaporation were tested at room temperature and at 100°C . Additionally, different Au-Ag solid-solution strengthened films were studied. The fracture toughness of the films is observed to increase with increasing yield stress. This is at first sight counterintuitive but can be explained by previously observed severe necking leading to a chisel-point type of fracture in freestanding metallic thin films.

ARTICLE HISTORY

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KEYWORDS

Fracture toughness; thin films; AuAg solid solutions; bulge test



IMPACT STATEMENT

Freestanding metallic thin films exhibit an extremely low fracture toughness. It is shown in this publication that K_{IC} can be significantly improved by increasing the yield strength of the films.

1. Introduction

Recent studies have shown that the fracture toughness of metallic thin films is about an order of magnitude lower than the fracture toughness of bulk materials [1–4]. It is likely that there is an intrinsic thickness effect on fracture toughness [4] as first suggested in the 1960s by Bluhm [5]. However, the abnormally low fracture toughness of the sub-micrometre metallic films is usually ascribed to their remarkably high strength [1,3,6], which is mainly a consequence of their fine microstructure [7–9]. This interpretation might hold for brittle materials but appears to be in direct contradiction with the common opinion that high strength usually results in high brittleness in metals. In opposition, the model of Knott predicts an increase of the fracture toughness with increasing yield strength for thin sheets under plane stress. It assumes a local mode-III fracture by sliding at an angle of 45° [10]. According to this model the fracture toughness K_{IC} is proportional to the square root of the yield stress σ_y with

t the sample thickness and E its Young's modulus:

$$K_{IC} = \sqrt{2tE\sigma_y} \quad (1)$$

This directly conflicts with the assumption that the higher yield strength of thin films could be the reason for their low fracture toughness. Although the number of studies focusing on the fracture properties of thin films and microscale specimens has strongly increased during the last years, the correlation between yield strength and fracture toughness has not yet been investigated systematically. The present study aims at elucidating the yield strength effect on fracture toughness for metallic thin films. For this purpose, the mechanical properties of freestanding gold thin films with columnar microstructure and similar thickness were characterized by bulge testing. The yield strength of the films was varied by two approaches: First, by increasing the testing temperature from room temperature to 100°C , and second by alloying with up to 56 at.% silver.

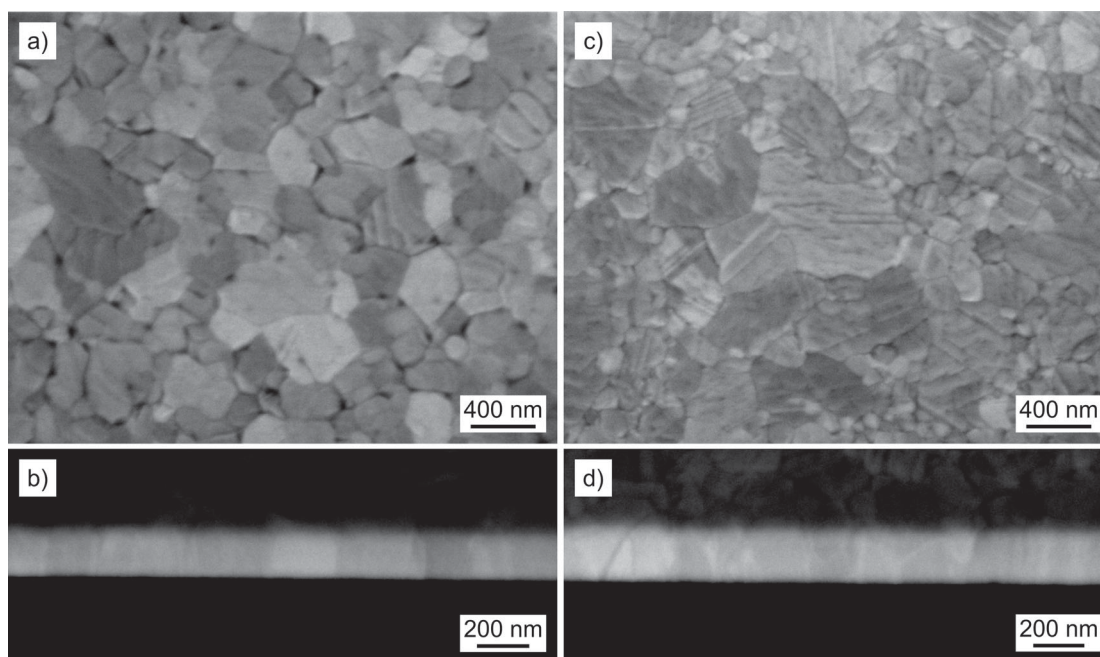


Figure 1. Top-view and cross sectional secondary electron micrographs of a pure gold reference film in (a) and (b) and a gold film containing 56 at.% silver in (c) and (d).

2. Materials and methods

2.1. Preparation of thin film samples

Pure gold films and AuAg films were produced by thermal evaporation onto sacrificial 100 nm thick and $1 \times 4 \text{ mm}^2$ large nonstoichiometric silicon-rich silicon nitride (SiN_x) membranes produced by low-pressure chemical vapour deposition (LPCVD) and supported by 525 μm thick silicon frames (Silson Ltd., Southam, UK). After deposition of Au or AuAg with a custom-built thermal evaporation unit, the SiN_x substrate layers were selectively removed by reactive ion etching (RIE) for 3 min by applying a 30 sccm flow of CF_4 at a pressure of 12.67 Pa and a RF power of 100 W.

The pure gold films (later used for tests at different temperatures) were deposited by thermal evaporation at a rate of 0.6 Å/s and at a substrate temperature of 120°C to a thickness of 100 nm. The average in-plane grain size of these films was about 150 nm, and the roughness average $R_a = 3.2 \text{ nm}$.

Three batches of AuAg solid-solution hardened films were prepared by co-evaporation of gold and silver. Additionally, a batch of pure gold reference films was deposited using the same parameters. The total deposition rate for all sample batches was kept constant at about 8 Å/s . During deposition, the substrate temperature was kept between 80°C and 86°C . Directly after deposition, the films were annealed for 70 min at 120°C under high vacuum. The thickness of the films ranged from 130 to 180 nm due to an imperfect control of the stoichiometry

during deposition, which led to a bias during in-situ thickness monitoring. Figure 1 shows a top-view electron micrograph and a FIB cross section of a pure gold reference film (a and b) and of a film with the highest silver content of 56 at. % (c and d). Both films exhibit a columnar microstructure and their mean grain size is in the order of the film thickness. The density of twin boundaries is much higher in the film with the highest silver content. The roughness average was $R_a = 2 \text{ nm}$ for all films.

Table 1 summarizes the most important properties of the six sample batches. The composition of the AuAg films was determined by energy-dispersive X-ray spectroscopy (EDX) after deposition. The film thickness was measured by AFM scanning across a previously introduced scratch reaching down to the substrate. The grain size was evaluated by the linear-intercept technique from several plane-view electron micrographs collected after the films had been released from the SiN_x carrier membranes by RIE.

2.2. Determination of the yield strength and fracture toughness by bulge testing

The mechanical properties of the films were measured by bulge testing on high aspect ratio rectangular membranes, as schematically shown in Figure 2. For this membrane geometry, the stress and strain along the short membrane axis are uniform and can be analytically determined from the applied pressure p , the resulting

Table 1. Properties of the six investigated sample batches (mean values). All specimens but ‘100°C Au’ were tested at room temperature. Specimens marked with asterisk were tested with the secondary setup designed for high temperature operation (see below).

Sample batch	Film thickness / nm	In-plane grain size / nm	Yield strength / MPa	Fracture strain / %	Fracture toughness / MPa.m ^{1/2}
pure Au	151	240	280	0.45	1.56
Au 4% Ag	134	294	283	0.47	1.60
Au 12% Ag	128	201	385	0.35	1.78
Au 56% Ag	183	236	398	0.31	1.92
pure Au *	103	148	233	0.66	1.49
100°C Au *	103	148	166	0.89	1.22

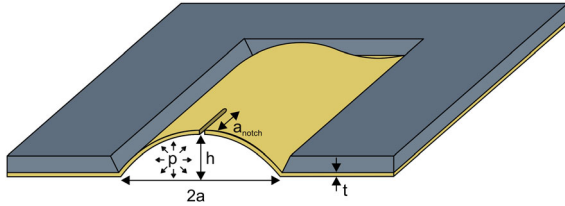


Figure 2. Bulge testing of a rectangular membrane that contains a centred through-thickness notch for determination of the fracture toughness.

deflection h in the centre of the membrane, and the membrane width a and thickness t [11]. Before the stress and strain were evaluated, the deflection data were corrected by subtraction of the machine contribution and by numerical adjustment of the initial height value. Latter was performed by fitting the bulge equation to the initial pressure-displacement data associated with the elastic deformation of the membrane as proposed by Small and Nix [12]. In order to determine the yield strength of the films, the obtained stress-strain curves that represent the mechanical behaviour in a plane-strain state were converted to uniaxial stress-strain curves. For this purpose, it was assumed that the material obeys the J_2 flow theory and the von Mises equivalent stress and plastic strain were calculated as suggested by Nix and Xiang [11]. The yield stress was finally taken as the equivalent von Mises stress at a plastic strain of 0.05%. For the determination of the fracture toughness, a 150 nm wide and 10 μ m long through-thickness notch was introduced into the centre of each membrane by focused ion beam (FIB) milling using an acceleration voltage of 30 kV and a Ga^+ current of 10 pA. Analogous to references [13] and [14] the fracture toughness K_{IC} was calculated from the fracture stress $\sigma_{xx, \text{fracture}}$ and the half length of the notch a_{notch} according to

$$K_{IC} = \sigma_{xx, \text{fracture}} \sqrt{\pi a_{\text{notch}}} \quad (2)$$

The main bulge-test setup used for testing the AuAg films is described in detail in reference [15]. For the tests at 100°C, a secondary setup with a larger pressure range (STJE pressure gauge by Honeywell Inc., USA) and a confocal chromatic deflection sensor (CHRcodile S by Precitec Optronic GmbH, Germany) was used. In this

setup, the bulge sample is mounted to the pressure cell by means of a steel plate or a heatable copper plate, whose temperature is controlled via a small ceramic heater and a thermocouple [16]. Before performing a bulge test at 100°C, the heated copper plate and sample were left to stabilize for 15 min.

3. Results

For each sample batch, a representative stress-strain curve of a notched membrane is shown in Figure 3. The y-axis intercept of the curves gives the residual stress of the film. It increases with increasing silver content for the solid-solution hardened AuAg films. This is expected since the coefficient of thermal expansion is higher for silver than for gold [17], which leads to the formation of higher thermal stresses upon cooling. The bulge test performed at 100°C shows slightly compressive residual stresses, which are due to the larger thermal expansion of the film than of the retaining silicon frame. The stress-strain curves of the gold films tested at room temperature and at 100°C evidence that raising the temperature increases the ductility of the films. From the stress-strain curves of the solid-solution hardened films it can be seen that increasing the silver content decreases the fracture strain.

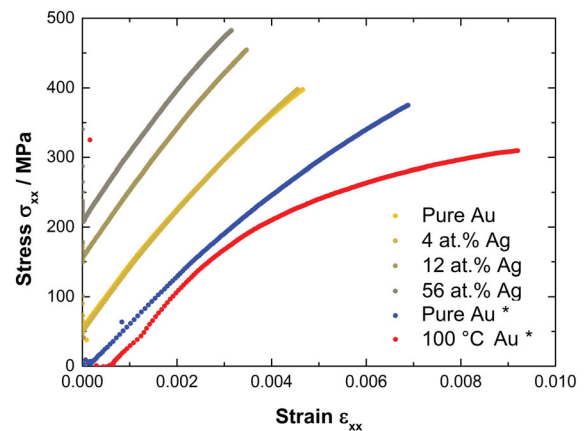


Figure 3. Stress-strain curves from bulge tests on notched membranes. All specimens but ‘100°C Au’ were tested at room temperature. The specimens marked with * were tested with the secondary setup designed for high-temperature.

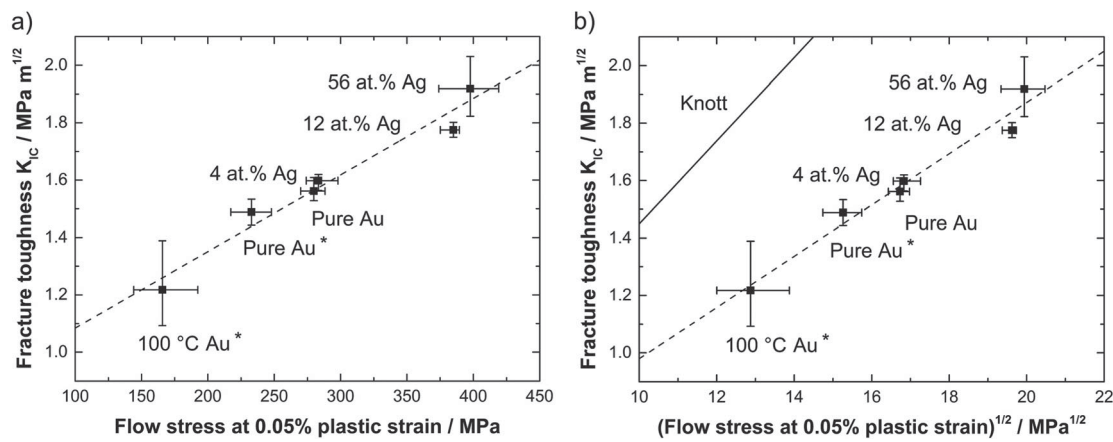


Figure 4. Average fracture toughness plotted versus (a) the flow stress at 0.05% plastic strain and (b) the square root of the flow stress at 0.05% plastic strain. The error bars represent maximum and minimum values out of at least three tests. A linear fit of the average values is given by the broken line. The continuous line in (b) represents the theoretical fracture toughness according to the model of Knott. All specimens but '100 °C Au' were tested at room temperature. The specimens marked with * were tested with the secondary setup designed for high-temperature.

In Figure 4 (a) the fracture toughness is plotted versus the flow stress at 0.05% plastic strain. The data were averaged from at least three different tests. The plot suggests a linear scaling of the fracture toughness with the yield strength. In Figure 4 (b) the same data is plotted versus the square root of the flow stress at 0.05% plastic strain. The regression line fits the data even slightly better than in Figure 4 (a) (sum of squared residuals 0.009 instead of 0.011) supporting a scaling of the fracture toughness with the square root of the yield strength as suggested by Knott. However, evaluation by the model of Knott according to equation (1) using the Young's modulus for bulk gold of 78.5 GPa [18] and the average film thickness of the investigated films of 133.7 nm results in much higher values than experimentally determined, as indicated by the continuous black line in Figure 4 (b). The effect of the different silver content on the Young's modulus of the films can be safely neglected, as the Young's modulus of silver is very similar (82.7 GPa [18]) to that of gold. As the regression coefficients for both of the fits shown in Figure 4 are much similar, it cannot be satisfyingly concluded from the limited data whether the scaling is linear or of square root form. Nevertheless, a strong correlation between the yield strength and the fracture toughness is evident.

4. Discussion

4.1. Yield strength variation

It is known from several studies that the yield strength of nanocrystalline materials and metallic thin films decreases significantly with increasing testing temperature. This is usually accounted for by the promotion of

thermally activated deformation processes like grain boundary sliding or diffusion-mediated dislocation climb [19–22]. The flow stress of the films was observed to decrease from 233 to 166 MPa when the temperature increased from 22 °C to 100 °C, which fits well to references [20–22] for gold thin films subjected to similar temperatures. It has been shown that the temperature effect sensitively depends on the strain rate [21] and the film thickness [22], which explains the huge variability of reported flow-stress data.

The increase of the yield strength with the silver content is at first of course understood as an effect of solid-solution strengthening. Although the lattice parameter of gold and silver is basically the same, solid-solution hardening has been repeatedly observed in this system [23,24]. Yet, considering the absolute yield strength increase, the strengthening effect is much stronger in our samples than in the single-crystals tested by Sachs [23] or from the extrapolation from the solid solution strengthening coefficient of $7.8 \text{ MPa}/c^{2/3}$ reported in the literature [24,25]. This is on the one hand in agreement with Rupert et al., who showed that the absolute effect of solid-solution hardening is much stronger in nanocrystalline materials than in their coarse-grained counterparts [26]. They ascribed this to an additional hardening contribution that becomes effective if dislocations are emitted from grain boundaries and have to bow out through the hardened matrix of the solid solution. On the other hand, alloying with Ag changes the microstructure of the gold films, as evidenced by the higher twin density visible in Figure 1(b). As twins can behave as strong obstacles to dislocation motion [27], this microstructural refinement is likely to account for a large part of the increase in yield strength.

4.2. Effect of yield strength on fracture toughness

The trend of increasing fracture toughness with increasing yield strength is consistent with experimental observations reported in literature. Hirakata et al. found that the fracture toughness of single-crystalline copper films was significantly lower than the one of polycrystalline reference films [28] that also had a much higher yield strength. A study by Singh et al. on copper films containing different densities of growth twins [29] also supports the trend of increasing fracture toughness with increasing yield strength.

This rather unexpected trend can be explained by the specific fracture mechanism of metallic thin films under plane stress. The fracture behaviour of similar polycrystalline gold films was investigated by in-situ deformation in the Atomic Force Microscope (AFM) in a related study [4]. The in-situ observations of crack propagation revealed strongly localized plastic deformation at the crack tip accompanied by successive necking and resulting in a chisel-point type of fracture (see Figures 8 and 9 in [4]). The essential difference to bulk samples is that the free surfaces allow for plastic deformation in the out-of-plane direction. Irrespective of the dominant deformation mechanism, out-of-plane plastic deformation will always result in a significant local reduction of the film thickness. The resulting stress concentration consequently leads to necking and finally to fracture. For materials with a high yield strength, necking is more difficult and the energy spent for necking will be larger. It follows that the fracture toughness increases with increasing yield strength. The paramount role of necking is confirmed by the following counter-intuitive behaviour: usually, with macroscopic samples, ductility enhances the fracture toughness because it increases energy dissipation by plastic deformation in front of the crack. On the opposite, the measurements on thin films evidence that the fracture toughness inversely scales with the fracture strain (Table 1). This is because, in these films, ductility primarily promotes necking in front of the crack tip, which in turn leads to catastrophic failure.

Although the experimental measurements point to a scaling relationship between fracture toughness and yield strength that is consistent with Knott's model, we have seen that the model and the data do not quantitatively agree. This is expected as the physical fracture mechanism assumed by Knott, i.e. local mode III fracture by shear separation under 45°, is different from the actually observed chisel-point fracture.

Other parameters that are likely to affect the fracture toughness of freestanding metallic thin films are the strain-hardening behaviour, the strain-rate sensitivity and the surface roughness, because these also influence

the energy required to form a through-thickness neck in front of the crack tip [30], which in turn governs the fracture toughness.

5. Conclusions

Our experimental results evidence that the low fracture toughness of metallic thin films cannot be ascribed to their increased yield strength. On the opposite, for thin films, an increase in yield strength results in an increase of fracture toughness. The likely reason is that a higher strength postpones the necking that usually leads to a chisel-point type of fracture (see [4]).

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Disclosure statement

No potential conflict of interest was reported by the authors.

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