Investigation of mechanical properties of gold thin films using a bulge test technique

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Abstract. A bulge test device has been built, with the aim to perform mechanical tests on membranes with a thickness in the 100 nm to 10 µm ranges, between room temperature and 900°C. Our set up a differential atmospheric pressure is applied across a membrane, while the deflection is measured using a laser interferometer. We show first results obtained from gold membranes with different thicknesses.

1. Introduction

Thin films are used in many application such as microelectronics and since a few years in MEMS devices. As these films are subjected to high stresses, either during industrial process or during device operation, their mechanical properties are particularly relevant.

However, these materials generally have microstructures (grain size, shape, texture...) and chemical compositions which differ from these of bulk materials. Moreover, their properties cannot be directly extrapolated from these of bulk materials: well-known scaling laws such as the Hall-Petch [1] equation (d⁻¹/₂ dependence of the yield stress on the grain size of polycrystalline materials) can be expected - and are indeed observed - to fail somewhere in the sub-micron range.

Among the different possible experimental techniques, from nanoindentation to direct testing of micron size specimens, the bulge test, first built by Beams in 1959 [2], offers a good compromise between two difficulties: specimen preparation and analysis of experimental data. Free-standing membranes are prepared from thin films deposited on a silicon substrate by opening a window within the substrate by classical wet or dry etching methods. The specimens are then loaded by use of a differential atmospheric pressure on both sides of the film.

The temperature dependence of the mechanical behaviour is of major importance. However, the range of currently existing bulge test devices does not exceed 450°C [3, 4, 5]. Other high temperature mechanical tests may be done by substrate curvature measurement, but require temperature variations during the test.

The aim of the present paper is to describe a bulge test apparatus currently being built, in order to test ductile and brittle films in a temperature range from room temperature to ~ 900°C, and to report first results obtained at room temperature on ductile gold thin films with a submicron thickness.
2. Description of the bulge test device.
A diagram of the device in room temperature configuration is shown in figure 1. A sample is clamped on a sample holder using a graphite screw and two graphite rings for an air-tight sealing. A differential pressure is applied using a syringe connected to a computer-controlled motor. A differential pressure gauge measures the applied pressure with a resolution of 0.1 Pa and a pressure range from 0 to 0.5 MPa. A graphite rod glued to a silicon mirror (total mass: 4 mg) is placed on the sample. Deflection of films induced by the applied pressure is measured using a Michelson interferometer with a λ=770 nm laser beam reflecting on the mirror. Pressure and deflection are measured continuously (figure 2), in order to command the pressure syringe for constant stress rate loading or tests under constant stress (i.e. creep tests).

![Figure 1. Diagram of the bulge test device.](image)

![Figure 2. Bulge test raw data (grey line) obtained for a rectangular gold membrane (7*1.5 mm\(^2\)) with a 200 nm thickness, and fit using (1) (dots) (image)]

The specimen can be heated using a pair of circular graphite resistors placed on top and bottom of the sample holder. The temperature is measured by two type K thermocouples and controlled by a PID system. The furnace is water-cooled and thermally insulated from the rest of the device. The whole is shut in a vacuum vessel which can be put under a secondary vacuum, in order to be under high-purity conditions before floating the vessel with pure argon. During pumping the device and heating, the specimen must be carefully kept at zero differential pressure, in order to avoid loading of the film.

2.1. Sample preparation
One of the major difficulties of this technique is the sample preparation. The films are deposited on 380 µm thick silicon substrates. A gold mask with a rectangular (7*1.5 mm\(^2\)) window is deposited by PVD on the substrate opposite to the film. The substrate is etched in a KOH solution at 80°C (wet etch) or by Deep Reactive Ion Etching. Both etch techniques leave perfect rectangular freestanding thin films.

2.2. Data analysis
The elastic deflection \( h \) of a square or rectangular membrane subject to a uniform pressure \( p \) is given by the following simplified equation [6, 7, 8]:

\[
p = C_1 \frac{Mt}{a^4(1-\nu)} h^3 + C_2 \frac{\sigma_0}{a^2} h
\]

(1)

Here \( C_1 \) and \( C_2 \) are two coefficient depending on the shape of the sample, \( M \) is the plane strain modulus, \( a \) the half side length, \( t \) the thickness of the film, \( \nu \) the Poisson’s ratio and \( \sigma_0 \) the residual stress of the film. The experimental data are fitted according to (1) and allow the extraction of basic parameters such as \( E \) and \( \sigma_0 \). Figure 2 provides an example of a pressure-deflection curve from a
rectangular gold thin film (t = 200 nm). The fluctuations of the pressure and deflection remain within the thickness of the curve. The dots are the fit of the initial data points (up to a 45 µm deflection) using (1).

For a rectangular membrane, we can assume a plane strain state in the centre of the membrane, the p, h data can be translated into σ, ε curves using the following relation [6, 7]:

\[
\sigma_{xx} = \frac{pa^2}{2ht}, \quad \varepsilon_{xx} = \frac{2h^2}{3a^2} + \varepsilon_0
\]

(2)

3. Experimental results

3.1. Microstructure

The microstructure of the film was analysed by X-ray diffraction, electron backscattering diffraction and transmission electron microscope.

The morphology and the grain size of both as deposited and annealed thin film were observed and determined from TEM observation. All films show a bimodal grain distribution with large grains (~ 1 µm) surrounded by smaller grains (~ 30 nm). The cross sectional view figure 3 e) reveals a columnar grain structure and shows that there is more than one grain stacked in the film thickness.

Figure 3. a)-d) Microstructure observed using TEM of gold thin films with different thickness deposited on silicon wafer, e) Cross section view of a 300 nm thick gold thin film.

Figure 4. X-ray diffraction from a θ/2θ set up. The scans of Au films deposited on Si showing a strong <1 1 1> texture for the raw film, and a dual <1 1 1>/<2 0 0> texture after annealing.

Figure 5. Stress-strain curves for a 7*1.5 mm² rectangular gold thin films with different thickness. The red arrow shows the beginning of microplasticity and the plane strain modulus was evaluated from the red lines.

Figure 3 a)-d) shows typical plane views from gold thin film with different thicknesses before and after annealing. The micrographs reveal the presence of defects such as growth twins and dislocations.
which can appear during the growth process. The average grain size was measured using the intercept method and varies from 50 to 70 nm for the as-deposited films. Annealing at 260°C of the free standing film for 20 h (after etching of the window) increases the average grain size to 200 - 300 nm [9].

The texture was determined by means of X ray diffraction. The results are shown in figure 4 and reveal a strong <111> texture [10]. A low (200) peak appears after annealing.

3.2. Stress strain curves

The free-standing gold thin films were initially buckled. The clamping of the specimens made them flat, except at their edges which were still wrinkled. Figure 5 shows typical stress vs. strain curves before and after annealing. The apparent initial stress (50 MPa for raw specimens and -2 MPa) for the annealed one thus do not represent the initial state of the film. The plane strain modulus M calculated from the slope of the elastic part (red line figure 5), lies between the value of M <111> (145 GPa) and M <100> (53 GPa) [10] (138, 122 and 90 GPa for thin films with a thickness of 300, 200, and 100 nm respectively). For the thinner film the reduced stiffness can be attributed to the film deposition technique. It can be related to a change in microstructure (porosity) of the film instead of a thin film effect. After annealing, the plane strain modulus is about 60 GPa. The decrease of M after a heat treatment is due to a change of texture (see the appearance of a <200> peak Fig 4).

A major source of systematic experimental error is the initial value of the deflection: For a film under a low initial stress, or a buckled film, the initial position of the mirror on rod device is very sensitive to its weight or to friction. This might explain the low modulus of the 100 nm film and the apparently erroneous behaviour of the annealed film.

Microplasticity begins approximately at \( \sigma = 150 \text{ MPa} \) (arrow Fig 5). This is probably the result of the heterogeneous microstructure, as the largest grains are expected to yield first, while the smaller ones still deform elastically. For each grain, we expect a different local stress depending on its size and orientation. The plastic domain of the curves is very short (less than 10\(^{-3}\)), and does not allow defining a classical 0.2% yield stress, as transition from purely elastic to plastic behaviour is smooth. The fracture strain happens approximately at \( \varepsilon = 0.3\% \) for as-deposited films while fracture was not reached for the annealed thin film. This difference may be the result of the increase in the average grain size.

4. Conclusion

A new bulge testing device has been built, aiming at strain membranes of brittle materials from room temperature up to 900°C. The first room temperature tests on gold films give results consistent with literature data. The variation of Young’s modulus can be attributed to changes in the volume fraction of the <111> fibre components. The yield stress remains insensitive to the film thickness. Further improvements will focus on non contact, full 2D measurements of the film shape, in order to get a realistic view of the stress and strain distribution within plastically strained membranes.

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