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A Blister-Test Apparatus for Studies on the Adhesion of Materials used for Neural Electrodes

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***Abstract*—A blister test apparatus has been developed, which allows a quantitative adhesion analysis of thin-film metallizations on polymers manufactured in cleanroom conditions suitable for micromachining of neural electrode arrays. The device is capable of pressurizing metallic membranes at wafer level, monitoring the pressure and the height of the developing blister while detecting the moment of delamination, allowing the calculation of the adhesion energy between metal film and polymer. The machine is designed for quantitative long-term studies of materials used in neural microelectrode arrays.**

I. Introduction

Adhesion is a parameter of crucial importance when it comes to any application of thin-film materials. Quantifying adhesion at a microscopic level or even in real life conditions presents a big challenge due to the influence of dissipative effects (e.g. plastic deformation) in the measurements [1]. More than 20 years ago, Ho [4] already stated that the combination of metals and polymers present a challenging problem. Microsystem engineering has allowed the fabrication of thin-film based neural electrodes and implants aiming for the therapy or restoration of lost body functions [3]. However, delamination of the layers in the wet body environment has been the major failure mode of these devices. A quote originated 1983 by White et al. [5] taking a closer look at the failure mechanism especially in the case of polyimide-metal electrodes states that "the principal difficulty encountered by scientist, and others attempting to fabricate polymer-metal-polymer electrodes, was that of delamination". Assuming, that the problem of delamination (between metallizations and polyimide layers) was encountered only at the beginning of the usage of polyimide is far wrong. Having a look at some recent literature on this topic reveals that the problem, reduced to a few important material of interest, persists: 1997 and 2009 Ti/Au, and 2007, platinum delaminating from polyimide [8]. Even though there are various approaches for adhesion improvement, the quantitative comparison has not been successfully assessed to date. The lack of such an important scientific analysis retains the theories and hypotheses on the adhesion mechanisms between the desired materials unconfirmed as such. To understand the failure mechanisms and subsequently improve the material adhesion it is mandatory to have a measurement system that builds up the basis for further comparative work. The blister test, introduced by Dannenberg in 1958, is the first test that allows an entirely quantitative analysis of adhesion, based only on fraction mechanic methods. Opposite to the peel-test for example, it only imposes relatively low strains on the material under test, avoiding complex nonlinear viscoplastic behaviours, which complicate the tests [2]. However, the complication arises with the sample fabrication. It is necessary to ensure, that the interface under load presents the weakest interface on the layer stack and a rupture of the coating is to be avoided before delamination occurs, which is a complicated task on only 300 nm thin metallic layers. Moreover, a peel-test cannot be undertaken on sputtered or evaporated thin-films due to the delicacy of the thin metal, which would be exposed to extremely high strains during the peel test. Peeling the polymer layer off metallic substrates does reveal the adhesion of an interface found in microelectrode fabrication, but it does not reveal the energetically higher adhesion achieved by the physical deposition process, which is of mayor importance. Furthermore, it is of great relevance to study the delamination process of thin-film electrode materials after aging the samples under the conditions at which the failure occurs: in a wet, ionic environment.

To assess this breach, the authors developed a device adapted from Dannenberg's 'blister adherometer' to undertake unconstrained wafer-level tests on thin-film metallizations fabricated in clean room conditions according to the fabrication processes of neural electrodes. The blister test apparatus is capable of applying a pressure on a metal membrane while monitoring the applied pressure and the height of the blister. The detection of delamination is done through analysis of the monitored parameters, leading to a calculation of the adhesion energy at the interface. The device has been designed parallel to a specific sample layout, which provides multiple tests on a single wafer, allowing the analysis of the adhesion degradation after sample accelerated aging under wet conditions or after thermal aging. The sample fabrication process can include adhesion promoter interlayers and can be compared to simple polyimide-metal combinations.

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II. Materials and Methods

A. Sample Layout

The measurement hardware depends on the sample layout and vice versa. Due to procedural reasons concerning the clean room possibilities and restrictions, the decision was made to first design a sample concept and based on this a model for the test device. The wafer layout is based on a 4" silicon wafer allowing a total amount of 14 test samples. For symmetry reasons, the wafer is split into 4 quarters, whereas each quarter has a different sample size ranging from 6 mm to 10.5 mm in diameter for the metallic circles. The sample distribution on each quarter is chosen in a way that the pressurization holes remain at the same position when rotating the wafer by 90°. This layout (Figure 1) enables a reduction of the pressure connections to the test areas and thus permits a high number of samples that can be addressed with a smaller device. A cross-sectional view of an individual sample shows the two possible operation modes: an unconstrained test mode where the lateral polyimide parts do not play a special role, and the constrained mode in which the lateral polyimide parts build the basis for the constraining layer. To ensure that the interface under load presents the weakest interface during test, the authors analysed different adhesion promoting layers between the wafer and the polyimide, leading to an extreme stable configuration when depositing diamond-like carbon (DLC) interlayers (*unpublished work)*.

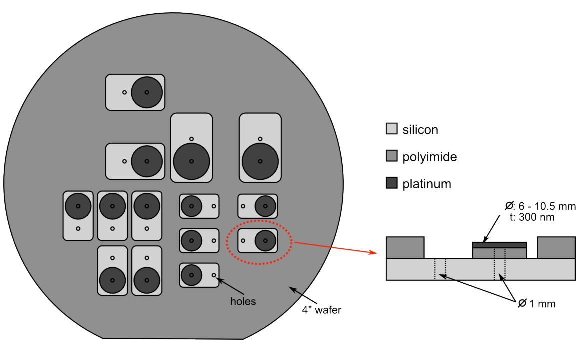

Figure 1: Sample layout for the blister test in top view (left) and in cross section, not to scale (right).

B. Design of the Sample Holder

In order to permit a pressurization of the single samples according to the layout given in Figure 1, a sample holder capable of (1) directing a pressurized fluid from a reservoir to the desired delamination site, (2) pressurizing each sample individually, (3) handling an entire wafer and accurately aligning the single sample on the fluid supply and (4) providing an easy mount and change of samples was developed (Figure 2). The chuck was split into three different parts to achieve all specifications. The device's interface to the fluid supply is provided by the lowest part, economizing the amount of external parts by only supplying a quarter of the chuck. The upper part consists of the probe holder, which connects individual samples on the wafer to the fluid supply on the bottom part. This allows a fixation and a safe handling of the wafer. Remounting the upper part after a 90° rotation can address a further quarter of the test positions on the wafer. A cover can be fixed via eight screws to the upper part, locking the wafer in the right position. Each channel contains a channel to flush the port previous to pressurization to remove air bubbles that would increase the compliance of the system.

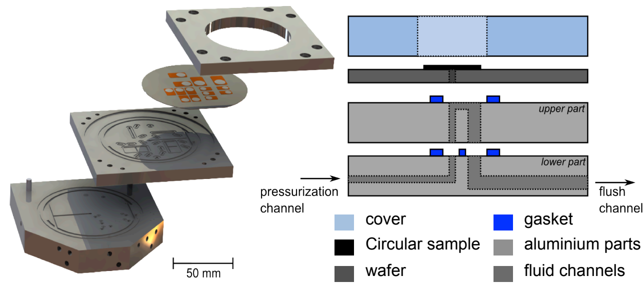

Figure 2: CAD model of the blister-test device (left) and a cross-sectional sketch of the device parts.

C. The Blister Equation

For calculating the adhesion energy it is necessary to find the correlation between the applied pressure and the resulting shape of the blister. As the baseline of the blister is circular, the overall shape of the developing blister will have the form of a hemisphere.

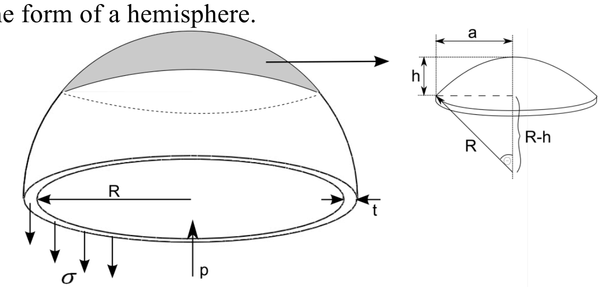

Figure 3: Hemisphere model for describing the blister shape.

Using this shape and building the balance of forces that act on the geometry one ends up with equation (eq. 1), where the left side represents the force that results from the pressure p acting on the area (πR^2) of the blister while the right term gives the stress σ in the thin-film of thickness t.

$$p\,\pi\,R^2 = \sigma\,t\,2\,\pi\,R \qquad \text{(eq.1)}$$

The more realistic hemisphere cap model (Figure 3, right) addresses the effect that a thin metal layer (as investigated in the present work) is supposed to bend slightly and not as much as given by the ideal hemisphere model. The radius in the hemisphere model can be described with the given parameters height h and cap radius a (radius of the pressurization opening). Using equation 1, the cap parameters description and some mathematics it is possible to elaborate an equation that relates the deformation of a membrane to the applied pressure (eq.2). Here, E represents the Young's modulus of the pressurized layer, v describes the Poisson ratio and σ_0 gives the residual stress in the layer. This stress matches the thermally induced stress caused by the difference in the coefficient of thermal expansion, which is supposed to cause the major internal stress during fabrication.

$$p = \frac{8}{3} \frac{E\,t\,h^3}{a^4\,(1-\nu)} + \frac{4\,\sigma_0\,t\,h}{a^2} \qquad \text{(eq.2)}$$

If the radius of the pressure inlet (parameter: a) is kept constant and the applied pressure is monitored in dependence of the square of the blister height (h^2) a straight line is expected as result (Figure 4). The intersection of this line with the pressure axis is denoted with i whereas the slope of the line is denoted with s. So if the values for the film-height, the pressurization diameter and the Poisson ratio are known, one can easily calculate the Young's Modulus and the residual stress in the membrane.

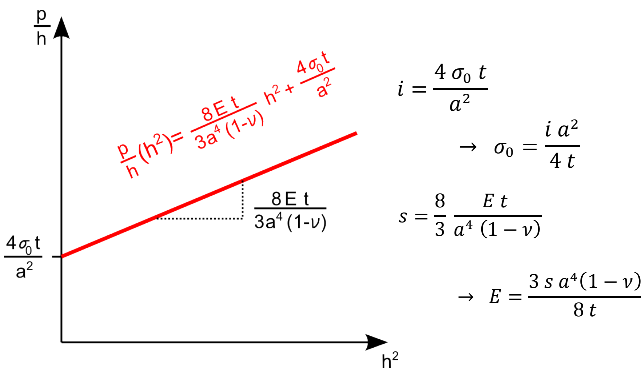

Figure 4: Usage of the blister equation for determining material properties.

After having an expression for the geometrical description of the blister it is mandatory to find a relation between the geometry and the adhesion of the blister material to the substrate. The basics of fracture mechanics state in a simple form that in order to keep the blister growing, the work performed by the pressure must exceed the total strain energy that can be stored in the thin-film material. The difference between the work done on the blister and the strain energy equals the amount of energy that can be used to delaminate the film and thus corresponds to the adhesion of the film (eq. 3).

$$\frac{\textit{work performed}}{\textit{by appl. pressure}} - \frac{\text{strain energy}}{\textit{stored in the film}} = \frac{\textit{free energy}}{\textit{for delamination}} \qquad \text{(eq.3)}$$

This free energy leads to the energy release rate, which is typically denoted with the letter G. For a circular blister with a circumference of $2\pi a$ the energy release rate according to the blister geometry is given by eq. 4 [9]. The derivation of this expression combined with some simplifications result in the final form for the description of the adhesive strength of different materials in the blister test (eq. 5). The only parameters that have to be observed in the measurement are the pressure under the film and the height of the film itself.

$$G = \frac{1}{2\,\pi\,a} \left. \frac{\partial W_{applied} - \partial U_{strain}}{\partial a} \right|_p \qquad \text{(eq.4)}$$

$$G \approx 0.5625 \cdot h\,p \qquad \text{(eq.5)}$$

This is an important finding for the sample preparation and the test layout, as only the geometry of the samples is of primary interest and additional layers (e.g. for reinforcement of the membrane) do not influence the blister measurement.

D. Hardware, Fluid Pressurization and Monitoring

A commercial syringe pump (Perfusor F, B.Braun, Melsungen, Germany) with the possibility of directly setting a desired flow rate (down to 10 µl/min) is used to push the fluid - along compliance-free brass tubing - into the system. The perfusor has been modified to achieve a maximal pressure of 400 kPa. The pressure is monitored in two different ranges (0 -100 kPa and 100 – 700 kPa) using two different pressure sensors (RBIP100DU & RBIP015DB, sensortechnics GmbH, Puchheim, Germany). Both sensors are differential pressure sensors but are operated in absolute mode by leaving the second port of the sensor unconnected. This enables an adaptation to the actual ambient pressure by calibrating the sensors prior to the measurement. The sensors are connected to a computer through a data acquisition device (NI-USB6009, National Instruments, Austin, TX, USA). Data acquisition, processing and display are done digitally through a graphical user interface based on LabView (National Instruments).

E. Height Measurement Monitoring

Even though a laser-interferometer based optical method for height monitoring has been proven feasible, a more elegant method requiring only the monitored parameters and no extra measurement devices is presented. The method is based on the mathematical description of the blister model and uses the height-dependence of the pressure equation. By re-stating the equation including the volume V under the blister and doing a simplification by neglecting the term involving the residual stress, a rather simple relation (avoiding numerical methods for solving) between the height h of the blister and the dispensed volume can be formulated (equation 6).

$$h = \sqrt[5]{\frac{3\,(1-\nu)}{8\quad E\,t} \cdot \frac{1}{{c_V}^2} \cdot p\,V^2} \qquad \text{(eq.6)}$$

F. Sample Testing

The blister test was performed on polymer tapes to proof the functionality of the devices as well as on cleanroom fabricated samples. Wafers with samples in the proposed layout to test pure platinum to polyimide adhesion and the adhesion using silicon carbide (SiC) as an inorganic adhesion promoter were fabricated (Figure 5). The polyimide base was adhered to the wafer through a DLC layer. A 10 µm polyimide layer on top of the membrane helped stabilizing the 300 nm thin pressurized metal.

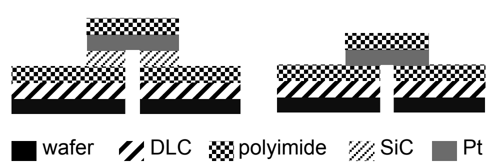

Figure 5: Cross-sectional sketch of the fabricated samples.

The samples were tested for their adhesion after fabrication. Subsequently they were sterilized in an autoclave (134 °C, 35 min, 2.06 bar) and tested for any changes in adhesion. The samples were then stored at 60 °C in phosphate buffered saline (PBS) and tested for their adhesion every 24 h. This temperature induces an accelerated aging of the samples by a factor of 4.9 compared to a body temperature of 37 °C.

III. RESULTS

A. The Height Measurement

For validating the accuracy of this method, the blister formation and propagation was monitored using a scanning acoustic microscope (SAM300, PVA Tepla Analytical System GmbH, Aalen, Germany) in a cross-sectional scanning mode (B-Scan). This allowed finding a correction factor of 3 to match the theoretical calculations to the practical blister height (Figure 6). The deviation of the theoretical calculation is attributed to approximations in the mathematical description and to the compliance of the syringe in the perfusor.

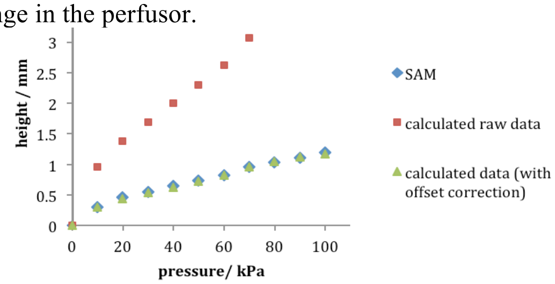

Figure 6: Validation data of the volume-approach for height calculation from monitored data.

B. Blister Test on Polymer Tapes

Analysing the pressure vs. time curve from the monitored data, a change in the slope of the curve could be observed. Through video analysis it could be demonstrated, that this change of slope is coincident with the onset of delamination. The intensity of the slope change is dependent on the dispensing velocity. Slower velocities increase the detectability of this point. Thus, evaluating the region of the alternating slope allowed the calculation of a critical pressure (32 kPa), finally resulting in an energy release rate of 4.5 J/m^2.

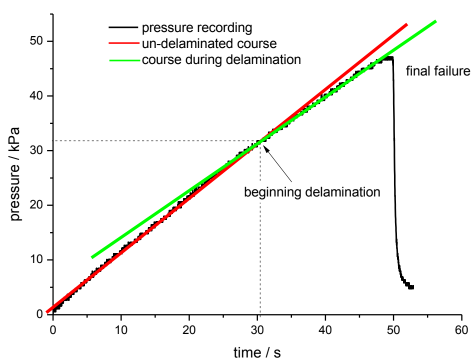

Figure 7: The pressure plot shows the instant in which delamination begins.

C. Blister Test on Fabricated Samples

The adhesion of platinum to polyimide and to SiC was found to be quite good as the samples ‘survived’ an initial pressurization (directly after cleanroom fabrication) up to 400 kPa without showing delamination. Also the subsequent sterilization process at 134°C did not affect the adhesion in a measurable way as the membrane still was able to withstand a pressure of 400 kPa (higher pressures were not used for not destroying the medical syringe pump). The repetitive measurements after 24 h aging up to 96 h showed the same results. However after 120 h in 60°C hot PBS the sample failed without showing delamination due to the breaking of the pressurized platinum membrane. Thereby the entire polyimide reinforcement circle on top of the platinum delaminated immediately (Figure 8).

IV. DISCUSSION AND CONCLUSION

A blister-test device to quantify adhesion forces between thin-film metallization and polymer substrates has been developed and tested on samples with different adhesion promoters. The failure of the samples in the test after aging 120 h can be assigned to an age-related full delamination of the top polyimide-platinum interface (Figure 8). The purpose of stabilization is not longer given and the membrane breaks. The samples of the next generation will be reinforced with SiC and polyimide to keep the top interface stable over the accelerated aging time. Since no load is applied to this interface, it is only necessary to ‘maintain’ the adhesion independent of the force of it.

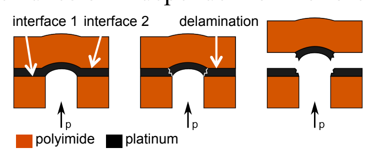

Figure 8: Failure of samples from delamination of stabilization membrane after 120 h incubation at 60 °C in PBS.

The proof of functionality for the presented device represents a large step into the analysis of delamination of materials applied for neural electrodes, building up a baseline for the comparative work of adhesion promoters to improve the long-time stability of these devices.

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