NUMERICAL AND EXPERIMENTAL ANALYSIS OF STRETCHING INDUCED INTERCONNECT DELAMINATION FOR STRETCHABLE ELECTRONIC CIRCUITS

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ABSTRACT

Stretchable electronics facilitate increased design freedom of electronic products. Representative applications can be found in health care, wellness and functional clothes, integrated electronics in stretchable parts and products. Typically, small rigid semiconductor islands are interconnected with thin metal conductor lines on top of a highly deformable substrate, such as a rubber material. A key requirement on these products is the ability to withstand large deformations during usage without losing their integrity (i.e., large stretchability). During stretching, the adhesion of the interconnects to the rubber substrate is of major importance from a reliability point of view. Experimental observations show that delamination between the metal conductor lines and the stretchable substrate may eventually lead to short circuits while also the delaminated area could result in cohesive failure of the metal lines. To characterize the copper-rubber interface, peel tests are performed. Experimental observations show that the rubber is severely lifted at the delamination front caused by its high compliance. When using the Environmental Scanning Electron Microscope (ESEM), actual fibrillation of the rubber at the peel front is observed at the micron scale. To quantify the interface properties, numerical simulations of the peel test have been performed by applying cohesive zone elements that describe the transient delamination process. The interface toughness is determined from the global parameters (i.e., forces and displacements) while the interface strength is defined by the local parameters (i.e., deformed rubber geometry at the delamination front and interconnect deformation). The thus quantified interface parameters are used to simulate the delamination behavior of a zigzag patterned interconnect three-dimensional structure. Furthermore, extensive in-situ failure mode analyses performed in scanning electron microscope are carried out. The occurring deformation behavior and failure mechanisms are characterized and used to validate the numerical model results.

KEYWORDS

Stretchable electronics, interfacial delamination, cohesive zones, in-situ failure analysis, peel test

INTRODUCTION

Stretchable electronics generally contain small rigid semiconductor islands interconnected with thin metal conductor lines, see Fig.1a. The required stretchability of the thin metal conductor lines is achieved by depositing the interconnects on compliant flat substrates using planar patterning technologies while applying spring-like conducting lines as is shown in Fig.1b. Experimental results demonstrate that the zigzag patterned interconnect provides stretchability up to 60\% without rupture, which is accommodated by in-plane rotation of arms and out-of-plane deformation of crests [1]. The structural integrity of the stretchable electronic circuits is one of the major concerns for stretchable electronics applications: (i) cohesive fracture of the interconnects directly leads to failure of the product due to the loss of its electrical functionality. This failure has been addressed by Li et al [2] and Gonzalez et al [3]. It is conclusively shown that a serpentine shape of the metal conductor permits large deformation of the substrate while small strains are present in the metal; (ii) adhesive fracture between the metal lines and the stretchable substrate is also of concern as the position of the delaminated conducting line is unsure and may lead to shorts, while also the delaminated region may be a source of cohesive fracture (i.e., necking [4, 5]) and thus electrical failure of the stretchable device.

In order to understand the mechanisms leading to failure of the conducting lines, insight into the key factors
leading to delamination is required. To this end, a three-dimensional numerical model is developed that can describe the entire delamination process using cohesive zone elements. More precisely, the cohesive zone user-element as formulated by Van den Bosch et al. [6] is employed. The mechanical properties of the PDMS rubber substrate and the copper conducting lines have been established from experiments while also the interface properties have been characterised by means of a dedicated peel test set-up. Although the adhesive toughness $\Gamma_c$ is rather straightforward to determine from these experiments [7], the adhesive strength $\tau_{\text{max}}$ is more challenging to obtain. In [8], the sensitivity of the numerical results to the cohesive zone parameters is illustrated by variational analysis. It appears that the results of most of the investigated test methods are quite insensitive to the interface strength value which renders this parameter more difficult to characterise. In this paper, it is proposed to use the local deformation of the compliant substrate at the delamination front. This is achieved by combining finite element results with experimentally measured local deformation of the substrate and the copper film. First results have been reported in [9]. Using the thus obtained material and interface properties, the delamination behaviour of a three-dimensional zigzag structure, as shown in Fig.1b, is determined. In-situ SEM and optical measurements are used to validate the numerically obtained results.

**PEEL TEST EXPERIMENTS**

As already mentioned in the Introduction, a 90° peel test is used to establish the adhesion properties. In this test a thin film is peeled-off a substrate at a fixed angle of 90° from the substrate. The force is measured as a function of the clamp displacement and during peeling the force reaches a more or less constant value, called the peel force. In general, this force is dependent on film thickness, peeling angle, materials and interface properties [10]. When the film deforms elastically and the substrate stiffness is high compared to the film stiffness, the peel force per unit sample width becomes a direct measure of the adhesion energy:

$$P_f (1 - \cos \theta) = \Gamma_c,$$

where $P_f$ is the peel force, $\theta$ the peeling angle and $\Gamma_c$ the adhesion toughness. A more detailed treatise of film peeling can be found in [7, 10].

The peel test samples consist of a rubber (Sylgard 186, PDMS) substrate with a copper (electrodeposited, grade TW-YE, Circuit Foil) film. The rubber is cast in a mould onto the copper film. After casting the samples are cured at room temperature. The copper film has a thickness of 18 ± 0.5 $\mu$m, and consists of an untreated shiny side with very low roughness and a treated side with high roughness values $R_z$ ranging from 6 to 8 $\mu$m. The high roughness value is used for improved rubber-copper adhesion; in addition an adhesion promoter is added. Prior to testing, the rubber side of the sample is glued on a metal plate that will be mounted in the test set-up. After mounting, a part of the copper is peeled-off manually for clamping purposes. In order to monitor the local geometry near the delamination front, a camera is added to the experimental set-up, as shown in Fig.2a. The camera is fixed with respect to the metal plate on which the sample is glued, and is positioned perpendicular to the side of the sample. The averaged peel force $P_f$ is used to calculate the adhesion energy or work of separation $\Gamma_c$ according to $\Gamma_c = P_f$, when assuming that the peeling angle remains approximately 90° during testing. The average adhesion energy
is $1343 \pm 51 \text{ J/m}^2$. An example of an image recorded with the camera system of the local geometry near the delamination front during steady-state peeling is shown in the right picture of Fig.2. For validation purposes, three geometry parameters are defined: the width of the lifted rubber $w$, the height of the lifted rubber $h$, and the radius of the copper foil $R$. The width is defined as the distance between the points where the lifted rubber crosses the original rubber upper plane. The height is defined as distance from the rubber upper plane to the top of the rubber peak. The averaged values over 15 measurements of different samples are: $h = 1.31 \pm 0.02 \text{ mm}$, $w = 2.1 \pm 0.3 \text{ mm}$, $R = 1.43 \pm 0.05 \text{ mm}$. The relatively large range in the width value is attributed to uncertainty of the position of the rubber upper plane in the images.

During peeling, fibrillation is observed at the copper-rubber interface, as illustrated by the ESEM picture in Fig.3. Moreover, surface analysis on the copper film reveals traces of rubber after peeling. This can be attributed to actual fracture of the rubber fibrils during peel testing. This rubber fibrillation and subsequent fracture might explain the high work of separation value.

**PEEL TEST MODEL**

The 2D plane strain finite element peel test model geometry is shown in Fig.4a (not to scale). The length is chosen as 15 mm instead of 84 mm, resulting in reduced calculation times while still reaching the steady-state plateau value $P_f$. At the bottom plane of the substrate all degrees of freedom are fixed, corresponding to the experiment where the sample bottom is glued to the test setup. The peeling of the copper film is simulated by prescribing a displacement in $y$—direction at the upper left corner point of the copper film: $U_y = 13 \text{ mm}$.

The rubber substrate is modelled with a neo-Hookean hyperelastic constitutive model, $W = C_{10} I_1$, where $W$ is the strain energy function and $I_1$ the first strain invariant. Uniaxial tensile experiments and planar extension tests are performed under room conditions ($23 \pm 2^\circ \text{C}$ and $50 \pm 5\% \text{ RH}$) on a Zwick 1474 tensile testing device. To check the validity of the hyperelastic constitutive model, hysteresis loops are performed.
by loading/unloading cycles. By fitting the Neo-Hookean model on the measured uniaxial and planar extension results, a value of $C_1 = 0.165$ [MPa] is established, see Fig.5a. Several other, more elaborate, hyperelastic models were used in the model but resulted in similar peel test results. For this reason, the simple neo-Hookean model is applied in all calculations.

To characterise the material behaviour of the copper foil uniaxial tensile experiments are performed. Again, these experiments are performed under room conditions using a Kammrath & Weiss 10 kN micro-loading stage equipped with a 100 N load cell. The displacements and strains are measured on the specimen surface by digital image correlation (DIC). A nonlinear elasto-plastic constitutive model is obtained from the measurements, as depicted in Fig.5b.

The delamination process is described by the cohesive zone user-elements as developed by Van den Bosch et al. [6]. Here, no distinction is made between normal and tangential direction as fibrils can only transfer loads along the fibril axis. Furthermore, a large displacement formulation is used that defines a single relation between the traction $t$ and separation $\delta$ according to

$$ t = \frac{\Gamma_c}{\delta_c} \left( \frac{\Delta}{\delta_c} \right) \exp \left( -\frac{\Gamma_c}{\delta_c} \right), \quad (1) $$

in which the traction vector $t = te$ and the opening displacement vector $\delta = \delta e$, where $e$ is the unit vector along the line between corresponding points of the interface, $\Gamma_c$ the interface work of separation, and $\delta_c$ is the separation value at which the maximum traction $t_{\text{max}}$ is reached, according to

$$ \delta_c = \frac{\Gamma_c}{t_{\text{max}}} \exp(1). \quad (2) $$
Figure 6: Deformation stages during peeling at a vertical displacement of (a) 2.5 mm; (b) 5 mm; and (c) 10 mm for $\Gamma_c = 1350 \text{ J/m}^2$ and $t_{\text{max}} = 2.5 \text{ MPa}$. For visualization purposes the cohesive zone elements are omitted from the pictures. The rubber deformation at the delamination front is obvious.

Irreversibility is taken into account by assuming linear elastic unloading to the origin. The cohesive zone elements are applied at the interface of the copper film and the rubber substrate, except at the position of the manually delaminated initial imperfection in the peel experiment (see Fig.4a). The geometry is discretized using a sufficient amount of linear quadrilateral finite elements that account for geometrical and material non-linearities, such that a unique, converged solution is obtained upon mesh refinement.

To determine the cohesive zone properties, the work of separation $\Gamma_c$ is varied between 1200 and 1400 J/m$^2$, while the interface strength $t_{\text{max}}$ is varied between 1.0 and 4.0 MPa. Fig.6 shows typical deformation stages during a simulation of the peel-test, in this case with $\Gamma_c = 1350 \text{ J/m}^2$ and $t_{\text{max}} = 2.5 \text{ MPa}$.

As rubber fibrillation and subsequent fracture occurs during peel testing (see Fig.3), these dissipative mechanisms are lumped in the cohesive zone elements. The following values have been established from the global and local scales: $\Gamma_c = 1330 \text{ J/m}^2$ and $t_{\text{max}} = 3.0 \text{ MPa}$. These values result in a peeling force of 24.33 N (Fig.4b, $h = 1.2 \text{ mm}$, $w = 1.4 \text{ mm}$, and $R = 1.5 \text{ mm}$). There are several reasons for the difference in local values: (i) experimental determination of the local geometry parameters is difficult due to the fact that a reference line has to be constructed which introduces inaccuracies; (ii) the neo-Hookean model does not fit the experiments exactly (see Fig.5a).

**APPLICATION TO THE THREE-DIMENSIONAL ZIGZAG STRUCTURES**

To predict the delamination behaviour of the zigzag interconnect structure, numerical simulations are performed on the structure illustrated in Fig.7. As boundary conditions, a uniaxial strain of 60% is prescribed, in accordance with the experiments reported in [1]. The geometry of the sample is as follows: $\theta = 60^\circ$, $l_{\text{Cu}} = 2 \text{ mm}$, $w_{\text{Cu}} = 0.1 \text{ mm}$, $W_S = 10 \text{ mm}$, $L_S = 8 \text{ mm}$, $T_S = 0.5 \text{ mm}$. Furthermore, the thickness of the interconnect is 18 $\mu\text{m}$, while its inner and outer radii are 0.1 mm.

Figure 7: Illustration of the patterned zigzag interconnection on top of the PDMS substrate.
In order to validate the numerical model, the deformation of the interconnects during sample loading is used. In Fig.8, the simulated deformation and the corresponding in-situ optical microscope images are depicted at different deformation stages.

From these pictures, it can be observed that a qualitatively good agreement has been obtained. To evaluate the accuracy in a quantitative way, three geometry parameters have been defined according to Fig.8a: $\theta$ is the angle of the zigzags, $b$ the length and $w$ the height of the zigzag structure. However, due to optical inaccuracies, no absolute comparison is allowed: for example, the measured initial value of $b$ is 1.91 mm from the optical images while according to the specifications, this value is 2.00 mm (which corresponds to the value in the numerical model). Hence, the absolute values of the measured geometry from the optical images do not represent the actual geometry accurately. Therefore, it is decided to use the relative change of the geometrical parameters between subsequent deformation stages at 25%, 50% and 60% strain. In Table 1, the results are given. It can thus be concluded that the model captures the deformation of the interconnects quite accurately. Remarkably, the numerically predicted values are higher than the experimental values, except during the strain change from 50% to 60%. Here, the numerical values are lower than the experimental values. During the elongation from 0 to 50%, the crests are deforming mainly in opening mode. During this stage, the zigzags are deforming in a symmetric way. It is observed that at around 60% deformation, the interconnect becomes unstable and consequently, the crests start to twist in order to be able to accommodate for the increasing deformation.

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Table 1: The relative changes of the geometrical parameters, defined in Fig.8a between different loading stages from experimental measurements (‘exp’) and numerical simulations (‘num’).
This can be seen in Fig.9a: here, the local deformation and rotation below the crest is clearly visible. This increased deformation explains the higher values of the geometry parameters compared to the numerical results, in which this instability is not taken into account. In Fig.9a, the fibrillation of the PDMS can be observed. As already mentioned, in the numerical model, this process is described by the cohesive zone elements. To illustrate this, the region of the deforming cohesive zone elements is identified in black in Fig.9b. From this, it can be concluded that the cohesive zone elements capture the fibrillation process in an averaged or macroscopic way. Furthermore, this fibrillation zone appears to be sensitive to the value of the interface strength. From Fig.9, it can be concluded that the fitted value of $\Gamma_{\text{max}}$ from the peel tests yields a good description of the fibrillation region. A study regarding the relation between fibrillation and work of separation is discussed in [11].

**DISCUSSION**

In order to predict the stretching induced delamination that occurs in stretchable electronic circuits, a combined experimental and numerical approach is pursued to establish interface properties from peel test experiments. The global force-displacement curve is used to identify the interface work of separation $\Gamma_c$, while local deformation measures at the delamination front are used to extract the interface strength value $t_{\text{max}}$. Consequently, all dissipation mechanisms occurring during peeling of the copper are lumped onto the cohesive zone elements, including experimentally observed rubber fibrillation and subsequent fracture.

To validate the obtained results from the three-dimensional delamination model containing zigzag interconnect structures, three geometry parameters are introduced that characterize the deformation of these zigzag structures. A good agreement is obtained between the experimentally measured and the numerically obtained geometry values. Furthermore, the experimentally observed region in which fibrillation occurs during loading of the structure agrees well with the numerically calculated process zone.

The local rubber geometry at the delamination front shown in Fig.2b suggests that delamination does not occur purely at mode 1 (opening mode). In fact, a relatively large mode 2 contribution is present which indicates that the interface is loaded in mixed mode. To account for this, a mode angle dependent work of separation $\Gamma_c(\psi)$ may have to be taken into account for more accurate prediction of delamination phenomena in arbitrary 3D stretchable electronics designs although experimental characterization of this mode dependency is far from trivial.
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