Interface integrity in stretchable electronics

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ABSTRACT

Stretchable electronic devices enable numerous futuristic applications. Typically, these devices consist of a (metal) interconnect system embedded in a stretchable (rubber) matrix. This invokes an apparent stretchability conflict between the interconnect system and the matrix. This conflict is addressed by shaping the interconnects in mechanistic patterns that bend and twist to facilitate global stretchability. Metal-rubber type stretchable electronic systems exhibit catastrophic interface delamination, which is investigated in this research. The fibrillation process occurring at the delamination front of the metal-rubber interface is investigated through *in-situ* SEM imaging of the progressing delamination front of peel tests of rubber on copper samples. Results show that the interface strength is dependent on the delamination rate and the interface roughness. Additionally, the fibril geometry seems highly dependent on the interface roughness, while being remarkably independent on the delamination-rate.

INTRODUCTION

To allow electronic devices to be used close to or inside the human body, the devices need to be able to comply with the body. Since the body is very flexible, and locally even stretchable it is desired that these electronic devices are also stretchable. These devices typically consist of rigid or flexible functional electronic components, embedded in a elastomeric (rubber) matrix material [1-3]. These functional islands then need to be connected, and for the interconnect design two paths can be followed; *(i)* start with a stretchable, typically non-conductive, material and add conductivity, or *(ii)* start with conductive, typically non-stretchable, material and add stretchability. Designs following the first path mostly use elastomers for the interconnect material filled with conductive particles, or even nanotubes [4]. These interconnects are often very stretchable but the conductivity is low which limits frequency of the electronic signals. Designs following the second path mostly use metals for the interconnects adding stretchability by structuring the interconnects in mechanistic patterns which locally reduce the deformation, when they are stretched globally, similar to a helical telephone cord [5-7]. Although, both design paths have reliability issues, this paper focuses mainly on issues related to the second design path.

In previous research is shown that interface delamination between the elastomer matrix and metal interconnect is a precursor to failure [8-9]. After delamination the device has not yet failed, but the matrix material no longer distributes the load over the metal structure, thereby causing localization in the delaminated part of the interconnect, which then quickly leads to failure. Improving the interface will also improve the stretchability and reliability of the device and thereby may reduce the requirements on the mechanistic design or even render it obsolete. The amount of energy an interface can dissipate before delamination determines the toughness of the interface. Previous research indicated to the rubber fibrillation which occurs at the delamination front as the dominant dissipative mechanism. Therefore in this research the fibrillation is investigated further focusing on the dependence of the interface delamination on the interface roughness and on the delamination rate.

EXPERIMENTAL SETUP

The interface delamination is investigated by means of a T-peel test, which is an established method for testing interface delamination properties [10]. In a T-peel test, the two layers of an interface are separated at an angle of 90° using a tensile stage

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(Figure 1). From the force measured at the steady-state part of the peel-force displacement curve the work of separation can be calculated as

$$G_c = \frac{U_e}{A} = \frac{2uF}{bu} = \frac{2F}{b}, \tag{0.1}$$

where U_e is the mechanical energy supplied to the system, A the peeled area, F the force measured on the load-cell, u the displacement of the clamps and b the width of the peel front.

Fig. 1 Schematic illustration of the sample layout, and loading conditions

For this research a Kammrath-Weiss micro-tensile stage is used which is especially designed for use in combination with optical or electron microscopes. The stage is fitted with a 100N load-cell and can operate at clamp displacement velocities between 0.1μm/s and 100μm/s. This tensile stage fits inside the vacuum chamber of the FEI Quanta 600, Environmental Scanning Electron Microscope (ESEM), thereby allowing for high magnification *in-situ* observation of the real-time progressing delamination front (Figure 2). The possibility to visualize the delamination front *in-situ* is proven to be invaluable for observing the fibrillation process, because the fibrils quickly relax when the experiment is stopped. Consequently, imaging the peel front in a paused or interrupted experiment would not have shown the correct fibril geometry. The same ESEM is also used to image both the new surfaces created by the peel-test experiment, where for the rubber side the Environmental mode was essential, due to the non-conductive nature of the material.

Fig. 2 The micro-tensile stage, placed in the vacuum chamber of the environmental scanning electron microscope

SAMPLES

The samples used in the test where provided by the TU-Berlin, courtesy of Thomas Löher. The samples consist of a 50 μm thick layer of TPU[1] laminated on a 35 μm thick layer of TW-YE grade printed circuit board copper, which is delivered with the interface side roughened, exhibiting a 3-5 μm deep fractal surface (Figure 2b). This combination is of materials is chosen because it is actually used in current stretchable electronic prototypes. To create a T-peel-test sample two of such (bi-layer) samples are laminated back to back, with the TPU in the center at a processing temperature of 200°, where a small part of the sample is not laminated to create a pre-crack. This effectively creates a 3 layer sample with on the outsides the copper layers and in the middle a 100 μm thick TPU layer with a pre-crack in the middle of the layer. This pre-crack is not at the correct interface, therefore before starting the "real" experiment, the sample is first peeled until the crack has propagated to either one of the two copper-TPU interfaces. To test the dependence of the interface delamination properties on the interface surface roughness the smooth side of the copper was treated with a chemical roughening step to create a 1 μm deep fractal surface (Figure 2a), from this point onwards called "smooth", the TPU was then laminated to this surface in the same way is for the rough surface, which is from this point onwards called "rough".

Fig. 3 Surface height maps measured with optical confocal profilometry for **(a)** the "smooth" sample type, and **(b)** the "rough" sample type

SURFACE ROUGHNESS DEPENDENCY

First the influence of the interface surface roughness on the fibrillation mechanics is investigated. The surface roughness is quantified by measuring "clean" pre-laminated copper surfaces of both sample types using a Sensofar plμ4200 optical confocal surface profilometer (Figure 3). From the surface height maps of both sample types the root mean squared roughness is calculated

[1] Thermo-Plastic Urithane, walopur by Epurex

$$R_q = \sqrt{\frac{1}{N}\sum_{i=1}^{N}\left(z_i - \bar{z}\right)^2}, \tag{0.2}$$

where N is the number of data points (i.e. pixels) in the surface height map, z is the height and $\bar{z}$ is the mean height of the entire measurement. Before calculating the roughness, the surface height map is leveled with a 3rd order plane, fitted through the data, to correct for any large scale deviations. The two sample types measured a roughness of 0.52 μm and 2.46 μm for the "smooth" and the "rough" sample respectively.

The peel-force displacement curves of peel-tests performed for both samples revealed that the "smooth" interface is a lot weaker than the "rough" interface (Figure 4). This is expected and already shown in [9], the reasons for a rougher interface to be stronger are threefold, a rougher interface *(i)* has ha larger area to chemically bond to, *(ii)* increases the amount of delamination mode-mixity, caused by the various angles of the local interface with respect to the peel-front opening angle, and *(iii)* the extreme asperities can also mechanically interlock the elastomer. However, the effect on the fibrillation process was not discussed in [9]. From the *in-situ* imaging of the progressing delamination front the fibrillation process can be visualized (Figure 5). From these micrographs can clearly be seen that the fibrils for the "rough" sample are much larger than the fibrils for the "smooth" sample. Furthermore, for the "rough" samples the fibrils seem to remain connected to the copper longer in the "valleys" of the copper roughness, while for the "smooth" sample no clear correlation is seen between the roughness pattern and the fibril pattern.

Fig. 4 Typical force displacement curves obtained from two peel tests for samples with different roughness, each curve consists of an initiation part, where the sample is loaded and starts to delaminate until a steady state peel-force is reached where the delamination rate is in balance with the loading rate, this is where the work of separation is determined.

Fig. 5 ESEM micrographs captured of the progressing delamination front for **(a)** the "smooth" sample and **(b)** the "rough" sample type, with copper shown on top and TPU shown on the bottom side. Note that for the "smooth" sample the fibrils are located in the darker band at the peel-front.

Movies where made from the *in-situ* ESEM images, which allows for further analysis of the fibrils. By evaluating the movies backwards an estimation of the average fibril length is acquired by measuring each fibril in the movie just before it fractures. This is method is performed for both sample types and the measured fibril lengths are approximately 12 ±3 μm and 23 ±3 μm for the "smooth" and "rough" sample respectively (Figure 6).

Fig. 6 A quantitative comparison of the fibril length as measured from movies, taken from both the "smooth" and "rough" sample

PEEL RATE DEPENDENCY

To investigate the rate dependency of the interface delamination, the peel-test experiments, using the "rough" copper surface sample type, were performed at various peel-rates. Where the peel-rates were chosen logarithmically over a regime as large as possible with the current setup, resulting in four peel-rates 0.1, 1, 10 and 100 μm/s. For the fastest peel-rate it was impossible to capture useful *in-situ* ESEM images due to the too low acquisition rate compared to the movements within an image.

Figure 7a shows typical peel-force displacement curves measured at all four peel-rates, showing that for increasing peel-rate the work of separation also increases. To reduce the scatter between different series of measurements, each measurement set is normalized with the 10 μm/s measurement of the set. Interestingly, if the normalized work of separation is plotted against the logarithm of the peel-rate, a linear relation is shown (Figure 7b).

Fig. 7 (a) Four typical force displacement curves obtained from peel tests performed at four different peel-rates. **(b)** Plotting the work of separation, normalized with the 10 μm/s measurement of that measurement set, against the logarithmic peel-rate reveals a linear peel-rate dependency.

Using the same method as described above to measure the fibril length, gives an approximation of the average fibril length for each peel-rate (Figure 8). From these fibril length measurements can be concluded that the fibrils remain remarkably constant over three decades of peel-rates (Figure 9). This indicates that while the peel-rate affects the interface strength, it does not affect the delamination micro-mechanisms. Therefore, to understand these mechanisms better, the surfaces created by the peel test can be investigated. The interface delamination can be subdivided in three failure modes, where the interface can fail *(i)* in the copper, leaving copper behind on the rubber surface and *(ii)* in the fibril, leaving rubber behind on the copper surface, *(iii)* at the interface.

Fig. 8 ESEM *in-situ* micrographs of the fibrillation process for various peel-rates, note that the fibril geometry is remarkably similar for all peel-rates

Fig. 9 A quantitative comparison of the fibril length as measured from movies, taken from both three different peel-rates

Figure 10 shows typical ESEM micrographs of the rubber surface after peeling for three peel-rates, each showing small copper particles on the rubber surface, confirmed with Energy-dispersive X-ray spectroscopy. From the area created by the peel test experiment for each respective peel-rate, five images are taken. For each image the amount of copper left behind on the rubber surface is quantified by means of segmentation, and plotted in figure 11, showing that there is an increasing trend for the amount of copper for increasing peel-rate. The “rough” copper surface is created by adding an extra electroplating step to the TW-YE grade copper foil. Resulting in a surface covered with various size (and strength) asperities. Consequently, it can be assumed that if more copper asperities are torn off the copper surface by the fibrils, that in the distribution of torn-off asperities, some were stronger. Following this reasoning, we can assume that the forces in these fibrils are also larger. Figure 8 shows that the fibril geometry does not change significantly as a function of peel-rate, leading to the conclusion that if the number of copper particles for a given peel-rate is larger, that then the stresses at the interface must also have been larger. However, it must be noted that the fibril geometry is actually very difficult to quantify or compare, therefore this conclusion must be taken lightly.

Fig 10 ESEM images of the rubber surface created by the peel-test experiment, the bright spots are copper particles torn off the copper surface.

Fig 11 The measured area of copper on the rubber surface after peeling for various peel-rates

Likewise, the copper area after peeling is also investigated, where rubber fibril fracture will leave rubber patches on the copper surface. For all four peel rates, 10 SEM images are taken from the area created by the peel-test for the respective peel-rates. Similarly to the rubber surface analysis the area covered with rubber is quantified using a threshold technique. The obtained rubber area fractions are compared in figure 12, which, due to large scatter, show an unclear trend between the rubber area fraction and the peel-rate. For all investigated copper surfaces the rubber area fraction was significant, which by itself shows that the rubber fracture mechanism plays a significant role in the delamination process, and thus possibly also in the peel-rate dependency.

Fig 12 The measured area of rubber on the copper surface after peeling for various peel-rates

CONCLUSIONS

In-situ visualizations are a powerful tool for investigating the real-time progressing delamination front, because it allows the visualization of the real fibrillation process. Due to the fast relaxation of the fibrils they are impossible to visualize for a paused experiment or after the experiment. The *in-situ* visualization shows that the fibril geometry depends strongly on the copper surface roughness, which can be expected since the onset of a fibril is a void in the interface which will probably occur in the weakest spot in the interface, which is determined by the micro-shape of the interface. This is confirmed by the measured work of separation which is larger for the rougher sample. Remarkably, the *in-situ* visualization also shows that the fibril geometry does not change significantly between different peel-rates, while the work of separation, or interface strength does depend logarithmically on the peel-rate.

Three interface failure modes are identified, *(i)* failure in the copper near the interface, *(ii)* failure in the rubber fibril, *(iii)* failure at the interface. Analysis of the rubber surface created by the peel-test revealed that for increasing peel-rates more copper fracture occurs, indicating that the fibril stresses are higher for faster peel-rates, which is in contrast with the apparent insignificant changes in fibril geometry for different peel-rates. Analysis of the copper surface after peeling showed that fibril fracture occurs frequent indicating that it will probably have a significant role in the rate-dependency of the interface. It remains a question if the observed constant fibril length is due to the fact the fibril fracture strain is actually constant, or that the fibrils are stronger for higher peel-rates, and therefore, pull in more bulk rubber material, where-after they fracture at a lower (local) strain.

Finally, these interfaces are typically modeled using Cohesive Zone (CZ) elements in a Finite Element Method (FEM) model. Such CZ elements model the traction response of the interface as a function of opening, using a Traction Separation Law (TSL), for which typically three parameters are required, *(i)* the maximum traction, *(ii)* the critical opening, and *(iii)* the work of separation. The work of separation can easily be determined from the peel-force displacement curve, and through the TSL all three are coupled, so only one more parameter is required to characterize the interface. Considering that the fibrils did not show significant geometrical changes for changes in peel-rate, indicates that if only one of the two remaining parameters would be time dependent it would probably be the maximum traction, since the critical opening of an interface is related to the maximum opening of an interface which is where the fibrils are imaged.

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