

Copper–rubber interface delamination in stretchable electronics

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Interface delamination in metal–rubber-type stretchable electronic systems leads to early failure. This paper reports an investigation of metal–rubber interfaces through in situ scanning electron microscopy imaging of the progressing delamination front of 90° peel tests of rubber on copper samples. The results show that the energy dissipated in the forming, elongation and rupture of ~50 µm long fibrils constitutes the major part of the work of separation. The experiments are characterized and modeled using a cohesive zone-enriched finite-element model.

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To open up a realm of new electronic devices with applications close to or inside the human body, flexibility and even stretchability of the device is required. Examples of such devices are smart clothing, sensitive skin for robots or prostheses, biomedical parameter monitoring, neural activity monitoring, and intraocular retinal prostheses [1,2]. These devices will most likely consist of rigid functional components connected via a stretchable interconnect system [1–4].

Some research groups have proposed a stretchable interconnect system by using metal (e.g. copper) interconnects embedded in a rubber matrix material [9–11]. This results in an apparent stretchability conflict between the highly stretchable rubber (~100%) and the metal which has a typical elastic stretchability of <1%. This conflict is typically addressed by mechanistic patterns which reduce the local stretch of the metal [6–11]. Nevertheless, some fundamental problems remain, including low maximum stretchability and limitations to miniaturization. A key factor in these problems is the interfacial integrity, as precursor to failure. For bilayer systems of metal adhered to rubber under extensional loading, debonding of the interface leads to localization of stress inside the metal interconnect, which accelerates necking and ultimately leads to failure [5].

A better understanding of these metal–rubber interfaces may improve the interface integrity and thereby improve the stretchability. In this paper, the microscopic delamination mechanics of two metal–rubber interface systems, explored by two collaborating research groups [9,11], are analyzed in detail through in situ environmental scanning electron microscopy (ESEM) imaging of the progressing delamination front in a 90° peel test. Additionally, the metal–rubber interface is characterized and simulated using a cohesive zone model, and the traction-separation law parameters of this model are confirmed using the experimentally obtained peel front deformation geometry.

Peel test experiments are carried out because it is an established method for measuring interface delamination properties such as the work of separation (WOS) [12]. A peel test experiment consists of separating the two layers of an interface at a specific angle while measuring the peel force and displacement. In this research, two bilayer samples are glued back-to-back and both bilayer interfaces are delaminated simultaneously using a T-type peel test, resulting in two stationary 90° peel fronts.

A microtensile stage (Kamrath & Weiss GmbH) is used, which fits in the vacuum chamber of an environmental scanning electron microscope (FEI XL30 ESEM-FEG) and under an optical microscope, to perform the peel tests, thereby allowing in situ imaging of the progressing delamination front. The setup is also placed inside a box, with controlled temperature and humidity, to assess the influence of environmental parameters.

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All samples consist of two layers, one copper and one rubber. Two types of copper surface roughnesses are investigated using printed circuit board grade copper foil, 35 μm thick, which is delivered with one side roughened, exhibiting a 3–5 μm deep fractal surface (Fig. 2a). Subsequently, the roughened side is covered by molding a ~ 1 mm thick layer of polydimethylsiloxane (PDMS, Sylgard 186, Dow Corning). The second copper surface roughness type is processed by roughening the same copper foil further with an electroplating technique, thereby yielding a significantly increased surface roughness with 5–10 μm sized protrusions (Fig. 2b). This surface is then also covered with a ~ 1 mm thick layer of PDMS. The as-received copper rough surface is hereafter called “rough” and the electroplated extra-rough surface is hereafter called “extra rough”.

In addition, two more types of rubber are investigated. To this end, the “extra rough” copper is laminated with a 50 μm thick layer of thermoplastic polyurethane (TPU, Walopur, Epurex). This lamination is performed at two different temperatures, 180 and 200 $^{\circ}\text{C}$, and the resulting interfaces are referred to hereafter as TPU180 and TPU200. The uniaxial elongation response of the prelaminated TPU, PDMS and copper is shown in Figure 1a.

Typical force–displacement curves from a peel test consist of an initiation regime, followed by a steady-state peeling regime during which the delamination velocity equals the clamp displacement velocity (Fig. 1b). From this steady-state force plateau, the peel energy G can be determined as $G = F/b$, where F is the peel force and b the width of the peel front [13].

As a first step, the influence of process temperature, relative humidity (RH), temperature (T) and velocity (v) on the delamination behavior is investigated by comparing the WOS to that of a reference peel test (“extra rough” copper with TPU180, RH = 25%, $T = 295$ K and $v = 5$ mm s $^{-1}$). The influence on the WOS of each parameter was determined: the relative humidity and velocity showed a negligible effect on the WOS, i.e. <2% over the tested ranges of $0 \leq \text{RH} \leq 75\%$ and $5 \leq v \leq 20$ mm s $^{-1}$, whereas the temperature showed a small influence of $\sim 4\%$ within the tested range of $295 \leq T \leq 318$ K. The TPU process temperature showed a significant influence of $\sim 23\%$ between the two tested processing temperatures of 453 and 473 K. The influence of the process temperature was therefore investigated further. First, the influence of the copper roughness was addressed.

In Figure 2, SEM images of the two types of copper roughness are shown for uncovered (prelaminated) cop-

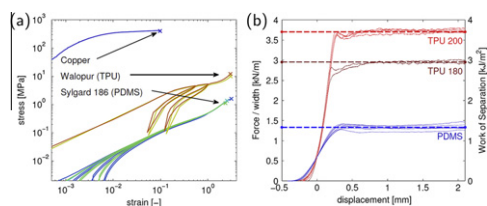


Figure 1. (a) Uniaxial stress–strain curves obtained from bulk samples of the as-received copper sheets and prelaminated rubber sheets. (b) Work of separation vs. clamp displacement for the three different rubber types deposited on the “extra rough” copper.

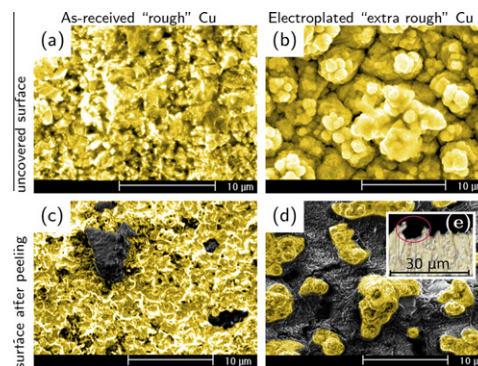


Figure 2. SEM images of the copper surface for two samples with different surface roughness. The bottom two images are made after delamination, showing PDMS (black) which fractured and remained on the copper (yellow) surface. Inset figure (e) shows a cross-section of a “extra rough” copper sample with rubber interlocked in the roughness valleys. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

per and copper after delamination of PDMS. Of the two types of copper roughness, the “extra rough” copper was found to have a 40% higher WOS. More remarkably, Figure 2c and d show that the rubber fractures during delamination, leaving rubber on the copper surface after delamination, which is confirmed by energy dispersive X-ray spectroscopy. Both surfaces were laminated/molded with the same rubber (PDMS); therefore no difference in the chemical bonding characteristics are expected. However, Figure 2d shows that much more rubber remains on the “extra rough” copper than on the “rough” copper (Fig. 2c).

Analyzing a cross-section of the “extra rough” copper samples (Fig. 2e) provides interesting details on this point. Even though macroscopically the interface is loaded under 90 $^{\circ}$, locally only part of the interface at the bottom of the roughness “valleys” is loaded in mode I, whereas the part of the interface at the side walls of the “valleys” are loaded close to mode II. In addition, the irregular surface morphology increases the surface area, effectively increasing the interface strength. Finally, Figure 2e shows that the “extra rough” copper surface contains some “valleys” that geometrically enclose the rubber, thereby mechanically locking it inside. This shows that rubber fracture is influenced by the fine-scale geometrical details of the bonded interface, resulting in a larger rubber fraction being left behind on the copper surface after delamination. The fact that the WOS is also 40% higher shows that a significant amount of energy is dissipated in fracturing the rubber (rather than in the interface debonding).

To investigate the influence of the rubber type on the delamination, the three above-mentioned rubber types were delaminated from the copper surface. The amount of rubber remaining on the surface after delamination, A_r , was determined by segmenting SEM images of the delaminated surface (top images of Fig. 3). Surprisingly, the results showed an inverse relation between the WOS and A_r , with the TPU200 samples having the highest WOS but the lowest rubber area fraction A_r , whereas the PDMS samples combine the lowest WOS with the highest remaining rubber area fraction.

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