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Multi-scale experimental analysis of rate dependent metal–elastomer interface mechanics

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ABSTRACT

A remarkable high fracture toughness is sometimes observed for interfaces between materials with a large elastic mismatch, which is reported to be caused by the fibrillar microstructure appearing in the fracture process zone. In this work, this fibrillation mechanism is investigated further to investigate how this mechanism is dissipating energy. For that purpose, thermoplastic urethane (TPU)-copper interfaces are delaminated at various rates in a peel test experimental setup. The fracture process zone is visualized *in situ* at the meso-scale using optical microscopy and at the micro-scale using Environmental Scanning Electron Microscopy (ESEM). It is shown that the geometry of the fracture process zone is insensitive to the delamination rate, while the interface traction scales logarithmically with the rate. This research has revealed that, the interface roughness is shown to be pivotal in initiating the fibrillation delamination process, which facilitates the high fracture toughness. The multi-scale experimental approach identified two mechanisms responsible for this high fracture toughness. Namely, the viscous dissipation of the TPU at the high strain levels occurring in the fibrils and the loss of stored elastic energy which is disjointed from the propagation due to the size of the process zone.

1. Introduction

Interfaces between materials with a large elastic mismatch are of increasing scientific and industrial interest, as they are more frequently used in advanced engineering materials and devices, including, e.g. composites (Kim and Sham, 2000), coatings (van den Bosch et al., 2008), microelectronics (Yao and Qu, 2002; van der Sluis et al., 2014), and flexible (Vella et al., 2009; Xu et al., 2011) and stretchable electronics (Loeher et al., 2006; Hsu et al., 2010a). The main benefit of the synergy between these materials lies in combining the high stiffness of one material with the high toughness and compliance of the other, usually soft material. Major improvements in material properties can be achieved if the interfaces are strong enough to transfer the applied loads and deformations. Consequently, interface failure often is a precursor to failure (Li et al., 2005; van der Sluis et al., 2008; Hsu et al., 2009). Improving the interface properties thereby generally improves the robustness of multi-layered devices.

Previously, a two elastomer-metal interface system is discussed with a large elastic mismatch and a large interface

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roughness, for which a remarkably high macroscopic fracture toughness was observed ($\mathcal{G}_c > 2\ \text{kJ/m}^2$) (Hoefnagels et al., 2010). The coarse scale toughness largely exceeds the adhesion energy of metal–rubber valence bonds, reported to be in the range of $0.1 < \Gamma_{ad} < 10\ \text{J/m}^2$ (Creton et al., 1992, 2001; Crosby et al., 2000). *In situ* observation of the delaminating peel front of these interfaces reveals the formation of microscopic fibrils. However, the exact micromechanical origin of the enhanced fracture toughness is so far poorly understood. To enable the engineering of interfaces with enhanced fracture toughness, it is important to understand the underlying mechanics that amplifies the fracture toughness of these interface systems.

Therefore, the goal of this paper is to unravel the delamination micromechanics which are responsible for dissipating the large quantities of energy in such ductile interfaces. To this end, the 'ThermoPlastic Urethane (TPU)'–copper interface system will serve as a model system for this investigation. The strategy departs from *in situ* high-magnification visualization of the delaminating TPU–copper peel front for different copper roughnesses. The interface micromechanics are evaluated at a large range of peel rates to investigate delamination rate sensitivity and to possibly activate different dissipation mechanisms without altering the interfaces.

2. Experimental methodology

The samples are created from commercially available laminated bi-layer TPU–copper foils (TPU: 50 μm Walopur, Epurex; copper: 37 μm TW-YE, ArcelorMittal), similar to what is used in stretchable electronic applications (Loeher et al., 2006; Hoefnagels et al., 2010). The TW-YE is a circuit grade, rolled copper foil with one relatively smooth untreated side ($R_a \approx 0.5\ \mu\text{m}$) and one rough side, which received an extra electroplating step ($R_a \approx 1.9\ \mu\text{m}$). Four-layer specimens are made by laminating two as-received TPU–copper bi-layer foils, enabling peel tests to be performed by clamping and pulling on a copper foil at each specimen arm, effectively creating a T-peel test. Such a T-peel test is chosen because of its optical accessibility and the stationary location of the peel front which is beneficial for imaging at high magnifications.

Sample type 'rough' is created by laminating TPU to TPU at 180 °C, with one part of the foil not laminated to form a pre-crack (Fig. 1, top figure). Sample type 'smooth' is created by laminating TPU to the smooth copper side (Fig. 1, bottom figure), using the same lamination procedure to induce as little variation with the rough sample type as possible. Fig. 2a shows a cross-section image taken with an optical microscope (Zeiss Discovery V20), which shows that the layer thicknesses are close to the specified thickness. Additionally, a cross-section of the copper was prepared by polishing a specimen which was embedded in epoxy, for optimal image quality. This cross-section is then imaged in a scanning electron microscope to show the complex roughness topology (Fig. 2b).

The topography of the smooth and rough side of TW-YE copper foil is measured directly using Confocal Optical Profilometry (Sensofar Plμ2300), see Fig. 3, clearly showing the contrast in roughness between the two surfaces. Nicely shown is the cauliflower-type topography of the rough surface, which is typical for electroplated metal.

The samples are delaminated in a T-peel test or 180° peel test configuration (Fig. 4a) using the micro-tensile stage (Kammrath & Weiss GmbH) shown in Fig. 4b. In such a configuration, the two arms of the pre-crack are clamped, while the laminated part of the sample is supported in a low-friction guide to enforce a perpendicular angle with respect to the loading direction, without significantly dissipating energy.

The tensile stage is small enough to fit underneath the objective of the optical microscope, or inside the vacuum chamber of an environmental scanning electron microscope (ESEM). By using the environmental mode of the ESEM, it is possible to prevent 'charging' effects, enabling high resolution imaging of the TPU. The tensile setup is such that the peel front propagates stationary allowing *in situ* high magnification ESEM imaging of the progressing delamination front, see e.g. Fig. 5. This image shows the complex micromechanics at the peel front, which is the focus of investigation in this paper. Note that the electron beam irradiation might easily alter the fibrillation process. This becomes clear by temporarily exposing the progressing delamination front with a typically used electron beam intensity, indicated by the rectangles in Fig. 5. Obviously, influence of the imaging technique on the delamination micromechanics is undesirable. Therefore, a preliminary study was carried out by careful comparison of exposed and unexposed adjacent surfaces, yielding optimized ESEM electron beam

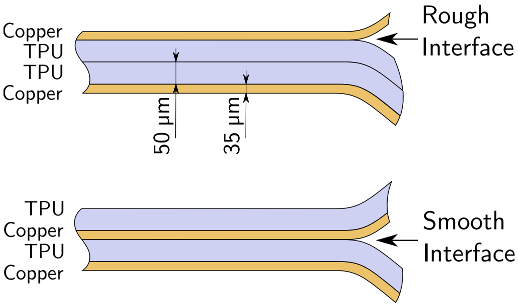

Fig. 1. Two sample configurations, where the rough interface type is created by laminating to TPU–copper sheets TPU side to TPU side and the smooth interface type is created by laminating two sheets TPU side to copper side, effectively creating four layer samples.