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On the role of fibril mechanics in the work of separation of fibrillating interfaces

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ABSTRACT

High values for the work of separation have been reported in peel tests on fibrillating interfacial systems. The exact origin of these high values is not properly understood, since it remains unclear which dissipative mechanisms related to fibrillation cause a significant increase in the work of separation. In this paper, the contribution of fibril mechanics to the work of separation is quantified. To this end, a micromechanical model of a single fibril is used, in which the growth of a nucleated fibril up to the moment of fracture is described. The initial geometry is varied to reflect the variability in the substrate profile. It is observed that the stresses and strains that occur in the model are well beyond typical bulk values. Given the large variation in measured stress–strain curves for rubber materials reported in literature, a small scale single fibril experiment is performed to assess the applicability of reported bulk material parameters to this problem. The obtained experimental response falls well within the model bandwidth for the range of material parameters from literature. Furthermore, the fibril fracture stress, which serves as the failure criterion in the model, is extracted from the experiment. From the micromechanical model results, it appears that, for the considered range of material properties, the work of separation is mainly influenced by the fracture stress. Furthermore the initial geometry has a profound influence on the obtained work of separation. It is concluded that the work of separation determined from the micro-model is significantly larger than the intrinsic adhesion energy, yet it remains still an order of magnitude smaller than the values reported in literature. For the considered system this indicates that, although it has a significant contribution, fibril deformation is not the only contribution to the high work of separation.

1. Introduction

A challenge in stretchable electronics is to accommodate the large difference in stiffness between the copper conductors and the compliant substrate, which is typically made of a rubber material. It is observed that the reliability of the device is dictated by the integrity of the copper–rubber interface, since failure of the interface triggers localization of deformation in the metal interconnect, as suggested by Lu et al. (2007), followed by fracture, and ultimately to loss of the electronical functionality. Furthermore, interface delamination may entail exposure of the interconnect to the environment, which can also lead to early failure (Hsu et al., 2010). Previous work has shown that interface

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delamination mainly occurs through fibrillation. This process involves the formation, growth, and failure of rubber fibrils (Hoefnagels et al., 2010).

Clearly, improving stretchable electronics reliability requires good understanding of the interface properties. Previously, macroscopic finite element models have been used to obtain the interface properties. In these models, the interface is described using cohesive zone elements and the parameters are obtained through fitting of the model response on the measured response in peel tests. Using this approach, high values of the work of separation were obtained (Neggers, 2013; van der Sluis et al., 2011) with an adequate correspondence between model and experiment. From a physical perspective, some discrepancies resulting from this approach persist, stemming from the fact that all dissipative mechanisms are lumped in the macroscopic interface properties. There is no clear relation between the obtained cohesive zone parameters and the observed fibrillation micromechanics: in fact, the cohesive zone critical opening length was approximately 6 times larger than the observed maximum fibril length (Hoefnagels et al., 2010). Furthermore, using a common traction-separation law (TSL), such as the exponential one, leads to spurious tractions on the interface, i.e. nonzero tractions are acting on parts of the interface where complete failure occurred in the experiment.

To overcome these concerns, a multi-scale interface model was recently developed (Vossen et al., 2014). In this model, the macroscopic interface is still modelled with cohesive zone elements. Yet, instead of using a predefined TSL, the traction-separation response (TSR) is obtained from an underlying micro-model. In the micro-model, relevant mechanisms such as fibril growth and debonding are incorporated. Mode I loading was imposed on a nucleated single fibril and the fibril growth was studied up to instantaneous debonding of the fibril from the substrate, for various values of the micro-scale interface properties. It was shown that for most values of the micro-scale interface parameters, the macroscopically observed work of separation consists mainly of elastically stored energy in the fibril, which is dissipated through dynamical release upon instantaneous fibril debonding. Using this approach, both of the mentioned shortcomings were addressed: the TSR showed a clear relation with the fibrillation micromechanics, and, as a result, spurious tractions acting on the interface at the macro-scale were absent.

Nevertheless, even though the developed model was a clear improvement over the classical single scale models in a qualitative sense, the results did not fully correspond to the experiments in a quantitative manner. Most importantly, the micro-model could not explain the origin of the high work-of-separation measured in peel tests. The macroscopic work-of-separation obtained from the micro-model was in the order of 2 to 3.5 $\mathrm{J/m^2}$, whereas experimental values of 300 $\mathrm{J/m^2}$ (Neggers, 2013) up to 1000 $\mathrm{J/m^2}$ (van der Sluis et al., 2011) have been reported. Even though the micro-model revealed a work-of-separation that was almost two orders of magnitude larger than the micro-scale adhesion energy, still two decades remain to be bridged in order to match the experiments.

Tack experiments have been used to gain insight into the mechanics of fibrillation and the corresponding work of separation in (typically) pressure sensitive adhesives, see for example Creton et al. (2001), Lakrout et al. (1999), Brown et al. (2002), Tanguy et al. (2014), Shull and Creton (2004) and Yamaguchi et al. (2007). In a tack test, mode I loading is imposed on the material. Upon loading, the adhesive fibrillates and the work of separation can be determined straightforwardly from the measured force–displacement response, e.g. Brown et al. (2002) and Lakrout et al. (1999). However, due to the difference in material behavior and in the size of the fibrils, the results of the tack experiments are of limited relevance for an improved quantitative understanding of the work of separation in the peel tests considered in this work.

In this paper, the energetic contribution of fibril mechanics to the work of separation will be analyzed in a rigorous way. Very large deformations are observed in the micro-model (principal true strain up to 2), which necessitates a material model that captures the strain hardening due to locking of the rubber network at large deformations. For this purpose, the Ogden model (Ogden, 1972) will be used to describe the mechanical behavior of the fibrils, since it accurately captures the stress upsweep.

Literature data on material parameters for rubber materials show a large variation. Furthermore, since the stresses and strains in the model are well beyond typical bulk values, it is imperative to critically assess the applicability of bulk material parameters from literature to this deformation regime. To this end, a single fibril experiment is proposed. In the experiment, the applied load, and the evolving geometry of a single rubber fibril are measured, which enables a detailed comparison with the model, providing the necessary data for a parameter assessment in the model.

Based on the resulting constitutive model, single fibril simulations are performed. The fibril response up to reaching the critical stress is analyzed and the work of separation is determined. Furthermore, the influence of the initial geometry and the material parameters on the work of separation is analyzed. The contribution of the dissipated energy directly resulting from fibril deformation to the overall work of separation is quantified.

The outline of this paper is as follows: first, the single fibril experiment used for the model assessment is introduced, along with the experimental results and the extracted failure criterion. Using the single fibril experiment, the adequacy of the constitutive model is analyzed. Based on the resulting model, the macroscopic work of separation is determined and the influence of the material parameters and the initial geometry is shown. Finally, the implications and limitations of the fibril model are discussed.

2. Single fibril experiment

The aim of the single fibril experiment is to process and test a fibril in a controllable way. Furthermore, the extension of the fibril should proceed in a similar manner as in peel