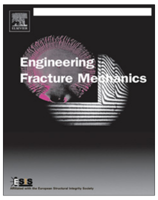
ARTICLE IN PRESS

Engineering Fracture Mechanics xxx (2016) xxx–xxx

Contents lists available at ScienceDirect

Engineering Fracture Mechanics

journal homepage: www.elsevier.com/locate/engfracmech



High toughness fibrillating metal-elastomer interfaces: On the role of discrete fibrils within the fracture process zone

B.G. Vossen [a], O. van der Sluis [a,b,*], P.J.G. Schreurs [a], M.G.D. Geers [a]

[a] *Department of Mechanical Engineering, Eindhoven University of Technology, P.O. Box 513, 5600 MB Eindhoven, The Netherlands*
[b] *Philips Research Laboratories, High Tech Campus 34, 5656 AE Eindhoven, The Netherlands*

ARTICLE INFO

Article history:
Received 25 February 2016
Received in revised form 16 May 2016
Accepted 20 May 2016
Available online xxxx

Keywords:
Fibrillation
Stretchable electronics
Delamination
Fracture process zone
Work-of-separation
PDMS

ABSTRACT

Fibrillating metal-elastomer interfacial systems, typically used in stretchable electronics applications, can exhibit remarkably high values for the interface fracture toughness. Consequently, a huge gap exists between the low adhesion energy at the microscopic scale and the measured macroscopic work of separation. This contribution aims to close this energy gap by unravelling the underlying dissipative mechanisms through a multi-scale approach. The first scale transition was established in earlier work, and concerned the formation and deformation of a single fibril at the copper-rubber interface up to failure. It was shown that the obtained work of separation was significantly larger than the small-scale interface adhesion, yet a decade too small with respect to the experimental values. In order to close the energy gap, in this contribution, the second scale transition is achieved by considering a finite number of elongating discrete hyperelastic fibrils within the fracture process zone. It is shown that the dynamic release of the stored elastic energy by fibril fracture that results from the spatial discreteness of multiple fibrils, the interaction with the adjacent deforming bulk elastomer material and the highly nonlinear behavior of the elastomer provides an explanation for the high work of separation values. In addition, an intrinsic shortcoming of cohesive zone formulations at the macroscopic scale is revealed. The results provide a mechanistic understanding of the physics involved with interface delamination through fibrillation in metal-elastomer interfaces.

1. Introduction

Stretchable electronics are nowadays finding applications in a variety of novel products, ranging from electronic textiles to biomedical applications such as epidermal electronic systems, stretchable optoelectronics and advanced surgical tools [2,9,10,18–20,24,33,38]. These applications benefit from the increased design freedom and comfort offered by devices that are deformable, as opposed to the conventional stiff semiconductor microelectronics. Stretchable electronics typically consist of semiconductor islands, embedded in highly compliant substrates, often made of a rubber material. The electrical connectivity is achieved through thin metal interconnect lines. The mechanical integrity of the conductor lines is one of the main reliability issues for stretchable electronics, since fracture of the conductor lines, as discussed by Gonzalez et al. [6] and Li and Suo [23], will lead to immediate failure of the product due to the loss of electrical functionality. It has been observed that

---

* Corresponding author at: Department of Mechanical Engineering, Eindhoven University of Technology, P.O. Box 513, 5600 MB Eindhoven, The Netherlands.

E-mail address: o.v.d.sluis@tue.nl (O. van der Sluis).

http://dx.doi.org/10.1016/j.engfracmech.2016.05.019
0013-7944/© 2016 Elsevier Ltd. All rights reserved.

Please cite this article in press as: Vossen BG et al. High toughness fibrillating metal-elastomer interfaces: On the role of discrete fibrils within the fracture process zone. Engng Fract Mech (2016), http://dx.doi.org/10.1016/j.engfracmech.2016.05.019

Nomenclature

G_c	work-of-separation
PDMS	poly(dimethylsiloxane)
H_f	fibril height
w	fibril width
h	spacing between fibrils
H	bulk height
W	strain energy density function
λ	principal stretch ratio
κ	bulk modulus
J	volume change ratio
$\mathbf{F}$	deformation gradient tensor
c_k	material parameter
m_k	material parameter
$\boldsymbol{\sigma}$	Cauchy stress tensor
p_c	critical first Piola-Kirchhoff stress
Γ_f	fibril energy loss
Γ_{bc}	bulk energy loss, homogeneous part
Γ_{bf}	bulk energy loss near bulk-fibril boundary

the reliability of the product is mainly dictated by the integrity of the interface between the conductor lines and the substrate. Lu et al. [26] discuss that, as long as the interface is intact, the strain in the conductor lines is delocalized by the substrate. Delamination may act as a precursor to necking of the metal and subsequently to fracture, thus leading to electrical failure. Furthermore, Hsu et al. [10] point out that delamination may cause exposure of the interconnect to the environment, possibly leading to premature metal failure due to the effect of for example humidity or salt corrosion. Clearly, understanding the interface behavior is of key importance to engineer reliable products.

Dedicated experiments on these particular metal-elastomer systems, Cu-poly(dimethylsiloxane) (PDMS) rubber, provide better understanding of the actual mechanisms in the fracture process zone. It has been observed that the interface delaminates through rubber fibrillation, a process that involves the formation, elongation and rupture of rubber fibrils, as shown in Fig. 1. From these peel experiments, notably high values for the work of separation have been reported [8,37], $G_c > 1\ \mathrm{kJ/m^2}$. This contribution aims to unravel the huge gap between the microscopic adhesion energy of a copper-rubber system and its measured macroscopic work of separation which is of key importance for interfacial engineering of stretchable electronics.

A vast body of literature on fibrillar structures exists, see for example the review paper by Jagota and Hui [17]. Here, fibrillar structures are studied that are in direct contact with the substrate, resulting in the loss of elastically stored energy in the fibril upon detachment, which is considered to be the main dissipative mechanism in these systems. Jagota and Bennison [16] used such a system to study adhesion enhancement induced by a fibrillar structure. Later, Glassmaker et al. [4], Hui et al. [12] studied the contact and adhesion between a PDMS fibrillar structure and a rigid substrate, mainly focusing on the relation between the pull-off stress and the properties of the fibrils. The adhesion enhancement has been studied in more detail by e.g. Glassmaker et al. [5], Shen and Soh [34], Tang et al. [39]. Typically, the energy loss is calculated analytically assuming linear elastic material behavior, which enables to directly establish the amount of energy in a fibril based on a given (simple) geometry loaded up to a certain stress level. More recently, Guidoni et al. [7] used finite strain elasticity to determine the energy contribution of the fibrils. The aforementioned studies do not account for the energy storage and loss in the bulk layer interconnecting the fibrillar structure. Although Glassmaker et al. [5] observed a discrepancy between their theoretical model and experimental data, which was attributed to the viscoelastic energy loss in the bulk region outside the fibrils, they

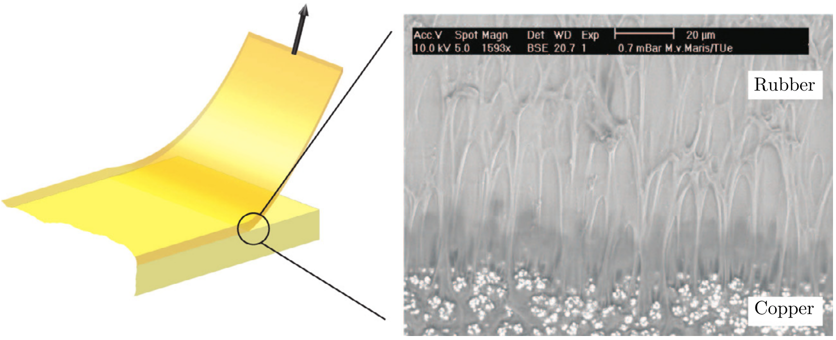


Fig. 1. *In situ* scanning electron microscopy observation of PDMS fibrillation during a Cu-PDMS peel test.

Please cite this article in press as: Vossen BG et al. High toughness fibrillating metal-elastomer interfaces: On the role of discrete fibrils within the fracture process zone. Engng Fract Mech (2016), http://dx.doi.org/10.1016/j.engfracmech.2016.05.019