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In situ observation of contact mechanisms in bioinspired adhesives at high magnification

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Abstract

We analyzed the contact mechanisms of bioinspired microfibrillar adhesives using in situ scanning electron microscopy. During adhesion tests we observed that (i) the superior adhesion of mushroom-shaped fibrils is assisted by the stochastic nature of detachment, (ii) the aspect ratio of microfibrils influences the bending/buckling behavior and the contact reformation, and (iii) the backing layer deformation causes the microfibrils to elastically interact with each other. These studies give new insights into the mechanisms responsible for adhesion of bioinspired fibrillar adhesives.

Introduction

During the last decade, researchers have developed sophisticated methods to fabricate adhesives inspired by the adhesion system of the gecko. Fibrillar structures were fabricated to mimic the natural system and to maximize adhesive performance.[1–6] Numerous experimental and theoretical studies were carried out to advance the understanding of the underlying contact mechanics.[4,7–13] However, to validate those models, in situ visualization of the contact phenomena at high magnification is necessary. Several groups have presented studies using optical microscopy combined with adhesion measurements.[14–17] These studies show, for example, the bending of fibrils, the actual contact area, or the attachment and detachment front. Although these experiments have given some insight into contact mechanisms, they are not suitable for investigating the processes at interfaces due to the limited magnification of the applied optical microscopy. Advanced bioinspired adhesives have features such as flaps (mushroom tips), which are in the sub-micrometer range.[1,18–20] To visualize the contact mechanisms at the length scale of these features, new measurement systems are required. Here, we propose scanning electron microscopy assisted contact mechanics visualization during an adhesion test with fibrillar surfaces.

Experimental methods

Microfibrillar arrays of polydimethylsiloxane (PDMS) with pillars of different aspect ratios (height/diameter) and tip geometries were fabricated using photolithography and soft-molding processes.[21] SU-8 resists (2010, 2025, Micro Resist Technology, Berlin, Germany) were used to prepare templates in a standard photolithography process. The templates were then silanized using hexadecafluoro-1,1,12,2-tetrahydrooctyltrichlorosilane and subsequently filled with PDMS (Sylgard 184, Dow Corning, 10:1 mixture). After cross-linking at 75 °C for at least 14 h, the samples were carefully demolded. For specific fabrication details, especially the fabrication of mushroom tip structures, see Refs.[4,21]

Contact experiments were performed in an environmental scanning electron microscope (ESEM, FEI Quanta 400F), extended with a micromanipulator (Kleindieck Nanotechnik, GmbH, Germany) and a self-constructed cantilever for force measurements. Probes were mounted on the tip of the micromanipulator and the sample was fixed to the cantilever. The low vacuum and the environmental modes of the ESEM allowed performing the experiments on non-conductive materials such as PDMS without further treatment.

In situ tests were performed by positioning the probe at the focal point using the micromanipulator. The sample was brought into contact with the probe using the ESEM stage and retracted again. This enabled high displacement with high magnification without losing the focus on the interface. At the same time, the deflection of the cantilever during the contact experiment was indicative of the resulting forces.

Results and discussion

Experiments were performed to investigate the following aspects of contact phenomena for fibrillar surfaces: influence of pillar tip shape on adhesion, bending and buckling of pillars under compression, and response of the backing layer to applied pressure.

Influence of pillar tip shape on adhesion

Figure 1 shows sequences of side images taken during contact experiments using a spherical probe (on top) on a PDMS fibrillar array with heights of 20 μm and aspect ratio 2. In the top

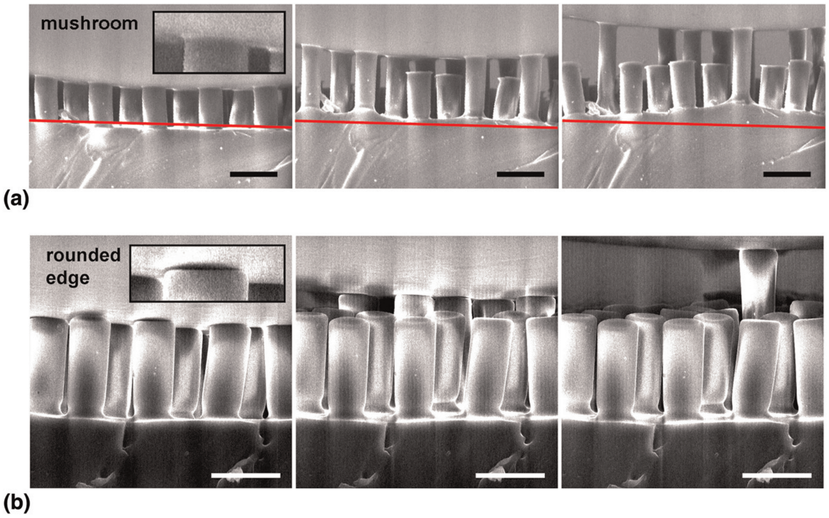

Figure 1. Sequences of ESEM side views of pillars with mushroom tips (top) and rounded edges (bottom). Pictures were taken at maximum compressive load (left), during retraction of the spherical probe (centre) and immediately before detachment. Inlays show interface during contact formation at high magnification, scale bar 20 μm.

sequence the pillars had tips with mushroom shape, while in the bottom sequence the pillars had rounded edges. For each sequence the first image shows the pillars at maximum applied compressive preload. The inlays show the interface at high magnification during contact formation of a single pillar. The subsequent images exhibit an intermediate tensile stress state during retraction of the sample and the last image was taken immediately before detachment. Both samples show adhesion to the spherical probe.

However, while most of the pillars with rounded edges detach at low applied tensile strain, a large fraction of the mushroom-shaped pillars sustain much higher strains before detachment. Due to the curvature of the spherical probe, it was expected that the pillars at the contact boundary, which experience the highest strain, would be the first to detach and the detachment front would proceed to the center of the contact area. Instead, the pillars detached in a stochastic manner rather than in an orderly fashion. In addition, individual pillars showed very different elongations prior to detachment due to different adhesive strengths of the individual pillars. The strain before detachment was found to be approximately 65% for mushroom tip pillars and 55% for pillars with rounded tips. While several mushroom tip pillars in this experiment were extended to maximum elongation, this was found only for a single pillar with rounded edges. This observation indicates that the tip shape, even if it may not significantly affect the adhesion of one single pillar, greatly influences the statistics of detachment in an array of pillars.[22]

Bending and buckling of pillars (pressure switching behavior)

Previous studies have pointed out that the adhesive strength for fibrillar arrays drops significantly at high preload; this effect has been exploited in the design of an adhesive system that allows

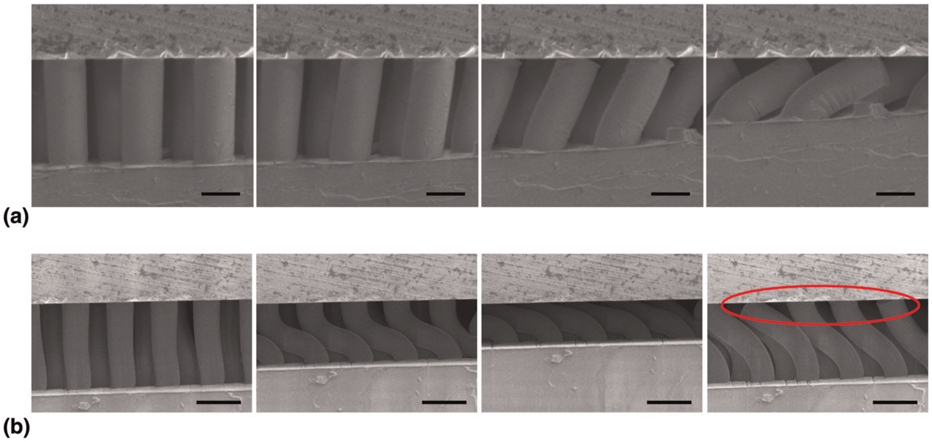

Figure 2. Switching behavior due to bending and buckling of pillars with mushroom tips having aspect ratio 3 (top), scale bar 10 μm and aspect ratio 5 (bottom), scale bar 20 μm.

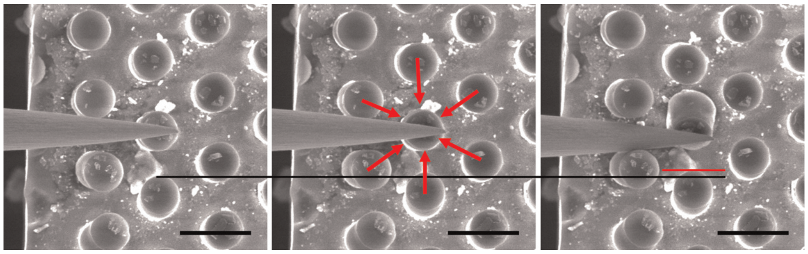

Figure 3. Experimental evidence for elastic interaction of the pillars through the backing layer: when the central pillar is compressed with a needle, the adjacent pillars bend towards the central pillar (see marker line), scale bar 20 μm.

switching between an adhesive and a non-adhesive state.[21,23] The switching was attributed to the loss of contact of the pillar tips due to bending and/or buckling. The sequences in Fig. 2 show the behavior of mushroom tip pillars with aspect ratios 3 (top) and 5 (bottom) under compressive preload. In the initial state, the pillars are in intimate contact with the flat probe. With increasing load, the low aspect ratio pillars in sequence start to bend, lose tip contact, and finally lie flat at maximum applied compressive preload. In the last image of the sequence, the bent pillars show surface wrinkles, which have not been observed before during adhesion tests on fibrillar surfaces and might result from the high compressive stresses.

The high aspect ratio pillars buckle with increased compressive preload and exhibit an "S" shape after buckling. Compared with the aspect ratio 3 pillars, the pillars in sequence (b) sustain tip contact even at large deformation and pillars lose tip contact only at very high preload. Unlike the pillars in sequence (a), the high aspect ratio pillars appear to regain tip contact during unloading. This new visualization technique enables us to precisely observe the deformation behavior of pillars under compressive loading and may lead to a better understanding of bending and buckling phenomena of small-scale polymeric materials.

Backing layer response to applied pressure

Whereas several mechanics studies acknowledge the importance of the backing layer contribution to the adhesion of fibrillar arrays,[24,25] the interaction between the pillars through the backing layer has not yet been observed experimentally. Figure 3 shows the response of six hexagonally packed pillars to a load applied to the central pillar. For this experiment, a sharp needle was attached to the micromanipulator. To better visualize the deformation, high-contrast particles were dispersed on the fibrillar surface. The applied load presses the central pillar into the backing layer, causing the neighboring pillars to be slightly bent towards the central pillar. The displacement of one of the surrounding pillars is highlighted by the guideline in Fig. 3. In addition to the top view, the backing layer interaction can also be visualized in side view as seen from Fig. 1 (a). There, the red guideline roughly indicates the initial position of the backing layer. It can be seen that the tension in the pillars causes a severe deformation of the backing layer, affecting the orientation of the adjacent pillars. This effect will be especially important for adhesion to rough or spherical surfaces. For example, if a pillar adheres to a spherical probe or an asperity of a rough surface, the surrounding pillars experience a bending moment away from the adhering pillar. This change in orientation of the neighboring pillars with respect to the contacting surface may exert significant influence on the adhesion. Besides this, the severe deformation of the backing layer greatly contributes to the stored elastic energy of the adhesive system and, thus, has to be considered in the energy balance of the contact mechanics models.

Conclusions

We presented a new setup for adhesion experiments with bioinspired fibrillar surfaces using in situ visualization in an environmental scanning electron microscope at high magnification. The influence of tip shape, bending, and buckling under compressive preload and the interaction of pillars through the backing layer on adhesion were investigated and gave additional insight not available otherwise.

Mushroom-shaped pillars did not show a notably higher elongation before detachment than pillars with rounded edges; however, the fraction of pillars retaining contact with the probe at maximum elongation was significantly larger. This points to the stochastic nature of the detachment process of fibrillar surfaces, which may be crucial for adhesive performance.

The aspect ratio of the pillars determined the deformation mode under compressive preload. In our experiments, pillars with aspect ratio 3 bent and lost tip contact. Aspect ratio 5 pillars buckled before losing tip contact and regained contact again during unloading. Such observations support the rational design of switchable adhesive devices exploiting an adhesive/non-adhesive transition.

With the new setup, it was possible to visualize the interaction of individual pillars through the backing layer in top and side view. Overall, these observations give new insight into the adhesion mechanisms of fibrillar adhesives and will, by improving the mechanistic understanding, support the design of bioinspired adhesive surfaces.

References

1. A. del Campo, C. Greiner, I. Alvarez, and E. Arzt: Patterned surfaces with pillars with controlled 3D tip geometry mimicking bioattachment devices. *Adv. Mater.* **19**, 1973 (2007).
2. M. Sitti and R.S. Fearing: Synthetic gecko foot-hair micro/nano structures as dry adhesives. *J. Adhes. Sci. Technol.* **17**, 1055 (2003).
3. B. Schubert, C. Majidi, R.E. Groff, S. Baek, B. Bush, R. Maboudian, and R.S. Fearing: Towards friction and adhesion from high modulus microfiber arrays. *J. Adhes. Sci. Technol.* **21**, 1297 (2007).
4. A. del Campo, C. Greiner, and E. Arzt: Contact shape controls adhesion of bioinspired fibrillar surfaces. *Langmuir* **23**, 10235 (2007).
5. L. Ge, S. Sethi, L. Ci, P.M. Ajayan, and A. Dhinojwala: Carbon nanotube-based synthetic gecko tapes. *Proc. Natl. Acad. Sci. USA* **104**, 10792 (2007).
6. H.E. Jeong, J-K. Lee, H.N. Kim, S.H. Moon, and K.Y. Suh: A nontransferring dry adhesive with hierarchical polymer nanohairs. *Proc. Natl. Acad. Sci. USA* **106**, 5639 (2009).
7. A. Jagota and S.J. Bennison: Mechanics of adhesion through a fibrillar microstructure. *Integr. Comp. Biol.* **42**, 1140 (2002).
8. E. Arzt, S. Gorb, and R. Spolenak: From micro to nanocontacts in biological attachment devices. *Proc. Natl. Acad. Sci. USA* **100**, 10603 (2003).
9. C. Greiner, A. del Campo, and E. Arzt: Adhesion of bioinspired micropatterned surfaces: effects of pillar radius, aspect ratio and preload. *Langmuir* **23**, 3495 (2007).
10. R. Spolenak, S. Gorb, H. Gao, and E. Arzt: Effects of contact shape on the scaling of biological attachments. *Proc. Math. Phys. Eng. Sci.* **461**, 305 (2005).
11. C.Y. Hui, N.J. Glassmaker, T. Tang, and A. Jagota: Design of biomimetic fibrillar interfaces: 2. Mechanics of enhanced adhesion. *J. R. Soc. Interface* **1**, 35 (2004).
12. A.V. Spuskanyuk, R.M. McMeeking, V.S. Deshpande, and E. Arzt: The effect of shape on the adhesion of fibrillar surfaces. *Acta. Biomater.* **4**, 1669 (2008).
13. R. Rong and C.Y. Hui: The effect of preload on the pull-off force in indentation tests on microfiber arrays. *Proc. R. Soc. A* **465**, 961 (2009).
14. N.J. Glassmaker, A. Jagota, C.Y. Hui, W.L. Noderer, and M.K. Chaudhury: Biologically inspired crack trapping for enhanced adhesion. *Proc. Natl. Acad. Sci. USA* **104**, 10786 (2007).
15. N.J. Glassmaker, A. Jagota, C.Y. Hui, and J. Kim: Design of biomimetic fibrillar interfaces: 1. Making contact. *J. R. Soc. Interface* **1**, 23 (2004).
16. A.J. Crosby, M. Hageman, and A. Duncan: Controlling polymer adhesion with “Pancakes”. *Langmuir* **21**, 11738 (2005).
17. S. Vajpayee, A. Jagota, and C.Y. Hui: Model independent extraction of adhesion energy from indentation experiments. *J. Adhes. Sci. Technol.* **86**, 39 (2010).
18. M.P. Murphy, B. Aksak, and M. Sitti: Gecko-inspired directional and controllable adhesion. *Small* **5**, 170 (2009).
19. S. Kim and M. Sitti: Biologically inspired polymer microfibers with spatulate tips as repeatable fibrillar adhesives. *Appl. Phys. Lett.* **89**, 261911 (2006).
20. J. Lee, B. Bush, R. Maboudian, and R.S. Fearing: Gecko-inspired combined lamellar and nanofibrillar array for adhesion on nonplanar surface. *Langmuir* **25**, 12449 (2009).
21. D. Paretkar, M. Kamperman, A.S. Schneider, D. Martina, C. Creton, and E. Arzt: Bio-inspired pressure actuated adhesive system. *Mater. Sci. Eng. C* **31**, 1152 (2011).
22. R.M. McMeeking, E. Arzt, and A.G. Evans: Defect dependent adhesion of fibrillar surfaces. *J. Adhes. Sci. Technol.* **84**, 675 (2008).
23. C.Y. Hui, A. Jagota, L. Shen, A. Rajan, N. Glassmaker, and T. Tang: Design of bio-inspired fibrillar interfaces for contact and adhesion — theory and experiments. *J. Adhes. Sci. Technol.* **21**, 1259 (2007).
24. S. Kim, M. Sitti, C.Y. Hui, R. Rong, and A. Jagota: Effect of backing layer thickness on adhesion of single-level elastomer fiber arrays. *Appl. Phys. Lett.* **91**, 161905 (2007).
25. R. Rong, C.Y. Hui, S. Kim, and M. Sitti: Modeling the soft backing layer thickness effect on adhesion of elastic microfiber arrays. *J. Appl. Phys.* **104**, 044301 (2008).