The role of the backing layer in the mechanical properties of micrometer-scale fibrillar structures

Griselda Guidoni[1], Dominik Schillo[1], Ude Hangen[2], Graciela Castellanos[1], Eduard Arzt[1], Robert McMeeking[1,3] and Roland Bennewitz[1]

1: INM – Leibniz Institute for New Materials, Campus D2 2, 66123 Saarbrücken, Germany.

2: Hysitron, Inc., 10025 Valley View Road, Minneapolis, MN 55344, USA.

3: Department of Mechanical Engineering, University of California, Santa Barbara, California 93106, USA.

Abstract

The contact mechanics of a micro-fabricated fibrillar surface structure made of poly(dimethylsiloxane) (PDMS) was studied in this work. The attachment and detachment of individual fibrils to and from a spherical indenter upon approach and retraction are detected as jumps in force and stiffness. A quantitative model describes the jumps in stiffness values by taking into account the deformation of the backing layer. The results emphasize the importance of long-range interactions in the contact mechanics of elastic materials and confirm the concepts underlying the development of fibrillar adhesive materials.

Introduction

Fibrillar microstructures of elastic materials interacting with substrates by short-range molecular forces, such as Van der Waals attraction, have recently attracted attention as dry adhesives [1-8]. Their development is motivated by the study of adhesion in biological systems such as the feet of some insects and geckos [5; 6; 9-12]. These fibrillar architectures exhibit high adhesive strengths for a variety of reasons, previously summarized by Majumdar, Sharma and Ghatak [13] and subsequently by Kamperman et al. [14]. For example, fibrillar surfaces resist peeling more effectively than those without textured patterning because the detachment of one fibril does not lead automatically to the detachment of its neighbors, whereas a smooth surface can peel continuously and relatively easily due to the strain concentration at the detachment front. In addition, surfaces with compliant fibrils can conform to rough surfaces more easily than monolithic pads, and so can adhere more strongly. Other benefits arise because fibrillar surfaces can be made with small dimensions (µm to nm scale), so that in some architectures the penalty due to elastic stresses that tend to drive the separation of surfaces is reduced compared to the adhesive tractions holding them together [5], a phenomenon known as strengthening due to contact splitting. Another benefit of small dimensions is that the pull-off force achieved can rise to levels comparable with the maximum Van der Waals adhesive strength times the area in contact [15], with advantages to adhesive strength realized for dense packing of such fibrils in comparison to undivided surfaces. Structures from µm to nm dimensions also limit the size of detached regions that can be present on the surfaces in contact, so that the extent to which large defects of this nature can undermine strong adhesion is limited [16-18].

Many of the systems presented thus far are produced by a molding process with elastomers. In these systems, fibrils and backing layer are made of the same elastic material. In this study, we quantify the combined elastic response of fibrils and backing layer and show that the measured stiffness of the fibrillar system is strongly influenced by elastic deformations of the backing layer. Such long-range deformation of the substrate is an essential ingredient in the recent modeling of the mechanical response of elastic materials [19].

In our experimental work, we focus on the approach of a stiff sphere into contact with the fibrillar system. The high sensitivity of a nanoindenter allows us to register the sequence of attachment of individual fibrils. Analyzing attachment rather than detachment avoids the non-linear effects of large strain and viscoelastic response which often hamper the quantitative analysis of adhesion experiments with elastomers. In our modeling we describe the combined deformation of fibrils and backing layer in a linear model at small strains. The combination of experiment and model allows us to understand the measured stiffness as a function of the number of fibrils in contact and to determine the elastic modulus of our material by an *in situ* measurement of individual fibrils on the backing layer.

Experimental

The PDMS (Sylgard 184) samples were prepared using a soft-molding process. The tested area consisted of an arrangement of 7 fibrils of radius 5 μm each and of height 18.7 μm. They were packed in a hexagonal pattern; separated by a distance 20 μm between the centre of neighboring fibrils (see Figure 1 for an image of a larger-scale model of the structure). Nanoindentation tests were carried out using a TI 900 instrument with a Perfor-mech controller (Hysitron TriboIndenter, Hysitron Inc., Minneapolis, MN, USA). A spherical sapphire indenter (radius R = 348 μm) was used. By means of an optical microscope, the indenter was located above the middle of the centre fibril. The approach was done from a height where there was no contact and where no attractive or repulsive forces were registered.

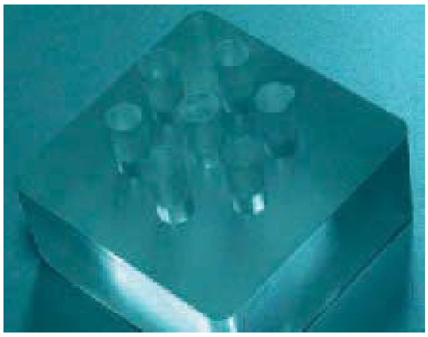

Figure 1: Model of the hexagonal arrangement of fibrils tested in this study.

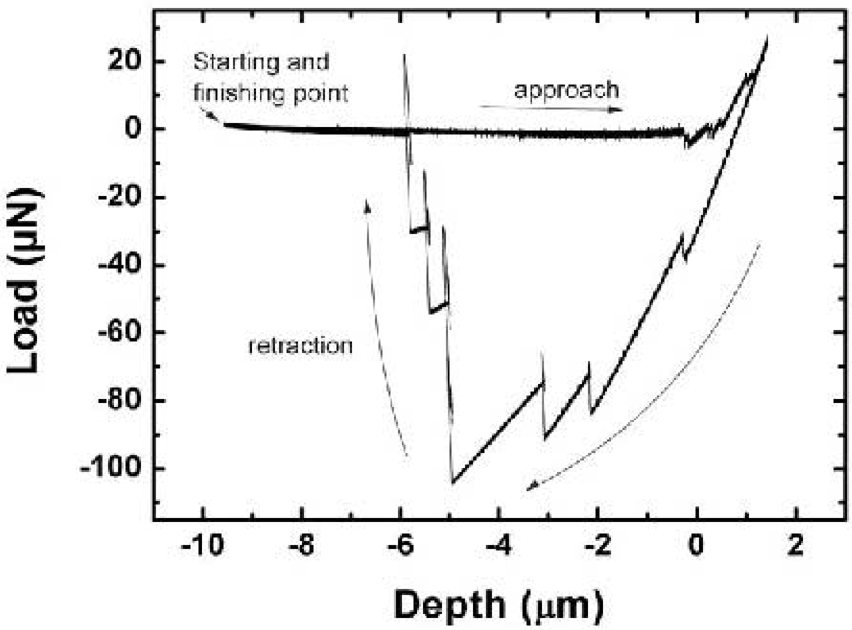

Figure 2: Force vs. depth curve during approach and retraction to and from the seven micrometer-sized PDMS fibrils on the backing layer of same material. Attachment and detachment can be recognized for each individual fibril.

All measurements were done under displacement control, at an approach and retraction rate of 200 nm/s using a close-loop feedback control.

Results

Figure 2 shows a typical load-displacement curve for approach and retraction. The individual attachment and detachment of the seven fibrils can be recognized through sudden jumps in the force. The attachment of the first fibril upon approach always generated a force drop into tension. The general shape of the approach-retract curve resembles the non-linear characteristics and the large hysteresis expected for indentation into an elastomer (see for example Figure 1 in [20]). However, the curve is composed of linear sections with different slopes separated by the force jumps. This observation reflects a step-wise increase in contact area, as we will discuss later in detail.

Figure 3 is a detailed view of the force data recorded upon approach. Each drop in force corresponds to the consecutive contact formation with single fibrils. The increasing slope of the linear sections quantifies the increase in overall stiffness with an increasing number of fibrils in contact. Correspondingly, a decrease in slope is observed in the unloading data (see Figure 2) as the number of fibrils in contact decreases.

The stiffness values (k_i), where i is the number of fibrils in contact, were determined for each section of the curve by linear fitting. Figure 4 summarizes the average stiffness values. While the stiffness does increase monotonically, it does not scale linearly with the number of fibrils in contact as it would be expected if the fibrils would act as independent identical springs in parallel. The stiffness of the nanoindenter is 1400 N/m, 35 times larger than the highest stiffness values measured in our experiments. Therefore, the deformation of the force sensor was neglected in the calculations.

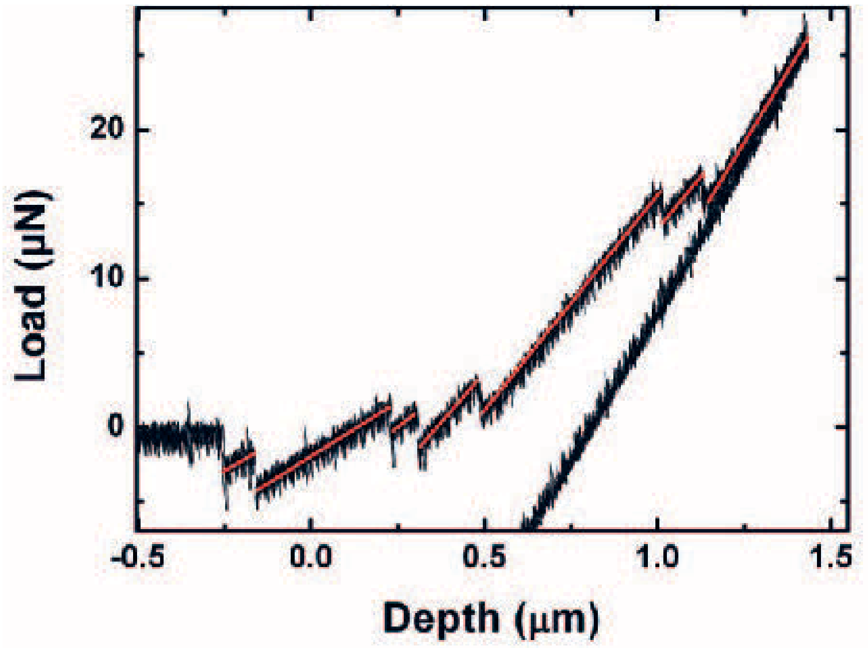

Figure 3: Detail of Figure 2, where the linear sections of the force curve between the attachment events are highlighted. The linear fitting yields the combined normal stiffness of the group of fibrils already in contact with the indenting sphere.

Discussion

For the calculation of the stiffness values k_i for i fibrils in contact a model has been developed which takes into account either the elongation or the compression of each fibril and the deformation of the backing layer. Details of the model will be published elsewhere. In short, the homogeneous stress in each fibril is assumed to act on a patch of the surface with the area of the fibril's cross section. This stress deforms the surface in a way predicted by Johnson through the methods of contact mechanics [21]. The surface deformation lifts or lowers the base of neighboring fibrils, thereby changing the stress in these fibrils. The balance between stresses in all fibrils in contact with the indenting sphere and the surface deformation defines the total stiffness of the system.

In this section we discuss the experimental results in tandem with the modeling result. We proceed chronologically with the observed events when approaching and retracting the indenter to the structured surface.

The indenter approaches the sample surface from a distance where interactions between sample and indenter cannot be detected. The first significant deviation from zero force is a sudden drop in load when the first fibril makes contact with the indenter (see Figure 2). The first contact always results in a tensile force, resulting from attractive forces between indenter and fibril. The linear increase of the force after the drop yields a value for the combined stiffness of the sample and the force sensor. The average value is k_1 = 7.7 N/m, almost two orders of magnitude lower than the stiffness of the force sensor. We conclude that the jump into contact of the first fibril occurs through a sudden stretching of the fibril and deformation of the backing. The stretching distance can be calculated as the force drop divided by the stiffness k_1. The values vary between 300 and 450 nm, i. e. typical strain in the fibril after jump into contact is of the order of 2 %.

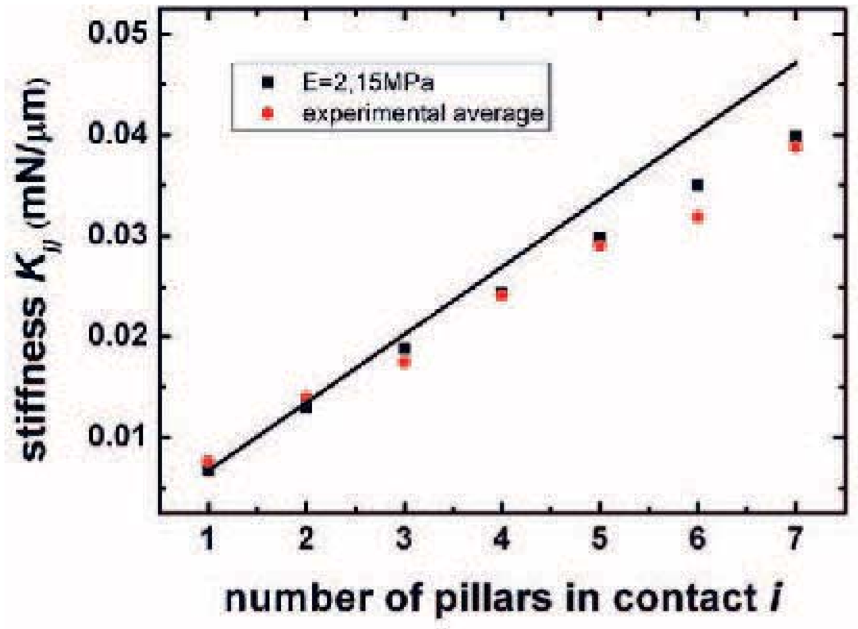

Figure 4: Stiffness k_i as function of the number of fibrils in contact with the indenter. Circles provide the average values for several experimental series, squares the results of our model. The straight line indicates the expected relation if no deformation of the backing layer was taken into account.

We estimate maximum distances for the jump-into-contact instability based on Van der Waals forces to be only 11 nm. The contribution of Van der Waals [10; 22] and capillary forces [6] to long range attractive forces has not been fully understood. Our results strongly indicate that long-range interactions like electrostatic forces have to be considered for the case of PDMS and sapphire.

The jump-into-contact events recur as the indenter moves toward the sample and the number of fibrils in contact with the indenting tip/sphere increases. At a certain indenter position, the force measured by the indenter will change from tension to compression since the stretching of the latest fibrils into contact is compensated by the compression of the earlier attached fibrils. The force vs. depth curve becomes steeper with each additional fibril in contact, reflecting the expected increase in stiffness. The linearity of the section between force drops indicates that each fibril immediately makes complete contact. Such increases in contact area in well-defined steps are very helpful for the quantitative evaluation of elasticity parameters. It has been shown that the dynamics of the stress singularities at the edge of the contact are a challenge in quantitative indentation studies in PDMS [20]. In our system, the gain in adhesion energy is large enough to elastically stretch and deform the fibrils such that full contact is established once the fibrils come close to the indenter. This observation is a clear demonstration of the optimization of contact formation by micro-structuring surfaces, a core concept of fibrillar biomimetic adhesive materials.

The stiffness increases sublinearly with the number of fibrils in contact. Our model explains this trend taking into account the deformation of the backing layer. Figure 4 compares the experimental k_i values with the model data. The straight line represents the stiffness assuming a rigid substrate. The model correctly predicts the stiffness relations. The only adjustable parameter is the elastic modulus E. It was found by minimizing the sum of square differences to the averaged experimental k_i data to be E = 2.15 ± 0.10 MPa. Note that the combination of our experimental method and our model allows for the unique *in situ* determination of the elastic modulus of PDMS fibrils within a given micro-structure. The elastic moduli of PDMS polymers similar to our material have been determined by a variety of methods and values are between 1.3 and 4 MPa [23-27]. Our method avoids the contact size problems usually encountered in indentation of flat elastomer samples [20; 24] and takes into account possible variations of the elastic modulus for a material which is cured in a confined geometry.

Upon retraction of the indenter, a significant adhesion hysteresis is observed in Figure 2. The seven fibrils are detached one after the other. However, detachment of the first of seven fibrils occurs far into the tensile regime. The shape of the adhesion hysteresis resembles the curves measured with similar indenters on flat PDMS surfaces [20]. However, for the micro-structured PDMS surface the adhesion hysteresis is far larger, emphasizing once more the function of the microstructured surface in the adhesive properties. While the maximum adhesion force on the flat PDMS surface is determined by the radius of the indenter, the adhesion force in the fibrillar system is determined by the sum of the adhesive forces of the fibrils, which is larger due to the effects discussed in the introduction. Additionally, the strength of adhesion compared to the softness of the fibrils at the micrometer scale yields perfectly flat contacts. Flat punch contacts however show a strong resistance against peeling as there is no stress singularity at the edge as in sphere-on-flat situations for frictionless contacts or a much reduced one for frictional contacts [18; 20].

Conclusion

In conclusion, we have performed contact mechanics experiments on a fibrillar micro-structured PDMS surface. Attachment and detachment of individual fibrils have been detected in force and stiffness measurements. Small deformations during approach of an indenter have been quantitatively modeled taking into account deformation of both the fibrils and the backing layer. The results emphasize the importance of long-range interactions via the substrate for the contact mechanics of elastic materials. The discrete growth of the contact area with increasing number of fibrils in contact allows an accurate *in situ* determination of the elastic modulus of the PDMS to be 2.15 MPa.

References

[1] S. Kim and M. Sitti, Appl. Phys. Rev. Lett. 89 (2006) p.261911.

[2] S. Gorb, M. Varenberg, A. Peressadko and J. Tuma, J. R. Soc.: Interfaces 4 (2007) p.271.

[3] A. del Campo, C. Greiner and E. Arzt, Langmuir 23 (2007) p.10235.

[4] C. Greiner, A. del Campo and E. Arzt, Langmuir 23 (2007) p.3495.

[5] E. Arzt, S. Gorb and R. Spolenak, Proc. Nat. Acad. Sci. 100 (2003) p.10603.

[6] G. Huber, H. Mantz, R. Spolenak, K. Mecke, K. Jacobs, S. N. Gorb and E. Arzt, Proc. Nat. Acad. Sci. 102 (2005) p.16293.

[7] A. Peressadko and S. N. Gorb, J. Adhes. 80 (2004) p.1.

[8] H. Lee, B. P. Lee and P. B. Messersmith, Nat. Lett. 448 (2007) p.338.

[9] K. Autumn, MRS Bull. 32 (2007) p.473.

[10] K. Autumn, Am. Sci. 94 (2006) p.124.

[11] K. Autumn, Y. A. Liang, S. T. Hsieh, W. Zesch, W. P. Chan, T. W. Kenny, R. Fearing and R. J. Full, Nature 405 (2000) p.681.

[12] W. Sun, P. Neuzil, T. S. Kustandi, S. Oh and V. D. Samper, Biophys. J.: Biophys. Lett. 89 (2005) p.L14.

[13] A. Majumdar, A. Sharma and A. Ghatak, Bio-Inspired Adhesion and Adhesives: Controlling Adhesion by Micro-nano Structuring of Soft Surfaces, Springer, USA, to appear.

[14] M. Kamperman, E. Kroner, A. d. Campo, R. M. McMeeking and E. Arzt, Adv. Eng. Mat. (to appear in 2010).

[15] H. Gao and H. Yao, Proc. Nat. Acad. Sci. 101 (2004) p.7851.

[16] C.-Y. Hui, Glassmaker N.J., Tang T. and A. Jagota, J. Royal Soc. Interface (2004) p.35.

[17] R. M. McMeeking, E. Arzt and A. G. Evans, J. Adhes. 84 (2008) p.675.

[18] A. V. Spuskanyuk, R. M. McMeeking, V. S. Deshpande and E. Arzt, Acta Biomat. 4 (2008) p.1669.

[19] B. Persson, J. Chem. Phys. 115 (2001) p.3840.

[20] D. M. Ebenstein and K. J. Wahl, J. Colloid Interface Sci. 298 (2006) p.652.

[21] K. L. Johnson, Contact Mechanics, Cambridge (UK), 2003.

[22] K. Autumn, M. Sitti, Y. A. Liang, A. M. Peattie, W. R. Hansen, S. Sponberg, T. W. Kenny, R.

Fearing, J. N. Israelachvili and R. J. Full, Proc. Nat. Acad. Sci. 99 (2002) p.12252.

[23] G. Bar, L. Delineau, R. Brandsch and M. Bruch, App. Phys. Lett. 75 (1999) p.4198.

[24] J. K. Deuschle, G. Buerki, H. M. Deuschle, S. Enders, J. Michler and E. Arzt, Ac. Mat. 56 (2008) p.4390.

[25] H. S. Gupta, J. Seto, W. Wagermaier, P. Zaslansky, P. Boesecke and P. Fratzl, P. Natl. Acad. Sci. USA 103 (2006) p.17741.

[26] J. Song, D. Tranchida and G. J. Vancso, Macromol. 41 (2008) p.6757.

[27] F. Schneider, T. Fellner, J. Wilde and U. Wallrabe, J. Micromech. Microeng. 18 (2008) p.065008.