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Langmuir, 2009, 25 (1), 611-617 • DOI: 10.1021/la803092d • Publication Date (Web): 08 December 2008

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Bioinspired Design of a Hierarchically Structured Adhesive

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Received September 20, 2008. Revised Manuscript Received October 28, 2008

The mechanism by which many creatures such as geckos can run at ease on a vertical wall and yet remain strongly adhered has been linked to hierarchically patterned microstructures: flexible pads, hairs, and subsurface fluidic vessels at their feet. Despite many advances, how these features of different length scales and the associated physical phenomena couple to engender this “smart” adhesive is yet to be understood and mimicked. In this context, we have designed elastomeric films of poly(dimethylsiloxane) embedded with stacks of planar microchannels, curved and straight, and channels with microscopically patterned walls. We have altered also chemically the adhesive surface including that of the microchannel walls by creating dangling chains. During indentation experiments, deformation and self-adhesion of these structures enhance the effective area of adhesion with a consequent increase in adhesion hysteresis over orders of magnitude. In addition, suitable orientation of these buried channels allows the generation of load dependent hysteresis and its spatial modulation.

Introduction

While man-made adhesives are traditionally a uniformly thin layer of viscoelastic glue confined between two rigid or soft adherents, natural adhesives at the feet of most animals are hairy, complex, hierarchically patterned and in many situations consist of subsurface microstructures, for example, air pockets and liquid filled vessels. Microstructures on the adhesive allow these animals to create not only strong adhesion on rough and smooth surfaces alike but also easy release, and also allow these animals to use their adhesive pads repeatedly without contamination or consequent decrease in adhesion strength. These natural mechanisms have inspired the design of patterned adhesives of different kinds implementing a variety of novel methods of adhesion: crack arrest and initiation on a surface patterned adhesive, enhanced compliance of fibers or hairs on an adhesive, local modulation of compliance of a microfluidic adhesive, and alteration of surface chemistry along with subsurface microstructures, for example, air pockets.

Embedded multilayered adhesives with stacked rectangular microchannels embedded in uniformly thick blocks of poly(dimethylsiloxane) (PDMS). Displacement controlled loading and unloading of a rigid indenter on this adhesive show that, in addition to adhesion between these two adherents, thin walls of the channels too buckle and adhere to each other, effectively increasing the surface area of adhesion and the cumulative hysteresis at each of these internal surfaces. In order to enhance the effect of hysteresis even further, we have extracted the sol portion of these elastomers using a suitable solvent. This process does not alter the modulus of the elastomer but creates surface bound tethered chains. When two such surfaces are brought in contact, a variety of intermolecular interactions come into play which enhance the adhesion strength. This mechanism is called “self-adhesion hysteresis”, which happens multiple times in our experiment at the interface of each internal layer, thereby enhancing the hysteresis enormously. The hysteresis increases with the extent of extraction and also with the depth of indentation. Furthermore, the specific geometry of the microchannel and the simple fabrication procedure allows the hysteresis to be varied spatially, resulting in modulated, directional, and anisotropic adhesion, which can be useful for many technological applications.

Materials and Methods

The model elastic adhesives were prepared by using poly(dimethylsiloxane) for which commercially available Dow Corning product Sylgard 184 elastomer was used. Thin flexible plates, that is, transparency sheets, glass coverslips, and aluminum foil of varying thickness, transparency sheets, glass coverslips, and aluminum foil of varying thickness = 10–100 μm and width = 2–8 mm were used as templates for embedding microchannels in the adhesive. Solvents such as chloroform were used for swelling the cross-linked elastomer. Microscope glass slides coated with a monomolecular layer of octadecyltrichlorosilane molecules were used for forming the blocks of PDMS of uniform thickness = 2.5–3 mm.

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used for generating the microchannels within the adhesive block as spacing were controlled. The templates of these various forms were sheet. The depth of the scratches (5 µm) were coated with a self-assembled monolayer of octadecyltrichlorosilane (OTS) molecules; the stack of templates was kept within linked between the top and bottom microscope glass slides which were kept separated by a gap of 2–3 mm using suitable spacers. Thin rectangular strips of transparency sheet, as shown in Figure 1a, were used as templates which were supported at desired separation heights from the bottom and the top plates by using spacers of known thickness. Multiple layers of channels were prepared by stacking several such layers of template with spacers of suitable heights placed in between. The prepolymer liquid was then cross-linked between the top and bottom microscope glass slides which were coated with a self-assembled monolayer of octadecyltrichlorosilane (OTS) molecules; the stack of templates was kept within the pool of the prepolymer liquid. Silanization ensures easy removal of the PDMS block from the glass slides after cross-linking. The cured sample was swelled in a solvent such as chloroform, following which the templates were withdrawn out of the swollen block by applying a gentle pull. The swollen block was deswelled by drying in it normal atmospheric conditions. Once the samples were prepared, these were extracted in a Soxhlet chamber using chloroform as solvent for about 8 h. The extracted samples were dried completely in normal atmospheric conditions for a sufficiently long period of time, for example, 24–48 h, following which these were subjected to contact mechanics experiments.

Figure 1g depicts the schematic of a monolithic stack of rectangular channels embedded inside a block of PDMS elastomer of height $h = 3$ mm which was used as a model adhesive. The channel height $h_c = 100$ µm, interchannel spacing $h_s$, skin thickness $t$, $h_s = t = 600–1200$ µm, and width of the channels $w = 5–8$ mm were controlled. Figure 2 shows the schematic of the experimental setup in which a cylindrical indenter was pressed against this adhesive bonded to a rigid substrate and a typical optical micrograph of the area of contact of the indenter and the adhesive. The adhesive remained strongly bonded to a rigid substrate (Figure 2a), while a glass cylindrical indenter of length $L = 2.7$ mm and radius of curvature $r = 2.24$ mm was brought in contact with it at a constant rate of 1.28 µm/sec using a motorized nanopositioner (Newport, PC actuator, 1 µstep =10 nm). In order to minimize nonspecific interactions such as hydrogen bonding, the indenter was coated with octadecyltrichlorosilane monolayer. The contact load $P$ was measured by using a weighing balance interfaced with computer, while the contact area was visualized using a microscope fitted with a digital camera.

**Results and Discussion**

For a soft elastic adhesive without any embedded structure, the load versus displacement, that is, $P$ versus $\Delta$ data (curve 1 of Figure 3a) is rationalized by using the JKR23,24 (Johnson, Kendall, and Roberts) contact mechanics theory. The contact of a rigid hemicylinder of radius $R$ and length $L$ with a semi-infinite elastic layer is depicted by the following equation:

$$\frac{(3\pi/8)a^{3/2}}{R} = \left(\frac{P}{KL^{1/2}}\right) + (6\pi W/K)^{1/2}$$

in which $2a$ is the contact width, $K$ is the elastic constant of the layer, and $W$ is the adhesion strength of the interface. Equation 1 suggests that the slope of $a^{3/2}/R$ versus $P/L^{1/2}$ yields the modulus

of the elastic layer, while from the intercept we obtain the work of adhesion, $W$. Curve 1 of Figure 3a represents typical plots obtained during loading and unloading of the indenter on a control layer (CL) without any internal structure, and curve 3b shows those for layers embedded with multilayer of channels (ML).

For curve 1 of Figure 3a, the data from experiments on CL fall on straight lines during both loading and unloading, yielding the shear modulus of the layer as $\mu = 1$ MPa. The work of adhesion is calculated from the intercepts as $W_{\text{loading}} = 45 \pm 4$ mJ/m$^2$ and $W_{\text{unloading}} = 75 \pm 3$ mJ/m$^2$ which results in adhesion hysteresis $\Delta W = 30$ mJ/m$^2$. It is appropriate to mention at this point that the rms roughness of the transparency sheets was found to be $\sim 5$ nm, which was about 1 order of magnitude higher than that of the microscope glass slides. In order to investigate the effect of this roughness on the adhesion hysteresis, elastic layers of thickness $2-3$ mm were prepared between two substrates, the bottom one of which was a microscope glass slide as before, while the top one was the strip of a transparency sheet backed by another glass plate. This top substrate was removed after curing the PDMS prepolymeric liquid. JKR contact mechanics experiments on these samples yielded a shear modulus of $1.0$ MPa and hysteresis of $\sim 30$ mJ/m$^2$ similar to that obtained on the controlled layers, thus signifying that the roughness did not have any significant effect on the hysteresis. Adhesion hysteresis, however, increases significantly (curve 2 of Figure 3a) when a soft, solvent extracted PDMS hemispherical lens used as an...
indenter is brought in contact with the adhesive block. This phenomenon, known as “self-adhesion hysteresis”, is attributed to intermolecular interactions of several types at the interface, for example, hydrogen bonding and entanglement and interdigitation of surface bound tethered chains. In essence, alteration of the chemical character of the surface without change in the bulk rheology. Following the procedure in ref 19, the adhesion hysteresis is found out to be quite significant, \( \Delta W = 180 \text{ mJ/m}^2 \). In fact, this value can be further increased by increasing the duration of extraction of the adherent in the solvent,25 although it remains significantly smaller than that in curve 1, because in this regime the indenter drives against the thin film, christened as 1–2, which remains supported only at the two sides of the microchannel. Thus, the internal microstructure of the adhesive leads to a loading condition in which the channel wall 1–2 stretches and bends like a thin membrane, but the surface 2 remains unloaded. The width of the contact between the interface of the indenter and the adhesive (optical micrograph 4A) increases with increasing indentation. Eventually, the vertical displacement of the film 1–2 gets close enough to surface 2 so that interference fringes begin to appear as in micrograph 4A. The contact between these two surfaces occurs at B, resulting in interface 2–2’ with the appearance of a second contact area (micrograph 4B) which remains somewhat elliptic. This particular shape of the contact area has to do with the local curvature and compliance of the adhering elastic surfaces. In fact, the effective radius of curvature of the indenter that contacts surface 2’ exceeds that for surface 1, which naturally results in a significantly larger contact area for interface 2–2’. After the contact of these two surfaces, the adhesive behaves like a single block with an increase in effective shear modulus; therefore, in range B → C, the rate of increase in load with displacement approaches what is observed for curve 1. In the reverse cycle, the unloading data follow closely the loading data within displacement range C → D, that is, until the adhesive continues to behave like a single continuous block (micrographs 4C and 4D). Beyond point D, the indenter separates from the adhesive, but due to hysteresis the walls of the microchannel do not peel off. In range D → E (micrographs 4D and 4E), the contact area of interface 1 decreases, although that of 2–2’ remains nearly unaltered which results in an increase in bending of the film 1–2. It continues to be so until the elastic energy penalty of the channel wall 1–2 exceeds the adhesion hysteresis. Beyond point E, the interface 2–2’ progressively separates with continuous unloading which results in a decrease in the contact area (micrograph 4F) of 2–2’ but an increase in area of interface 1. This later effect occurs because of the increase in compliance of the wall 1–2 following its separation from the surface 2’. Correspondingly, the contact load \( P \) increases slightly until the interface 2–2’ separates completely at F. Finally, in F → G, the unloading data superimpose the loading data, because of debonding of interface 1. The net hysteresis of the adhesive is estimated by computing the shaded area and dividing it by the maximum contact area of interface 1 at C, as 277 \( \pm 48 \text{ mJ/m}^2 \), which is a significant enhancement over that of the control adhesive. Importantly, insignificant hysteresis is observed in the BCD and AAFG portion of the load–displacement curve, whereas maximum hysteresis occurs within A’BDEF. Effectively, the hysteresis corresponds to very small contact load on the surface 2’, signifying surface rather than bulk dissipation in the adhesive. Naturally, the surface hysteresis does not come into effect if the interface 2–2’ does not separate during unloading. For example, a decrease in the skin thickness of the above microchannel system to \( t = 440 \mu m \) results in a load–displacement

Figure 5. Adhesion hysteresis as a function of depth of indentation and number of layers. (a) Graph showing the hysteresis for the channel height \( h_c = 100 \mu m \), with skin thickness \( t = 1000 \mu m \) and 1000 \( \mu m \) interchannel spacing. The symbols \( \bigcirc, \Delta, \), and \( \bigotimes \) represent the data for one, three, and five layers of channels, respectively. The solid lines are a guide to the eye. (b) Maximum value of hysteresis for different thicknesses of the adhesive layer and different numbers of channels plotted against \( \xi = (1/n) \sum 800 (1 + id/f)^{0.01} \) to obtain a straight line with the result: \( \Delta E_{\max} = 8746 \xi - 10866 \).

Figure 6. Indentation experiment as in Figure 4c on an adhesive block embedded with a stack of \( n \) number of layers of microchannels. The graph shows that the data of contact width corresponding to zero load on each internal layer increase linearly with the number of embedded layers of microchannels.

Adhesive Embedded with a Single Layer of Microchannels. Curve 1 of Figure 4a shows that the \( P \) versus \( \Delta \) data for the control layer superimpose, yielding negligible hysteresis, \( \Delta E = 40 \text{ mJ/m}^2 \), very similar to that obtained by the JKR theory. However, these curves change for an adhesive with internal structures. Curve 2 of Figure 4a of skin thickness \( t = 800 \mu m \), channel height \( h_c = 100 \mu m \), channel width \( w = 8 \mu m \), and interchannel spacing \( h_b = 800 \mu m \) shows that the load \( P \) neither increases continuously with indentation depth \( \Delta \) during loading nor decreases monotonically during unloading. Initially, in displacement range \( A \rightarrow B \), \( P \) increases with \( \Delta \) but remains significantly smaller than that in curve 1, because in this regime the indenter drives against the thin film, christened as 1–2, which remains supported only at the two sides of the microchannel. Thus, the internal microstructure of the adhesive leads to a loading condition in which the channel wall 1–2 stretches and bends like a thin membrane, but the surface 2 remains unloaded. The width of the contact between the interface of the indenter and the adhesive (optical micrograph 4A) increases with increasing indentation. Eventually, the vertical displacement of the film 1–2 gets close enough to surface 2 so that interference fringes begin to appear as in micrograph 4A. The contact between these two surfaces occurs at B, resulting in interface 2–2’ with the appearance of a second contact area (micrograph 4B) which remains somewhat elliptic. This particular shape of the contact area has to do with the local curvature and compliance of the adhering elastic surfaces. In fact, the effective radius of curvature of the indenter that contacts surface 2’ exceeds that for surface 1, which naturally results in a significantly larger contact area for interface 2–2’. After the contact of these two surfaces, the adhesive behaves like a single block with an increase in effective shear modulus; therefore, in range B → C, the rate of increase in load with displacement approaches what is observed for curve 1. In the reverse cycle, the unloading data follow closely the loading data within displacement range C → D, that is, until the adhesive continues to behave like a single continuous block (micrographs 4C and 4D). Beyond point D, the indenter separates from the adhesive, but due to hysteresis the walls of the microchannel do not peel off. In range D → E (micrographs 4D and 4E), the contact area of interface 1 decreases, although that of 2–2’ remains nearly unaltered which results in an increase in bending of the film 1–2. It continues to be so until the elastic energy penalty of the channel wall 1–2 exceeds the adhesion hysteresis. Beyond point E, the interface 2–2’ progressively separates with continuous unloading which results in a decrease in the contact area (micrograph 4F) of 2–2’ but an increase in area of interface 1. This later effect occurs because of the increase in compliance of the wall 1–2 following its separation from the surface 2’. Correspondingly, the contact load \( P \) increases slightly until the interface 2–2’ separates completely at F. Finally, in F → G, the unloading data superimpose the loading data, because of debonding of interface 1. The net hysteresis of the adhesive is estimated by computing the shaded area and dividing it by the maximum contact area of interface 1 at C, as 277 \( \pm 48 \text{ mJ/m}^2 \), which is a significant enhancement over that of the control adhesive. Importantly, insignificant hysteresis is observed in the BCD and AAFG portion of the load–displacement curve, whereas maximum hysteresis occurs within A’BDEF. Effectively, the hysteresis corresponds to very small contact load on the surface 2’, signifying surface rather than bulk dissipation in the adhesive. Naturally, the surface hysteresis does not come into effect if the interface 2–2’ does not separate during unloading. For example, a decrease in the skin thickness of the above microchannel system to \( t = 440 \mu m \) results in a load–displacement


channels in a stack including the one buried deepest within the adhesive come in contact and adhere. Thus, hysteretic effects of all these channels are accumulated, resulting in maximum enhancement of adhesion.

The dependence of hysteresis on the indentation depth is systematically studied in Figure 5a in which adhesives embedded with up to $n = 5$ layers of microchannels with $h_i = 100 \, \mu m$ and $r = h_i = 1000 \, \mu m$ are used. Here, $\Delta E$ increases with both $n$ and $\Delta$ until a plateau value $\Delta E_{max}$ is reached at $\Delta_{max}$ (as shown by the dotted lines); $\Delta E_{max}$ increases with both $n$ and $h_i$, reaching a value of $\Delta E_{max} = 4412 \pm 90 \, mJ/m^2$ for five embedded channels within the adhesive. We can rationalize this huge enhancement in adhesion hysteresis mediated by subsurface structures by adopting results from the JKR analysis, since in our experiments the contact width $a$ is at least 1 order of magnitude smaller than the radius of curvature $r$ of the indenter. For example, for micrographs A–G in Figure 4, the maximum contact width of interface 1, $a = 200 \, \mu m$, is significantly smaller than the radius of the indenter, $r = 2.24 \, mm$. Therefore, for a small load, the JKR analysis of the $P$ versus $\Delta$ data can yield the work of adhesion and an effective modulus which will however be significantly underestimated. Nevertheless, as a first approximation, we will assume that, for each internal layer, the channel wall is contacted by a cylindrical indenter of appropriate radius. For example, the surface $2'$ of layer 1 channel is contacted by an indenter of radius $r + h_i$, so that the contact width $a_{2'-i}$ ($a_{2'-i}$: the superscript $i$ represents $i$th layer and subscript $2'$ represents the interface of contact between surfaces 2 and 2') at zero load is expressed
from JKR theory:$^{19} \quad d_{2} \sim (256W_{2}-\pi E_{c})^{1/3}(r + h_{c})^{2/3}$. Here, $W_{2}$ is the adhesion strength during unloading of the interface 2–2’ which is intrinsic to all interfaces within the adhesive; the effective Young’s modulus $E_{c}$ too is assumed to be the same for different layers, and for $h_{c} \approx 100 \mu m$, $h_{c} \approx 1000 \mu m$, and $h_{c} \ll h_{i}$ the wall thickness is assumed to remain unaltered because of bending. Therefore, at the interface of the channel in the $i$th layer, the contact width can be approximated as $d_{2} \sim 256W_{2}/\pi E_{c}(r + h_{c})^{2/3}$. Now, hysteresis is essentially contributed by the self-adhesion of the internal walls of the channels; furthermore, for a sufficiently large debonding strength, $W_{2}$ is nearly equal to the hysteresis of the interface. Therefore, the total hysteresis for an $n$-layered system is obtained by summing up the contact areas of all the interfaces and multiplying it with $W_{2}$:

$$\Delta E_{\text{max}} \sim (256W_{2}/\pi E_{c})^{1/3} \sum_{i=1}^{n} (1 + ih_{i}/r)^{2/3}.$$

Substitution of this expression yields

$$\Delta E_{\text{max}} \sim (W_{2}/W_{1})^{1/3} \sum_{i=1}^{n} (1 + ih_{c}/r)^{2/3}$$

which shows that $\Delta E_{\text{max}}$ has a non linear dependence on the number of layers of channels in the adhesive and other geometric parameters, for example, the skin thickness or interchannel spacing.

Experiments using adhesives with a different number of embedded layers of channels and skin thickness $t = h_{i} = 800 - 1200 \mu m$ show that the maximum hysteresis indeed scales linearly with the quantity $\xi = (1/n)\sum_{i=1}^{n} (1 + ih_{c}/r)^{2/3}$ as depicted in Figure 5b. The coefficient of this fit is however significantly larger than that predicted from eq 2, for example, for $W_{2} - W_{1} = 180 \text{ mJ/m}^{2}$ and $W_{1} = 30 \text{ mJ/m}^{2}$, and the coefficient $(W_{2}^{2} - W_{1})^{1/3}$ is calculated to be $327 \text{ mJ/m}^{2}$. This discrepancy could be due to interfacial shear at the 2–2’ interfaces, which was not accounted for in calculation.

Adhesive Embedded with Spatially Varying Skin Thickness of the Microchannels. The effect of skin thickness of the microchannel was further studied using adhesives in which $t(\Delta x)$ was varied spatially by embedding channels which remain differently oriented within the adhesive. For example, in Figure 7a, we show a rectangular channel formed in the shape of an inverted “V” embedded inside an adhesive with various degrees of inclination $\theta$. The adhesive was then indented at different spatial locations $\Delta x$ from the point of minimum skin thickness, $O$. Interestingly, the secondary contact at interface 2–2’ occurs not vertically beneath the primary contact at interface 1, but unlike channels oriented parallel to the surface at a lateral distance $\Delta a$ toward $O$. As a result, from the top view, we observe two different contact areas superimposing partially as in Figure 7b or appearing distinctly side by side as in Figure 7c. In some experiments in which $\Delta x$ is close to the middle of the two vertices, the 2–2′ interface appears at two neighboring sides of the inverted “V”. Here, too, the contact area of the 2–2’ interface appears significantly larger than that of 1 as observed earlier. In fact, the area $A_{2} - 2$ of the 2–2’ interface increases as the indentation is done further away from $O$, that is, with an increase in local skin thickness $t(\Delta x) \sim h(\Delta x)$; this effect gets increasingly pronounced with the angle of inclination $\theta$. Experiments with $\theta \sim 6 - 60^\circ$ and spatial location $\Delta x \sim 0.5 - 3.5 \text{ mm}$ show that hysteresis depends only on $t(\Delta x)$. In fact, by putting $n = 1$ and $h_{i} = t(\Delta x)$ in eq 2, $\Delta E_{\text{max}}$ is observed to vary as $\Delta E_{\text{max}} \sim (1 + (c_{1} + c_{2}\Delta x))/r^{2/3}$ remaining independent of $\theta$ as shown in Figure 7d. The error bar represents the standard deviation $\Delta E_{\text{max}}$ obtained from similar $t(\Delta x)$. In essence, linear variation in skin thickness, $t(\Delta x) = c_{1} + c_{2}\Delta x$, yields a power-law type of spatial variation in hysteresis: $\Delta E_{\text{max}} \sim (1 + (c_{1} + c_{2}\Delta x))/r^{2/3}$. This result signifies that the hysteresis can be modulated to different other functional forms by suitably varying the skin thickness $t(\Delta x)$. For example, Figure 7e depicts the schematic of the side view of an adhesive in which a curved channel is embedded such that the skin thickness assumes the general form $t(\Delta x) = c_{1} \exp(c_{2}\Delta x)$; the channel height is kept constant at $h_{i} = 100 \mu m$. Indentation as in Figure 1b is carried out on these adhesives at different spatial locations and to the maximum indentation depth of $\Delta = 200 \mu m$. The resultant hysteresis on the adhesive with a different combination of channel parameters $c_{1}$ and $c_{2}$ increases linearly with the dimensionless quantity $\xi = (1 + c_{1} \exp(c_{2}\Delta x))/r^{2/3}$ as shown in Figure 7f. Furthermore, the intercepts in these figures can be understood by considering the limiting cases for these experiments. For example, in the context of Figure 5b, one limiting situation occurs if we substitute $n = 1$ and $h_{i} = 0$, that is, when $\xi = 1$. Physically, this situation corresponds to “self-adhesion” between an indenter and the elastomeric layer, for example, a rigid indenter coated with an elastomeric film used against the adhesive layer without any embedded structure, both extracted in a Soxhlet chamber using solvent. In the context of Figure 7d and f, this situation occurs when skin thickness $t(\Delta x)$ asymptotically goes to zero, $t(\Delta x) \sim 0$. The hysteresis of these interfaces are then estimated from the experimental data by substitution of $\xi = 1$. For example, in Figure 7d and f, $\xi = 1$ yields $\Delta E_{\text{max}} \sim 140 \text{ mJ/m}^{2}$ which is very similar to $W_{2} - W_{1} = 180 \text{ mJ/m}^{2}$ observed for self-adhesion of interfaces 2 and 2’. However, in Figure 5b, the intercept yields an unphysical negative value of the hysteresis, signifying that.
for multilayer channels, the error in calculation increases and can only be resolved by a more rigorous analysis.

Adhesive Embedded with Microchannels Having a Patterned Wall. Thus, the effect of “self-adhesion hysteresis”, a macroscopic manifestation of interactions involving surface bound molecules, gets coupled with a macroscopic geometric feature of the adhesive, for example, the stacked microchannels, not only to enhance the adhesion hysteresis but also to vary it spatially as desired. We now show that we can incorporate microscopic features of intermediate length scale into this system. Figure 8 shows a typical example in which one surface of a planar channel remains smooth while the other is patterned by incorporating uniformly spaced hairlike protrusions spanning the width of the sample. Such features not only increase the effective area of adhesion but also result in “arrest and initiation” of surface cracks as observed with incision patterned and fibrillar adhesive surfaces. Furthermore, these effects are amplified by stacking several such surface patterned microchannels within the adhesive block.

In Figure 8b, we summarize the results from indentation experiments and compare them with the corresponding data from an adhesive with smooth, featureless channels. The bar chart shows that the hysteresis increases for the surface patterned channels. Patterns of different other dimensions and spacing can be incorporated on the walls of the channel to achieve better manipulation of the adhesion hysteresis.

Summary

We have presented here the design of a novel hierarchically structured adhesive in which we have introduced features of three different length scales: stacked channels of dimension 400–1500 µm, microscopic patterns of 15–50 µm at the internal walls of the channels, and finally the molecular length scale associated with the chemical alteration of surfaces following solvent extraction of the adhesive. In each of these dimensional regimes, adhesion hysteresis increases because of a distinct physical phenomenon which can be modulated by altering the dimensions and spatial density of the features. Importantly, these length scales remain orders of magnitude apart, which ensure that the dominant physical mechanisms become effective independently without influencing the others. For example, the hierarchical structures of this type are not plagued by “self-adhesion”, agglomeration, and consequent loss of adhesion, unlike many others, for example, fibrillar adhesives. In contrast, we take precisely the advantage of self-adhesion of the walls of microchannels to enhance the adhesion. Furthermore, the hierarchical structure allows the hysteresis of the same adhesive to be controlled by varying the contact load. We have introduced also methods to achieve spatially varying hysteresis by suitably orienting the channels and by varying the number of layers in a stack. The simple methods adopted here to incorporate structures of different length scales within the adhesive should help incorporate many other additional characteristics and manufacture designer adhesives useful for many different applications.

Acknowledgment. A.G. acknowledges the research grants of the Department of Science and Technology, India and Council of Scientific and Industrial Research, India for this work. A.G. and E.P.A. thank the anonymous referee for useful suggestions.