Microfabrication of Bio-Inspired Adhesive Systems

A dissertation submitted in partial satisfaction of the requirements for the degree Doctor of Philosophy in Mechanical Engineering

by

Sathya Shrinivas Chary

Committee in charge:
Professor Kimberly L. Turner, Chair
Professor Jacob N. Israelachvili
Professor Matthew R. Begley
Professor Carl D. Meinhart

June 2013
The dissertation of Sathya Shrinivas Chary is approved.

____________________________________________
Jacob N. Israelachvili

____________________________________________
Matthew R. Begley

____________________________________________
Carl D. Meinhart

____________________________________________
Kimberly L. Turner, Committee Chair

April 2013
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Figure I – 1 in Chapter I is reprinted with permission from Y. Tian, et al., "Adhesion and friction in gecko toe attachment and detachment," *Proceedings of the National Academy of Sciences of the USA*, vol. 103, no. 51, p. 19320–19325, 2006. (© 2006 National Academy of Sciences, USA)


VITA OF SATHYA SHRINIVAS CHARY
April 2013

EDUCATION

Bachelor of Engineering in Mechanical Engineering, Birla Institute of Technology and Science, Pilani (India), August 2006

Bachelor of Engineering in Electrical and Electronics Engineering, Birla Institute of Technology and Science, Pilani (India), August 2006

Master of Science in Mechanical Engineering, University of California, Santa Barbara, March 2009

Doctor of Philosophy in Mechanical Engineering, University of California, Santa Barbara, June 2013 (expected)

PROFESSIONAL EMPLOYMENT

2006-07: Teaching Assistant, Department of Mechanical Engineering, University of California, Santa Barbara

2011: Teaching Assistant, Department of Mechanical Engineering, University of California, Santa Barbara

2007-2013: Graduate Student Researcher, Department of Mechanical Engineering, University of California, Santa Barbara

PUBLICATIONS


**AWARDS**

Graduate Student Excellence Fellowship, Department of Mechanical Engineering, University of California, Santa Barbara, Summer 2012

French Lower-Division Courses Outstanding Student, Department of French and Italian, University of California, Santa Barbara, Spring 2009

**FIELDS OF STUDY**

Major Field: Micro/Nanoscale Systems

ABSTRACT

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Sathya Shrinivas Chary

Most geckos can rapidly attach and detach from most surfaces using the weak but universal van der Waals forces. This ability is attributed to the hierarchical structure of their feet (involving macroscale toe pads, arrays of ‘seta’ microfibers, and nanoscale spatula tips), and how these anisotropic structures are moved (articulated) to generate strong adhesion and friction forces on gripping that rapidly relax on releasing. Inspired by the gecko adhesive system, various structured surfaces have been fabricated suitable for robotic applications.

First, vast arrays of both vertical and tilted rectangular PDMS micro-flaps were fabricated using micro-electromechanical systems (MEMS) fabrication techniques. Friction and adhesion force properties were investigated using both a Surface Forces Apparatus (SFA) and a custom-built tester with larger contact area. Demonstrating the importance of both asymmetric tilted structures (such as the fibers in geckos) and an optimal articulation mechanism, it was found that the anisotropic structure of the tilted micro-flaps resulted in highly anisotropic adhesion and friction forces when
articulated along different directions: high friction and adhesion when sheared along the tilt direction, and low friction and adhesion when sheared against tilt.

Then, the friction and adhesion properties of the tilted micro-flaps were characterized against five different rough glass substrates with non-uniformly spaced asperities. It was found that while adhesion and friction anisotropy was maintained on all substrates, the magnitudes showed an unexpected increase at intermediate height RMS roughness due to a geometric match between the dimensions of the flaps and the average spacing between asperities, leading to interlocking. This both demonstrates the importance of substrate surface structure as well as indicates how fibrillar adhesives could be tailored to achieve higher forces on specific surfaces.

Finally, the importance of both tilt angle and fiber shape was demonstrated with tilted half-cylinder PDMS microfiber arrays. With suitable articulation, the adhesion force was switched from a maximum of 9.4 kPa for strong attachment to a minimum of zero for easy detachment. Practical applications require adhesives to be highly durable in addition to being anisotropic – this material retained up to 77% of the initial adhesion after 10,000 test cycles.
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I. Introduction

A. The Gecko Adhesive System

Geckos – and several species of insect – rely on a fibrillar adhesive system to effectively traverse a wide variety of surfaces encountered in nature – rough, smooth, horizontal and vertical. Most of the approximately 850 species of gecko possess fibers on the ventral side of their toes, although tokay geckos (*Gekko gecko*) have the best studied system with a multi-scale hierarchical structure, as shown in Figure I – 1. Each tokay gecko foot has 5 toes; each toe bears an expanded digital pad carrying about 20 flaps (called scanners or lamellae), each lamella bearing an array of micron-scale fibers (setae), with each seta fiber being ~4.7 µm in diameter and 30-130 µm in length. Each seta in turn branches out several times before splitting into a total of 100-1000 roughly triangular nano-scale spatulae of ~200 nm length and width at the tip [1]. The plate-like terminal spatulae are just 5-10 nm in thickness [2]. Each lamella is only attached to the toe at the lamella’s proximal end, allowing rotation about the base [3]. This makes it compliant in the direction normal to the surface, useful for conforming to a substrate surface and ensuring that all the terminal features make contact. The action of fluid-filled sacs underlying the lamellae and tendons in the gecko toe in promoting good contact and an equal load distribution have also been suggested [4].
Several examples of non-adhesive keratinous fibers exist in nature – for example, human hair made primarily of α-keratin, and hair on the bodies and in the feathers of birds that are made primarily of β-keratin. Even gecko feet have a default non-sticky state, and do not generate strong adhesion unless suitably articulated to engage the fibers. When articulated with a lateral shearing motion, the hierarchical structure enables the fibers made of stiff and durable β-keratin to conform to the micro and nano scale asperities of rough surfaces – resulting in intimate contact between the
surfaces – and thus to utilize the weak and short ranged van der Waals forces for adhesion along with other types of non-covalent forces such as capillary forces [6, 7]. Although the van der Waals force is weak for a single small contact such as a spatula, this force can be substantial when summed over the large contact area created by all the available spatulæ, especially since setae are packed at an areal density of \( \sim 14,400 \text{ mm}^{-2} \) [8, 9]. Measurements on the gecko have reported that the surface area of the two front feet is able to generate a total friction force of \( \sim 20.1 \text{ N} \), an average shear force per seta of \( \sim 6.2 \mu \text{N} \) [10]. Tests on single setae showed that the shear force could reach as high as 194 \( \mu \text{N} \) [11]. Since van der Waals forces are universal, gecko setae thus also have the property of material independence – they can adhere strongly to a wide range of materials, largely independently of surface chemistry. Recent experiments demonstrate that under certain environmental conditions the gecko adhesive pads, by changing their hydrophobicity under humid conditions, also have the ability to exploit environmental conditions to maximize their adhesion and stabilize their friction forces [7, 12]. Other studies indicate that the influence of humidity on gecko adhesion may be indirect – the mechanical properties of setae could be influenced by an increase in humidity, reducing elastic modulus while increasing the loss tangent, and thereby resulting in more energy dissipation and higher adhesion [13, 14].

Key characteristics of the natural system [9] include high and anisotropic normal adhesion and lateral shear forces during attachment, low detachment force, a high adhesion to initial preload force ratio (\( \mu' \)) of 8-16 [6, 11], lack of inter-fiber self-
adhesion, and operation over more than 30,000 cycles without significant loss of adhesion performance [15]. The highly reversible and controllable adhesion and friction in the gecko adhesive system enables both sticking and easy, rapid peeling, with foot detachment possible in just ~15-20 ms with no measurable detachment forces [9], allowing geckos to run rapidly on walls and ceilings, at speeds even greater than 1 m/s [16]. Recent experimental and theoretical findings have demonstrated that the anisotropic structure of gecko setae results in anisotropy in their adhesion and friction forces when engaged and displaced along opposing directions [17, 18]. The setae start at an angle of ~45° with respect to the skin of the underlying toe pad [19], and are curved at their distal spatula-bearing ends. High net friction and adhesion forces on the whole gecko are obtained by gripping inward, shearing the setae proximally, bringing large numbers of spatulae in intimate contact with the substrate with small pulling angles $\theta$ between them. Tests on both the whole animal and single setae have indicated that for $\theta > 30^\circ$, spontaneous detachment occurs [11, 20]. Therefore, to detach, the high adhesion/friction is rapidly reduced by over three orders of magnitude to a very low value by rolling the toes upward and backward (digital hyperextension), which, mediated by the lever function of the setal shaft, peels the spatulae off perpendicularly from the substrate [5]. Thus, gecko setal arrays already exhibit desirable directional behavior of strong adhesion and friction in the gripping direction, and almost zero adhesion and low friction in the releasing direction [17].
An important point to note about the gecko adhesive system is the interplay between, and simultaneous action of, friction and adhesion forces [20]. In a manner analogous to the action of the fingers in picking up a beach ball by squeezing it, the toes and feet of the gecko act together in a Y-configuration so that a resolved component of the friction force also contributes to adhesion. Even at the level of the nanoscale spatulae, the pulling force of a spatula along its shaft with an angle $\theta$ between 0 and 90° to the substrate has a normal adhesion force contribution produced at the spatula-substrate bifurcation zone, and a lateral friction force contribution from the part of spatula still in contact with the substrate. Yu Tian et al. have theoretically shown that the friction component contribution to the adhesion is greater than the direct normal adhesion force contribution at the angles typically subtended by the spatulae and setae during gecko locomotion [5].

Because they are soft and sticky, conventional pressure-sensitive adhesives also tend to degrade, self-adhere, attach accidentally to unintended surfaces, and get fouled after just a few uses. In contrast, the natural gecko adhesive survives tens of thousands of cycles between successive skin molts that may be several months apart – this without individual fiber wear, contamination from dust or other contaminant particles, or inter-fiber self-adhesion and matting. In durability tests on gecko setal arrays isolated from the animal, up to 95% of the initial shear force was retained after 30,000 continuous cycles, and a 25% increase in adhesion force generated was reported [15]. Since gecko setae are made of stiff $\beta$-keratin fibers, with a reported Young’s modulus of ~1.6 GPa [21], their wear resistance is enhanced, while their
slender structure and tilt angle result in a low effective Young’s modulus for setal arrays, of the order of ~100 kPa [22], enabling them to still conform to asperities over a wide range of roughness. Unlike several species of adhesive foot pad-bearing insects, geckos have not been observed to actively groom and clean their feet – yet, their adhesive system does not get fouled by dust or other particles from the natural environment. The self-cleaning properties of other surfaces in nature, for example the lotus leaf, have been explained based on the micro-rough structure of an exposed wax layer that both prevents intimate contact with and adhesion to contaminant particles, and also enhances surface hydrophobicity, thus allowing external water droplets to function as a cleaning agent when they easily roll over the hydrophobic surface and collect the weakly adhering particles [23]. While gecko setal arrays have also been shown to be highly hydrophobic with a water contact angle of ~161° [24], it is the fibrillar structure of the gecko adhesive system that has been used to explain their non-fouling property. When contaminant particles (ceramic microspheres of radius = 2.5 μm) were intentionally introduced in tests on both single gecko toes and isolated setal arrays against a glass substrate, the adhesive system was found to self-clean, with particles left behind on the substrate surface, resulting in a quick recovery of measured adhesion to initial levels after just a few simulated steps [25]. The self-cleaning seen with setal arrays isolated from the animal indicates that this property is one that results from the fiber array structure itself, and does not require any specific foot articulation or toe peeling to function. A simple model was also developed to explain how it might be more energetically favorable for dust particles
to stick to the substrate surface rather than to gecko spatulae. Further, matting of adjacent setal stalks and adhesion of facing arrays have not been observed even with setal arrays used for repeated testing. This anti-self-adhesion property may result from a principle similar to that of self-cleaning: it is unlikely that the number of spatula–spatula bonds will yield forces exceeding the forces acting to pull two facing arrays of setae apart. Experimental observations indicate stronger adhesion and friction forces between a setal array and a glass substrate than between two setal arrays [19]. An interesting corollary of the anti-self-adhesion property is that maximum attainable setal areal density is limited not by clumping between adjacent seta, but by the finite seta diameter resulting in ‘crowding’ with adjacent setae at low seta-substrate angles [19].

Several potential applications for reusable dry adhesives exist, including climbing robotic systems [26, 27], mobile sensor platforms, reversible bandages and other medical skin patches [28], non-fouling pick-and-place handling systems for wafers in nanofabrication facilities, and in liquid crystal display fabrication facilities [29]. The material independence of van der Waals forces means that gecko-inspired synthetic adhesives can be made from a wide range of materials which could in the future be designed to resist extreme environments such as high temperatures. A wider choice of materials also means that they can be made non-toxic, biocompatible, or biodegradable, depending on the application. Theoretical approaches have also shown that both the van der Waals adhesion and the friction forces of flexible, tilted, and optimally spaced setal stalks or synthetic pillars are
high enough to support a human being on rough surfaces such as ceilings (adhesion) and walls (friction) with an estimated effective area of 230 cm², close to the total area of a human being’s hands and feet [30].

In order to better understand the underlying mechanisms responsible for the unique adhesion and friction properties of the gecko fibrillar adhesive system, a review of some important concepts relating to adhesive contact between elastic bodies, the origin of van der Waals forces and friction fundamentals is presented in the next three sections.

**B. Adhesion Mechanics**

1. Hertz, JKR and DMT Theories

The work of adhesion $W_{12}$ is the free energy change or reversible work done to separate unit surface areas of two bodies 1 and 2 from contact to infinity in vacuum. If the surface energy, or the energy required to create unit area of surface, of these two surfaces is given by $\gamma_1$ and $\gamma_2$, $W_{12}$ is given by the Dupré equation [31]:

$$W_{12} = \gamma_1 + \gamma_2 - \gamma_{12}$$

(1 - 1)

Where $\gamma_{12}$ is the energy per unit area of the interface between them. This analysis has not yet taken into account any elastic energy involved in the deformation of the bodies due to contact stresses arising from the adhesive forces and any externally applied loads.
For two deformable spheres in contact, the area in contact varies with the applied external load $P$. Looking first at the case of a non-adhesive contact, this problem of elastic contact between two spheres has a solution first established by Hertz [32]. With the assumptions that the contact radius is small compared to the sphere radii; that the contact is frictionless; that the surfaces are continuous, smooth and non-conforming; and that no tensile stress exists within the area of contact, Hertz derived an equation for the contact radius $a$ as:

$$a^3 = \frac{3PR^*}{4E^*}$$  \hspace{1cm} (1-2)

Where $P$ is the external load, and $E^*$ is the reduced Young’s modulus for the contact between spheres of two homogeneous and isotropic materials 1 and 2 with Young’s moduli $E_1$ and $E_2$ and Poisson’s ratios $\nu_1$ and $\nu_2$ respectively, and is given by:

$$E^* = \left(\frac{1 - \nu_1^2}{E_1} + \frac{1 - \nu_2^2}{E_2}\right)^{-1}$$  \hspace{1cm} (1-3)

And $R^*$ is the reduced or effective radius defined as:

$$\frac{1}{R^*} = \frac{1}{R_1} + \frac{1}{R_2}$$  \hspace{1cm} (1-4)

The penetration $\delta$ of the indentation is given by:

$$\delta = \frac{a^2}{R^*} = \left(\frac{9P^2}{16E^{*2}R^*}\right)^{1/3}$$  \hspace{1cm} (1-5)
The assumption that $a \ll R$ is satisfied in most practical situations since for larger indentations materials will often be no longer within their elastic limit and the Hertz theory does not apply. The indentation of an elastic half-space by a spherical indenter with radius $R$ has been studied by Sneddon [33] and Ting [34], who derived equations for the relationships between external load $P$, penetration depth $\delta$ and contact radius $a$ without the Hertzian approximation of $a \ll R$, and a comparison of their results with those of Hertz for values of $a/R$ of up to 0.4 show the Hertz theory to be a good approximation.

Though the Hertz theory allows the calculation of the contact shape and compressive stress distribution between externally loaded spheres, it does not include any surface forces and therefore does not include the possibility of adhesion between the two spheres with a non-zero contact area for zero external load. An extension of the Hertz theory taking into account adhesive interactions and their influence on the contact shape was introduced by Johnson, Kendall and Roberts [35], and has become known as the JKR theory. Taking into account the adhesive interactions only within the contact area, they predicted an increase in contact radius due to a reduction in interfacial energy, with the contact radius $a_{\text{JKR}}$ given by:

$$a_{\text{JKR}}^3 = \frac{R}{K} \left[ P + 3\pi R^*W_{12} + \sqrt{6\pi R^*W_{12}P + (3\pi R^*W_{12})^2} \right]$$ (1 - 6)

Where $W_{12}$ is the interfacial surface energy per unit area, and the effective stiffness, $K = 4E^*/3$. The first term in this equation is identical to the Hertzian
contact radius in Equation (1 – 2), while the second and third are due to the adhesive interaction and lead to an increased contact radius compared to the adhesionless case. Also, in a JKR-type contact, the inner circle of the contact area is under compression, while the outer annular area is under tension. Also, while the surfaces meet tangentially for a Hertzian contact, in the JKR case, the spheres deform to a right angle with a small neck at the edge of the contact zone.

Unlike the Hertz theory, where the contact area reduces to zero as the external load is reduced to zero and the spheres simply separate for negative external loads, the JKR theory predicts a finite contact radius $a_0$ even for zero external loads, as can be seen by substituting $P = 0$ in Equation (1 – 6). Further, it predicts a pull-off event at a negative external load, yielding an adhesion force $F_{JKR}$ given by:

$$F_{JKR} = \frac{3}{2} \pi W_{12} R^*$$

(1 – 7)

With the contact radius at pull-off $\approx 0.63a_0$. Note that the expression for $F_{JKR}$ is independent of the reduced Young’s modulus $E^*$. However, this is only true for the special case of contacting spheres – for other contact geometries such as sideward contact of a cylinder on a flat surface, the adhesion force could also depend on $E^*$ [36, 37].

The JKR theory has been used to model the adhesive interactions of both natural and synthetic fibrillar adhesives [17, 38, 39]. This theory also offers a simple explanation for the use of fibrillar structures in biological adhesive systems through
the phenomenon of ‘contact splitting’ [38, 40]. As seen in Equation (1 – 7), the adhesion force varies linearly with the fiber dimension. Hence, fibers of smaller radii will result in a linear decrease in adhesion per fiber. However, since the number of fibers possible per unit area varies inversely with the square of the radius, the overall adhesion force increases for small fibers. As explained in [40], the advantage is maintained even if all fibers do not pull off at once and a peeling action propagates through a forest of fibers. Also, on rough surfaces, hierarchical fibers like gecko setae and spatula can better conform to a range of substrate surface roughness. Further, an adhesive mechanism utilizing an array of fibers is more insensitive to flaws at individual contact points, as opposed to the catastrophic drops in adhesion that a single large contact would experience with such a flaw.

Another theory advanced by Derjaguin, Muller, and Toporov, since called the DMT theory, included the effect of attractive surface forces in an annular ‘cohesive zone’ region around the contact [41]. The additional load due to the surface forces is then simply added to the external load $P$ to obtain the correct contact radius from the Hertzian equations. Since it was assumed that the surface forces do not deform the surfaces outside the contact area, the surface profile thus remains the same as for the Hertzian contact case. The pull-off event in the DMT model occurs at zero contact radius, and the adhesion force $F_{DMT}$ was predicted to be given by:

$$F_{DMT} = 2\pi W_{12}R^*$$  \hspace{1cm} (1 – 8)
This value is in agreement with the value expected for perfectly rigid bodies in adhesive contact as derived by Bradley [42] and as expected from the Derjaguin approximation [31]. However, there is a discrepancy between the JKR and DMT theories, which caused some debate. Also, since the expression for \( F_{\text{JKR}} \) does not depend on \( E^* \), it should be valid even for rigid spheres, but instead, the DMT prediction is the one that agrees for this case. Both theories have limitations – the JKR theory does not account for surface forces outside the contact area but allows deviations from the Hertzian contact shape, while the DMT model takes into account the surface forces outside the contact area but not the deformations due to these forces. Therefore, it is expected that the JKR model is more appropriate for the case of large soft spheres with high surface energies, while the DMT model is more appropriate for the case of small rigid spheres and low surface energies, since deformations close to the contact area should remain small in the latter case. The apparent discrepancy was resolved by David Tabor [43], who recognized that a consequence of the neck formation in the JKR theory is that the gap width increases steeply outside the contact zone, rendering surface forces outside this zone ineffective. At the critical contact just before pull-off, the gap width \( \delta_c \) is given by:

\[
\delta_c = \left( \frac{9}{4} \right)^{2/3} \frac{(R^*)^{1/3}(W_{12})^{2/3}}{(E^*)^{2/3}}(1 - 9)
\]
Tabor concluded that the JKR theory should apply if the neck height is of the order or larger than the range of action of the surface forces (set equal to the interatomic separation $\sigma$), and introduced the following Tabor Coefficient $\mu_T$ [44]:

$$\mu_T = \frac{(R^*)^{1/3}(W_{12})^{2/3}}{\sigma(E^*)^{2/3}} \quad (I \text{ } - \text{ } 10)$$

For values of $\mu_T << 1$, the DMT theory should be valid and for $\mu_T >> 1$, the JKR theory should be applicable.

However, both theories assume perfectly smooth surfaces, and most particle surfaces are rough. Since asperities limit the average distance of closest approach between two bodies, and thus severely reduce short-range intermolecular interactions, asperities as small as 1 – 2 nm can significantly lower adhesion. However, no satisfactory theory exists for such real world situations as yet. In this research, the optically smooth spherical glass surfaces used in SFA testing of the synthetic adhesives were found by AFM analysis to have an RMS roughness value of $11 \pm 8$ nm. For larger scale testing with a flat-on-flat geometry in the Bio-F apparatus, the glass puck was found by optical profilometry to have an RMS roughness of $\sim 160$ nm.

2. Kendall Peel Model

An experimentally verified energy balance analysis by Kendall [45] showed that the energy required to peel an elastic film from a rigid substrate depends not only on
the adhesive energy $R$, but also on the elastic energy of deformation. Including the
potential energy term due to the externally applied peel force $F$, there are thus three
main contributors to the energy change during peeling. For a thin film of thickness $d$,
width $b$, and Young’s Modulus $E$ peeling from a substrate at a fixed angle $\theta$, a
simple energy balance may be rewritten as a quadratic equation in $F/b$ as follows:

$$\left(\frac{F}{b}\right)^2 \frac{1}{2dE} + \left(\frac{F}{b}\right) (1 - \cos \theta) - R = 0 \quad (I - 11)$$

This quadratic equation may be solved to obtain the peel force $F$ required:

$$F = bdE \left( \cos \theta - 1 + \sqrt{\cos^2 \theta - 2 \cos \theta + 1 + \frac{2R}{dE}} \right) \quad (I - 12)$$

Previous analyses by others have modeled the adhesion of both natural gecko
spatulae [25] and synthetic fibrillar arrays [29, 46] using this model for the peel
force. In this study, the adhesion behavior (for tests against a flat glass puck
substrate) of both vertical and angled PDMS micro-flaps, as well as those of angled
half-cylinder PDMS microfibers, was found to be well described by the Kendall Peel
Model. However, it is important to note here the main assumptions in the Kendall
Peel Model – a) the film is represented as a membrane with no bending stiffness; b)
the film in contact with the substrate is not allowed to slide during peeling; and c)
the elastic deformations of the film during peeling are small.
C. Origin of Adhesive Forces

Johannes Diderik van der Waals showed the need to take into account the finite volumes of gas molecules as well as the attractive interactions between them in his correction of the ideal gas equation. Even in the absence of permanent electric charges and covalent bonds, attractive forces exist between all molecules in vacuum, as evidenced by the fact that even non-polar molecules condense to liquids at sufficiently low temperatures. ‘Van der Waals forces’ is a term used to describe the effect of three separate intermolecular interactions: 1) the orientational or Keesom interaction between two dipoles free to rotate, that on average is attractive because the dipoles preferentially orient themselves with their opposite charges facing each other; 2) the induction or Debye interaction that arises due to the polarization of a molecule without a static dipole moment by another with a dipole moment; and 3) the dispersion or London interaction between two non-polar molecules [31]. The last of the three arises due to the instantaneous dipole created by a shifting electron distribution around the nucleus of an atom, and always exists between any two molecules. Dispersion forces generally exceed the dipole-dependent induction and orientation forces except for small highly polar molecules. All three components of the van der Waals force have an interaction energy that varies with the inverse sixth power of distance $D$ for intermolecular interactions. Thus, we can write:

$$w_{VDW}(D) = -C_{VDW}/D^6 = -\frac{[C_{ind} + C_{orient} + C_{disp}]}{D^6}$$  \hspace{1cm} (I - 13)
It must be noted that since dispersion forces are dependent on molecular polarizability, and all but spherically symmetric molecules are anisotropic, the dispersion force between molecules becomes dependent on their mutual orientation, which is more important in solids where their orientations are relatively fixed compared to liquids where this constantly changes due to Brownian motion. Also, van der Waals forces are not generally pairwise additive since the force between any two molecules is affected by the presence of other neighboring molecules. This means the calculation of the effective van der Waals force between larger particles and surfaces is a many-body problem. Another effect to consider is the retardation effect – the faster decay (to the inverse seventh power of distance) of dispersion forces at large interatomic separations, when the time taken for the electric field of the first atom’s instantaneous dipole to reach the second and return can become comparable to the period of the fluctuating dipole itself, resulting in a less favorable attractive interaction between them.

At very small interatomic separations, the electron clouds of atoms overlap, and a strong repulsive force arises. Unlike the attractive interactions discussed so far, no general equation exists that describes the distance dependence of these repulsive forces, and a number of empirical potential energy expressions have been used to describe them that all share the quality of having a steep rise in repulsion at small separations. The total interatomic pair potential is obtained by adding the attractive and repulsive potentials, the best known of which is arguably the Lennard-Jones potential given by the expression:
\[ w(D) = A/D^{12} - B/D^6 \] (1–14)

The inverse twelfth power term in this potential is chosen to model the steeply rising repulsion at low values of the separation \( D \).

Using the assumption of pairwise additivity of the attractive inverse sixth power interatomic interactions, the total van der Waals interactions between larger bodies such as spherical particles and flat surfaces may be calculated by integration [42, 47, 48, 49]. Neglecting any retardation effects for larger separations simplifies the analysis, and closed form expressions for the distance dependence of interactions between larger bodies may be obtained. A list for the interaction laws for some common geometries is given in [31]. An important point to note for these interactions is that the distance dependence for larger bodies falls much more slowly with separation \( D \) than for two atoms – for example, the van der Waals interaction energy between a sphere and a flat surface has an inverse first power dependence on \( D \), while the force per unit area between two flat surfaces falls with the inverse second power. Hence, the separation range over which these interactions are significant is greatly increased. This also means that the van der Waals forces between macroscopic objects can reach significant values if intimate contact is achieved, as best exemplified in nature by the gecko.

Although the power laws for distance dependence as previously derived remain valid, a more accurate method to derive the constants in the equations for interactions between larger bodies is possible with the Lifshitz theory. In this
approach, the problem of pairwise additivity is avoided since the atomic structure of the bodies is ignored with them being treated as continuous media, and the forces are derived in terms of easier to access bulk material properties such as dielectric constants and refractive indices. This theory also allows taking the influence of an intervening medium into account. Hence, based on the Lifshitz theory, it is even possible to accurately predict that the van der Waals interactions between two dissimilar media 1 and 2 in a third intervening medium are repulsive for the case when its dielectric properties are intermediate in value to those of media 1 and 2 [31].

In the presence of humidity, capillary forces could provide an additional attractive force mechanism. The Laplace equation relates the pressure difference between media on two sides of a curved fluid interface to its curvature. Neglecting gravitational effects for small objects, the Laplace equation states that this ‘Laplace pressure’ difference \( \Delta P \) is given by [44]:

\[
\Delta P = \gamma_L \left( \frac{1}{r_1} + \frac{1}{r_2} \right)
\]

Where \( \gamma_L \) is the surface energy of the liquid, and \( r_1 \) and \( r_2 \) are the two principal radii of curvature. The pressure inside both a drop and a bubble is higher than that outside. This then results in a change in the vapor pressure of a curved liquid surface from a flat surface. The raised Laplace pressure inside a drop causes the molecules to evaporate more easily, raising the vapor pressure from the flat surface case. The
converse if true for a liquid surrounding a bubble – since the Laplace pressure in the bubble is higher, this makes it more difficult for molecules from the surrounding liquid to evaporate into it. A quantitative expression for the change of vapor pressure for curved liquid surfaces is described by the Kelvin equation as follows [44]:

$$RT \cdot \ln \frac{P}{P_0} = \gamma_L V_m \cdot \left( \frac{1}{r_1} + \frac{1}{r_2} \right)$$  \hspace{1cm} (I - 16)$$

Where $P$ is the equilibrium vapor pressure of a liquid with surface curvature $1/r_1 + 1/r_2$, $P_0$ is the equilibrium vapor pressure of a liquid with a planar surface, and $V_m$ is the molar volume of the liquid. An important consequence of this relationship is capillary condensation, that is, the condensation of vapor into capillaries and fine pores at vapor pressures below $P_0$.

Two particles in contact could create a narrow slit around the contact area, and liquids that wet or have a small contact angle on these surfaces will spontaneously condense from vapor into this region as described by the Kelvin equation. The meniscus formed then creates an attractive force between the particles for two reasons: first, the direct action of the surface tension forces around the periphery of the meniscus pulls the particles together; and second, the curved liquid surface results in a Laplace pressure as described by Equation (I – 15), which is lower than the pressure outside the meniscus. The force arising from the resolved surface tension is always small compared to the Laplace pressure contribution except for contact angles close to 90° [31]. Considering spheres as an approximation for small
particles in contact, the expression for the adhesion force $F_{\text{cap}}$ due to capillary effects may then be derived to be [50]:

$$F_{\text{cap}} = 2\pi \gamma_L R^* (\cos \theta_1 + \cos \theta_2) \quad (1-17)$$

Where $\theta_1$ and $\theta_2$ are the liquid contact angles on the two spheres, and $R^*$ is the effective radius. Capillary forces are also affected by surface roughness, especially at low vapor pressures when capillary bridges are only formed between the very few asperities in close contact [51].

The extent and manner in which humidity and water in the environment affect gecko adhesion and friction continue to be debated. Initial reports indicated a strong influence of humidity on the normal adhesion of gecko spatulae when tested with an AFM tip [7], with large increases in adhesion at high humidity. This was also confirmed with tests on setal arrays in a SFA [12], when considerably higher normal adhesion forces were measured at 100% relative humidity in comparison with dry N₂ conditions. It was suggested that a change in the superhydrophobic properties of gecko setae on prolonged exposure to water for over 20 minutes might have led to a switch in the conformation of surface proteins in the beta-keratin, thereby increasing the surface energy and the adhesion. Based on mechanical tests on setal arrays, others [13] have instead suggested that increased humidity changes the mechanical properties of the setae, making them more compliant, thereby resulting in better contact with the substrate and higher normal adhesion. It should also be noted that very low adhesion was measured in both SFA tests on setal arrays and AFM tests on
spatulae when completely submerged in water. For the SFA tests, the results were shown to be roughly consistent with estimates from the Lifshitz theory of van der Waals forces for two media interacting across a third medium (in this case, water) with a large dielectric constant.

At high loads in the SFA tests on setal arrays, the friction force did not seem to be affected by environmental humidity or immersion under water [12]. No shear forces were measured in the tests on single spatulae [7], but recently published whole animal tests contradict the SFA results and show a large decrease in shear force supported by initially dry toe pads on both a water-misted glass surface and a completely submerged glass surface [52]. Further, initially soaked toe pads also showed lower friction forces against a dry glass substrate, and did not recover even after four consecutive steps of the animal. This apparent loss of shear force performance under wet conditions has however not yet been correlated with any reported variations in the activity patterns of geckos during rainstorms in their natural habitat. Further, a reported ambient temperature dependence of the effect of humidity on gecko friction force is yet to be fully understood [53].

**D. Friction**

Friction is the force resisting the relative sliding motion between two bodies. Closely related phenomena are wear and lubrication. Wear is the progressive loss of material from a body caused by contact and relative motion. The aim of lubrication is to reduce friction and minimize wear. The surface interactions are complex, and
involve several factors such as surface roughness over several length scales down to the nanoscale, plastic deformation, wear and lubrication. Also, these interactions take place at a buried interface that is hard to access during experiment. However, over the past few decades, with the development of new experimental methods such as the surface forces apparatus (SFA) and the atomic force microscope (AFM), it has become possible to study friction and lubrication at the molecular scale. Several important empirical observations have been made, and theories advanced to account for these results.

The classic laws of friction as discovered by Leonardo da Vinci and then later experimentally rediscovered by Guillaume Amontons are that the friction force does not depend on the apparent area of contact of the two bodies in relative sliding motion, and that the friction force is proportional to the normal load [44]. These laws are described by the equation:

\[ F_\parallel = \mu F_\perp \]  

(1 – 18)

Where \( F_\perp \) is the normal load; and \( \mu \) is the coefficient of friction, that should be constant and independent of the contact area between two bodies. Strictly, this equation, resulting from the two laws of Amontons, should apply only to dry and unlubricated surfaces, since these were the systems originally studied. However, even for such surfaces it does not apply since there is always some, and in some cases, even strong adhesion (for example, between clean, dry, unoxidized and smooth metal surfaces) [31]. The equation, although now known to be incorrect, is
still widely used in many situations for approximations of the expected friction force in a sliding contact [31].

Coulomb added a third law (also incorrect) based on his observations that the friction force during sliding (kinetic friction) does not depend on the sliding velocity. Coulomb also realized that surfaces are not perfectly smooth, and instead have numerous asperities, resulting in a hill and valley structure. He proposed that rigid interlocking asperities could be a source of the friction force [54]. However, a major limitation of this model is apparent when a perfectly sinusoidal interface is considered – for such an interface, sliding would not require any energy dissipation since once the top of the first asperity has been surmounted, the system will slide down the other side and continue to do so, needing no additional force once set in motion. Bowden and Tabor proposed another mechanism. They pointed out that the real area of contact $A_{\text{real}}$ between two solids is only a small fraction of the apparent contact area $A_{\text{app}}$, due to surface roughness [55]. Even optically smooth surfaces might simply have a surface roughness below the wavelength of light, which could still be large in comparison with molecular dimensions. Due to this rough surface topography and since the range of intermolecular forces is very small, the two surfaces achieve intimate contact only at some microscopic contacts. Recognizing the importance of surface forces, they proposed that adhesion occurs at the asperities in contact, and shearing motion requires these adhesive forces to be overcome. The friction force is equal to the force necessary to shear these junctions –since the
number of microscopic contact ‘bonds’ is proportional to the real area of contact, the friction force is given by:

\[ F_{\parallel} = \tau A_{\text{real}} \]  

(1 – 19)

Where \( \tau \) is the interfacial shear stress, that is, the maximum lateral stress that the contact can bear. This equation has also been used to describe nanoscale contacts [56].

Bowden and Tabor suggested that plastic deformation at the contacting asperities could explain the linear dependence of \( A_{\text{real}} \) on load \( (A_{\text{real}} \propto F_{\perp}) \), since for a fully plastic contact, the normal stress stays constant and equal to the material yield stress. Therefore, for non-adhering surfaces, based on Equation (1 – 19) and the linear area-load relationship, the dependence of friction on normal load takes the form: \( F_{\parallel} = \mu F_{\perp} \). This simple model has however been criticized since it does not explain how engines can sustain relative motion of surfaces for several years without a significant change in friction behavior if continuous plastic deformation of the contacting surfaces occurs throughout this period [44]. If we instead assume only elastic deformation at the contacting asperities and model each asperity as a sphere, then using the Hertz theory for contact, the real area of contact is predicted to depend on load according to: \( A_{\text{real}} \propto F_{\perp}^{2/3} \). This situation, however, only considers the deformation of a single asperity. Greenwood and Williamson [57] developed a model for a population of asperities on interacting surfaces by using a Gaussian distribution for asperity heights, with all asperities taken as spherical caps with
identical radii of curvature. This model gave the result that, for rough surfaces, $A_{\text{real}}$ is, in fact, approximately proportional to normal load, resulting once again in a linear dependence between real area of contact and normal load. Thus, for non-adhering surfaces, both the Bowden-Tabor and Greenwood-Williams models lead from the linear dependence of friction on real area of contact to Amontons’ Law of the linear dependence of friction on normal load.

Previous experimental results have shown that in general the friction force can be split up into separate and additive (external) load dependent and (internal) adhesion-dependent contributions [58]. Extending Coulomb’s interlocking asperity theory of friction to include both adhesive interactions and energy dissipation, a ‘cobblestone’ model of friction has been developed [58]. This results in a general equation that describes the friction force, $F_{\parallel}$, between two adhesive surfaces:

$$F_{\parallel} = \mu F_{\perp} + S_c A_{\text{real}}$$  \hspace{1cm} (I – 20)

Where $F_{\perp}$ is the normal load, $\mu$ is the friction coefficient, $S_c$ is the shear strength, and $A_{\text{real}}$ is the real contact area. The first term of Equation (I – 20) is Amontons’ law for non-adhering surfaces, and it dominates the friction at high loads. The second term is the adhesion-dependent contribution, which is proportional to the real molecular contact area. It exists at zero and even negative loads so long as two surfaces are still in contact over a finite area. For adhesive surfaces, this gives rise to a positive offset in the $F_{\parallel}$ vs. $F_{\perp}$ curve; while for repulsive surfaces, it gives rise to a negative offset. At the opposite extreme of very high loads, since $A_{\text{real}} \propto F_{\perp}^{2/3}$ for
both JKR and Hertzian contacts, the first linear term in $F_\perp$ eventually dominates the sub-linear adhesive term even for adhesive systems, as observed experimentally [58].

Sliding motion between bodies often proceeds in jerks known as ‘stick-slip’ instead of smoothly. Several models have been advanced to explain this behavior in a variety of systems, including both dry, unlubricated solids as well as solids with thin lubricating film between them [59]. In the velocity dependent model, dependence of friction force on sliding velocity may result in stick-slip. Static friction is defined as the force that must be overcome to start the movement between two bodies initially at rest, while dynamic or kinetic friction is the force between sliding bodies that resists the relative motion. If the dragging force is coupled elastically to a body, a difference in static and kinetic friction values may lead to so-called ‘stick-slip’ motion [55]. With a body initially at rest, the spring force transmitted by the elastic coupling between the body and the externally applied force will not cause sliding until the static friction force has been exceeded (this is the stick phase). Then, if the kinetic friction force during sliding is lower than the static value, the body is accelerated due to the force imbalance (this is the slip phase). This increase in the velocity of the body relaxes the elastic coupling, resulting in a lower force transmitted to the body, causing it to come to rest and ‘stick’ again. This stick-slip motion occurs repeatedly. Common examples of stick-slip motion in everyday life are the squeaking of vehicle brakes and door hinges, the excitation of a violin string by a bow, and earthquakes. In many practical applications, stick-slip motion is
detrimental in terms of increased wear, vibrations and noise, energy loss, and loss of precision in movement.

**E. Gecko-inspired Synthetic Adhesives**

To design dry, responsive adhesive systems inspired by the gecko, various kinds of patterned surfaces with arrays of vertical and tilted fibers, with tips of different shapes and materials, and with sizes spanning both the micron and nanometer scales, have been fabricated [46, 60–63]. These structures mainly focus on enhancing adhesion properties. While some other attempts focus on both friction and adhesion, they commonly use cylindrical micro-fibers, which only provide a line contact with the adhering surface when sheared, limiting the real surface area of contact for van der Waals forces [29, 64–66]. This provided the motivation to fabricate vast arrays of micron-sized rectangular flaps composed of polydimethylsiloxane (PDMS) using massively parallel micro-electromechanical systems (MEMS) techniques to increase the surface area in contact after lateral shearing/sliding. Using a surface forces apparatus (SFA), the friction and adhesion forces of both vertical (symmetric) and angled/tilted (x-y-z asymmetric) micro-flaps were measured under various loading, unloading and shearing conditions against a spherical glass probe. It was found that the anisotropic structure of tilted micro-flaps resulted in very different adhesion and friction forces when articulated along different x-y-z directions: high friction and adhesion when articulated in the y-z plane along the tilt, and weak friction and adhesion forces when articulated against the tilt. These results demonstrate that
asymmetric angled structures, such as those that occur in geckos, are required to enable the gecko to optimize the requirements of high friction and adhesion on gripping, and low frictional-adhesion on releasing. This work has been described in detail in Chapter III, and previously published in [67].

Small scale tests performed with a glass sphere contacting the flap structures using the Surface Forces Apparatus (SFA) had contact areas of $0.1 - 1 \text{ mm}^2$, depending on the applied normal load. The encouraging results from SFA tests necessitated larger testing sizes since small test areas can overestimate the forces achieved when patch sizes are scaled up. The discrepancy, as was shown between single seta [11] and whole feet [10] force measurements for the natural system, as well as for varying patch sizes of a synthetic adhesive [68], can be substantial and are likely due to incomplete or poor attachment at large testing sizes. To verify the scalability, a new ‘Bio-Forces’ or ‘Bio-F’ tribometer with larger testing area was used to investigate the material [62]. The Bio-F’s test contact area is between 12 and 120 times greater than the SFA with an available area of $12.6 \text{ mm}^2$. This value is roughly constant for the tests shown in the report because the testing geometry is flat-on-flat where changes in load change contact area with the tips of the fiber arrays only at small loads. Although loading of the fibers is more uniform over the entire contact area with the Bio-F, a significant challenge is careful alignment of the two flat surfaces with one another for good contact. For tests involving sliding to measure friction forces, the sample must also be aligned with the direction of sliding.
motion. Tests on angled flaps using the Bio-F apparatus are reported in detail in Chapter IV, and results from these tests have been previously published in [69].

In a natural environment, almost all surfaces are rough, usually containing different scales of roughness. The gecko frequently encounters surfaces such as bark and rock, which can contain varying degrees of roughness. Surface roughness can significantly reduce the real contact area of two surfaces, and therefore reduces the adhesion force between surfaces [70, 71]. Although much attention has been focused on the fabrication of various structures that mimic the functionality of the hierarchical structure of a gecko foot, yet no systematic effort, in experiment or theory, has been made to quantify the effect of surface roughness on both the adhesion and friction performance of these gecko-inspired structures. Using a modified Surface Forces Apparatus, the adhesion and friction forces between micro-fabricated tilted PDMS flaps and optically smooth glass and rough glass surfaces were measured. The SFA test probes were roughened by a plasma etching process to create substrate surfaces with a range of RMS roughness values from 11 nm to 308 nm. Anisotropic adhesion and friction forces were measured when sliding the top glass surface both along and against the tilted direction of the flaps. Increasing the surface roughness first increased the adhesion and friction forces measured between the flaps and the rough surface due to topological matching of the two surfaces, but then led to a rapid decrease in both of these forces. These results demonstrate that the surface roughness significantly affects the performance of gecko mimetic adhesives, and that different surface textures can either increase or decrease the
adhesion and friction forces of the fabricated adhesives. These results are discussed in detail in Chapter V, and have been previously published in [72].

In addition to high and controllable adhesion and friction, most practical applications for reversible synthetic adhesives require them to also demonstrate high reusability. Although several synthetic adhesives capable of supporting large adhesion and/or shear forces have been reported, either no extended durability testing was done [67, 69, 73–74], or they exhibited very limited lifetime [75–77], or no systematic study into durability was performed beyond just 50-100 cycles [29, 66, 78–79]. A highly reusable synthetic adhesive has been developed using tilted Polydimethylsiloxane (PDMS) half-cylinder micron-scale fibers, with a reduction in adhesion of just 23% of the initial value over 10,000 repeated test cycles against a flat glass puck. Although a bio-inspired synthetic fibrillar adhesive system with a long lifetime comparable to gecko setae has been reported in [68], the wedge-shaped fibers used did not exhibit any gecko seta-like adhesion or friction anisotropy on sliding, and supported maximum shear loads far inferior to the gecko. The only other study thus far with tests over 10,000 cycles reported a large reduction in normal adhesion for high-density polyethylene (HDPE) fibers, and a complete loss of adhesion for polypropylene (PP) fibers [80]. Large reductions in shear performance were also reported over these repeated tests. The ratio of adhesion to initial preload force (μ’) is also especially important for climbing applications, with higher values resulting in increased operational stability on walls and ceilings due to a low corresponding reaction force from the substrate surface, as well as enhancing
reusability by preventing damage to fibers during preloading. Results from Bio-F tests using the half-cylinder tilted fibers also show a large improvement in the value of $\mu'$ over other high-cycle adhesives. These results are discussed in detail in Chapter VI, and have been previously published in [81] and [82].
II. Microfabrication

A variety of vertical and tilted PDMS microstructures exhibiting anisotropic adhesion and friction properties were fabricated using a molding technique. In order to repeatably fabricate large arrays of several tens of thousands of microstructures over cm-scale patch areas with very few defects, the negative molds used were created using massively parallel microfabrication techniques. Conventional projection lithography and Bosch Deep Reactive Ion Etching (DRIE) processes were used to fabricate molds in silicon for the vertical structures, while a novel angled lithography technique with a photoresist bilayer was used to fabricate molds for the tilted structures. These methods are described in detail in this chapter.

A. Fabrication of Vertical Flaps

To fabricate the vertical flaps, a negative silicon mold was prepared by first defining the flap lateral dimensions by lithography in an i-line stepper (GCA 6300, Refurbished by RTS Technologies, Doylestown, PA), and then etching into a single crystal silicon wafer up to the desired depth using the Bosch DRIE process. To increase (double) the areal density of the flap structures using the same lithographic mask, this process was slightly modified (Figure II – 1).
Figure II - 1: Schematic of the microfabrication process used to fabricate a negative silicon mold for vertical (symmetric) PDMS micro-flaps. A two stage lithography process was used to increase the area coverage of the flaps.

A 0.5 µm thick layer of silicon oxide was deposited on a clean silicon wafer using a plasma-enhanced chemical vapor deposition (PECVD) system. An approximately 1.0 µm thick layer of AZ 5214, an image reversal photoresist (AZ Electronic Materials, Branchburg, NJ), was spin coated over the wafer, and slots for flaps, 4 µm by 10 µm in lateral dimension, were defined by lithography. The oxide layer exposed in these slots was etched away in a CHF₃ plasma using an inductively coupled plasma reactive ion etching system (ICP-RIE). The photoresist layer was
now stripped away using Microposit 1165 photoresist stripping solution. A fresh coat of AZ 5214 photoresist was applied to the cleaned and dried wafer and a second set of slots for flaps was subsequently defined using lithography, the second set of flaps being offset symmetrically from the first in the interstitial spaces. The exposed oxide layer was again etched away using the same ICP etch process. After stripping off the photoresist from the second lithography step, the cleaned and dried wafer was ready for the silicon etch step in the Bosch DRIE system, with the patterned oxide layer acting as an effective etch mask. The micro-scale rectangular slots were vertically etched to a depth of 15 µm in the Bosch DRIE system.

The negative mold fabricated by this method was then prepared for PDMS molding by vapor depositing a silane layer. PDMS (Sylgard 184, Dow Corning, Midland, MI) was mixed in a 10:1 ratio by weight with a crosslinker, and poured over the negative mold. This was degassed and then cured at 100 °C for 15 minutes in a convection oven to fully cure the PDMS. The two materials were then separated by carefully peeling off the cured PDMS, resulting in cm-scale PDMS samples with micro-flaps (Figure II – 2) and a silicon mold that could be reused.
Figure II - 2: Scanning electron microscope (SEM) images of large arrays of microfabricated vertical PDMS flaps 15 µm in height, 10 µm wide in the ±x direction, and 4 µm thick in the ±y direction. The –z direction is the (vertical) loading direction, and +z is the unloading direction.

B. Fabrication of Angled Flaps

An initial attempt to fabricate tilted microstructures used a two-step molding procedure (Figure II – 3). First, silicon masters were created using lithography and Bosch DRIE, and used to create a negative PDMS mold. The PDMS was only partially cured before being peeled off from the master mold, and was then sheared before being fully thermally cured to set in an angle of tilt. The sheared PDMS negative mold with angled slots was then used to mold tilted structures in poly-mercaptopropyl methyl siloxane (PMMS).
Figure II - 3: (a) Schematic diagram of the two-step molding process initially used to fabricate tilted PMMS microstructures. repeatability and angle control were major issues with the shearing technique. The parallel plate device used to perform shearing is shown in Figures (b) and (c).

Although it was possible to create tilted cylindrical microfibers with the shearing technique (Figure II – 4(a)), repeatable fabrication with uniform tilt angle over cm-scale patch size areas could not be achieved. Further, it was not possible to fabricate tilted rectangular micro-flap structures with this method – shearing the partially cured PDMS mold resulted in collapse of these slender structures (Figure II – 4(b)).
Figure II - 4: (a) SEM image of tilted PMMS cylindrical microfibers, fabricated using a two-step molding technique with shearing of a partially cured PDMS negative mold. (b) SEM image of a PDMS negative mold for micro-flaps, with walls collapsed after shearing.

Another shearing technique that was investigated involved direct mechanical deformation of vertical PDMS flaps to obtain tilted structures. The mechanical device used to shear PDMS negative molds was modified to include a moat for a lubricating silicone oil, and a glass window in the top surface to enable continuous observation of the shearing process under a microscope (Figures II – 5(b) and II – 5(c)). Partially cured vertical PDMS flaps were peeled off from a negative silicon mold and carefully sheared after alignment with the top surface using 4 spring mounted screws. The lateral shearing motion of the lower surface was enabled by a manual linear stage controlled by a micrometer. The sheared flaps were then allowed to fully cure at room temperature for 24 hours. Although micro-flaps tilted at ~13° from the vertical were observed in small areas of the sample (maximum patch size
Further, large areas of the sample exhibited either unsheared vertical flaps or fully bent over flaps adhered to the base due to difficulty in flat-on-flat alignment in the shearing device.

Figure II - 5: (a) Schematic for fabrication of tilted micro-flaps by directed mechanical deformation. (b) The angle of tilt obtained was \( \sim 13^\circ \) from the vertical, but uniform fabrication over areas greater than \( \sim 0.5 \, \text{mm}^2 \) was not possible. (c) Large areas with flaps completely bent over were observed due to flat-on-flat alignment issues during the shearing step.

Since Bosch DRIE is a vertical etching process, fabrication of a negative mold with angled slots for tilted flaps necessitated development of a different microfabrication technique. Although direct angled exposure of photoresists in i-line
stepper or contact aligners has been reported elsewhere in the literature [64, 68], the existing facilities at UCSB allow only a maximum tilt angle of ~5° from the vertical with such methods (Figure II – 6).

**Figure II - 6:** Schematic diagrams representing two possible angled exposure techniques to create angled slots for molding tilted PDMS structures. In Figure (a), a prism placed over a photomask mounted in a contact aligner bends incident light to expose the photoresist at an angle. In Figure (b), the photoresist-coated wafer (clipped to the photomask) is itself mounted at an angle in the contact aligner for angled exposure. However, due to space limitations in all contact alignment systems at UCSB, the maximum angle of tilt from the vertical that could be generated with such schemes is limited to ~5°.
To reliably fabricate large arrays of angled micro-flap structures over cm-scale areas, a novel angled exposure technique using a photoresist bilayer was developed (Figure II – 7), requiring the use of a non-reflective glass substrate wafer. This technique was later also used to fabricate tilted structures with a semicircular lateral geometry, as described further in Section C of this chapter.

Figure II - 7: Schematic of the microfabrication process used to fabricate a negative mold for angled (tilted, asymmetric) PDMS micro-flaps. A two stage lithography process was used, the lateral features being defined by the first stage (steps 1 to 3 in the schematic), the angle $\phi$ being set by the second stage (steps 4 and 5). A glass wafer was used as the substrate to minimize reflections during the angled exposure step 4.
A clean and dry glass wafer was coated with a photoresist bilayer – first with a 
~10 µm thick layer of polymethylglutarimide (PMGI) positive tone photoresist 
(PMGI SF-15, MicroChem Corp., Newton, MA), and then with a top layer of AZ 
5214 image reversal photoresist approximately 1.4 µm thick. The desired thickness 
for the PMGI SF-15 layer was obtained by applying two coats of the photoresist at a 
low spin speed of 1000 rpm, with a short soft bake of 1 minute at 200 °C between 
the two coats.

The two photoresist materials are sensitive to different wavelengths of UV light, 
PMGI to the deep UV range of wavelengths between 200 and 260 nm, and AZ 5214 
to the i-line UV wavelength of 365 nm. The flap lateral dimensions were defined in 
the i-line lithography step used to pattern the AZ 5214 imaging resist layer on top. 
After developing this resist in AZ 400K developer (at 1:4 dilution), rinsing in DI 
water and drying, the sample was mounted at an angle in a deep UV flood exposure 
system. The angle chosen for mounting the sample is related to the final angle 
obtained in the PMGI layer by Snell’s law for refraction at the air-PMGI interface 
(refractive index for PMGI ~1.54). The patterned AZ 5214 layer atop the PMGI sets 
the lateral dimensions of the angled flaps by acting as a mask during the deep UV 
flood exposure step. The entire process sequence for negative mold fabrication with 
this angled lithography method has been included in the Appendix (Table A – 1). 
Process development undertaken to improve the process has been summarized 
below.
Figure II - 8: Tuning the total deep UV exposure dose as well as post-exposure development time resulted in an improvement in micro-flap shape, as seen in Figures (a) and (b). Splitting the exposure and develop steps into a number of shorter steps and cycling through resulted in further improvements in micro-flap geometry, as seen in Figures (c) and (d).

Both the total exposure time (at a constant lamp power of 1000 W) and the total develop step time (in PMGI 101 developer – MicroChem Corp., Newton, MA) were tuned to increase the depth to thickness aspect ratio of the angled slots while minimizing undercut (Figure II – 8). These parameters were further improved by
repeating the PMGI exposure step for 300 seconds each time, and splitting the develop step of 7 minutes into a series of six equally long develop steps. Photoresist undercut still resulted in a bulged out region on the micro-flaps, as seen in Figure II – 8(c) (marked by the red oval). This was further improved by using eight PMGI develop steps instead of six, while still keeping the total development time fixed at 7 minutes. This process then enabled fabrication of angled flaps ~10 µm in height (10.8 µm measured along the tilt) and ~3.5 µm thick, with a tilt angle of ~20° from the vertical (Figure II – 8(d)).

Before molding PDMS flaps using the PMGI negative molds, the durability of the molds were increased (to facilitate reuse) by hard baking the patterned PMGI resist at 110 °C for ~15 minutes. The molds were also then exposed to an oxygen plasma for 3 minutes, followed by deposition of a silane layer to facilitate easy peel-off. PDMS molding was then done using the same scheme described earlier for molding vertical flaps with silicon negative molds, taking care to only peel with the direction of tilt to avoid damage to the angled flaps and to the photoresist mold. It was observed that the molds could be reused ~15–20 times before significant damage.

However, before repeatable fabrication of durable molds could be achieved, two other issues relating to thick photoresist film handling remained to be resolved. The first was the formation of deep cracks in the photoresist bilayer film from the top surface right down to the glass wafer substrate. These cracks, extending for several millimeters in length across the sample, occurred during the top photoresist layer
(AZ 5214) soft bake step, and resulted from the different thermal expansions of the two different photoresist materials. On molding, the cracks gave rise to a fairly dense network of tall ridge-like structures, in most cases either at least equal to or even greater than the height of the angled flaps (Figure II – 9).

Figure II - 9: Cracking of the thick photoresist bilayer film during the final soft bake step resulted in the formation of undesirable ridge-like structures, in most cases of height greater than the flaps, which interfered in sample adhesion and friction properties characterization.

This meant that reliable tests over large patch areas could not be conducted without observing artifacts due to the presence of the ridges. To eliminate crack formation, the soft bake step for the AZ 5214 photoresist was modified to include a gradual ramp up to the soft bake temperature of 95 C from a base temperature of 50
C over a period of 3 minutes, and a gradual cool down from 95 C after soft baking for an additional 3 minute period.

The second thick photoresist film issue that was resolved related to the limited depth of focus of the GCA 6300 projection stepper alignment system used to define the lateral feature dimensions in the top AZ 5214 layer (step 3 in Figure II – 10). While the stepper aligner is a high precision instrument that is routinely used by several other cleanroom users to define challenging features such as isolated lines with a resolution of ~0.5 \( \mu \text{m} \), it was unable to reliably define the 2 \( \mu \text{m} \) thick flap dimensions in the top AZ 5214 layer (as seen in an optical microscope image in Figure II – 10(a)). This resulted in final micro-flap structures with a poor height to thickness aspect ratio and a very rough surface finish after angled lithography and PDMS molding were performed (as seen in an SEM image in Figure II – 10(b)).

Poor resolution was due to the fact that the stepper aligner is optimized for exposures on thin photoresist films (thickness < 3 \( \mu \text{m} \)) coated on reflective silicon wafers, whereas our process requires the utilization of a 11 \( \mu \text{m} \) thick photoresist bilayer stack over a non-reflective glass substrate. The quality of focus obtained from an exposure in the stepper aligner also tends to vary, being most sharp near the center of any 5 inch square photomask. Thus, an improvement in lateral feature resolution was achieved (Figure II – 10(c)) by drawing a new mask with small arrays of the features of interest clustered around the center. Large continuous arrays were then obtained during lithography by exposing the features in a rectangular array with a small 1 \( \mu \text{m} \) overlap to eliminate any empty spaces in between. This enabled repeatable
fabrication of tilted PDMS micro-flaps with high aspect ratio and good lateral surface finish over cm-scale patch size areas (Figure II – 10(d)).

Figure II - 10: Poor resolution in rectangular features defined in the top photoresist layer (Figure (a)) yielded micro-flaps with poor dimensions and surface finish (Figure (b)) after angled lithography and PDMS molding. An improvement in lateral feature resolution (Figure (c)) resulted in a significant improvement in angled micro-flap geometry and surface quality (Figure (d)).

C. Fabrication of Angled Half-Cylinder Fibers

To fabricate large arrays of tilted PDMS half-cylinder microfibers over cm-scale patch areas, a negative mold with angled slots was fabricated out of photoresist polymer material using an angled photolithography technique similar to that used for
angled flaps, as shown in Figure II – 7. However, instead of rectangular slots in the top imaging photoresist layer, semicircular slots are defined.

Figure II - 11: Schematic diagram showing a glass wafer with a top AZ 5214 photoresist layer (pre-patterned with semicircular features) being mounted and exposed at an angle to create angled slots in the underlying deep UV photoresist layer (PMGI SF-15). The wafer is mounted so as to ensure that the final PDMS half-cylinder microfibers fabricated from the photoresist negative mold will have the flat side tilted to face towards any contacting substrate, and the curved side facing away, in order to maximize anisotropy in contact area on lateral shearing.

Also, in the angled exposure step (step 4 in Figure II – 7), the wafer was mounted so as to ensure that the final PDMS structures fabricated from this mold
would have the flat side of the half-cylinder fibers tilted so as to face towards any contacting substrate, and the curved side of the fibers face away, in order to maximize anisotropy in contact area on lateral shearing. This has been schematically shown in Figure II – 11.

For the half-cylinder fibers, it was found that step 4 for PMGI exposure and step 5 for PMGI development (as shown in Figure II – 7) were required to be repeated 12 times each in order to fully develop the photoresist, with each exposure step being for 5 minutes at a constant deep UV lamp power of 1000 W, and each develop step being for 25 seconds in PMGI 101 developer.

Similar to previous molding processes used, the negative mold was hard bakes, silanized, and used for casting PDMS structures, resulting in cm-scale PDMS samples with tilted microfibers and a photoresist mold that could be reused up to 15-20 times. The tilted fibers were ~13 µm in height, with a semicircular lateral profile of average diameter ~8.5 µm, although this varied with z-axis height as seen in Figure II – 12(b) due to greater development of parts of the photoresist mold that were repeatedly directly exposed to deep UV light and the developer solution. The angle of tilt of the flat side was observed by SEM to be ~28° from the vertical, while the angle of tilt of the curved side was ~15° from the vertical (Figure II – 12(a)).
Figure II - 12: (a) SEM image of PDMS angled half-cylinder microfiber arrays, with the fiber lateral geometry outlined, and coordinate system used indicated. In all images, the $+y$ axis points towards the flat face of the fibers (tilted at 28° from the vertical), while the $-y$ axis points towards the curved face (tilted at 15° from the vertical). The $\pm x$ axis is parallel to the straight edge of the top face. The $-z$ axis is the normal loading direction, while the $+z$ axis is the normal unloading direction. Articulation of the adhesive occurs in the $y$-$z$ plane. (b) SEM image showing the curved face of the microfibers. The width of a single fiber varies along both the $\pm x$ and $\pm y$ axes due to the fabrication process. Both images were taken at a 70° mounting angle in the SEM.

**D. Fabrication of Non-Uniformly Rough Substrate Surfaces**

In order to test the roughness adaptation of the angled micro-flap adhesive system, rough substrate surfaces that could be easily integrated into the SFA test apparatus were required. Initial attempts to roughen SFA pucks included HF etching,
sand blasting, and etching with a commercial glass etching kit (Armour Etch Cream, Armour Products, Hawthorne, NJ, USA). While no roughening was observed after etching with both buffered HF and 48% HF, the roughness asperities generated by both the sand blasting and Armour etching techniques were \( \sim 5\text{-}20 \mu m \) in height, nearly an order of magnitude greater than the level of roughness desired for this study. SEM images of the rough surfaces produced by sand blasting are shown in Figures II – 13(a) and II – 13(b), and images of the rough surfaces produced after Armour etching are shown in Figure II – 13(c) and II – 13(d). Hence, a plasma etching technique to roughen the glass surfaces was investigated.

Micromasking is a phenomenon whereby surface roughening can occur during reactive ion etching due to the deposition of sputtered material from several possible sources in the reaction chamber, most commonly from the patterned mask material itself. This debris results in a slower etch rate due to masking in unintended regions, with microhillock formation and a final rough etched substrate surface [83, 84]. Dry etching of glass substrates using fluorine containing plasmas has been previously shown to cause significant surface roughening in certain etch parameter ranges, both in experiments with a \( \sim 10 \) nm thin metal layer (such as Au or Cu) deposited as a seed layer for micromasks [85, 86] and in experiments without any externally deposited layer over the glass material [86]. The latter phenomenon, whereby surface roughening occurs even without any masking layer, has been attributed to the production of less volatile metal halogen compounds on the glass surface during etching. The uncoated SFA glass disks used were manufactured with Schott N-BK7
517/642 material, which is a borosilicate glass (Schott North America, Inc., Elmsford, NY, USA) with a refractive index of 1.517. In addition to SiO$_2$, the BK7 borosilicate glass material has been reported to contain oxides of sodium, aluminum and barium [87, 88]. It has been postulated that the creation of less volatile species such as NaF, AlF$_3$, and BaF$_2$ from these materials in a fluorine chemistry RIE plasma may lead to a roughening of the glass substrate by micromasking [85, 89].

Unetched and uncoated SFA disks were found by AFM (Tapping Mode) analysis to have a polished surface with an RMS roughness value of ~11 ± 8 nm. Through trial and error, a dry etch recipe using a mixture of O$_2$, SF$_6$, CF$_4$ and Ar as process gases in an RIE plasma was developed to etch these optically smooth SFA glass disks and generate non-uniformly rough test substrate surfaces, as shown in Figures II – 13(e) and II – 13(f). Initially smooth SFA glass pucks (Product No. NT63-476, Edmund Optics Inc., Barrington, NJ, USA) were etched in a Materials Research Corporation MRC 51 reactive ion etching system (Praxair Surface Technologies, Indianapolis, IN, USA). However, these surfaces were not found to be robust enough for extensive adhesion and friction testing, and flaked off after just a few test cycles.
Figure II - 13: SEM images of rough glass puck surfaces produced by sand blasting (Figures (a) and (b)), Armour etching (Figures (c) and (d)), and by reactive ion etching in a CF$_4$/SF$_6$/O$_2$/Ar plasma (Figures (e) and (f)).

However, it was found that using the same RIE plasma composition with lower flow rates for all gases (3/4 the flow rates in the initial recipe for all gases) and a reduced chamber pressure of 10 mTorr (compared with the initial recipe value of 80 mTorr), it was possible to generate roughness asperities that were robust enough for
repeated adhesion and friction testing. These asperities were roughly cylindrical, distributed in a non-uniform pattern, and had a maximum height lower than 3 µm. No cracks or line textures were observed on the pucks after etching. Surfaces of increasing RMS values of roughness were generated by simply exposing smooth SFA disks to the RIE plasma for increasing lengths of time (Table II – 1), resulting in roughly cylindrical asperities of larger radii and heights (Figure II – 14).

**Table II - 1: Etching times for the creation of non-uniformly rough surfaces on smooth glass disks**

<table>
<thead>
<tr>
<th>Etching time$^a$ (min)</th>
<th>RMS height roughness and standard deviation (nm)</th>
<th>Average distance between asperities and standard deviation (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>11 ± 8</td>
<td>N/A</td>
</tr>
<tr>
<td>35</td>
<td>73 ± 5</td>
<td>8.6 ± 4.5</td>
</tr>
<tr>
<td>48</td>
<td>133 ± 20</td>
<td>8.1 ± 3.2</td>
</tr>
<tr>
<td>65</td>
<td>192 ± 9</td>
<td>4.9 ± 3.3</td>
</tr>
<tr>
<td>120</td>
<td>308 ± 56</td>
<td>4.8 ± 2.7</td>
</tr>
</tbody>
</table>

$^a$Process conditions: $O_2$ flow rate = 30 standard cubic centimeter per minute (sccm), $SF_6$ flow rate = 30 sccm, $CF_4$ flow rate = 3 sccm, $Ar$ flow rate = 30 sccm, chamber pressure = 10 mTorr, Bias Voltage = 500 V.
Figure II - 14: SEM images of the rough surfaces fabricated with different height RMS roughness values: (a) 11 ± 8 nm; (b) 73 ± 5 nm; (c) 133 ± 20 nm; (d) 192 ± 9 nm; and (e) 308 ± 56 nm.

From the SEM and AFM images, it was also possible to quantify the average lateral distance between asperities on these rough surfaces. As seen in Table II – 1, increasing the etch time resulted in a continuous decrease in the average spacing, although not as uniformly as the increase in height RMS values. The fact that the average spacing for both the 73 nm and 133 nm height RMS surfaces is close to the
width of the PDMS angled flaps tested on these surfaces (~10 µm) had a very significant effect on their adhesion and friction behavior, as discussed in detail in Chapter V.
III. Vertical and Angled Micro-Flap Structures for a Gecko-Inspired Dry Adhesive

A. Introduction

Most geckos can rapidly attach and detach from almost any kind of surface. This ability is attributed to the hierarchical structure of their feet (involving toe pads, setal arrays and spatulae), and how they are moved (articulated) to generate strong adhesion and friction forces on gripping that rapidly relax on releasing. Inspired by the gecko’s bioadhesive system, various structured surfaces have been fabricated suitable for robotic applications. In this chapter, x-y-z asymmetric, micron-sized rectangular flaps composed of polydimethylsiloxane (PDMS) were fabricated using massively parallel micro-electromechanical systems (MEMS) techniques with the intention of creating directionally responsive, high-to-low frictional-adhesion toe pads exhibiting properties similar to those found in geckos. Using a surface forces apparatus (SFA), the friction and adhesion forces of both vertical (symmetric) and angled/tilted (x-y-z asymmetric) micro-flaps under various loading, unloading and shearing conditions were investigated [67]. It was found that the anisotropic structure of tilted micro-flaps results in very different adhesion and tribological forces when articulated along the different x-y-z directions: high friction and adhesion forces when articulated in the y-z plane along the tilt (+y) direction, which is also the direction of motion, and weak friction and adhesion forces when articulated against the tilt (-y) direction. These results demonstrate that asymmetric
angled structures, as occur in geckos, are required to enable the gecko to optimize the requirements of high friction and adhesion on gripping, and low frictional-adhesion on releasing. These properties are also intimately coupled to the articulation mechanism used.

**Figure III - 1: SEM images of some polymer microstructures fabricated at UCSB, including A) angled cylindrical PMMS microfibers; B) hierarchical vertical cylindrical PDMS microfibers with a flap on top; and C) vertical rectangular PDMS micro-flaps. The increase in real surface area of contact on shearing is mostly limited to a line contact in A and B (indicated by the red lines), while the micro-flaps in C are capable of providing a large surface of contact, as indicated by the blue square.**

The extraordinary climbing ability of geckos has inspired extensive studies in designing dry, responsive adhesive systems and robots. Various kinds of patterned surfaces of different shapes and materials with micro- to nano-scale structures have been fabricated [46, 60–63]. These structures mainly focus on enhancing adhesion properties. While some other attempts focus on both friction and adhesion, they commonly use cylindrical micro-fibers, which only provide a line contact with the
adhering surface when sheared, limiting the real surface area of contact for van der Waals forces [29, 64–66]. The expected increase in real area of contact (after shearing/sliding) by switching from cylindrical fibers to rectangular flaps is shown in Figure III – 1.

The lateral force or the friction force is very important when the gecko climbs on walls. As previously described in Chapter I, Section D, a general equation that describes the friction force, $F_\parallel$, between two adhesive surfaces is [58]:

$$F_\parallel = \mu F_\perp + S_c A_{\text{real}},$$  \hspace{0.5cm} (III – 1)

Where $F_\perp$ is the normal load, $\mu$ is the friction coefficient, $S_c$ is the shear strength, and $A_{\text{real}}$ is the real contact area. The first term of equation (III – 1) is Amontons’ law for non-adhering surfaces, and it dominates the friction at high loads. The second term is the adhesion-dependent contribution, which is proportional to the ‘real’ molecular contact area. It exists at zero and even negative loads so long as two surfaces are still in contact over a finite area. For the gecko adhesion system, the load contribution is very small, and the adhesion contribution therefore dominates the friction force.

In this study, vertical (Figure III – 2(a)) and pre-angled (Figure III – 2(b)) PDMS micro-flap structures mimicking the angled setae structure of the gecko feet were manufactured using micro-fabrication techniques. A new SFA 3D force-displacement attachment has been used to investigate the adhesion and friction
properties of both micro-flaps. It was found that the tilted flap-structure could provide strong frictional adhesion that mimics the gecko adhesion system.

**B. Experimental Methods**

1. Microfabrication of Vertical and Angled Micro-Flaps

Large arrays of both vertical and angled micro-flap structures have been fabricated using microfabrication techniques. These techniques have been described in detail in Chapter II, Sections A and B. To very briefly summarize these techniques, negative molds were prepared to cast PDMS micro-flap structures. For the vertical flaps, a negative silicon mold was prepared by first defining the flap lateral dimensions by lithography, and then etching into a single crystal silicon wafer up to the desired depth using the Bosch Deep Reactive Ion Etching (DRIE) process. The negative mold fabricated by this method was then prepared for PDMS molding by vapor depositing a silane layer. PDMS (Sylgard 184, Dow Corning, Midland, MI) was mixed in a 10:1 ratio by weight with a crosslinker, and poured over the negative mold. This was degassed and then cured at 100 °C for 10 minutes in a convection oven to fully cure the PDMS.

This process enabled the fabrication of vertical micro-flap arrays over cm-scale patch size areas with almost no defects. The flaps tested were 15µm in height (z axis), 10 µm wide (x axis), and 4 µm thick (y axis). An SEM image (Figure III – 2(a)) shows the fabricated flaps along with the coordinate system used to discuss adhesion and friction test results.
Figure III - 2: Scanning electron microscope (SEM) images of (a) vertical PDMS flaps of 15 µm height, and (b) ~20° angled PDMS flaps of ~10 µm height, with a bulged out region due to undercut in PMGI negative mold (marked by the red oval). In all images, the ±x directions are the directions parallel to the long edge of the flaps; the +y direction is the direction of tilt of the flaps, also the “gripping” direction, and the direction of motion; the –y direction is the “releasing” direction; –z is the (vertical) loading direction, and +z is the unloading direction. Articulation of the feet (toe pads and setae) occur in the y-z plane.

The pre-angled PDMS flaps were cast from negative molds with angled slots made out of photoresist materials. In order to fabricate arrays of angled structures of uniform height and tilt angle over cm-scale patch size areas, a new angled lithography fabrication technique was developed. This is described in detail in Chapter II, Section B. This process enabled fabrication of angled flaps ~10 µm in height (10.8 µm measured along the tilt) and ~4 µm thick, with an angle of ~20° from the vertical. Fabrication was subsequently tuned to both increase the height-to-
thickness flap aspect ratio and reduce the undercut in the PMGI layer, which translates as a bulged out region in the PDMS flaps (as marked by the red oval in Figure III – 2(b)). The angled flaps are spaced in a regular hexagonal pattern with the side of the hexagon, \( t = 25 \mu m \). Hence, the areal density of the angled flaps is:

\[
\sigma = \frac{2}{\sqrt{3} \cdot t^2} = 1847.6 \text{ mm}^{-2}
\]

Due to the two exposure steps in the fabrication of the molds for the vertical flaps, their areal density is: \( 2\sigma = 3695.2 \text{ mm}^{-2} \)

2. Adhesion and Friction Testing in a Surface Forces Apparatus (SFA)

In order to quantify the adhesion and friction properties of the fabricated structures, a 3D displacement and force sensing probe attachment for the surface forces apparatus (SFA) 2000 was developed (Figure III – 3). The new attachment can generate both normal and lateral movement of surfaces, and measures the resulting normal and lateral forces independently [90]. It was designed to permit both load/pull and load/drag/pull tests on the fabricated structures on a small scale with a contact area of around \( \sim 0.1 \text{ -- } 1 \text{ mm}^2 \). The actual contact area depends on the applied normal load. The bottom disk is mounted in a normal load sensor in the SFA 2000. The sensor has 4 foil strain gauges (Vishay Micro- Measurements) glued symmetrically to the bending arms of the double cantilever spring, forming a Wheatstone bridge strain gauge system. When a normal force is applied to the surfaces, the strain gauges are used to measure the deflection of the spring with a signal conditioning amplifier (Vishay Measurements, 2300), which outputs the
signal to either a computer data acquisition system or a chart recorder. The voltage signal is then calibrated against the weight.

**Figure III - 3:** Schematic of the experimental set-up illustrating the main features of the 3D force-displacement sensor attachment to the SFA 2000. This attachment allows 3D translation and (independent) force sensing.
The top surface is held by a friction device that can move laterally with a sliding distance of between 200 and 500 \( \mu \text{m} \). Driven by a reversible DC motor, the friction device can slide the upper disk back and forth smoothly with respect to the lower disk at different constant or variable speeds using a function generator. With the same force sensing mechanism as the normal load sensor, the friction device can measure the lateral shearing force (friction) during the sliding of the top surface.

The adhesion and friction tests were done with a spherical glass disk compressed/sliding against an opposing micro-flap/polymer substrate. Before the experiment, a thin PDMS sheet with fabricated PDMS micro-flaps was glued onto a flat glass disk. The disk was then mounted into the normal load sensor of a SFA box for further measurement. A spherical glass disk with a radius of 2 cm was mounted to the top friction device. Prior to use, the glass disks were soaked in chloroform (EMD Chemical Inc., Gibbstown, NJ) for 1 day and then rinsed thoroughly with ethanol followed by drying in dry nitrogen gas.

In the SFA experiments, the spherical glass disk was first pressed against the PDMS micro-flaps at a constant speed of about 0.06 \( \mu \text{m/s} \) until the desired normal preload was achieved. To measure the adhesion forces, the two surfaces were separated from each other at an angle perpendicular to the bottom surface at the same speed. To measure friction forces, a square wave voltage function with a peak-to-peak voltage of 20 V was applied to the DC motor to shear the upper surface at a constant velocity of \( \pm 20 \mu \text{m/s} \). The spherical disk slid a distance of 200 \( \mu \text{m} \) forwards.
and backwards in the direction perpendicular to the long or short edges of the fabricated flaps. Friction tests were repeated eight to ten times at the same spot. When the sliding concluded, the adhesion after sliding was measured during the separation of two surfaces.

C. Results and Discussion

1. Friction and Adhesion of Vertical Micro-Flaps in SFA Tests

The vertical PDMS micro-flaps (Figure III – 4(a)) exhibit strong adhesion against the glass surface, as shown in Figure III – 4(b). During testing the vertical pillars were compressed axially until they either buckle or are slightly bent over due to the curvature of the sphere. Upon separation, strong adhesion was evident by the rapid jump of the output voltage of the normal load sensor in the SFA setup. No adhesion was measured with a zero preload, which is due to the geometry of our experimental setup: the flaps need to deform elastically in order to make a larger contact area with the upper sphere disk, and a finite preload is therefore required.

As shown in Figure III – 4(b), at low normal preloads ($F_p < 5 \text{ mN}$), the adhesion force increases rapidly with increasing load – a larger $F_p$ gives rise to a larger elastic deformation of the PDMS flaps, resulting in a higher number of contacts as well as larger contact area of a single contact and therefore greater adhesion. The increase in adhesion continues for higher loads, but becomes much less rapid. For a spherical puck in contact with a fibrillar surface, saturation in adhesion force at high preloads
has been previously reported from experiments by others [73] and modeled [91] by assuming that the individual fibers behave as linear springs.

**Figure III - 4: Adhesion forces and friction forces of the vertical (symmetric, not tilted) PDMS flaps.** (a) Schematic of a single vertical flap under a normal load and shear force. (b) Adhesion forces, $F_{ad}$ of the vertical flaps as a function of the preload force $F_p$. (c) Friction forces in both the ±x and ±y directions as functions of the real normal load. The vertical flaps exhibit isotropic friction in both the ±x and ±y directions, but these are different (anisotropic) in the x- versus the (tilted) y-directions. The friction is purely ‘load-controlled’ in the ±x directions (along the long flap edges), while in the ±y directions the friction is ‘adhesion-controlled’ at low loads (< 15 mN), becoming more load-controlled at higher loads.

Strong anisotropic frictional behavior was observed while sliding the upper glass disk along different directions of the vertical flaps. Sliding in the ±x direction, perpendicular to the smaller face, shows only load-controlled friction where the friction force is directly proportional to the normal load with a friction coefficient of
While sliding towards the large edge of flaps, in the $\pm y$ direction, friction force $F_{\parallel}$ is adhesion-controlled at loads less than 15 mN, indicated by the nonlinear relationship between $F_{\parallel}$ and the normal load $F_{\perp}$. When $F_{\perp} > 15$ mN, a linear dependence of $F_{\parallel}$ on $F_{\perp}$ is measured with a load controlled friction coefficient of 4.0. Higher $F_{\parallel}$ was measured when sliding along the y direction at low $F_{\perp}$, which also demonstrates the adhesion contribution on the friction. Since preload is required to generate adhesion, as indicated in the adhesion test, no $F_{\parallel}$ is measured when $F_{\perp} = 0$.

The width of the micro-flaps is about 2.5 times larger than the thickness and therefore the stiffness along the smaller face of the flaps is much higher. The SFA results demonstrate that this anisotropic stiffness of the PDMS micro-flaps in different directions is crucial to both the friction and adhesion properties of the vertical flaps. For a small deflection, assuming that deformation is governed by small-deflection cantilever bending, the stiffness of a vertical flap is:

$$k = \frac{3EI}{L^3} \quad (III - 2)$$

Where $E = 1.8$ MPa is the elastic modulus of the crosslinked PDMS [92], $L = 15$ $\mu$m is the height of the flap, and $I$ is the area moment of inertia. $I$ can be calculated by:

$$I = \frac{bh^3}{12} \quad (III - 3)$$
Where \( b \) is the width along the bending direction and \( h \) is the thickness in the bending direction. Using the size of the vertical PDMS flaps fabricated, we get \( I_x = 0.333 \times 10^{-21} \text{ m}^4 \), and \( I_y = 0.053 \times 10^{-21} \text{ m}^4 \). The stiffness of the vertical flaps is proportional to the area moment of inertia, \( k \propto I \). A single vertical flap is therefore 6 times stiffer in the \( \pm x \) direction than in the \( \pm y \) direction. Due to this higher stiffness, the flaps are harder to deform elastically in the \( \pm x \) direction. The high stiffness prevents a large contact area between the spherical glass disk and the micro-flaps, and therefore eliminates strong adhesion between the surfaces, which would explain the pure load-controlled behavior of the friction when sliding along the \( \pm x \) direction (Figure III – 4(c)). For sliding along the \( \pm y \) axis, the smaller stiffness along the large face makes the flaps easier to bend over during sliding, which gives rise to a larger contact area and thus causes the adhesion-controlled friction seen at low normal loads.

2. Friction and Adhesion of Angled Micro-Flaps in SFA Tests

The angled flaps tested in the SFA are \(~10 \mu\text{m} \) in height and are tilted by about \( 20^\circ \) from the vertical (Figure III – 2(b)). Highly anisotropic frictional behavior was observed during the friction tests performed. As for the vertical flaps, the friction of the angled flaps sliding along the \( \pm x \) direction is again mainly load-controlled (Figure III – 5(b)). \( F_\parallel \) is proportional to \( F_\perp \) with a friction coefficient of \(~3\). The large width of the flaps gives high bending stiffness and therefore a small contact area during sliding, which limits the adhesion between the flaps and the glass disk. The
SEM image also shows that the top surface of the flaps is very rough, which further eliminates the real contact area and the adhesion. On the other hand, strong adhesion-controlled friction was evident while moving the glass surface in the forward $y$ direction, $+y$ direction. The tilt angle makes the flaps easier to bend over elastically along the $+y$ direction while sliding with the angle of tilt. This elastic deformation results in a larger contact area, therefore resulting in stronger adhesion. Because of the adhesion contribution to the total friction force, sliding along the $+y$ direction had higher $F_{||}$ at low $F_{\perp}$. However at high $F_{\perp}$ ($F_{\perp} > 10$ mN), the sliding friction in the $x$ direction increases faster than in the $+y$ direction (Figure III – 5(b)). The easier bend over of the flaps in the $+y$ direction leads to a smaller resistance while sliding, resulting in a lower friction coefficient, $\mu_{+y}$, than the friction coefficient, $\mu_x$, in the $\pm x$ direction. At low load, although sliding along the $\pm x$ direction gives a higher friction coefficient, the load contribution is small, and the adhesion controlled friction dominates the total friction force, which results in an overall higher friction force for the $+y$ direction sliding. At high $F_{\perp}$, however, the load contribution wins out over the adhesion contribution, $\mu_x F_{\perp} > \mu_{+y} F_{\perp} + S_c A_{\text{real}}$, and therefore the friction force in the $\pm x$ direction is higher in this regime. The friction force in the $y$ direction is also very anisotropic: sliding along the $+y$ direction gives a much higher friction force than sliding in the $-y$ direction (backward direction) under the same preload (Figure III – 5(b)). Similar behavior was also found in friction tests on isolated gecko setal arrays [17].
Figure III - 5: (a) Schematic of a single angled micro-flap. The block arrows show the normal load and the shear force in the ±\(y\) directions as functions of the load. (b) The friction forces of the angled flaps when sliding in the ±\(x\) and ±\(y\) directions. (c) Sliding along the ±\(x\) directions gives almost a constant but weak adhesion regardless of the preload. In the \(y\) direction, the tilt angle strongly affects the adhesion: sliding along the +\(y\) direction enhances the adhesion force, while moving in the –\(y\) direction gives the lowest adhesion values, even lower than in the \(x\) direction.
Unlike the vertical flaps, which showed strong adhesion after applied normal loads without sliding, the angled flaps did not show strong adhesion in the stationary adhesion test. The adhesion force was very low and never exceeded 0.1 mN for preloads between 0.8 and 6.5 mN. The small adhesion in the stationary adhesion test could be due to the effect of surface roughness. SEM images show that the top edge of the flaps is very rough. Although it is not possible to estimate exactly the roughness of the top edge, the SEM images clearly show asperities on the order of hundreds of nanometers. This surface roughness could eliminate the real contact area. Although the real adhesion probably slightly increases with increasing preload, the increase is very small and could not be measured by the experimental setup used.

Sliding of the two surfaces, meanwhile, gave much stronger adhesion forces. To test the effect of sliding, the upper glass disk was first slid back and forth for 8 cycles, and the friction force was measured during the sliding. After stopping the sliding of the upper disk, the two surfaces were separated slowly by a motor and the adhesion force was recorded. The results show that the adhesion strongly depends on the sliding direction (Figure III – 5(c)). The adhesion force measured after sliding along the ±x direction increases slightly with increasing normal preload $F_p$ and then levels off at high $F_p$ with a maximum adhesion of ~0.7 mN. There is no directional dependence while sliding in the ±x direction: stopping sliding along either the forward direction (+x direction) or the backward direction (−x direction) gave the same adhesion force under the same load. Since in the ±x direction the flaps are not angled, this isotropic behavior is expected. On the other hand, sliding in the ±y
direction, the adhesion force exhibits strong directional dependence. Sliding in the +y direction significantly enhanced the adhesion force even at low preloads. Sliding forward at a small preload of $F_p = 1.7$ mN already gave an adhesion force of ~0.72 mN, a value that is even higher than the largest adhesion values measured at higher loads for both the cases of no sliding and of sliding along the ±x direction. Under higher loads, sliding forward resulted in even higher adhesion before the force reaches a plateau at around 1.6 mN (Figure III – 5(c)). However, only weak adhesion forces (less than 0.4 mN) were measured during the separation after sliding in the –y direction, and the forces were almost a constant for the different loads tested.

The role of the tilted angle has been explained by Autumn et al. [22]. The tilted angle gives rise to an anisotropic stiffness of the flaps. The displacement of an angled flap with a tilted angle $\varphi$ under a normal load $F_\perp$ and a shear force $F_\parallel$ could be estimated by:

$$\Delta_{\text{approx}} = \frac{L_3^2}{3EI} \left[ F_\perp \cos^2(\varphi) \pm F_\parallel \sin(\varphi) \cos(\varphi) \right]$$  \hspace{1cm} (III – 4)

The stiffness of the flaps is therefore:

$$k = \frac{F_\parallel}{\Delta_{\text{approx}}} = \frac{F_\parallel}{\frac{L_3^2}{3EI} \left[ F_\perp \cos^2(\varphi) \pm F_\parallel \sin(\varphi) \cos(\varphi) \right]}$$  \hspace{1cm} (III – 5)

In the above equations, take the positive sign if sheared in the direction of tilt (the +y direction), and negative if sheared in the opposite direction (the –y direction). This simple model predicts the anisotropic stiffness of the angled flaps: sliding against the
direction of tilt gives higher stiffness, while sliding in the tilt direction results in lower stiffness.

The direction dependent stiffness indicated by Equation III – 5 strongly affects the adhesion properties of the flaps (Figure III – 6). For only the normal adhesion test without shearing, due to the top surface roughness of the flaps, the flaps only make point contacts with the glass surface. The small contact area results in small adhesion that is insensitive to preload. This observation also agrees with previous SFA experiments on randomly rough surfaces, which demonstrated that the adhesion between a randomly rough surface and a molecularly smooth bare mica surface decreased exponentially with increasing roughness and the adhesion did not increase significantly with increasing loads at high root mean square roughness (RMS) [70, 93].

During testing with sliding in the +y direction, the flaps, pre-angled in the fabrication process, bend down easily as they slide over the glass surface, leading to a large contact area. This increase of contact area can give rise to both high adhesion and friction. In the SFA tests, this increase of contact area was observed to lead to a two to eight fold increase of adhesion over the pure adhesion test result. At low loads, strong adhesion causes adhesion-controlled friction, which gives much higher friction forces. While sliding in the –y direction, the flaps are sheared backward. The tilted angle of the flaps prevents large bending over to the backward direction, and
therefore the flaps are pulled away from the surface, leading to a smaller contact area and smaller adhesion and friction.

**Figure III - 6: Schematic of the frictional adhesion mechanism of the angled flaps.** (a) The glass surface makes only point contact with the flaps due to the surface roughness of the flaps when only a normal load is applied. (b) Sliding in the gripping direction (the \( +y \) direction) causes the flaps to bend over. The flaps are pulled towards the glass surface, resulting in a large contact area and high adhesion. (c) Sliding in the releasing direction (the \( -y \) direction), the flaps are pulled away from the glass surface.

The magnitude of the adhesion force obtained with the angled flaps after sliding can be understood semi-quantitatively with the JKR theory. In SFA tests with a camera attached to the microscope, videos of the vertical unloading/retraction phase of the load/slide/retract test sequence employed showed a sudden final jump away from contact at an apparent contact area of \( \sim 0.3 \text{ mm}^2 \) for previous sliding in both the \(+y\) and \(-y\) directions. For an angled flap areal density of \( \sigma = 1847.6 \text{ mm}^{-2} \), this results in approximately 550 flaps in contact just before the jump. As seen in Figure III – 5(c), the adhesion force at this jump is 1.8 mN after sliding in the \(+y\) direction, and
~0.3 mN after sliding in the $-y$ direction. Thus, the contributing force per flap is ~3.3 µN after $+y$ sliding, and ~0.6 µN after $-y$ sliding. As previously stated in Equation (I – 7), the JKR theory states that for adhesion energy $W$ between contacting surfaces, and a sphere of radius $R$, the magnitude of this adhesion force jump, $F_{JKR}$, should be given by: $F_{JKR} = (3/2)\pi RW$. Assuming each contacting flap to be approximated by a sphere, and $W = 0.1$ Jm$^{-2}$ for the PDMS-glass interface [94], this results in an effective radius, $R_{+y}$, of 7.0 µm after sliding with tilt in the $+y$ direction, and $R_{-y} = 1.3$ µm after sliding against tilt in the $-y$ direction. This has been illustrated in Figure III – 7, where it can be seen that sliding with tilt is expected to result in a surface with a larger effective radius (due to the large deflection predicted by Equation (III – 4)) than sliding against tilt.

**Figure III - 7:** Schematic showing a single angled PDMS flap after sliding in the $+y$ direction (a), and after sliding in the $-y$ direction (b). The anisotropic adhesion forces obtained may be understood using the JKR theory based on the different effective radii of the flaps after sliding in the two directions.
Frictional adhesion is the unique mechanism applied by the gecko to achieve its extraordinary climbing ability [20]. In many robotic applications, the problem is making adhesives that can rapidly detach from the surface rather than simply strongly adhering to the surface. By careful design of the geometrical and mechanical properties of the micro-flaps, the same mechanism could be designed and optimized with artificial dry adhesives.

However, based on the current design, the real area in contact between the puck and the angled flaps has a fill factor of just ~18% even at full flap deflection with the entire flap face in contact with the puck. Adhesion and friction forces achieved could be considerably increased from the values obtained so far by using a mask design with higher fill factor. SFA tests on angled micro-flap arrays with a significantly higher fill factor of ~65% are ongoing.

**D. Conclusion**

The experimental results presented in this chapter demonstrate how surfaces with directionally oriented or tilted structures can generate anisotropic adhesion and friction when articulated in a specific manner – the tilted micro-flaps give rise to both strong friction and adhesion forces when sheared along the tilt direction, and low friction and adhesion forces when sheared against the tilt direction. This also appears to be the method employed by geckos allowing them to run on (vertical) walls and (horizontal) ceilings: the anisotropic, directionally tilted setae provide both strong adhesion and friction for attachment (in the gripping stage) and weak
adhesion and friction for detachment (in the releasing stage) during a step cycle. The anisotropy built into the micro-flap design allows for controllability and switchability by varying the flap dimensions, density, tilt angle, and shearing direction. These results may be the first step towards fabricating optimized gecko-mimetic feet structures for responsive adhesives suitable for climbing robotics applications.
IV. Millimeter Size Patch Behavior of a Gecko-Inspired Reversible Adhesive

A. Introduction

A synthetic adhesive inspired by the gecko fibrillar system should replicate the important properties found on the animal, but does not need to be a direct copy of its system. Rectangular polymer micro-flap arrays have been chosen in order to create large areas of contact on the face of the structures after shearing, similar to the animal’s terminal structures. Additionally, an angle of tilt has been incorporated into the fiber design in order to generate anisotropy in the system based on lateral shearing direction. Small scale tests, with contact areas of $0.1 - 1 \text{ mm}^2$, using the surface force apparatus (SFA) have been previously performed with a glass sphere contacting the flap structures [67]. Results from these tests have also been described in detail in Chapter III. The encouraging results necessitated further tests on larger patch sizes since small test areas can overestimate the forces achieved when patch sizes are scaled up. This discrepancy, as was shown between single setae [11] and whole feet [10] force measurements for the gecko, and for PDMS micro-wedge synthetic adhesives of various patch sizes [68], can be substantial and are likely due to incomplete or poor attachment at large testing sizes. To obtain insight into the performance of the adhesive at larger patch size scales and help more fully evaluate its performance for future possible real world applications, a custom-built tribometer, called the ‘Bio-Force’ or ‘Bio-F’ apparatus, with a larger test area was used to investigate the adhesion and friction properties of the material.
This chapter describes the adhesion and friction force testing of micron-sized vertical and tilted rectangular PDMS flaps over a patch area of \(~12.6 \text{ mm}^2\)\), i.e. \(~12\) times larger than patch sizes previously tested with an SFA. The Bio-F apparatus utilizes a flat-on-flat testing geometry to characterize adhesive performance against a glass puck that is first carefully aligned with both the sample and the direction of sliding. The results obtained are compared with and shown to be in broad agreement with those from the SFA, and show that the micro-flap adhesive system continues to successfully mimic key characteristics of the gecko adhesive system when patch size is significantly scaled up. Further, the experimental results for adhesion after shearing were found to approximate theoretical estimates from a Kendall Peel Model.

\textbf{B. Experimental Methods}

1. Microfabrication of Vertical and Angled Micro-Flaps

Fabrication of the microstructures is similar to that previously carried out for tests in the surface forces apparatus [67], and is described in detail in Chapter II. For the fabrication of the angled flaps, refinement to the shape of the flaps was achieved by splitting the development time of the polymethylglutarimide (PMGI) into a larger number of equal parts, eight instead of six as used initially for the flaps tested in Chapter III. SEM images of the flaps can be seen in Figure IV – 1. The vertical flaps are 10 \(\mu\text{m}\) wide, 4 \(\mu\text{m}\) thick, and 15 \(\mu\text{m}\) tall while the angled flaps are tilted in the y-
direction at an angle of ~20° from the vertical and are 10 µm wide, 3.6 µm thick, and 10.7 µm tall.

Figure IV - 1: SEM images of vertical rectangular PDMS micro-flaps (a) and angled rectangular PDMS micro-flaps tilted at 20° from the vertical (b). The vertical flaps are 10 µm wide, 4 µm thick, and 15 µm in height; the angled flaps are 10 µm wide, 3.6 µm thick, and 10.7 µm in height. The coordinate system used throughout this chapter is indicated. For both sets of flaps, the –z direction is the vertical loading direction, and the +z direction is the vertical unloading direction. The ±x axis is parallel to the 10 µm long edge of the flaps, and the ±y axis is perpendicular to this edge. For the angled flaps, the +y direction points in the direction of tilt, while the –y direction points against the tilt.

2. Adhesion and Friction Testing in the Bio-F Apparatus

Small scale tests often report high friction and adhesion forces for natural and synthetic adhesive systems that are often not attainable in large scale tests because of
the inability of all the micro/nano fibrillar structures to make sufficient contact with test substrates over larger meso-scale areas. The use of spherical test pucks is common for easier alignment of the puck with the adhesive surface [39, 64, 67, 73, 75, 95]. For short micro-flap structures with a total height of approximately 10 µm, however, the limited vertical travel of the surfaces relative to each other limits the area of the substrate in contact during testing with a sphere on flat geometry. For example, the area tested in the SFA using a spherical glass puck of radius 2 cm is of the order of 0.1 – 1 mm². Also, the loading of microstructures contacting the puck in different regions is non-uniform in such a setup.

Figure IV - 2: Schematic diagrams showing the sphere-on-flat testing configuration used in an SFA (a) and the flat-on-flat configuration used in the Bio-F (b). The vertical PDMS flaps have not been drawn to scale. Patch sizes tested with the Bio-F are ~12 – 126 times larger than those tested with the SFA. Both the SFA and the Bio-F are capable of performing both normal and lateral relative motions of the test probe and adhesive samples in order to perform load/drag/pull tests to characterize material adhesion and friction properties.
Figure IV - 3: (a) Bio-F adhesion and friction test apparatus designed and built in the Turner Lab. (b) Details and relative placement of the various tilt adjustment stages, motion control stages, force sensor, sample and microscope objective to view samples tested through the transparent glass puck test surface.

Testing over larger patch areas was performed using a custom-built tribometer with a four millimeter diameter flat glass puck as the opposing surface. The Bio-F’s test area is therefore around 12.6 mm$^2$, between 12 and 126 times greater than the test area in the SFA. The schematic in Figure IV – 2 shows a comparison of the
testing geometry. Further, the test patch area value is roughly constant for the tests shown in this section of the report because the testing geometry is flat-on-flat where changes in load do not change the apparent contact area.

Details of the Bio-F apparatus are shown in Figure IV – 3. Movement of the sample is generated by two motorized linear stages (Newport Corporation, Irvine, CA) with incremental movements of 100 nm in the vertical direction and 500 nm in the horizontal direction. Force sensing is provided by a load cell (ATI Industrial, Apex, NC) with a force resolution of approximately 3 mN. A rotational stage (Newport Corporation, Irvine, CA) was used to align the direction of in-plane motion parallel to the small vertical face of the flap. Two goniometers (Newport Corporation, Irvine, CA) oriented the sample parallel to the plane of movement in order to minimize force differences during shearing. A three-point leveling mechanism allowed the glass puck to be aligned to the sample, ensuring even force distribution during loading. The sample was viewed during testing by looking through the glass puck onto the top of the sample using a CCD camera attached to a high magnification zoom lens (Navitar Inc., Rochester, NY). Control and data acquisition software was all written in LabVIEW (National Instruments Corporation, Austin, TX) with one program allowing images for later processing to be obtained using an image acquisition board (National Instruments Corporation, Austin, TX).

Position, force, and torque data was recorded during the two types of tests performed on the samples. The adhesion force generated is obtained using a load/pull test. In this test, the sample is moved towards the glass surface at a vertical
speed of 1 µm/s until a given compressive load, the preload, has been reached. The adhesive sample is then moved away from the glass at the same speed until separation has occurred. The maximum tensile load supported by the sample is the adhesion force.

The second test investigates the material’s response to shear. Shear forces and the shear adhesion force are both obtained from a load/drag/pull test. Again, the sample is moved toward the glass puck at the same speed until the preload has been reached. Before the surfaces are separated, the sample is moved in the in-plane direction perpendicular to the large face of the flap at a horizontal speed of 3 µm/s. The positive direction will generate movement with the angle of the flap while the negative direction will be against the angle. Once the desired distance has been achieved, the sample will move away from the glass puck at a vertical speed of 1 µm/s. To separate adhesion forces generated with shearing, which can be quite different from those obtained during a load/pull test, the forces obtained with this additional movement are called shear adhesion forces, although they are calculated in the same manner. Shear forces are generated in-plane and the maximum values for differing lengths are given in the results section.

C. Results and Discussion

1. Tilt Angle Necessary for Anisotropic Adhesion and Friction

Testing on the structures revealed remarkably different results between the vertical and angled designs. As can be seen in Figure IV – 4, the vertical flaps are
able to create roughly 60 mN of adhesive force above preloads of 85 mN. Between 25 and 60 mN preloads, maximum contact area has yet to be achieved across the entire puck, as can be seen by the increasing adhesion values. Below 25 mN preloads, only small areas of contact have been created which result in negligible adhesion values.

Figure IV - 4: Adhesion test performed on vertical flaps and angled flaps. The angled flaps exhibit little adhesion without shear due to the tilt and the roughness asperities on the top surface which result in poor contact with the test surface.

One would expect that the adhesion values would plateau once full adhesion has been achieved; however, the values start to fall above preloads of 193 mN. This slight drop with increasing preloads can be explained as a result of overloading and
buckling of the flaps which produces a change in orientation over a portion of the flaps on the sample. This could result in a decrease in contact area from the flat top face of the flaps to just the top edge, accounting for the drop in adhesion observed. Reorientation due to buckling is supported by the experimental observation that during separation, there is another reorientation of the flaps, evident by an increase in compressive force, and then a final pull off event (adhesion) while in tension. It is believed that the reorientation during loading is responsible for the second orientation change observed during retraction, and the loss of contact area with the puck results in the material being able to support less tensile force.

The minimum compressive load needed to induce a buckling instability in the vertical PDMS micro-flaps may be verified using Euler’s buckling formula for a column with one end clamped (attached to the PDMS base), and the other end pinned and thereby free to rotate (this rotation allowing in a change in orientation of the contact with the puck from the top face to just the top edge) while restricting lateral motion at the point of application of the force. Approximating each vertical flap to be a beam with clamped-hinged end conditions, the critical buckling load per micro-flap, $P_{\text{crit}}$, is then given by [96]:

$$ P_{\text{crit}} = \frac{2.04\pi^2EI}{L^2} \quad (\text{IV} - 1) $$

Where the Young’s modulus of PDMS, $E = 1.8 \text{ MPa}$ [92]; $I = \text{area moment of inertia of the micro-flap rectangular cross section} = 53.33 \times 10^{-24} \text{ m}^4$, as previously
calculated in Chapter III using Equation (III – 3); and vertical micro-flap height, \( L = 15 \ \mu \text{m} \). From Equation IV – 1, the critical buckling load per flap, \( P_{\text{crit}} = 8.59 \ \mu \text{N} \).

For an areal density of ~3694 flaps/mm\(^2\) and a puck area of ~12.6 mm\(^2\), the total number of flaps in contact, \( N \sim 46,500 \). Hence, the critical buckling load for the entire patch = \( N \times P_{\text{crit}} = 399 \ \text{mN} \). The experimentally observed value of 193 mN is of the same order of magnitude, although considerably lower. A major reason for this discrepancy could be that the value of \( E \) used for PDMS, chosen from the literature, may not match the material used in these experiments. Also, the glass puck was found by optical interferometry (Wyko NT1100 Optical Profiling System) to have a surface RMS roughness ~160 nm. This roughness could have resulted in non-uniform contact and loading of the flaps, with some fraction of the flaps reaching a critical buckling load before others. This could explain both the lower critical buckling load observed experimentally as well as the gradual drop in adhesion at high preloads due to reduction in contact area over only some fraction of the micro-flap array.

The vertical flaps were tested for durability by repeating the load/pull test 50 times without any intermediate cleaning, at a preload of 80 mN. The flaps retained up to 97% of the initial adhesion value after 50 tests, as shown in Figure IV – 5.
Figure IV - 5: The vertical PDMS flaps were subjected to 50 continuous load/pull test cycles at the same patch location and without intermediate cleaning in order to gauge durability. Up to 97% of the initial adhesion force value, $F_{ad}$, was retained after 50 cycles.

The angled flaps do not respond in the same manner to load/pull tests. Preloads under 200 mN did not produce significant amounts of adhesion, as shown in Figure IV – 4. Only when the force was above this value and flap contact area was increased, were small values observed. Surface roughness on the top of the flap, in addition to tilt, is likely responsible for the small adhesion values seen during load/pull tests. Previous tests in the SFA against a spherical glass puck also showed no adhesion of angled flaps for simple load/pull tests without any shearing articulation motion (Chapter III, Section C – 2). While no adhesion may appear to be a poor result, it is in fact one of the key properties identified by Kellar Autumn for a gecko-like adhesive [9]. This non-sticky default state at small preloads allows for the possibility to control the adhesive and only stick when desired. Decreases in
expend energy to detach and increased robotic stability during foot removal are two prominent advantages of this property.

To discern the effect of shearing on the vertical flaps, they were tested at preloads representative of partial and full contact, 45 and 80 mN, as can be seen in Figure IV – 6(a). It was believed that shearing would cause a contact area increase with the glass puck, from the flap tops in the vertical orientation to the large face after the flap has been bent over. At zero shear length, the test is exactly the same as the load/pull test and adhesion values should be, and are in fact, very similar. As the shear lengths increase, the flaps are bent until at a shear length around 12.5 μm, only the edge of the top face of the flap is in contact and small loads can be supported. Contact then increases on the large face until a maximum contact area is reached at a shear length of approximately 90 μm. In both preload cases, the highest shear adhesion force achieved in either direction is higher than the pure adhesion force at zero shear length. In the positive direction, the highest value is 1.4 and 3.3 times the value created without any shear for 80 and 45 mN preloads. The force increases are due to higher contact areas as can be seen in optical microscope images obtained during shearing.

After the maximum, the adhesion forces supported slowly fall to a steady state value as flaps gain and lose contact with the glass surface. For shear lengths much greater than the individual flap dimensions, it is likely that shearing results in a loss of contact due to slipping of some flaps. The results should be symmetric for both
directions (as previously seen in SFA tests on these flaps in Chapter III), and small misalignments are likely responsible for the differences.

Figure IV - 6: (a) Shear adhesion tests performed on the vertical flaps at preloads of 45 and 80 mN. Shearing the flaps in either direction increases the shear adhesion force due to higher contact area. (b) Shear adhesion tests performed on the angled flaps at preloads of 40, 60, and 100 mN. Only shearing with the flap tilt direction (+y) resulted in high shear adhesion forces.
The same shear adhesion test was then performed on flaps with a 20° angle from the vertical to discover other characteristics of tilt addition. The results, as can be seen in Fig. IV – 6(b), display anisotropic shear adhesion properties depending on shear direction. When sheared against the angle of tilt (in the \(-y\) direction), small areas of contact are created as the trailing edge of the flap is dragged across the glass puck. When sheared with the angle of tilt (in the \(+y\) direction), increased contact area on the leading face of the flap results in significant shear adhesion forces. The maximum shear adhesion values obtained, 46 mN, 42 mN and 28 mN for preloads of 100 mN, 60 mN, and 40 mN, may appear to be small based on the vertical flap values of 86 mN and 53 mN for preloads of 80 mN and 45 mN, but differences in flap areal density, not geometry, are likely responsible for this discrepancy. The vertical flaps have twice the areal density due to differences in fabrication, and also have a larger face available for contact.

A theoretical estimate for the shear adhesion forces expected from both vertical and tilted PDMS flaps can be obtained by considering the Kendall peel model for the force \(F_{\text{peel}}\) required to peel a thin elastic adhesive film from a surface [45]:

\[
F_{\text{peel}} = bdE \left( \cos \theta_{\text{peel}} - 1 + \sqrt{\cos \theta_{\text{peel}}^2 - 2 \cos \theta_{\text{peel}} + 1 + \frac{2w}{dE}} \right) \quad (IV - 2)
\]

Where \(\theta_{\text{peel}}\) is the angle of peel between the film and the substrate, \(w\) is the adhesive energy for the contact, \(b\) is the contact width of the film, \(d\) is the thickness of the film, and \(E\) is the Young’s modulus of the film material. For the Sylgard 184 formulation of PDMS (1:10 crosslinker to base elastomer ratio by weight), \(E = 1.8\)
MPa [92], and the adhesive energy of the contact for PDMS in contact with glass, \( w = 100 \text{ mJ/m}^2 \) [94]. To estimate the adhesion force from the peel force, only the normal component is considered. Also, Equation IV – 2 gives an estimate of the peel force per flap, and would need to be multiplied by the maximum number of flaps \( N \) in contact with the puck before comparisons with experimental data can be made.

In the absence of a mechanism to directly observe the peel angle \( \theta_{\text{peel}} \) for the flaps, we must take an estimate to fit the experimental data. For the vertical flaps, \( b = 10 \mu\text{m} \), and \( d = 4 \mu\text{m} \), as observed in SEM images. Also, \( N \approx 46,500 \). Then, using Equation IV – 2, the peel angle required to match the experimental adhesion value of 53 mN for the 45 mN preload case is 82°, and the peel angle required to match the adhesion value of 86 mN for the 80 mN preload case is 55°. A decrease in peel angle at higher preloads as indicated by this simple model may be expected since the flaps are bent over further at high preloads.

This decrease in peel angle – leading to higher peel forces – is also predicted by the model for the shear adhesion of angled flaps. The number of flaps in contact is half the number for vertical flaps, \( N/2 \). For preloads of 40, 60, and 100 mN, and for shearing in the \(+y\) direction, the peel angles to fit the maximum experimental values obtained are 79°, 56°, and 45° respectively. For shearing in the \(-y\) direction, a low adhesion force of 5 mN can be modeled assuming a peel angle of 156°.

It should also be noted that there is adhesion at shear lengths of zero for the later tests of angled flaps. Over 240 tests had been performed before adhesion was seen for a preload of 100 mN, and 300 tests performed before adhesion was obtained for a
preload of 40 mN. It is possible that over this time there had been a change in tilt angle of the flaps. Supporting this potentiality, inspection was performed after testing was finished (i.e. after approximately 1600 tests), and flaps were observed to be stuck to the PDMS base in certain areas.

Shear tests performed on both sets of flaps, shown in Figure IV – 7, display additional advantages of tilted structure. At positive shear lengths, i.e. the gripping direction, for the distances shown, the angled flaps are able to support higher shear forces despite having half the density of flaps. In the negative direction, the angled flaps still outperform the vertical ones at shear lengths less than 30 µm and differences at higher lengths are not overly pronounced for comparable preloads. Following the trends to longer shear lengths, vertical flaps should be capable of providing higher force in both directions, but they lack a simple release mechanism. Although trying to minimize contact by shearing until an edge is in contact would be possible as discussed in [68], the simplicity of shearing against the tilt direction to a range of shear lengths enhances the viability of using angled rectangular flaps. It can also be seen in Figure IV – 7 that at small shear lengths the tilt addition results in anisotropic shear behavior for improved controllability.
2. Comparison with Results from SFA Tests

A comparison of results from tests on both the SFA and Bio-F apparatuses shows good qualitative agreement for several adhesion properties critical to gecko-inspired adhesives, as shown in Table IV – 1. In both test apparatuses, the vertical PDMS flaps adhered strongly after shearing, but showed no anisotropy based on shearing direction. For the angled PDMS flaps, highly anisotropic adhesion was measured in both systems after shearing along the ±y axis. The non-sticky default state of the angled flaps was maintained in tests on both the SFA and Bio-F testers. This property is crucial for increased controllability, and enables adhesion to be switched
on only when needed, based on a specific shearing articulation scheme (in this case, shearing along the ±y axis). Tests against a flat glass puck in the Bio-F apparatus tended to result in higher µ’ values than against a spherical glass puck in the SFA, likely due to more uniform loading against a flat substrate surface leading to both a reduction in preload required for good contact as well as higher pull-off adhesion force.

**Table IV - 1: A comparison of test results for normal adhesion from both SFA and Bio-F tests on vertical and angled micro-flap arrays.**

<table>
<thead>
<tr>
<th></th>
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<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Vertical PDMS Flaps</td>
<td>3.2 (SFA)</td>
<td>0.23 (SFA)</td>
<td>No (SFA)</td>
<td>No (SFA)</td>
<td>No (SFA)</td>
<td>&gt;8 (SFA)</td>
</tr>
<tr>
<td>Angled PDMS Flaps</td>
<td>1.8 (SFA)</td>
<td>0.36 (SFA)</td>
<td>Yes (SFA)</td>
<td>Yes* (SFA)</td>
<td>Yes (SFA)</td>
<td>&gt;8 (SFA)</td>
</tr>
</tbody>
</table>

* Low detachment force of 0.25 kPa measured in SFA after sliding in releasing direction.

A comparison of results from tests on both the SFA and Bio-F apparatuses also shows good qualitative agreement for friction properties, as shown in Table IV – 2. Although the shear stresses in both the gripping (‘forward’) and releasing
('backward') directions are significantly higher in SFA tests than in Bio-F tests, both confirm the anisotropy in friction forces that may be obtained with vertical PDMS flaps and flaps tilted at 20º from the vertical. Higher friction/shear forces during sliding in SFA tests may be understood from the fact that these forces are directly proportional to the real area in contact [56]. In small-scale patch tests such as the SFA sphere-on-flat geometry, it is possible to obtain better contact between the flaps and the substrate than in larger scale Bio-F tests. This results in higher shear stresses for SFA tests when these forces are divided by the apparent area of contact. Although the vertical PDMS flaps show a large (order of magnitude) difference in the shear forces per unit patch area that may be obtained when patch sizes are scaled up, the angled flaps show a much smaller ~50% reduction in shear stress, indicating good operation for potential real-world applications.

**Table IV - 2: A comparison of test results for shear forces along various axes from both SFA and Bio-F tests on vertical and angled micro-flap arrays.**

<table>
<thead>
<tr>
<th>Adhesive System</th>
<th>Anisotropic Friction?</th>
<th>Forward Shear Stress (kPa)</th>
<th>Backward Shear Stress (kPa)</th>
<th>Anisotropy Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td><em>Vertical PDMS Flaps</em></td>
<td>No* (SFA)</td>
<td>*365 [+Y], 275 [+X] (SFA)</td>
<td>*365 [−Y], 275 [−X] (SFA)</td>
<td>1.0 (SFA)</td>
</tr>
<tr>
<td></td>
<td>No (Bio-F)</td>
<td>16 [+Y] (Bio-F)</td>
<td>16 [−Y] (Bio-F)</td>
<td>1.0 (Bio-F)</td>
</tr>
<tr>
<td><em>Angled PDMS Flaps</em></td>
<td>Yes (SFA)</td>
<td>45 [+Y] (SFA)</td>
<td>28 [−Y] (SFA)</td>
<td>1.6 (SFA)</td>
</tr>
<tr>
<td></td>
<td>Yes (Bio-F)</td>
<td>22 [+Y] (Bio-F)</td>
<td>14 [−Y] (Bio-F)</td>
<td>1.6 (Bio-F)</td>
</tr>
</tbody>
</table>
D. Conclusion

The results from Bio-F testing have shown that the angled micro flap structures are capable of achieving highly controllable adhesion and friction properties over mm-scale areas against a rough glass substrate surface, demonstrating both patch size scale up and some measure of roughness adaptation. Further, no loss of adhesion was observed after 50 pure adhesion test cycles when tested at the same location on the sample, demonstrating reusability for practical applications. However, further durability testing over several thousand test cycles is required to more fully characterize the adhesive system. Although the results for using asymmetric rectangular microstructures as synthetic gecko-like adhesives are encouraging, further scalability tests and improvements in flap geometry and density are required to increase the force values. The experimental results for shear adhesion forces are in broad agreement with both previous test results on smaller patch sizes in an SFA and with theoretical predictions from a Kendall Peel Model.
V. Friction and Adhesion of Gecko-Inspired PDMS Micro-Flaps on Rough Surfaces

A. Introduction

Surface roughness is an extremely important parameter that affects the friction and adhesion forces between two surfaces. In a natural environment, almost all surfaces are rough, usually containing different scales of roughness. Surface roughness can significantly reduce the real contact area of two surfaces, and therefore reduces the adhesion force between surfaces [70, 71]. Gecko hierarchical foot structure (Figure I – 1) – a gecko toe has tens of arrays (lamellae) of micron-scale hair (setae), with each seta splitting into 100-1000 nanoscale spatulae on the top – is used to overcome this problem. This hierarchical structure, by conforming to both micro- and nano-scale asperities, can cover most of the scales of the surface roughness that geckos encounter in nature, achieving a large true area of contact, so that geckos can adhere to different surfaces via the weak van der Waals force.

In developing gecko-inspired adhesives, much attention has been focused on fabricating different structured surfaces that mimic the hierarchical structure of gecko feet and their adhesion and friction properties on model smooth surfaces [29, 61, 62]; however, less attention has been paid to the effect of surface roughness, one of the most crucial parameters in the study of adhesion and friction. Tests on geckos and beetles have shown that surface roughness significantly affects the adhesion of
those animals on model substrates [97–99]. Both the geckos and beetles showed stronger adhesion on smooth surface. On rough substrates, both tests showed that minimum adhesion was measured at a certain intermediate value of roughness, while increasing or decreasing the roughness led to an increase in adhesion.

The effect of surface roughness on the adhesion of elastic plates has been mainly studied theoretically [2]. Various theoretical works using both continuum theory and molecular simulation have shown that increasing the roughness can significantly reduce the adhesion between two surfaces [100–102]. Molecular simulation studies have also shown that the surface topology is one of the most important parameters affecting sliding friction [103]. Anisotropic friction was observed when sliding a topologically anisotropic surface against a flat surface [104]. The effects of roughness on the adhesion and sliding friction between two polymeric surfaces of 1-10 nm roughness and between a rough surface and a smooth mica surface have been investigated using the surface forces apparatus, showing that the adhesion between two surfaces decreased exponentially with increasing surface roughness [93]. The SFA experiments clearly demonstrate the importance of surface roughness on the adhesion and sliding friction; however, the RMS roughness studied is very small (up to 10 nm) and significantly lower than the surface roughness of many practical applications. Previously, some work on characterizing synthetic bio-inspired adhesive surfaces has been reported. However, this work was seriously limited in that the rough substrate surface consisted of a regular pattern [29, 105]. When tests were reported against real world surfaces such as smooth granite and wood, the
variation in roughness values between surfaces was large and abrupt, and not fully characterized [106]. Thus, despite various theoretical attempts at modeling adhesion and friction on rough surfaces [107], systematic experimental investigations are lacking. A recent experimental study on the effect of nano and sub-micron scale substrate roughness focused only on the adhesion of a gecko-inspired adhesive, and friction was not studied [108]. Further, the vertical polymer fiber array adhesive tested was not structurally anisotropic with controllable adhesion and friction properties like the angled micro-flaps reported in the current study. Also, the fibers were not sheared and simply tested using a load/pull scheme – thus, it was not possible to ascertain if sliding a patterned microfiber array against a non-uniformly rough surface could significantly affect the adhesion pull-off force.

The lateral force or the friction force, $F_{\parallel}$ that both balances the body weight of the gecko on walls and contributes to normal adhesion via a resolved component along the gecko’s tilted fibers, is very important [5]. A general equation that describes the adhesive friction force between two surfaces is [31]:

$$F_{\parallel} = \mu F_{\perp} + S_c A_{\text{real}} , \quad \text{(V – 1)}$$

Where $\mu$ is the friction coefficient, $F_{\perp}$ is the normal load, $S_c$ is the shear strength, and $A_{\text{real}}$ is the real contact area. In Equation (V – 1), the first term is the simple Amontons’ law for a non-adhering surface, representing the load contribution to the total friction force. The second term gives the contribution of adhesion, which is proportional to the ‘real’ molecular contact area. This ‘real’ molecular contact area
rapidly decreases with increasing the surface roughness, and therefore the adhesion contribution is very small, almost negligible in many practical situations [109]. In many biological adhesive systems, however, this adhesion contribution is essential and dominates the friction force [2, 110].

Most of the surfaces that the geckos and gecko-inspired robots encounter have different topological surface profiles with various length scales of roughness. In order to better design gecko-inspired structures for real applications, understanding the performance of the fabricated structure on surfaces with different roughness is of extreme importance. In this study, gecko-inspired tilted PDMS flaps (Figure V – 1) were tested against a series of non-uniformly rough glass substrate surfaces (with increasing values of height RMS roughness) prepared by a plasma etching technique. Using a modified surface forces apparatus (SFA), adhesion and friction forces were measured between the microfabricated tilted PDMS flaps and both optically smooth and unetched glass as well as rough plasma-etched glass surfaces. On both smooth and rough substrate surfaces, anisotropic adhesion and friction forces were measured for sliding of tilted flaps with tilt (+\(\gamma\)) and against tilt (-\(\gamma\)). Increasing the surface roughness first increased the adhesion and friction forces measured between the flaps and the rough surface due to a topological match between the two surfaces, but then led to a rapid decrease in both of these forces. These results demonstrate that surface roughness significantly affects the performance of gecko-mimetic adhesives, and that different surface textures can either increase or decrease the adhesion and friction forces of the fabricated adhesives.
Figure V - 1: SEM Image (a) and schematic orthographic top-view (b) of the angled PDMS micro-flaps tested against smooth and rough glass surfaces. The 10 µm wide flaps are arranged in a regular hexagonal array of side $a = 25$ µm.
B. Experimental Methods

1. Plasma Etching and Inspection

In order to test the roughness adaptation of the angled micro-flap adhesive system, rough substrate surfaces that could be easily integrated into the SFA test apparatus were required. Smooth SFA glass pucks (Product No. NT63-476, Edmund Optics Inc., Barrington, NJ, USA) were etched in a Materials Research Corporation MRC 51 reactive ion etching system (Praxair Surface Technologies, Indianapolis, IN, USA) using a CF<sub>4</sub>/SF<sub>6</sub>/O<sub>2</sub>/Ar plasma to generate a non-uniform surface roughness pattern with maximum asperity heights lower than 3 µm. No cracks or line textures were observed on the pucks after etching. Process parameters for this etching process as well as results from other experimental trials with other surface roughening techniques are included in Chapter II, Section D.

Unetched and uncoated SFA disks were found by AFM (Tapping Mode) analysis to have a polished surface with an RMS roughness value of ~11 (± 8) nm. Surfaces of increasing RMS values of roughness were generated by exposing smooth SFA disks to the RIE plasma for increasing lengths of time (Table II – 1).
Figure V - 2: AFM and SEM images of the rough surfaces with different height RMS roughness. (a), (c), (e), (g), and (i): AFM images of the etched SFA glass surfaces with $11 \pm 8$ nm, $73 \pm 5$ nm, $133 \pm 20$ nm, $192 \pm 9$ nm, and $308 \pm 56$ nm RMS roughness. (b), (d), (f), (h), and (j): SEM images of the same surfaces.
Examination of the surface topography after etching using an optical microscope, SEM (XL-30, FEI Company, Hillsboro, OR), and AFM (MFP-3D SA, Asylum Research) revealed circular mesa-like structures. The RMS roughness of each etched surface was measured using tapping mode AFM scanning over various areas of the surface and a scan area of 50 µm by 50 µm. AFM images showed that the size and density of the asperities increase with increasing etching time, while the gap distance between two neighboring asperities becomes smaller with a longer etching time. Similar features were also confirmed by the SEM measurements (Figure V – 2).

**Table V - 1: Height RMS roughness values and average distance between asperities for both the smooth and rough substrate surfaces used in adhesion and friction testing.**

<table>
<thead>
<tr>
<th>Etching time (min)</th>
<th>RMS height roughness and standard deviation (nm)</th>
<th>Average distance between asperities and standard deviation (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>11 ± 8</td>
<td>N/A</td>
</tr>
<tr>
<td>35</td>
<td>73 ± 5</td>
<td>8.6 ± 4.5</td>
</tr>
<tr>
<td>48</td>
<td>133 ± 20</td>
<td>8.1 ± 3.2</td>
</tr>
<tr>
<td>65</td>
<td>192 ± 9</td>
<td>4.9 ± 3.3</td>
</tr>
<tr>
<td>120</td>
<td>308 ± 56</td>
<td>4.8 ± 2.7</td>
</tr>
</tbody>
</table>
2. SFA Measurements of Normal and Friction Forces

The adhesion and friction properties of the tilted PDMS flaps structure on different rough substrates were tested using a 3D displacement and force sensing probe attachment conjugated surface forces apparatus 2000 (SFA) in a previously reported sphere-on-flat surface configuration [67, 90]. A CCD camera mounted on a microscope was incorporated into the system, which allowed viewing of the local contact during sliding and adhesion testing. The bottom surface was a flat glass disk with the fabricated PDMS flaps glued on top. The top glass surface, either smooth or with different roughness, was a spherical glass disk with a radius of 2 cm. The top surface can slide laterally over a distance of 200 to 500 µm with different sliding speeds.

During microfabrication, testing setup, and measurements, all work was performed in a clean, sealed environment (laminar flow hood or sealed SFA). The surfaces were stored in a sealed container before testing, protecting them from dirt or dust from the atmosphere. The adhesion tests were reproducible throughout the measurements as long as the structured surface was not damaged, or only damaged in a few places, which only occurred after prolonged high-load and sliding. This reproducibility shows that the effects observed could not be attributed to removal of a surface layer or ‘dirt’ particles.
C. Results

1. Low Adhesion Force Without Sliding on All Rough Substrates

The static adhesion forces of the tilted PDMS flaps against the surfaces with different roughness were investigated using a 3D surface force apparatus. The static adhesion was measured by first applying a desired preload and then separating two surfaces at a constant speed of ~5 µm/s without applying any lateral motion. On the smooth surface with a RMS roughness of 11 ± 8 nm, no adhesion was measured with preloads lower than 1 mN (Figure V – 3). When the preload was higher than 1 mN, adhesion force was detected during the separation of two surfaces. The asperities on the top surface of the flaps were observed to have a peak-to-valley height of less than 250 nm. The asperities on the sidewalls are of lower height than the asperities on the top surface (Figure V – 4). Due to this nano scale roughness of the flaps, under small normal preloads (< 1 mN), the flaps only make point contacts with the opposing surface, and only a small number of flaps can be in contact with the top spherical surface. So although the opposing surface is smooth, the real contact area is small and the adhesion force is under the detection limit of the apparatus. Increasing the preload gives rise to a larger real contact area per single flap as well as a larger number of flaps in contact with the spherical glass surface, and thereby higher adhesion forces.
Figure V - 3: Static adhesion (no prior shearing) test of tilted PDMS flaps on glass surfaces with different RMS roughness values. Strong adhesion was only measured on the smooth glass surface (RMS roughness = 11 ± 8 nm). No adhesion force was detected on the other surfaces with higher roughness values. Higher adhesion forces were measured even at lower preloads after sliding against the top glass surface. The colored regimes represent the trend of the adhesion forces and the experimental errors.

Increasing the roughness significantly decreased the static adhesion of the tilted PDMS flaps on the glass surface. No adhesion force on surfaces roughness higher than 11 nm could be detected by the current setup. The asperities of the rough surface greatly reduce the real contact area and therefore the adhesion force between two surfaces. Previous SFA measurements also showed that the adhesion force of a
randomly rough surface against a smooth mica surface decreased exponentially with increasing the roughness, with no adhesion force detected with a RMS roughness higher than 10 nm [70]. This result obtained with tilted PDMS flaps once again demonstrates that the roughness is one of the most important parameters influencing the adhesion force between two surfaces.

Figure V - 4: Detailed view of the top and sidewall surfaces of the angled micro-flaps. The asperities on the top surface were observed to have a peak-to-valley height of less than 250 nm. The asperities on the sidewalls are of lower height than the asperities on the top surface. The asperities on the top surface limit contact with the glass surface when no prior sliding is performed.

2. High Adhesion Force at Intermediate RMS Roughness After Sliding

Sliding significantly enhanced the adhesion performance of the PDMS flaps against the opposing glass substrate (Figure V – 3). Shearing deformable surfaces,
structures on surfaces or trapped liquids have been shown to cause alignment between the surfaces, affecting both the adhesion and friction forces [111, 112]. For the smooth glass surface (RMS roughness = 11 nm), a strong adhesion force of 0.95 mN was measured after sliding the top glass surface with just a 1 mN preload, whereas no adhesion force was detected under a similar preload without sliding. Although the final saturation values of the adhesion forces on the smooth glass surface for both static adhesion and the adhesion measured after sliding are almost the same (~1.2 mN), sliding reduced the required normal preload, $F_p$, to reach the maximum adhesion force.

Sliding also significantly enhanced the adhesion of the tilted flap arrays to the glass surfaces with various roughness: adhesion forces were measured for all the rough surfaces during the separation of the two surfaces after first sliding the top glass surface in the direction along the tilt, the $+y$ direction, or in the direction against the tilt, the $-y$ direction, and then stopping sliding but keeping the shear stress between the surfaces unreleased (Figure V – 5). After sliding in the $+y$ direction, except for the roughest surface (RMS roughness = 308 ± 56 nm), the adhesion forces between the PDMS flaps and the rough surface first rapidly increased with load at low loads ($F_p < 3$ mN), and then slowly increased with further increasing load (Figure V – 5(a)). Interestingly, increasing the RMS roughness from 11 ± 8 nm to 133 ± 20 nm did not lead to smaller adhesion forces, especially at high loads ($F_p > 3$ mN, highlighted regime in Figure V – 5). The adhesion force first increased with increasing roughness, with stronger adhesion forces measured on the
surfaces with 73 ± 5 nm and 133 ± 20 nm RMS roughness under similar loads. Further increasing the RMS roughness to 192 ± 9 nm and 308 ± 56 nm rapidly decreased the adhesion forces, with the lowest adhesion forces measured with the surface of 308 ± 56 nm RMS roughness. Clearly, surface roughness had a pronounced effect on the adhesion force between the tilted PDMS flaps and the glass surface measured after sliding. These results, however, show that higher roughness does not always lead to lower adhesion forces.

Strong anisotropy was also noticed in the adhesion measurement after sliding in either +y or –y direction, Figure V – 5(a), (b). For each individual rough surface, sliding in the +y direction resulted in a 50% to 100% higher adhesion force than for sliding in the –y direction under the same or similar preloads. While the weakest adhesion forces were measured with the roughest surface (RMS roughness = 308 ± 56 nm) tested, it showed the strongest anisotropic adhesion after sliding. Almost no adhesion force (less than 0.1 mN) was detected after sliding in the –y direction under various loads up to 9.3 mN, whereas an adhesion force of 0.4 mN was detected after sliding the top surface in the +y direction under a preload of ~9.8 mN.
Figure V - 5: Adhesion measured during the separation of glass surfaces (with various height RMS roughness values) and the tilted PDMS flaps after sliding the glass surface in $+y$ direction (a) and $-y$ direction (b). The adhesion forces measured after sliding the top surface in the $+y$ direction are significantly higher than the values measured after sliding in the $-y$ direction. The solid black lines with arrows serve to illustrate the trends observed with surfaces of increasing RMS roughness values.
3. Anisotropic Friction Forces on Rough and Smooth Substrates

Anisotropic friction force was also measured when sliding the tilted flaps against surfaces with various RMS values of roughness, as seen in Figure V – 6(a), (b). For each rough surface tested, sliding in the +y direction always resulted in higher friction forces than sliding in the –y direction. This asymmetry perfectly correlates with the adhesion measurement: the higher adhesion force while sliding in the +y direction results in higher adhesive friction force.

Increasing the RMS roughness led to an initial increase and then a subsequent decrease in the friction force. When sliding the tilted flaps against the glass surface with a RMS roughness of 73 ± 5 nm, in both +y and –y directions, the friction force, initially lower than that of the smooth surface (RMS = 11 nm) at lower load ($F_\perp < 2$ mN), rapidly increased to similar magnitudes in the middle load regime ($2 < F_\perp < 5$ mN, highlighted). Further increasing the load, however, again led to lower friction force in both directions in comparison to the friction force measured when sliding the flap against the smooth glass surface. The friction force was more enhanced on the surface with a RMS roughness of 133 ± 20 nm: the highest friction force was measured in both +y and –y directions on this surface for all the loads tested. The effect of roughness is more pronounced at higher RMS roughness. Not only were smaller friction forces measured while sliding the flaps against the surfaces with 192 ± 9 nm and 308 ± 56 nm RMS roughness under similar applied loads, but also the friction forces showed a fairly linear dependence on the applied load in both the low
load regime and the high load regime with no friction force measured at zero load. These two surfaces also showed the lowest adhesion forces under different loads.

The high friction forces measured with surfaces of relatively low RMS roughness are probably due to the adhesion between two surfaces during sliding. This is clearly demonstrated by the strong adhesion force previously measured after sliding two such surfaces. When the flaps were sliding against the surfaces with 11 ± 8 nm, 73 ± 5 nm, and 133 ± 20 nm RMS roughness, finite friction forces were measured with zero even negative loads when sliding in both the +y and −y directions, another indication of strong ‘adhesive controlled’ friction. These results, at least at first glance, contradict the common knowledge in tribology. It is usually assumed that increasing surface roughness leads to smaller contact area between two surfaces during sliding, and therefore smaller adhesion and friction forces. However, increasing the surface roughness also changes the surface topology, which could significantly alter the friction behavior of the flaps on different rough surfaces. With certain “matching” geometries, this interlocking of flaps can even lead to higher adhesion and friction forces, which will be discussed further in the discussion section.
Figure V - 6: Anisotropic friction forces of the tilted PDMS flaps sliding against glass surfaces with various height RMS roughness values in the +y direction (a) and the –y direction (b). The solid black lines with arrows serve to illustrate the trends observed with surfaces of increasing RMS roughness values.
D. Discussion

1. The Effect of Roughness

One particular difficulty in studying rough surface friction and adhesion is that quantification of roughness is not trivial. The root mean square roughness (RMS roughness) is the most widely used parameter in describing the surface roughness, and the only parameter used in many studies. However, RMS roughness is just an averaged value of the whole surface topological profile, and it alone gives no information about the shape of the surface profile: different surface topological profiles could result in the same RMS roughness value but totally different adhesive and tribological behaviors [113]. For example, two surfaces – one with cone-shaped peaks and the other with cone-shaped valleys – could have exactly the same RMS roughness value, whereas the adhesion and friction properties of these two surfaces against a smooth substrate surface are totally different. When compressing two rough surfaces, they only make contacts at a few of the highest asperities at the beginning, and the number of contacts increases with increasing loads. Therefore, adding a few asperities significantly higher than all others on one surface could totally change the adhesion and friction between two surfaces without significantly changing the RMS roughness. To accurately define the surface roughness, we need to characterize a series of parameters including different length scales of the surface topology, the distribution of the size and shape of asperities [109, 114].
This difficulty was encountered also in this study. Although the RMS value has been used to represent the roughness of the surfaces studied, it indeed did not fully capture the nature of the rough surfaces used. The rough surfaces used in this study have spatially randomly distributed asperities, whereas the height and the size of the asperities, due the limit of the etching technique, are quite uniform rather than randomly distributed. Increasing etching time just increases the height and the size of the mesa-shaped asperities, and decreases the gap between nearest neighbor asperities.

In general, the RMS height roughness is not a sufficient characterization of structured surfaces, which must also include the width roughness between asperities. Table V – 1 shows both the height and width roughness values of the surfaces studied. As discussed further below, both roughness parameters exhibit similar commensurability trends, both for the adhesion and friction forces. For example, with the $73 \pm 5$ and $133 \pm 20$ nm RMS (height roughness) surfaces, the average gap distances between neighboring asperities (width roughness) were $8.6 \pm 4.5$ µm and $8.1 \pm 3.2$ µm, respectively. Therefore, the gap distances between neighboring asperities were close to 10 µm, which is very close to the width of the tilted PDMS flaps.

In the static adhesion test, because of the asperities on the top glass surface, the flaps mainly made point contacts with some pillar-like asperities, leading to a small real contact area with no adhesion being measured during separation. Sliding the top
rough surface along the $+y$ or $-y$ directions, however, gave the flaps the chance to slide past or over different asperities. The low modulus PDMS flaps could also deform during this process, and therefore could penetrate into the gaps between randomly distributed asperities, Figure V – 7(b). This penetration gave rise to interlocking-like structures between the flaps and asperities, which led to much stronger friction and adhesion forces during and after sliding. Interestingly, this interlocking mechanism is widely used in many biological systems, such as insects, plant spreading, and bird feathers, and has also inspired the engineering design of structured adhesives [115–118]. When the RMS roughness of the surface was further increased to $192 \pm 9$ nm, the size of the asperities grew bigger, and most of the sizes of the gaps between different neighboring asperities were smaller than $10 \mu$m (the average distances between neighboring asperities was $4.9 \pm 3.3 \mu$m). During sliding, the flaps, now not being able to penetrate into the gaps between asperities, mainly stayed on top of the asperities, Figure V – 7(c). This effect was even more pronounced with the surface of $308 \pm 56$ nm RMS roughness, leading to the smallest friction forces and almost no adhesion forces measured under different loads.
Figure V - 7: (a) A schematic shows the flaps on the smooth glass surface (RMS = 11 ± 8 nm). (b) The geometric interlocking mechanism that results in high adhesion and friction forces when sliding the PDMS flaps on the etched rough surfaces with 73 ± 5 nm or 133 ± 20 nm RMS roughness values. (c) Increasing the roughness increases the size of the asperities while decreasing the gap distance between neighboring asperities, preventing the interlocking mechanism.

Previous SFA experiments on random rough surfaces have shown that the adhesion of two surfaces decayed exponentially with increasing the RMS roughness [93]. This conclusion, although also confirmed by computer simulation, cannot be
directly applied to the current study on the tilted PDMS flaps. This once again shows that fully characterizing the surface profile is extremely important in order to study the adhesion and friction between rough surfaces. Unlike ideal surfaces, many real surfaces contain different length scales of roughness, which may have a correlative effect on the adhesion and friction between surfaces. Different geometries of the opposing surfaces can either enhance or diminish the adhesion and friction between two surfaces. Computer simulation and experimental studies have also shown that with an anisotropic surface, the adhesion and friction forces could be significantly different while sliding along different axes of the surface [104].

Another important parameter affecting the adhesion and friction forces between the flaps and the rough surface is the elastic moduli of the two materials that the surfaces are made of, especially at the contact area [113, 119]. In the system tested, the elastic modulus of SiO$_2$ (~70 GPa) is much higher than the elastic modulus of PDMS (~1.8 MPa), therefore the rough surface can be considered to be non-deformable, whereas the flaps are highly deformable. With low surface roughness, high elastic deformation may lead to higher contact area, similar to the function of setae on gecko feet. This could also be a reason for the higher adhesion and friction forces measured on the surfaces with lower RMS roughness than on the control smooth surface. However, the elastic deformation of the flaps can only cover a small range of surface roughness. Beyond this range, increasing the roughness rapidly reduces the contact area of the surfaces, and thereby the adhesion and friction forces.
2. The Effect of Anisotropic Structure

In the gecko adhesion system, the setae are naturally tilted, with a tilt angle of 45° [19]. This anisotropic structure enhances the adhesive friction during the attachment of the foot (in the gripping motion) and reduces the adhesive friction during the detachment (in the peeling motion) [17, 18]. Mimicking this anisotropy is crucial for developing gecko-inspired robots: the robot should be able to achieve strong adhesion and friction when it attaches to a surface, and should also be able to easily peel its feet off with suitable articulation. By adding an angle of tilt to the micro-flaps, the anisotropy of the gecko setae has been successfully mimicked. The flaps, tilted by about 20°, have a much smaller stiffness in the +y direction compared to the –y direction, and bend over in the +y direction easily [67]. Made of PDMS with an elastic modulus of ~1.8 MPa, the flaps are also able to achieve large deformations during compressing and sliding, resulting in a larger contact area with the opposing surface. When sliding backwards, in the –y direction, the geometry of the flaps does not allow them to bend over naturally, leading to a smaller total contact area. The van der Waals adhesion force between two surfaces is proportional to the total contact area, and therefore the sliding in the +y direction results in much higher adhesion forces and adhesive friction forces under different loads. The strong anisotropy of the adhesion and friction forces measured on all the substrate surfaces demonstrated that this anisotropy held fairly well on not only on a smooth surface but also on rough surfaces with various values of RMS roughness.
E. Conclusion

Natural surfaces are usually rough, containing various scales of roughness. Surface roughness greatly affects the adhesion and friction properties of almost all tribological and adhesive systems, including gecko-inspired adhesives, yet not much attention has been paid thus far. This work presents the first experimental effort to systematically study the effect of nano-scale surface roughness on both the adhesion and friction of gecko-inspired structured adhesives. The results presented demonstrate that suitable surface roughness could effectively enhance the adhesive friction between tilted PDMS flaps and a rough/patterned substrate in comparison to a smooth surface, most likely due to a good geometric match between the flaps and the surface structure. Thus, for certain normal load ranges, maximized forces could be achieved when the mean space between asperities equals the width of flaps.

Although surface roughness strongly affected the magnitude of the adhesion and friction forces, anisotropic friction and adhesion properties of the tilted flaps were still preserved while sliding along different directions on surfaces with various roughness values. Although the results of this work cannot be directly applied to all gecko-inspired adhesive structures, they clearly illustrate the importance of load, surface roughness and structure, adhesive fiber anisotropy, and direction of sliding. In order to develop better structured adhesives for gecko robots, additional systematic studies on the effect of surface structure are required.
VI. Angled Half-Cylinder Microfibers for Controllable and Reusable Adhesion

A. Introduction

Key characteristics of the natural gecko adhesive system [9] include high and anisotropic normal adhesion and lateral shear forces during attachment, low detachment force, a high adhesion to initial preload force ratio ($\mu'$) of 8-16 [11], lack of inter-fiber self-adhesion, and operation over more than 30,000 cycles without significant loss of adhesion performance [15]. Unlike conventional pressure-sensitive adhesives which are easily fouled after a few uses, the natural gecko adhesive survives several thousand cycles between successive skin molts. Potential applications for reusable dry adhesives exist, such as climbing robotic systems [26, 27], mobile sensor platforms, non-fouling pick-and-place handling systems for wafers and liquid crystal displays in industrial nanofabrication facilities [29], and human wall climbing [30], require them to demonstrate high reusability in addition to controllable adhesion and friction.

To design dry, responsive adhesive systems inspired by the gecko, various kinds of patterned surfaces with arrays of vertical and tilted fibers, with tips of different shapes and materials, and with sizes spanning both the micron and nanometer scales, have been fabricated [29, 66, 68, 72–78, 80, 120]. Although several synthetic adhesives capable of supporting large adhesion and/or shear forces have been reported, either no extended durability testing was done [67, 69, 73–74], or they
exhibited very limited lifetime [75–77], or no systematic study into durability was performed beyond just 50-100 cycles [29, 66, 78–79]. In this study, vast arrays of angled half-cylinder Polydimethylsiloxane (PDMS) microfibers (Figure VI – 1) were used to create an adhesive material with high adhesion anisotropy, high $\mu'$, high shear force, and high reusability. In order to rigorously test the adhesive for durability, it was subjected to 10,000 repeated test cycles without intermediate cleaning, and the material was found to maintain high adhesion and friction forces without any significant wear of the fibers.

Although a bio-inspired synthetic fibrillar adhesive system with a long lifetime comparable to gecko setae has been reported in [68], the wedge-shaped fibers used did not exhibit any gecko seta-like adhesion or friction anisotropy on sliding, and supported maximum shear loads far inferior to the gecko. The only other study thus far with tests over 10,000 cycles reported a large reduction in normal adhesion for high-density polyethylene (HDPE) fibers, and a complete loss of adhesion for polypropylene (PP) fibers [80]. Large reductions in shear performance were also reported over these repeated tests. The ratio of adhesion to initial preload force ($\mu'$) is also especially important for climbing applications, with higher values resulting in increased operational stability on walls and ceilings due to a low corresponding reaction force from the substrate surface, as well as enhancing reusability by preventing damage to fibers during preloading. Results from the current study also show a large improvement in the value of $\mu'$ over other high-cycle adhesives.
Figure VI - 1: (a) SEM image of angled PDMS semicircular microfibers, with scale bar, coordinate axes, angles of tilt of the flat face (28°) and curved face (15°), and fiber lateral geometry (white semicircle) indicated. Articulation occurs in the y-z plane. The +y direction (towards the flat face, and with fiber tilt) is the gripping direction, the −y direction (towards the curved face, and against fiber tilt) is the releasing direction, −z is the vertical loading direction, and +z is the vertical unloading direction. (b) SEM image showing the curved face of the microfibers. The width of a single fiber varies along both the ±x and ±y axes due to the fabrication process.

B. Experimental Methods

Fabrication of angled microfibers over cm-scale patch sizes was performed using a previously described angled photolithography and molding technique (Chapter II, Section C). The fibers obtained were ~13 µm in height, with a tilt angle of 28° from the vertical for the flat face, and a tilt angle of 15° for the curved face.
Adhesive testing was performed using a previously reported flat-on-flat testing geometry with a custom-built microtribometer in which careful relative alignment of the two surfaces before testing is possible with manual goniometer and tilt stages [69, 74]. After alignment, ‘load-drag-pull’ tests were performed to normally preload, laterally shear, and then pull off from the adhesive material. In these tests, a 4-mm diameter flat glass puck is first brought into contact (test contact area = 12.6 mm$^2$, with a maximum of ~64,440 microfibers in contact over this area) with the adhesive at a vertical speed of 1 µm/s (puck motion in the $-z$ direction as defined in Figure VI – 1) until a specified preload is reached. Then relative sliding motion of the puck and sample at a lateral speed of 3 µm/s is performed with a motorized linear stage. Sliding motion of the puck in the $+y$ direction (as defined in Figure VI – 1) brings the flat face of the semicircular fibers into contact, and is the ‘gripping direction’ with high contact area, and high adhesion and friction forces. Conversely, sliding in the $-y$ direction brings the fiber’s curved face into contact, and is the ‘releasing direction’ with low contact area, and low adhesion and friction forces. After sliding for the desired distance, the two surfaces are separated at a vertical speed of 1 µm/s. Position, force, and torque data are continuously recorded during all tests.

The maximum tensile loads supported by the sample after lateral sliding and normal pull-off are called shear adhesion forces, while the in-plane forces generated during sliding are referred to simply as shear forces. Load-drag-pull tests were performed on the sample at three different preload values of 0.025 N, 0.1 N, and 0.18 N over a wide range of sliding lengths greater than 0.15 mm (up to 0.2 mm for
the 0.18 N preload case) in both positive and negative directions. Each test was performed 6 times, and the average values and standard deviations calculated.

The sample was subsequently tested for durability by performing 10,000 repeated tests in a single location without intermediate cleaning of either sample or glass puck surface. The testing procedure for durability was identical to the load-drag-pull procedure previously described, except that all durability tests were performed at the maximum preload value of 0.18 N and maximum positive (+y) shear length of 0.2 mm.

Temperature and humidity measurements were not taken during the testing period; however, later measurements taken every five minutes over a 2 month period showed an average laboratory temperature of 23.5 °C with a standard deviation of 0.3 °C, and a relative humidity average of 49.6% with a standard deviation of 5.3%.

C. Results and Discussion

1. Anisotropic Adhesion Behavior

The tilted half-cylinder microfibers exhibited some adhesion even without lateral sliding for all the three preloads tested (0.025N, 0.1 N, and 0.18 N), as seen in Figure VI – 2. The maximum adhesion pressure obtained without lateral sliding was 3.0±0.1 kPa, which is due to contact between the flat top face of the microfibers (as seen in Figure VI – 1), and the flat glass puck. In optical microscope images taken after preloading and before lateral sliding (Figure A – 1(a), a part of which is shown as Figure VI – 6(a) for clarity) for a preload of 0.1 N, the contact area between the puck
and the tops of the microfibers may be clearly observed as the darker regions. The outlines of the tilted flat face of the microfibers are also visible, and observed to be not in significant contact without sliding. This situation has also been illustrated in the schematic diagram in Figure VI – 6(d).

**Figure VI - 2: Adhesion force as a function of the applied normal preload.** Some adhesion is obtained without lateral shearing due to contact between the flat top face of the microfibers and the flat glass puck. All data points depict average values with ±1 standard deviation error for 6 repeated tests.
Figure VI - 3: Shear adhesion force as functions of the preload and shear length along the ±\(y\) axis. Shear adhesion forces can be controlled based on shear direction. A large \(\mu'\) value enables the material to generate normal adhesion of up to 4.7 times the preload with suitable shearing articulation. All data points depict average values with ±1 standard deviation error for 6 repeated tests.

Figure VI – 3 illustrates how normal adhesion force may be easily controlled by laterally shearing the material along a preferred axis – either increasing adhesion after sliding (‘shear adhesion’) to a maximum of 9.4±0.1 kPa in the fiber tilt direction (+\(y\)) from the zero-shear value of ~3.0 kPa, or decreasing it to a steady state value of 1.6±0.2 kPa in the direction against tilt (−\(y\)). Thus, the anisotropy in shear adhesion is 5.9, considerably higher than the anisotropy values previously obtained
with purely vertical half-cylinder microfibers [74]. It is also important to note that the adhesion force after sliding in the \(-y\) direction repeatedly showed a minimum force value of zero for a shear length of \(-0.01\) mm at all three values of the applied preload. Thus, with suitable articulation, it is possible to use this adhesive for strong attachment but very easy detachment in a manner similar to the gecko.

The maximum ratio of shear adhesion to preload force \((\mu')\), obtained for a preload of 0.025 N for sliding in the \(+y\) direction, is 4.7, a 571% increase when compared with 0.7 obtained with similar tests on both tilted micro-flaps [69] and vertical half-cylinder fibers [74]. This large reduction in initial preload required contributes to increased operational stability in climbing applications on walls and ceilings due to a low corresponding reaction force from the substrate surface during preloading. A high value of \(\mu'\) is also likely to enhance reusability by preventing damage to fibers during the preloading step.

The normal forces recorded during initial puck-sample approach and preloading up to 25 mN, subsequent lateral sliding over a distance of 150 \(\mu\)m, and final retraction to separate the two surfaces are shown in Figure VI – 4. As shown in Figure VI – 4 by the blue trace, sliding in the gripping direction \((+y)\) results in the normal force becoming tensile and negative from the initial compressive positive preload force of 25 mN. On retraction after sliding, the normal force rapidly falls back to zero, and a maximum shear adhesion force, \(F_{ad, grip}\), of 107 mN is obtained.
In contrast, as shown by the red trace in Figure VI – 4, sliding in the releasing direction (−y) results in an initial increase in repulsive force between the puck and the sample, until a maximum value $F_{\text{repulsion}} = 61$ mN, is attained. Further sliding results in an eventual switch to an attractive tensile force. However, the maximum shear adhesion obtained, $F_{\text{ad, release}}$, is just 20 mN.

Figure VI - 4: Dynamic measurements of the anisotropic normal force generated by the adhesive when preloaded to 25 mN, and subsequently sheared a distance of 150 µm. The trace in the gripping direction (+y) is marked in blue, and the trace in the releasing direction (−y) in red.
Dynamic measurements of the normal forces measured with an initial preload of 25 mN, and shear length of 70 µm in the gripping direction, are shown in Figure VI – 5(a). This shear length results in the maximum adhesion force on pull-off – a value of 118 mN, or 9.4 kPa over the area of the puck. Dynamic measurements of the normal forces measured with an initial preload of 25 mN, and shear length of 10 µm in the releasing direction are shown in Figure VI – 5(b). Similar to the red trace shown in Figure VI – 5(a), shearing in the releasing direction after initial approach and preloading resulted in an increase in repulsive force between the puck and the sample to 61 mN. However, in the case shown in Figure VI – 5(b), sliding was stopped before the normal force could switch from compressive to tensile. Retraction then resulted in zero shear adhesion being observed at complete separation, facilitating easy release in a manner similar to the gecko.

Figure VI - 5: Dynamic measurements of the normal forces generated by the adhesive for a preload of 25 mN, and shear lengths of 70 µm in the gripping direction (Figure a) and 10 µm in the releasing direction (Figure b).
Figure VI - 6: Optical microscope images (figures (a), (b), and (c)) and corresponding schematic diagrams (figures (d), (e), and (f)) illustrate the role of the tilt angle and fiber geometry in obtaining adhesion anisotropy after lateral shearing along the ±y axis. The optical microscope images were taken at the end of normal preloading in figure (a), and at the end of lateral sliding (itself performed after normal preloading) in figures (b) and (c). Areas of the fibers in contact with the glass puck are seen as darker regions in these images.
The anisotropic adhesion behavior of the tilted microfibers can be explained by considering the anisotropic fiber shape in conjunction with the Kendall peel model for the force required to peel a thin elastic adhesive film from a surface [45]:

\[
F_{\text{peel}} = b d E \left( \cos \theta_{\text{peel}} - 1 + \sqrt{\cos^2 \theta_{\text{peel}} - 2 \cos \theta_{\text{peel}} + 1 + \frac{2w}{dE}} \right) \quad (VI - 1)
\]

Where \( \theta_{\text{peel}} \) is the angle of peel between the film and the substrate, \( w \) is the adhesive energy for the contact, \( b \) is the contact width of the film, \( d \) is the thickness of the film, and \( E \) is the Young’s modulus of the film material. Tilted half-cylinder microfibers may be expected to have different values of contact width \( b \) as well as peel angle \( \theta_{\text{peel}} \) for peeling of the flat face from a substrate versus peeling of the curved face from the same substrate, resulting in anisotropy in the peel force.

The half-cylinder microfibers are expected to have a larger area in contact with a substrate surface on the flat face when compared with the curved face, and this was experimentally observed in optical microscope images taken after lateral sliding and before normal pull-off for the 0.1 N preload case (Figures VI – 6(b) and VI – 6(c)). Destructive interference of reflected white light at the glass–PDMS interface enabled visualization of the fiber areas in contact as darker regions. Figure VI – 6(b) shows the area in contact between the puck and the sample after sliding towards the flat face of the fibers, in the \(+y\) direction. For clarity, only a portion of the entire image taken has been shown in Figures VI – 6(a-c). The full snapshots taken are included in the Appendix, with Figures A – 1(a), A – 1(b), and A – 1(c) corresponding to
Figures VI – 6(a), VI – 6(b), and VI – 6(c) respectively. Analysis of the full image in Figure A – 1(b) indicates that the average contact width with the flat face after sliding was ~8.2 µm. This is in good agreement with the value obtained from analysis of SEM images of the microfibers (e.g. Figure VI – 2(b)) which indicate an average value of the contact width of ~8.5 µm. The curved face of the microfibers was designed to minimize contact area with a substrate. Although a cylinder forms only a lateral line contact with a flat surface, since PDMS has a low Young’s modulus, the curved face of the microfibers is expected to significantly deform and have a finite contact width. An approximate estimate for the contact width on the curved face may be obtained by considering the equilibrium contact width \( c \) of a cylinder in contact with a flat surface [121, 122]:

\[
c = 8 \left[ \frac{WR^2}{\pi E^*} \right]^{1/3}
\]

Where \( W \) is the work of adhesion between the two surfaces, \( R \) is the radius of the cylinder, and \( E^* \) is the effective Young’s modulus of two materials 1 and 2 in contact, given by:

\[
\frac{1}{E^*} = \frac{(1 - \nu_1^2)}{E_1} + \frac{(1 - \nu_2^2)}{E_2},
\]

\( \nu \) denoting the Poisson’s ratio. For PDMS (\( E_1 = 1.8 \) MPa) [92] in contact with glass (\( E_2 > 50 \) GPa) [123], \( E^* \approx \frac{E_1}{(1 - \nu_1^2)} \). Also, \( W = 100 \) mJ/m\(^2\) for PDMS in contact with glass [94], and \( \nu_1 = 0.5 \) for PDMS. Thus, Equation (VI – 2) predicts that the contact width with the curved face is 5.0 µm. This value is close to the observed value for the 0.1 N preload
case – analysis of the optical microscope image in Figure A – 1(c) (part of which has been shown in Figure VI – 6(c)) yields an average contact width of 4.7 µm.

Since the Kendall model in its original form applies to a film with a rectangular cross section of width \(b\) and thickness \(d\), and the microfibers in this case have a semicircular cross section of radius \(R\), a small modification is made to correct the cross sectional area, and the equation for peel force is re-derived as originally done in [45]. Also, taking the vertical component of the peel force as the adhesion force, and multiplying the equation by the maximum number of fibers \(N\) in contact with the puck, we may write an equation for the shear adhesion force \(F_{\text{ad}}\) as follows:

\[
F_{\text{ad}} = N \left\{ \sin \theta_{\text{peel}} \frac{\pi R^2}{2} E \left( \cos \theta_{\text{peel}} - 1 \right) + \sqrt{\cos^2 \theta_{\text{peel}} - 2 \cos \theta_{\text{peel}} + 1 + \frac{4wb}{\pi R^2 E}} \right\} \quad (VI – 3)
\]

For the adhesive tested, \(N \sim 64,440\). For the contact width, we may take \(b = 2R\) for the flat face, while \(b = c\) as calculated earlier using Equation (VI – 2) for the curved face.

In the absence of a mechanism to directly optically observe the peel angles of the fibers after sliding in various directions, estimating \(F_{\text{ad}}\) from Equation (VI – 3) requires the use of an estimate for the peel angle, \(\theta_{\text{peel}}\). From the geometry of the fibers, as shown in the schematic diagram in Figure VI – 6(e), taking the peel angle
for the flat face $\theta_{\text{peel}}^f$ as the complementary angle of the tilt angle of this face from the vertical ($\theta_{\text{peel}}^f = 62^\circ$), we get a theoretical estimate of 89 mN for the adhesion force after sliding from Equation (VI – 3), a discrepancy of 17% from the experimental steady state shear adhesion value of ~107 mN for the $+y$ direction as seen in Figure VI – 3. The exact experimental value is obtained by using $\theta_{\text{peel}}^f = 52^\circ$ in this model. Similarly, as seen in the schematic diagram in Figure VI – 6(f), taking the peel angle for the curved face $\theta_{\text{peel}}^c$ as the supplementary angle of the tilt angle of this face from the horizontal ($\theta_{\text{peel}}^c = 105^\circ$), we get a theoretical estimate of 24 mN for the adhesion force after sliding from Equation (VI – 3), a discrepancy of 26% from the experimental average value of 19 mN for the $-y$ direction as seen in Figure VI – 3. The exact experimental value may be obtained by using $\theta_{\text{peel}}^f = 119^\circ$ in this model.

The reasons for differences between theory and experiment are likely due to the fact that both the microfiber surface (as seen in Figures VI – 1(a) and VI – 1(b)) as well as the glass puck surface are rough, while the theory is for smooth surfaces in contact. The glass puck surface was found to have an RMS roughness value of ~160 nm by optical profilometry (Wyko NT1100 Optical Profiling System). Also, the actual peel angles with the adhesive engaged may not exactly match the geometric values as obtained by SEM observation of the fibers when not engaged. Further, values in the literature for the Young’s modulus of PDMS [92, 94, 124–128], as well as for $w$ for the PDMS-glass interface [94, 128], span a range of values, and the
values chosen may not perfectly match the system. The Kendall model also assumes a constant film thickness, whereas the fibers do not have this property in both the $\pm x$ and $\pm y$ directions, as seen in Figures VI – 1(a) and VI – 1(b) – instead, average values for the cross sectional area and contact width as obtained from SEM analysis have been used in Equation (VI – 3).

**Table VI - 1: A comparison of the effect of peel angle (due to fiber tilt) and contact width (due to fiber anisotropic geometry) on the theoretically predicted adhesion force using the modified Kendall peel model from Equation (VI – 3).**

For the same peel angle, variations in contact width $b$ from the curved face ($b = 4.97 \mu m$) to the flat face ($b = 8.5 \mu m$) result in a significant change in adhesion force. Also, for a fixed value of contact width, a change in peel angle $\theta$ from a value of 62° (shearing in the gripping direction) to a value of 105° (shearing in the releasing direction) also results in a significant change in adhesion. The theoretical estimates are also compared with experimental values where available.

<table>
<thead>
<tr>
<th>Contact Width, $b$ (µm)</th>
<th>Peel Angle, $\theta$ (°)</th>
<th>Calculated Adhesion Force (mN)</th>
<th>Experimental Adhesion Force (mN)</th>
</tr>
</thead>
<tbody>
<tr>
<td>8.5</td>
<td>62</td>
<td>89</td>
<td>107</td>
</tr>
<tr>
<td>8.5</td>
<td>105</td>
<td>42</td>
<td>N/A</td>
</tr>
<tr>
<td>4.97</td>
<td>105</td>
<td>24</td>
<td>19</td>
</tr>
<tr>
<td>4.97</td>
<td>62</td>
<td>52</td>
<td>N/A</td>
</tr>
</tbody>
</table>
The analysis, however, clearly provides certain guidelines for obtaining high adhesion force anisotropy, and provides an approximate estimate of the values that may be obtained. Using Equation (VI – 3) to obtain a theoretical estimate for the adhesion force, it is possible to show that both angle of tilt and fiber shape have a significant effect on adhesion anisotropy. This has been shown in Table VI – 1, where a switch in peel angle $\theta_{\text{peel}}$ from 62° to 105° for the same value of contact width $b$ results in a significant change in adhesion force. A change in contact width $b$ from a value corresponding to the flat face to a value corresponding to a curved face for the same peel angle $\theta_{\text{peel}}$ (whether 62° or 105°) also results in a significant change in adhesion force. Thus, both semicircular fiber shape (influencing the contact width) and angle of tilt (influencing the peel angle) were found to have a significant effect on adhesion anisotropy for the reversible adhesive material.

From the model used, use of half-cylinder microfibers with a higher Young’s modulus is expected to further lower the contact width $c$ on the curved face from Equation (VI – 2), thereby increasing anisotropy. Use of larger tilt angles (from the vertical) is expected to result in a higher normal component of peel force on the face with the angle of tilt until a maximum is reached in accordance with Equation (VI – 3), before reducing again. Simultaneously, the normal component of the peel force on the face against the angle of tilt is predicted to continuously decrease for larger tilt angles, resulting in higher adhesion anisotropy.
2. Anisotropic Friction Behavior

The lateral or friction force that may be generated by a synthetic bio-inspired adhesive is very important for applications requiring wall climbing. The adhesive material fabricated generates large and anisotropic in-plane shear forces during sliding along the ±\(y\) axis (Figure VI – 7) for all three initial preloads tested. The anisotropy in shear forces are 2.0, 1.6, and 1.4 for preloads of 0.025 N, 0.1 N, and 0.18 N respectively. The shear force generated increased with sliding distance until it saturated in a manner similar to that seen with other tests on gecko setal arrays [17], and a large maximum shear pressure of 78±0.1 kPa was supported for a preload of 0.18 N after sliding in the +\(y\) direction with the tilt of the fibers. Shear force values for the natural system reported in the literature vary, and range from 88 kPa from measurements taken on two whole gecko feet of area = 227 mm\(^2\) [10], 226 kPa from measurements on a single gecko digit of area = 19 mm\(^2\) [9, 25], 184±23 kPa from measurements on isolated setal arrays of area = 0.93 mm\(^2\) [15], 550 kPa from measurements on setal arrays of total area = 0.54 mm\(^2\) [65], up to 4585 kPa from measurements on a single seta of area = 43.6 µm\(^2\) [9, 11]. The adhesive in the current study, tested over an area of 12.6 mm\(^2\), thus generates a shear pressure approaching the 88-226 kPa range reported for gecko toes and feet.
Figure VI - 7: (a) Maximum shear force generated along the ±y axis as a function of the shear length for various preload values. The synthetic adhesive, tested over a 12.6 mm$^2$ patch area, was able to sustain a maximum shear pressure of 78±0.1 kPa, approaching the 88-226 kPa range for natural gecko feet and digits. (b) Dynamic measurements of the anisotropic shear forces generated for a preload of 180 mN, and shear length of 200 µm, in both the +y gripping direction (blue trace) and −y releasing direction (red trace).
A general equation describing the friction/shear force, $F_{\parallel}$, between two adhesive surfaces is [56]:

$$F_{\parallel} = \tau A_{\text{real}} \quad (\text{VI} - 4)$$

Where $\tau$ is the interfacial shear strength, and $A_{\text{real}}$ is the real area of contact between two surfaces. For non-adhering surfaces, $F_{\parallel}$ is a linear function of the load, $L$, and can take the common form $F_{\parallel} = \mu L$, where $\mu$ is the coefficient of friction. The friction force is also influenced by the adhesion force between the adhesive material and the substrate. For adhering surfaces, Equation (VI – 4) remains the same; however, the friction force is no longer a linear function of the load because of the adhesive component of the load. The above equation also demonstrates that a flat surface in contact with a flat substrate is expected to generate higher friction forces than a curved surface against the same substrate. As is the case with adhesion anisotropy, it is important to note that this approach can be extended to higher modulus materials as well – since the contact area on the curved face is expected to be lower with higher modulus, it should be possible to achieve higher shear force anisotropy ratios.

It is also to be noted that nearly all throughout the duration of sliding, the shear force in the gripping direction stays consistently higher than in the releasing direction (Figure VI – 7(b)). Similar anisotropic friction behavior has also been observed on tests with natural gecko setal arrays in a Surface Forces Apparatus [17]. While transient anisotropy in friction forces with tilted hierarchical polyurethane
microfibers has been recently reported, dynamic measurements showed a complete loss of anisotropy with continued sliding when forces attained steady state values, with forces in the releasing direction becoming comparable to or even higher than those in the gripping direction [75]. In the case of the current study, a combination of tilt angle and anisotropic fiber shape ensures lower real area of contact and forces in the releasing direction even after prolonged sliding.

For PDMS in contact with glass, the interfacial shear strength \( \tau \) has been experimentally estimated to be \(~0.3\) MPa [129]. For a friction/shear force of \( 0.66 \) N, as obtained for sliding in the \( +y \) direction with an initial preload of \( 0.1 \) N (Figure VI – 7(a)), Equation (VI – 4) thus predicts a real area of contact of \( 2.2 \) mm\(^2\). Sliding in the \( -y \) direction for the same initial preload resulted in a shear force of \( 0.41 \) N, suggesting a real area of contact of \( 1.4 \) mm\(^2\). However, analysis of the optical microscope images in Figures VI – 6(b) and VI – 6(c) shows an area of contact of \(~6\) mm\(^2\) after sliding in the \( +y \) direction, and \(~1.7\) mm\(^2\) after sliding in the \( -y \) direction. A major reason for this discrepancy could be the fact that these optical microscope images were taken at the *conclusion* of sliding, and just before normal pull-off, whereas the shear forces plotted in Figure VI – 7 were measured *during* sliding. Although sliding was smooth without any macroscopic stick-slip, considering that the sliding/shear lengths for the steady-state friction values in Figure VI – 7 far exceeded the dimensions of the individual fibers, it is possible that random small amplitude stick-slip events of some fraction of the fibers occurred, and thus, despite individual fibers possibly experiencing sliding irregularities, their collective motion
is one of smooth sliding. Smooth sliding with stochastic stick–slip of a population of individual fibers has also been previously suggested for tests on gecko setae [15], and optically observed for tests on both PDMS angled wedge-shaped microfibers [15] and Polyvinylsiloxane (PVS) vertical mushroom-tipped microfibers [130]. For the PVS microfibers, a significantly lower area of contact was observed during sliding than in the static case. A smaller reduction in real area of contact between Polymethylmethacrylate (PMMA) blocks in relative sliding motion with stick-slip characteristics was also observed at the onset of slipping [131, 132]. A reduction in area of contact during the slip event has also been reported for dissimilar metals in sliding contact [55]. Thus, Figures VI – 6(b) and VI – 6(c), taken after cessation of sliding, would be expected to only provide an upper bound on the contact area during sliding, although the decrease in contact area may be expected to be less drastic for tilted half-cylinder microfibers than for the vertical mushroom-tipped microfibers in [130]. The test apparatus is currently being modified to be able to include continuous video capture of all stages of load-drag-pull testing in the future to verify the behavior of the fibers during both lateral sliding and normal pull-off.

3. High Durability

Durability is an important concern for synthetic adhesives for most practical applications in climbing robotics and industry. Several synthetic adhesives capable of supporting large adhesion and/or shear forces have been reported in the literature, but either no extended durability testing was done [67, 73–74], or they exhibited
very limited lifetime [75–77], or no systematic study into durability was performed beyond just 50-100 cycles [29, 66, 78–79]. In [75], intermediate cleaning of the test probe was required to regain the maximum value of the friction force after just 8 test cycles. In this study, an adhesive has been fabricated that demonstrates very high durability when tested over 10,000 continuous cycles.

10,000 repeated load-drag-pull tests were performed, all at the maximum preload of 0.18 N and maximum sliding distance in the +y direction of 0.2 mm, sufficient for the adhesive to attain large shear adhesion and maximum shear force values. Adhesion and friction forces were recorded in each test, and the results are plotted in Figure VI – 8(a), showing a 23% decrease in adhesion after shearing and an 18% increase in shear force after 10,000 cycles without intermediate cleaning of the glass puck or sample. Although it could be argued that the increase in shear force supported with repeated use is beneficial, the decrease in normal adhesion could be problematic in applications that require secure attachment to both vertical and inverted surfaces. The adhesive material was inspected in an SEM after durability testing, and no significant damage to individual fibers, or any inter-fiber self-adhesion, was observed over the entire test area, as seen in Figure VI – 8(b).

Dynamic measurements of the normal and shear forces during durability testing are shown in Figure VI – 9, where traces for every intermediate 500th test cycle are plotted to show the evolution of the forces over 10,000 continuous cycles.
Figure VI - 8: (a) Repeatability tests for shear adhesion and shear forces at a preload of 0.18 N and a shear length of 0.2 mm over 10,000 continuous testing cycles without intermediate cleaning. The adhesive retained 77% of its initial shear adhesion while shear force increased by 18% from the initial value. (b) No damage to individual fibers, or inter-fiber self-adhesion, was observed in SEM images of the sample test area after durability testing.
Figure VI - 9: Dynamic normal and shear forces generated by the adhesive for a 180 mN preload and 200 µm shear length in the gripping direction, measured over 10,000 test cycles (Figure (a)). The shear force trace for test cycle #1 is shown with a bold dashed red line, for cycle #10,000 with a bold continuous red line, and for every 500th intermediate test cycle with a continuous green line. Details of the normal force adhesion jump are shown in Figure (b). The normal force trace for test cycle #1 is shown with a bold dashed blue line, for cycle #10,000 with a bold continuous blue line, and for every 500th intermediate test cycle with a continuous black line.
Similar qualitative behavior - with strong adhesion and shear force generation after sliding in the gripping direction - is observed throughout Figure VI – 9(a). It is to be noted that the value for shear force increases from 85 kPa (for cycle #1) to 100 kPa (for cycle #10,000), bringing it within the range of values reported for gecko toes and feet [10, 25]. The higher initial rate of decrease in adhesion shown by traces for cycles 1 and 500 than between subsequent traces (Figure VI – 9(b)) is consistent with a dynamic equilibrium material transfer mechanism between the PDMS fibers and the glass puck over a repeated contact [133].

Even though tests were performed in ambient condition, almost no fouling due to dust particles was observed after two weeks of testing (Figure VI – 10).

**Figure VI - 10: Optical microscope image of a portion of the sample test area after 10,000 continuous test cycles in ambient air. Almost no fouling due to dust particles is observed, with nearly all defects observed being pre-existing periodic lithographic defects from the microfabrication process.**
In general, a high value of $\mu'$ (= 4.7 for this adhesive) could play an important part in enhancing reusability by preventing damage to the fibers during preloading. Even at the high preloads intentionally applied during durability testing, PDMS being an elastomer with a low Young’s modulus of 1.8 MPa, but with a large elongation at break of 160% and toughness of 4.77 MJ/m$^3$ [92], high durability could result from the material’s ability to bend and deform without fracturing or tearing. Further, as stated earlier in Section C.2, no macroscale stick-slip was observed during sliding. Macroscale stick-slip is typically an undesirable behavior that often results in the damage and wear of materials in sliding systems [31]. As suggested in [15] to explain the high wear resistance of both natural and synthetic fibrillar adhesives, the absence of macroscale stick-slip with just uncorrelated random microscale stick-slip events could explain the durability of the adhesive in this study. The slight decrease in adhesion, as mentioned earlier, could be associated with modification of the surface properties of PDMS and the glass puck due to material transfer over a repeated contact [133]. Others, [65], have reported an increase in shear force generated by vertical polypropylene microfiber arrays over repeated test cycles, and suggested that this could be due to increased side contact between the fibers and the substrate due to angling of some of the fibers with use. However, the angling of the polypropylene fibers was reported to be non-permanent, with the fibers recovering to a near-vertical state within several hours of unloading. In this study, SEM examination of the PDMS fibers after durability testing was performed after a lapse of one week. Although no permanent change in tilt angle was observed,
a mechanism similar to the one reported in [105] could be responsible for the small increase in shear force supported over 10,000 test cycles.

The durability of the adhesive in this study is compared with adhesives from two other references that were also tested over 10,000 cycles or more [68, 80] in Table VI – 2. While the percentage change in adhesion and shear pressures are greater than those observed for the PDMS wedge shaped adhesive in [68], the material in this study generates 184% the shear adhesion pressure, 312% the shear pressure, and 224% the value of $\mu'$ reported in [68]. Also, the wedge shaped adhesive material showed no anisotropy in adhesion or friction behavior when sheared towards and against the wedge tilt angle despite structural anisotropy. In [80], large adhesion and shear pressures were supported by both polypropylene (PP) and HDPE fiber adhesives. However, tests were performed with all fiber samples mounted in a loop-against-flat configuration to increase compliance. For a similar PP fibrillar material, this configuration was found to yield a 3 to 4 fold increase in maximum shear stress generated when compared with a previously used flat-on-flat configuration [65]. Thus, a direct comparison with the adhesive in the current study, tested in a flat-on-flat configuration, is not straightforward. However, despite the increased compliance of the loop, the value of $\mu'$ for HDPE fibers was 49% lower than the current study. Large losses in adhesion and shear forces were observed over 10,000 test cycles, with significant wear seen after just 300 cycles. For the PP fiber adhesive, a complete loss of adhesion was reported, along with poor repeated shear force performance. Further, the value of $\mu'$ for PP fibers was very low at ~0.3.
Table VI - 2: A comparison of the performance of gecko-inspired adhesives from the current study and from references [68] and [80]. In [80], significantly poorer durability than the current study was reported for both polypropylene (PP) and HDPE fibers. While higher shear pressures were reported for both HDPE and PP fiber loops, this was at significantly lower values of \( \mu' \) than the current study. Higher durability was reported in [68] than in both [80] and the current study, but maximum adhesion, shear and \( \mu' \) values were all lower than in the current study.

<table>
<thead>
<tr>
<th>Material [Reference]</th>
<th>Young's Modulus (MPa)</th>
<th>( \mu' )</th>
<th>Maximum Adhesion Pressure (kPa)</th>
<th>Maximum Shear Pressure (kPa)</th>
<th>No. of Cycles Tested</th>
<th>Wear Observed?</th>
<th>Force Retained After 10000 Cycles</th>
</tr>
</thead>
<tbody>
<tr>
<td>PDMS, Sylgard 184 [Current Study]</td>
<td>1.8</td>
<td>4.7</td>
<td>9.3</td>
<td>78</td>
<td>10000</td>
<td>No</td>
<td>77%</td>
</tr>
<tr>
<td>Polypropylene [80]</td>
<td>1500</td>
<td>~0.3</td>
<td>~13</td>
<td>300</td>
<td>10000</td>
<td>Yes</td>
<td>0%</td>
</tr>
<tr>
<td>HDPE [80]</td>
<td>400</td>
<td>2.4</td>
<td>36</td>
<td>286</td>
<td>10000</td>
<td>Yes</td>
<td>~36%</td>
</tr>
<tr>
<td>PDMS, Sylgard 170 [68]</td>
<td>1.75</td>
<td>2.1</td>
<td>5.1</td>
<td>25</td>
<td>30000</td>
<td>No</td>
<td>~86%</td>
</tr>
</tbody>
</table>

Since no inter-fiber self-adhesion was observed after 10,000 cycles in this study, it is therefore possible to increase the fiber areal density in future designs to further increase both adhesion and friction forces without compromising fiber integrity. The current microfiber density is 5128 mm\(^{-2}\), chosen as a conservative first iteration, and significantly less than 14,400 mm\(^{-2}\) for gecko setae [6, 134]. However, the durability of the synthetic adhesive in this study is still lower than that of natural gecko setae as
reported in [15], where a 25% increase in adhesion and just 5% decrease in shear force were observed after 30,000 load-drag-pull tests. In addition to its effect on adhesion and friction anisotropy, the use of polymers of varying elastic moduli in future designs will be studied for its effect on, and to maximize, adhesive durability.

D. Conclusion

Arrays of angled semicircular PDMS microfibers, when coupled with a suitable articulation mechanism, may be used as a gecko-inspired reversible dry adhesive that displays high and anisotropic dynamic shear and adhesion forces, a high ratio of normal adhesion to initial preload, and high durability exceeding 10,000 continuous operating cycles. The normal adhesion forces obtained experimentally are close to theoretical approximations from the Kendall peel model, quantitatively demonstrating that both fiber anisotropic shape and tilt angle are crucial in obtaining highly anisotropic adhesion after lateral sliding with synthetic adhesives, thereby offering clear guidelines for future design of adhesives with even higher anisotropy. The lack of wear or inter-fiber self-adhesion makes this adhesive a good candidate for future use in supporting large payloads in high-cycle applications such as robotics, industrial grippers, and mobile sensor platforms. Ongoing work is aimed at achieving high forces in real-world wall hanging tests with cm-size scale patches, before integration into a climbing robot prototype.
VII. Conclusions and Future Work

This thesis presents a novel microfabrication method to fabricate vast arrays of tilted micron-scale polymer structures over cm-scale patch areas with very few defects using an angled photolithography technique. Two types of tilted structures have been fabricated – rectangular flap-like structures and half-cylinder microfibers. Experimental results demonstrate how adhesion and friction anisotropy can be obtained with both types of synthetic microfiber arrays, with strong friction and adhesion forces when sheared along the tilt direction, and low friction and adhesion forces when sheared against the tilt direction. This also appears to be the method employed by geckos allowing them to run on (vertical) walls and (horizontal) ceilings: the anisotropic, directionally tilted setae provide both strong adhesion and friction for attachment (in the gripping stage) and weak adhesion and friction for detachment (in the releasing stage) during a step cycle [17, 18].

This work also presents the first experimental effort to systematically study the effect of nano-scale surface roughness on both the adhesion and friction of gecko-inspired structured adhesives. The results presented demonstrate that suitable surface roughness could effectively enhance the adhesion and friction between tilted PDMS flaps and a rough/patterned substrate in comparison to a smooth surface, most likely due to a good geometric match between the flaps and the surface structure. Although the results of this work cannot be directly applied to all gecko-inspired adhesive structures, they clearly illustrate the importance of load, surface roughness and
structure, adhesive fiber anisotropy, and direction of sliding. In order to develop better structured adhesives for gecko robots, additional systematic studies on the effect of surface structure are required.

The angled semicircular PDMS microfiber arrays displayed very high durability exceeding 10,000 continuous operating cycles in addition to high and anisotropic shear and adhesion forces, and a high ratio of normal adhesion to initial preload. The normal adhesion forces obtained experimentally were found to be close (< 25% error) to theoretical approximations from the Kendall peel model, quantitatively demonstrating that both fiber anisotropic shape and tilt angle are crucial in obtaining highly anisotropic adhesion after lateral sliding with synthetic adhesives, thereby offering clear guidelines for future design of adhesives with even higher anisotropy.

A comparison of the natural gecko adhesive system with several key synthetic gecko-inspired systems has been shown in Table VII – 1 (for adhesion performance) and Table VII – 2 (for shear/friction performance). The natural system outperforms all polymeric synthetic adhesives with a high value of $\mu'$ and very high durability (Table VII – 1). Higher values of $\mu'$ with synthetic adhesives have thus far been only reported for carbon nanotube-based systems – however, these systems do not exhibit the crucial properties of controllable adhesion and anisotropic friction, and have hence been excluded from the tabular comparison [135]. While the angled microflaps presented in this thesis have lower values of normal adhesion pressure and $\mu'$ than other synthetic alternatives, they have the advantage of having both a non-sticky
default state as well as directional adhesion (controlled by direction of shearing articulation). The angled half-cylinder microfiber arrays combine the advantages of a large $\mu'$ value with directional adhesion and high durability. While the adhesive system reported by the Sitti Group [78] has both a high normal adhesion pressure as well as a large $\mu'$ value, it has not been tested for durability beyond 100 cycles. Also, tests for normal adhesion were performed using a hemispherical puck over a small patch area that is just $\sim 3.5\%$ of the test contact area using the Bio-F tester [82]. Adhesion values for small patch areas often tend to be higher for small patch areas due to ease of making good contact between two surfaces over small areas [10, 11, 68].

Table VII – 2 shows a comparison between natural and synthetic systems for shear/friction force behavior. In tests on setal arrays in an SFA, the ratio between shear stress for the gripping and releasing directions was found to be 5.0 [17]. They also survived over 30,000 test cycles with just a 5\% reduction in shear force [15]. The polypropylene nanofiber adhesive reported in [65] supported a high shear force, but did not exhibit any anisotropy based on shearing direction. Further, some fibers were observed to be plastically deformed after just 50 test cycles. While the PDMS micro-wedge fibers reported in [68] had very high durability, they also did not exhibit any anisotropy for shearing articulation with and against the micro-wedge tilt angle. The PUA hierarchical microfiber adhesive reported by the Suh Group was not tested for durability beyond 50 cycles [29]. The angled micro-flaps exhibited anisotropic friction in tests in both the SFA and Bio-F apparatuses. In the Bio-F
tests, it was observed that the adhesive would lose its non-sticky default state after 240 cycles [69]. Since this is a crucial property of the natural gecko adhesive, the durability of the adhesive is taken to be approximately 240 cycles. The angled half-cylinder microfibers combine the advantages of high and anisotropic friction with durability over 10,000 cycles [82]. No fiber wear was observed after durability testing using both an optical microscope and SEM.

The anisotropy built into the tilted PDMS micro-flap design allows for controllability and switchability by varying the flap dimensions, density, tilt angle, and shearing direction. The fill factor for the initial design presented in Chapters III and IV, however, is low at ~18% for flap deflection with the entire face in contact with the substrate. Adhesion and friction forces achieved could be considerably increased from the values obtained so far by using a mask design with higher fill factor. SFA tests on angled micro-flap arrays with a significantly higher fill factor (Figure VII – 1(a) & (b)) of ~65% are ongoing, and initial adhesion test results show a 7 to 8 fold increase in pull-off force over the previous low fill factor micro-flap arrays (Figure VII – 1(c)).
Table VII - 1: A comparison of the performance of several natural and synthetic adhesives for normal adhesion.

Adhesives reported in this thesis are highlighted with grey-shaded boxes.

<table>
<thead>
<tr>
<th>Adhesive System [References]</th>
<th>Material; Structure</th>
<th>Adhesion Pressure (kPa)</th>
<th>$\mu' = \text{Pull-off force to preload ratio}$</th>
<th>Area tested (cm²)</th>
<th>Durability (cycles)</th>
<th>Non-sticky Default State?</th>
<th>Directional Adhesion?</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sitti Group [78]</td>
<td>PUA (E = 3 MPa); Angled microfibers with mushroom tips</td>
<td>80</td>
<td>7</td>
<td>0.0044 [9]</td>
<td>100</td>
<td>No</td>
<td>Yes</td>
</tr>
<tr>
<td>Cutkosky Group [68]</td>
<td>PDMS (E = 1.75 MPa); Micro-wedge fibers</td>
<td>5.1</td>
<td>2.1</td>
<td>1</td>
<td>$&gt;30,000$</td>
<td>Yes</td>
<td>No</td>
</tr>
<tr>
<td>Gorb et al. [61]</td>
<td>PVS (E = 3 MPa); Vertical microfibers with mushroom tips</td>
<td>60.6</td>
<td>7.1</td>
<td>0.066</td>
<td>Not tested</td>
<td>No</td>
<td>No</td>
</tr>
<tr>
<td>Turner &amp; Israelachvili [67, 69]</td>
<td>PDMS (E = 1.8 MPa); Angled micro-flaps</td>
<td>1.8 [67]</td>
<td>0.33 [67]</td>
<td>0.01 [67]</td>
<td>Not tested [67]</td>
<td>Yes [67]</td>
<td>Yes [67]</td>
</tr>
<tr>
<td></td>
<td></td>
<td>3.7 [69]</td>
<td>0.7 [69]</td>
<td>0.126 [69]</td>
<td>Not tested [69]</td>
<td>Yes [69]</td>
<td>Yes [69]</td>
</tr>
<tr>
<td>Turner Group [81, 82]</td>
<td>PDMS (E = 1.8 MPa); Angled half-cylinder microfibers</td>
<td>9.4 [81, 82]</td>
<td>4.7 [81, 82]</td>
<td>0.126 [81, 82]</td>
<td>$10,000$ [81, 82]</td>
<td>No [81, 82]</td>
<td>Yes [81, 82]</td>
</tr>
</tbody>
</table>
Table VII - 2: A comparison of the performance of several natural and synthetic adhesives for shear/friction force.

Adhesives reported in this thesis are highlighted with grey-shaded boxes.

<table>
<thead>
<tr>
<th>Adhesive System [References]</th>
<th>Material; System</th>
<th>Anisotropic Friction?</th>
<th>Forward Shear Stress (kPa)</th>
<th>Backward Shear Stress (kPa)</th>
<th>Area tested (cm²)</th>
<th>Durability (cycles)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fearing Group [65]</td>
<td>Polypropylene (E = 1.0 GPa); Vertical nanofibers</td>
<td>No</td>
<td>20</td>
<td>20</td>
<td>2</td>
<td>Fiber plastic deformation seen after 50 cycles</td>
</tr>
<tr>
<td>Sitti Group [78]</td>
<td>PUA (E = 3 MPa); Angled microfibers with mushroom tips</td>
<td>Yes</td>
<td>74</td>
<td>20</td>
<td>1 (Patch on glass)</td>
<td>&gt;100</td>
</tr>
<tr>
<td>Cutkosky Group [68]</td>
<td>PDMS (E = 1.75 MPa); Micro-wedge fibers</td>
<td>No</td>
<td>25</td>
<td>25</td>
<td>1</td>
<td>&gt;30,000</td>
</tr>
<tr>
<td>Suh Group [29]</td>
<td>PUA (E = 19.8 MPa), Vertical microfibers with angled tips</td>
<td>Yes</td>
<td>90</td>
<td>20</td>
<td>3 (Patch on glass)</td>
<td>&gt;50</td>
</tr>
<tr>
<td>Turner &amp; Israelachvili [67, 69]</td>
<td>PDMS (E = 1.8 MPa); Angled micro-flaps</td>
<td>Yes [67]</td>
<td>45 [67]</td>
<td>28 [67]</td>
<td>0.01 [67]</td>
<td>Not tested [67]</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Yes [69]</td>
<td>22 [69]</td>
<td>14 [69]</td>
<td>0.126 [69]</td>
<td>~240 [69]</td>
</tr>
<tr>
<td>Turner Group [81, 82]</td>
<td>PDMS (E = 1.8 MPa); Angled half-cylinder microfibers</td>
<td>Yes [81, 82]</td>
<td>78 [81, 82]</td>
<td>53 [81, 82]</td>
<td>0.126 [81, 82]</td>
<td>&gt;10,000 [81, 82]</td>
</tr>
</tbody>
</table>
Figure VII - 1: (a) SEM image of PDMS angled micro-flaps with a higher areal density (6410 flaps/mm$^2$) than previously used (1850 flaps/mm$^2$). (b) Schematic top-view orthographic drawing of the flaps in the array. (c) Tests on the high areal density flap array in an SFA show a 7 to 8 fold increase in adhesion when compared with the lower areal density array, while still maintaining anisotropy.
After 10,000 continuous operating cycles, the angled half-cylinder microfiber array retained up to 77% of the initial normal adhesion and also showed an 18% increase in shear force supported, making it a good candidate for future use in repeatedly supporting large payloads. Also, since no individual fiber wear or inter-fiber self-adhesion was observed after 10,000 cycles, it is therefore possible to increase the half-cylinder fiber areal density in future designs to further increase both adhesion and friction forces, in a manner similar to the angled micro-flaps. The current half-cylinder microfiber density is 5128 mm\(^2\), chosen as a conservative first iteration, and significantly less than 14,400 mm\(^2\) for gecko setae [6, 134].

The tests in the Bio-F apparatus confirmed that even at larger patch sizes, the angle micro-flap adhesive was able to achieve strong adhesion while retaining its desirable anisotropic properties. However, even these patch sizes are small at 12.6 mm\(^2\), and significantly larger patch sizes will be needed to support large payloads in real-world applications. Tests are currently being conducted on both angled micro-flaps as well as angled half-cylinder microfibers using patch sizes of >100 mm\(^2\) against a glass slide in the Turner Lab to verify further patch size scale-up. Further, in order to adhere well to a variety of real-world surfaces like the gecko, the synthetic adhesive will have to be able to conform to several scales of roughness, from the mm-scale to the nm-scale. Previous work in the Turner Group has also experimentally shown the advantage in combining at least two length scales (micro- and nano-scales) of features for higher adhesion than simply with either one [39]. In that study, nano-scale features were easily integrated on to micro-scale features by
growing nanorod structures in an unpatterned SPR 220-7 photoresist layer exposed to an ICP oxygen plasma [136]. With process modifications such as plasma chemistry, bias power, and chamber pressure, this process has since been extended to yield high areal density (~10^6 nanorods/mm^2) vertical nanostructures in three new materials – NR9-8000, AZ 9245 and SU-8 photoresists (Figure VII – 2).

**Figure VII - 2: SEM images of vertical polymer nanorod structures grown in a previously unpatterned NR9-8000 photoresist (a) and SU-8 photoresist (b). The nanorods have a height ~3 µm and radius ~300 nm.**

Nanostructure growth has also been previously reported in PDMS [137]. However, nanorods in PDMS have not been integrated over micron-scale structures and used as an adhesive. Angled flap or angled half-cylinder PDMS microstructures would provide controllability based on shear direction as previously shown, while the nanorods could help enable the hierarchical adhesive to better conform to nano-scale substrate roughness.
Before future integration into a climbing robot prototype, a further level of hierarchy at a larger size scale than the microfibers can be added by fabricating a dual-layer adhesive. A previously reported dual layer adhesive was fabricated by directly gluing an array of microscale PDMS wedges over another array of mm-scale tilted polymer stalks, and showed significantly higher adhesion than either of the single layers on rough real-world surfaces such as granite and wood [106]. A simple dual-layer adhesive has also been fabricated at UCSB using PDMS to glue the two layers together (Figure VII – 3), with features on the size scale of both a few microns and hundreds of microns. Vertical rectangular micro-flap structures have been used for the larger size-scale supporting layer so far. Tests are planned on vertical glass slides as well as other real-world surfaces using this material.

Figure VII - 3: (a) SEM image showing a dual-layer adhesive, with vertical PDMS flaps of ~120 µm height supporting a top layer with a backing thickness of ~110 µm. (b) The top layer consists of an array of vertical semicircular fibers of ~19 µm height.
In addition to focusing on adhesive structure, optimal performance of the adhesive will also depend on the use of an optimized articulation mechanism. Tests performed thus far in the SFA have all used a normal preloading step (with the direction of puck motion being perpendicular to the fiber array backing layer), followed by lateral shearing at 10-20 μm/s (when shear/friction force is measured), and then normal retraction to measure the pull-off force. This is also the case with tests on the larger scale Bio-F tester, except that the speed during shearing is 3 μm/s.

The effect of articulation on adhesive performance has not received much attention in the literature either. Recent tests on vertical half-cylinder microfiber arrays in the Bio-F apparatus using various angles of approach from a minimum of 2.5° to a maximum of 177.5° have shown that it is possible to significantly reduce the initial repulsive forces during preloading while still making good contact with the test substrate. This is done by using a low angle of approach, and results in a large increase in the value of $\mu'$ that can be achieved, by up to 38 fold, while still generating high and anisotropic adhesion and friction forces [138]. The extent to which other articulation parameters such as retraction angle and sliding speed influence the adhesion force, shear force, and $\mu'$ value need to also be fully experimentally characterized. An optimal stick-and-peel articulation strategy could then be programmed into the motion controller of a climbing robot to achieve high performance.

Tests are also currently ongoing to understand the effect of sliding speed on the friction force generated by angled micro-flap arrays using an SFA. In these tests,
sliding speed is being varied over several orders of magnitude, from 40 nm/s to 100 µm/s. In addition to information on the influence of sliding speed on friction force *magnitude*, these tests also provide information on the friction force *regime*, whether smooth or stick-slip. Recent experimental results from SFA tests on articular cartilage showed that normal load and sliding speed influence the nature of the friction regime, and that damage generally occurs in the stick-slip regime [139]. Characterization of the angled micro-flap adhesive in a similar manner could yield information on optimal load-speed regimes that limit damage and promote reusability.

**Figure VII - 4: Friction map showing the stick-slip and smooth sliding regimes for PDMS angled micro-flap arrays for sliding in both the direction with tilt (+y) and the direction against tilt (–y).**
Beetles utilize two layers of interlocking microscale hair – one layer on their backs and the other on the underside of their wings – to lock their wings in place with high shear forces when not in use [116]. However, to free their wings for use, separation of these interlocking fibrillar layers is achieved with low normal pull-off forces, unlike Velcro (itself also bio-inspired) which requires high peel forces. Inspired by this natural system, others have fabricated interlocking arrays of microscale polyurethane fibers with isotropic [117] and anisotropic [140] shear properties. A directional, reversible interlocking system is potentially useful for many applications including a fastener-type adhesive, and for switchable adhesion on specially prepared substrates. However, the method used thus far to fabricate interlocking adhesives with anisotropic shear properties requires a complex process.

Figure VII - 5: SEM images of PDMS chevron micro-flaps (a) and PDMS micro-pillars (b) designed to interlock with anisotropic shear forces. Shearing the pillars towards the open face of the chevron flaps (+y direction) results in trapping and high shear forces, while shearing the pillars towards the spine of the flaps (−y direction) does not result in trapping, and lower shear forces.
In order to simplify the process and fabricate an interlocking adhesive with anisotropic shear forces, vertical PDMS microfiber arrays with anisotropic fiber shape have been fabricated (Figure VII – 5). When the cylindrical micro-pillar layer is sheared in the $+y$ direction (towards the open face of the chevron-shaped flaps), the pillars could be ‘trapped’ due to the anisotropic shape of the flaps, resulting in high shear forces. When sheared in the $-y$ direction, however, this trapping effect does not occur, and shear forces are expected to remain low. Microfabrication is complete, and tests are planned in the SFA using a crossed-cylinder geometry.

In summary, a variety of bio-inspired polymer microstructure arrays have been fabricated and tested to show how suitable articulation can effectively switch adhesion and friction from a high value for gripping to a low value for easy release. Further improvements are planned that incorporate higher areal density features as well as several levels of structural hierarchy to enable adhesion on a wide variety of real-world rough surfaces. In order to better model the synthetic adhesives, suitable models incorporating the effect of fiber beam bending energy and interfacial slip will be considered in addition to the Kendall peel model [141, 142]. In addition to the effect of substrate surface roughness on adhesion and friction, the effects of articulation trajectory, sliding speed and normal load are being investigated to determine an optimal stick and release strategy before integration into a climbing robot prototype.
References


[48] J. H. de Boer, "The influence of van der Waals' forces and primary bonds on binding energy, strength and orientation, with special reference to some artificial resins," *Transactions of the Faraday Society*, vol. 32, pp. 10-37,
1936.


12, pp. 4559-4563, 1996.


IEEE, Limerick, Ireland, 2011.


### Appendix

Table A - 1: Process sequence – fabrication of a bilayer photoresist negative mold for casting angled micro-flap arrays.

<table>
<thead>
<tr>
<th>Step</th>
<th>Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>Clean: 1165/PRX-127/DI Rinse/Bake</td>
<td>Bake for 5 min @ 115 C</td>
</tr>
<tr>
<td>Spin Coat HMDS</td>
<td>4000 rpm, 30 sec</td>
</tr>
<tr>
<td>Spin Coat PMGI SF-15</td>
<td>1000 rpm, 45 sec</td>
</tr>
<tr>
<td>1st PMGI Coat Soft Bake</td>
<td>200 C, 1 min</td>
</tr>
<tr>
<td>Spin Coat PMGI SF-15</td>
<td>1000 rpm, 45 sec</td>
</tr>
<tr>
<td>2nd PMGI Coat Soft Bake</td>
<td>200 C, 2 min</td>
</tr>
<tr>
<td>Spin Coat AZ 5214</td>
<td>4000 rpm, 30 sec</td>
</tr>
<tr>
<td>Soft Bake AZ 5214</td>
<td>Ramp to 95 C from 50 C, Hold at 95 C for 2 min, Ramp Down to 75 C</td>
</tr>
<tr>
<td>Stepper (GCA 6300) UV Lithography</td>
<td>Exposure for 0.5 sec, Focus offset = 0</td>
</tr>
<tr>
<td>Post Exposure Bake</td>
<td>Ramp to 110 C from 50 C, Hold at 110 C for 1 min, Ramp Down to 85 C</td>
</tr>
<tr>
<td>UV Flood Exposure</td>
<td>1 min, any contact aligner may be used (dosage not exact, to exceed threshold)</td>
</tr>
<tr>
<td>Develop</td>
<td>AZ 400K Developer, Diluted 1:4, 1 min</td>
</tr>
<tr>
<td>Post Develop Bake</td>
<td>Ramp to 110 C from 50 C, Hold at 110 C for 1 min, Ramp Down to 85 C</td>
</tr>
<tr>
<td>1st Angled Deep UV Exposure</td>
<td>300 sec @ 1000 W</td>
</tr>
<tr>
<td>1st PMGI Develop</td>
<td>SAL 101 Developer, 70 sec</td>
</tr>
<tr>
<td>2nd Angled Deep UV Exposure</td>
<td>300 sec @ 1000 W</td>
</tr>
<tr>
<td>2nd PMGI Develop</td>
<td>SAL 101 Developer, 70 sec</td>
</tr>
<tr>
<td>3rd Angled Deep UV Exposure</td>
<td>300 sec @ 1000 W</td>
</tr>
<tr>
<td>3rd PMGI Develop</td>
<td>SAL 101 Developer, 70 sec</td>
</tr>
<tr>
<td>4th Angled Deep UV Exposure</td>
<td>300 sec @ 1000 W</td>
</tr>
<tr>
<td>4th PMGI Develop</td>
<td>SAL 101 Developer, 70 sec</td>
</tr>
<tr>
<td>5th Angled Deep UV Exposure</td>
<td>300 sec @ 1000 W</td>
</tr>
<tr>
<td>5th PMGI Develop</td>
<td>SAL 101 Developer, 70 sec</td>
</tr>
<tr>
<td>6th Angled Deep UV Exposure</td>
<td>300 sec @ 1000 W</td>
</tr>
<tr>
<td>6th PMGI Develop</td>
<td>SAL 101 Developer, 70 sec</td>
</tr>
<tr>
<td>PMGI Hard Bake</td>
<td>Ramp to 110 C from 50 C, Hold at 110 C for 10-20 min, Ramp Down to 85 C</td>
</tr>
<tr>
<td>Oxygen Plasma Surface Activation</td>
<td>100 W, 300 mTorr, 3 min (PE-II Sys.)</td>
</tr>
<tr>
<td>Silane Layer Deposition and Bake</td>
<td>Program ‘FDTS’ on MVD (10 min dep.) Bake @ 100 C, 3 min</td>
</tr>
</tbody>
</table>
Figure A - 1: Top view optical microscope images of the angled half-cylinder microfibers in contact with a flat glass puck. The darker regions in the images are in contact. Figure (a) shows the contact before lateral sliding, Figure (b) after sliding in the $+y$ direction (towards the flat face and with tilt), and Figure (c) after sliding in the $-y$ direction (towards the curved face and against tilt).