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Effect of nano- and micro-roughness on adhesion of bioinspired micropatterned surfaces

Natalia Cañas [a], Marleen Kamperman [b,*], Benjamin Völker [c], Elmar Kroner [a], Robert M. McMeeking [a,c,d,e], Eduard Arzt [a]

[a] INM – Leibniz Institute for New Materials, Functional Surfaces Group and Saarland University, Campus D2 2, 66123 Saarbrücken, Germany
[b] Wageningen University, Physical Chemistry and Colloid Science, Dreijenplein 6, 6703 HB Wageningen, The Netherlands
[c] Department of Mechanical Engineering, University of California, Santa Barbara, CA 93106, USA
[d] Materials Department, University of California, Santa Barbara, CA 93106, USA
[e] School of Engineering, University of Aberdeen, King's College, Aberdeen AB24 3UE, UK

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ABSTRACT

In this work, the adhesion of biomimetic polydimethylsiloxane (PDMS) pillar arrays with mushroom-shaped tips was studied on nano- and micro-rough surfaces and compared to unpatterned controls. The adhesion strength on nano-rough surfaces invariably decreased with increasing roughness, but pillar arrays retained higher adhesion strengths than unpatterned controls in all cases. The results were analyzed with a model that focuses on the effect on adhesion of depressions in a rough surface. The model fits the data very well, suggesting that the pull-off strength for patterned PDMS is controlled by the deepest dimple-like feature on the rough surface. The lower pull-off strength for unpatterned PDMS may be explained by the initiation of the pull-off process at the edge of the probe, where significant stress concentrates. With micro-rough surfaces, pillar arrays showed maximum adhesion with a certain intermediate roughness, while unpatterned controls did not show any measurable adhesion. This effect can be explained by the inability of micropatterned surfaces to conform to very fine and very large surface asperities.

1. Introduction

Many insect and lizard species possess adhesive organs on their feet that allow them to adhere to a wide variety of surfaces. The key strategy to control adhesion in these natural systems is the incorporation of fibrillar structures [1–6]. In the particular case of the gecko foot, each fibril or seta is ~100 μm long, has a diameter of a few microns and branches into an array of hundreds of spatula structures. These structures terminate in a triangular plate tip with dimensions of ~0.2 μm in length and a thickness of 10 nm [1]. The gecko uses non-covalent surface forces to achieve adhesion, which relies primarily on van der Waals forces [7].

Because the strength of van der Waals forces strongly decreases with increasing distance between the surfaces, an important aspect in adhesion is the true area of contact. Although surface area is increased by the surface roughness, more elastic strain energy is needed for the adhesion structure to conform to the rough surfaces and make contact. Macroscopic solids normally do not adhere on rough surfaces; a root-mean-square (RMS) roughness of ~1 μm is sufficient to result in negligible adhesion between rubber and a hard flat surface [8]. For purely elastic materials, only very compliant materials (Young's modulus E ~100 kPa) can adhere well on hard rough surfaces, because the elastic energy stored during deformation of the compliant material is low compared to the energy gained by forming a contact [8,9].

Geckos show high adhesion to rough surfaces in spite of the stiff structural material (β-keratin: E ~1 GPa) [10–12]. In this case adhesion is possible, because the hierarchical build-up of the fibrillar structure results in a low effective modulus and allows conformation to rough surfaces by fiber bending and buckling [5,8,13–15]. Despite the ability of geckos to conform to rough surfaces, observations of living geckos show that adhesion strongly decreases for certain roughness values [10–12]. This may explain why geckos seem to have an over-redundant attachment system [16].

Significant decreases in adhesion were also found in the few studies published on biomimetic adhesives using technologically relevant rough surfaces [17–19] or model surfaces with well-defined roughness [19,20]. In all cases, the adhesion decreased with increasing roughness [19,20] and hierarchical structures outperformed single-level structures, but only on rough surfaces [18,20].

* Corresponding author. Tel.: +31 (0)317 482358; fax. +31 (0)317 483777.
E-mail address: marleen.kamperman@wur.nl (M. Kamperman).

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In this work we study the adhesion of polydimethylsiloxane (PDMS) pillar arrays with mushroom-shaped tips on “nano-rough” and “micro-rough” surfaces. These surfaces have RMS roughness values in the nano- and micro-range, respectively. PDMS pillar arrays were fabricated by molding on lithographic molds and roughened Si wafers and sandpaper substrates were used as counter surfaces in adhesion measurements. The results provide new insights on the effects of roughness on the nano- and the micron scale on adhesion of patterned surfaces.

2. Materials and methods

2.1. Sample fabrication

Micropatterned structures were fabricated by demolding PDMS (Sylgard 184, Dow Corning, USA) from structured templates. SU-8 templates (SU-8 from Micro Resist Technology, Berlin, Germany; Si wafers from Crystec Berlin, Germany) with holes of different radii and lengths were obtained by a modified photolithography technique, in which quenching was used to control the pillar tip shape. Process parameters can be found in previous publications [21,22]. Quenching the template, i.e. rapid cooling from 90 °C to room temperature after the photoresist hard-baking step, caused delamination of the SU-8 at the edges of the holes. Silanization with hexadecafluoro-1,1,2,2-tetrahydrooctyltrichlorosilane and subsequent molding PDMS on these templates resulted in pillars with a small thin cap on the tip (mushroom shape). For molding, a 10:1 ratio of Sylgard 184 prepolymer and cross-linker were mixed and degassed in a desiccator for 30 min to eliminate bubbles. The mixture was poured on the template and cured for 24 h at 75 °C and 600 mbar. PDMS samples were then carefully peeled off from the mold and characterized with light microscopy (Olympus BX51) and scanning electron microscopy (SEM) (FEI Quanta 400 ESEM operating at energy between 1 and 15 kV). Micropatterned PDMS adhesives with different pillar lengths (20 and 42 μm) and aspect ratios (length/diameter) (1 and 2) were fabricated (see Table 1). The specimens had a cross-sectional area of 8×8 mm^2.

Fig. 1 shows representative SEM images of PDMS-1 (Fig. 1a and b), PDMS-2 (Fig. 1d) and PDMS-3 (Fig. 1c). The pillars have a ~500 nm ring around the tip (Fig. 1b), resembling a mushroom profile.

2.2. Preparation and characterization of rough surfaces

Silicon wafers and sandpaper with different roughness were selected as probe surfaces. As-received nominally flat wafers, with an RMS roughness of about 2 nm, were chosen as flat probes (probe A). Further, Si wafer pieces, with a square area of 9 mm^2, were roughened with diamond particles and with sandpaper. This resulted in probes with randomly distributed scratches and grooves on the surface (probes B through E). The surfaces were characterized with light microscopy (see Fig. S1 in the Supporting information) and roughness parameters were measured by white light interferometry. The RMS roughness and the peak to valley distance (PV) are listed in Table 2 and Gaussian height distributions are shown in Supporting information, Fig. S2.

Table 1
Micropatterned PDMS characteristics.

Sample	Length (μm)	Diameter (μm)	Aspect ratio	Tip-shape
PDMS-1	20	10	2	Mushroom
PDMS-2	20	20	1	Mushroom
PDMS-3	42	20	2	Mushroom
PDMS-unpatterned	–	–	–	–

Atomic force microscopy (Jeol JSPM 5200) was used to characterize the surfaces with higher resolution using smaller areas (10 μm × 10 μm), see Fig. 2.

The sandpaper substrates (Buehler GmbH, Düsseldorf, Germany) that were used as micro-rough probes were cut into 9 mm^2 pieces and glued on the cantilever without further treatment. Their average particle or asperity diameters were provided by the company and are listed in Table 3.

2.3. Adhesion measurements

Adhesion measurements were performed with a home-built adhesion tester, as previously described in Ref. [23]. The PDMS samples were placed on a stage, while the probe (Si wafer or sandpaper) was glued onto the spring with cyanoacrylate glue (Cyanolube, HK Wentworth Ltd., Derbyshire). The sample was loaded against and retracted from the probe using a hexapod, i.e. a six-axis positioning system that allows controlled displacement with an accuracy of 100 nm. The deflection of the spring was measured with a laser interferometer. The cantilever stiffness was 1095 N m^{-1} and the velocity for each measurement was 5 μm s^{-1}. The temperature and relative humidity (RH) were controlled during experiments and set at ~23 °C and ~50% RH. Since the measurements were performed using a flat probe, a precise alignment procedure had to be carried out to obtain representative and reproducible data [23]. The sample was scanned for maximum pull-off force values by tilting the hexapod along the x-axis and y-axis to determine the parallel configuration. When the position for maximum pull-off force was identified, the pull-off forces were measured for various compressive pre-stresses. The probe was cleaned with ethanol and brought into contact with the sample several times before the actual experiment because the pull-off force measured on PDMS is known to change with the number of contact formations [23].

3. Results

3.1. Nano-rough surfaces

For the experiments on nano-rough probes, patterned PDMS-1 was used. Figs. 3 and 4 show the results of adhesion measurements of unpatterned and micropatterned specimens on Si surfaces with five different roughness values. In Fig. 3, the adhesion results are presented as pull-off strength values as functions of compressive pre-stresses, both of which are derived by dividing the measured force by the nominal area of the probe (9 mm^2). The pull-off strength increased rapidly with increasing pre-stress and plateaued at higher pre-stresses. The highest adhesion was found on the smooth Si wafer (probe A) at a pre-stress above ~3 kPa. Fig. 4 compares adhesion of patterned and unpatterned PDMS: the adhesion strength of patterned PDMS-1 on probe A was nearly five times higher than that of unpatterned PDMS (~20 kPa). Compared to the smooth Si wafer (probe A), a decrease in adhesion by more than 75% was observed for the surface with the lowest roughness (probe B). With increasing roughness, the adhesion dropped further. In all cases, the micropatterned PDMS sample showed higher adhesion than unpatterned PDMS.

3.2. Micro-rough surfaces

Micropatterned PDMS adhesives with different pillar lengths (20 and 42 μm) and aspect ratios (1 and 2) were tested on sandpa-