Supporting Information

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SI Text

Angled Etching System. Fig. S1*A* shows a schematic diagram of the inductively coupled plasma (ICP) etching system used in this study. Separate 13.56-MHz radio-frequency (rf) power generators were used to independently ignite a plasma and control the ion energy. Shown in Fig. S1*B* is a schematic diagram of the Faraday cage system, which consisted of a 15-mm-high cylindrical copper sidewall and a stainless-steel grid top plane. The diameter and the pitch of grid wires were 0.0254 mm and 0.254 mm, respectively. The Faraday cage was fixed on the cathode to have a good thermal and electrical contact. The ion-incident angle (θ) is defined as the angle between the direction of an incident ion and the normal to the substrate surface (Fig. S1*B*).

Angled Etching of polySi Substrates. The polySi wafer was cleaved in a coupon with a dimension of 13 × 39 mm. The polySi substrate was attached on the substrate holder inside the Faraday cage using a silver paste which provided a good thermal and electrical contact between the substrate and the holder. The slope angles of the substrate holders were 30°, 45°, and 60°. The substrate was subjected to isotropic etching in SF_6/Ar plasma at a pressure of 10 mTorr for 210 s. The source power and the self-bias voltage were 450 W and −10 V, respectively. Subsequently, the substrate was etched by using the gas chopping process, which consisted of alternating etching and deposition steps. The deposition process was carried out in C_4F_8/CH_2F_2/Ar plasma at a pressure of 50 mTorr. The source power and the self-bias voltage were 450 W and 0 V, respectively. The flow rates of SF_6, C_4F_8, CH_2F_2, and Ar were 30, 15, 10, and 4 SCCM, respectively. The condition of the etching process was the same as that for the isotropic etching except for a self-bias voltage of −35 V. Each cycle of the gas chopping process consisted of the deposition step for 8 s and the etching step for 80 s. In the case of etch profiles shown in Fig. 1*F*, 16 cycles of the gas chopping process were repeated. After the gas chopping process, fluorocarbon residues were removed from the substrate by ashing in a furnace at 600 °C for 1 h. Finally, the SiO_2 mask and the etch-stop layer were removed by wet etching in an HF solution (HF/H_2O = 1/10). The process time of the wet etching was varied from 15 to 40 s, depending on the desired area of the bottom surface.

Shear Adhesion Tests. The macroscopic shear adhesion strength of the angled nanohairs was evaluated by a hanging test. A flexible adhesive patch (thickness: 50 to ≈60 μm, area = 1 × 3 cm^2) was attached onto the surface under a preload of ≈0.3 N/cm^2, and then the hanging weight was increased until an adhesion failure occurred. To investigate the directional adhesion properties of the adhesive, weight was applied along (forward) and against (reverse) the angled directions of the slanted hairs (see Fig. S2).

Effective Modulus of a Surface with Slanted Nanohairs. In the presence of a slanted angle of nanohairs, the effective modulus of the hairy surface can be reduced. According to a previous study, the effective modulus should be <100 kPa for ensuring a tacky surface (so called "Dahlquist criterion"), which is given by (1)

$$E_{\text{eff}} = \frac{3EID\sin(\theta)}{L^2\cos^2(\theta)[1 \pm \mu\tan(\theta)]} \quad \textbf{[s1]}$$

where E is the elastic modulus, I is the moment of inertia ($\pi R^4/4$, R is the radius of hair), D is the hair density, L is the hair length, μ is the friction coefficient (typically 0.25 for polymer), and θ is the slanted angle. For our slanted structures (E = 19.8 MPa, R = 200 nm, L = 3 μm, and θ = 60°), the effective modulus decreases to ≈26.3 kPa, which meets the Dahlquist criterion (see Fig. S3). This result indicates that our dry adhesive with slanted nanohairs can make intimate contact with the substrate, rendering a tacky surface.

Repeatability of Shear Adhesion. To investigate the durability of dry adhesive, the shear adhesion of the adhesive was measured by repeating cycles of attachment and detachment (thickness: 50 to ≈60 μm, size: 3 × 1 cm^2) against a flat Si surface under a preload of 0.3 N/cm^2. As shown in Fig. S4, the adhesion force was maintained even after more than 50 cycles of attachment and detachment. The average adhesion was 21.6 N/cm^2 and the standard deviation was 2.3 N/cm^2.

Fabrication of Micro-/Nanoscale Hierarchical Nanohairs. Our fabrication method is based on a sequential application of capillary molding for generating micro-/nanoscale hierarchical structures. For fabricating polymer microhairs, a PDMS mold having micropatterns was placed onto the spin-coated, UV-curable PUA resin onto the flexible PET substrate (50 μm thickness). The PDMS mold with micropatterns was prepared by casting PDMS (10% curing agent) against a silicon master having a complementary relief structure prepared by photolithography (Fig. S6*A*). Then the PUA resin was partially cured by UV exposure for 50 s (λ = 250–400 nm, dose = 100 mJ/cm^2). After fabricating a partially cured microstructure, the PUA mold with angled nanopatterns was placed on the preformed microhairy structures under a slight pressure (≈10 g/cm^2), followed by additional UV exposure for 10 s. The PUA mold with angled nanoholes was prepared by self-replication of the angled, PUA nanohairs that had been replicated from the Si master prepared by angled etching (Fig. S6*B*). For a uniform pressure distribution, a thin PDMS block was placed as a buffer on top of the PUA mold before the application of pressure. The resulting hierarchical hairs are very similar to real gecko foot hairs as shown in Fig. S7.

1. Autumn K, Majidi C, Groff RE, Dittmore A, Fearing R (2006) Effective elastic modulus of isolated gecko setal arrays *J Exp Biol* 209:3558–3568.

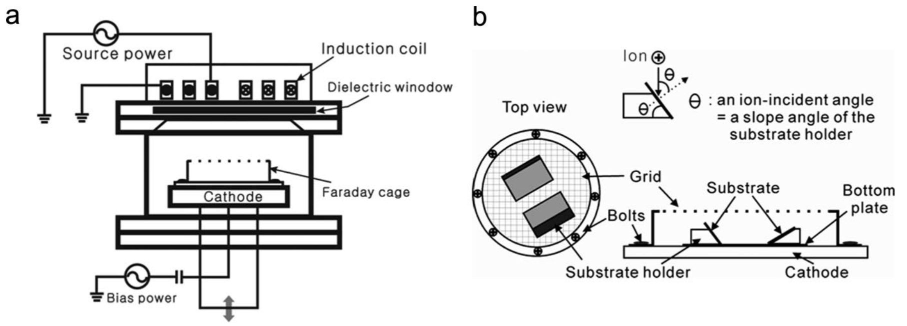


Fig. S1. (*A*) A schematic diagram of the inductively coupled plasma (ICP) etcher. (*B*) A schematic diagram of the Faraday cage system. The Faraday cage was fixed on the cathode, as shown in *A*.

Fig. S2. Schematic illustrations of macroscopic, shear adhesion test, with the definitions of ''forward direction'' and ''reverse direction'' with respect to the angle of slanted hairs.

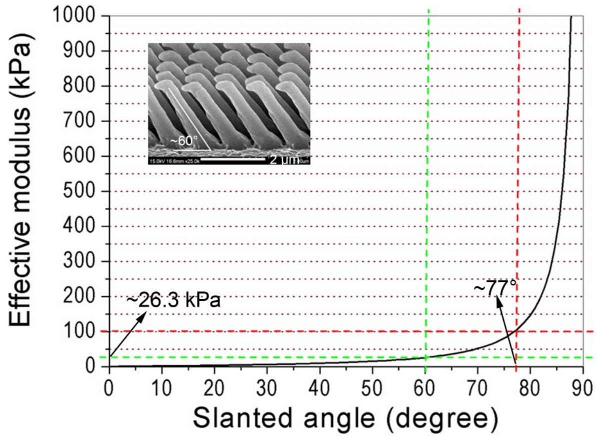


Fig. S3. Effective modulus of nanohairy structure as a function of slanted angle. When the slanted angle reduces to ≈77°, the effective modulus becomes <100 kPa, which meets is the Dahlquist criterion. For leaning angle of 60°, the effective modulus decreases more to ≈26.3 kPa.

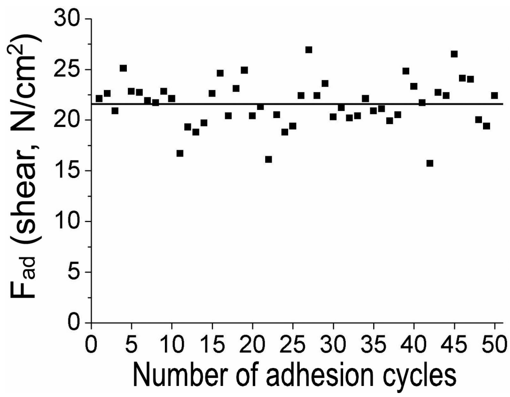


Fig. S4. Measurements of shear adhesion after multiple cycles of attachment and detachment. The adhesion force was remained the same even after 50 cycles.

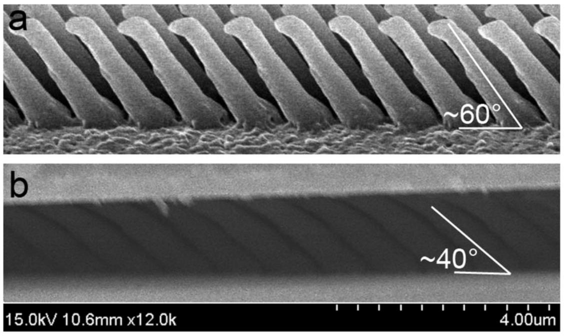


Fig. S5. Titled SEM images of slanted nanohairs before (*A*) and after (*B*) preloading.

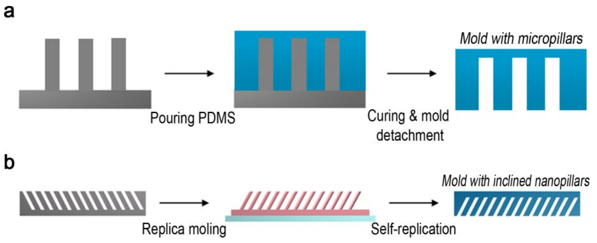


Fig. S6. (*A*) A schematic illustration of fabrication of a micromold used for fabrication of micro-/nanoscale combined hierarchical hairs (*B*) A schematic illustration of the self-replication procedure for generating a nanomold having recessed, slanted nanoholes.

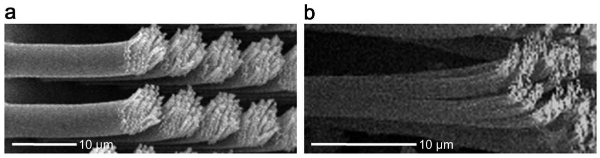
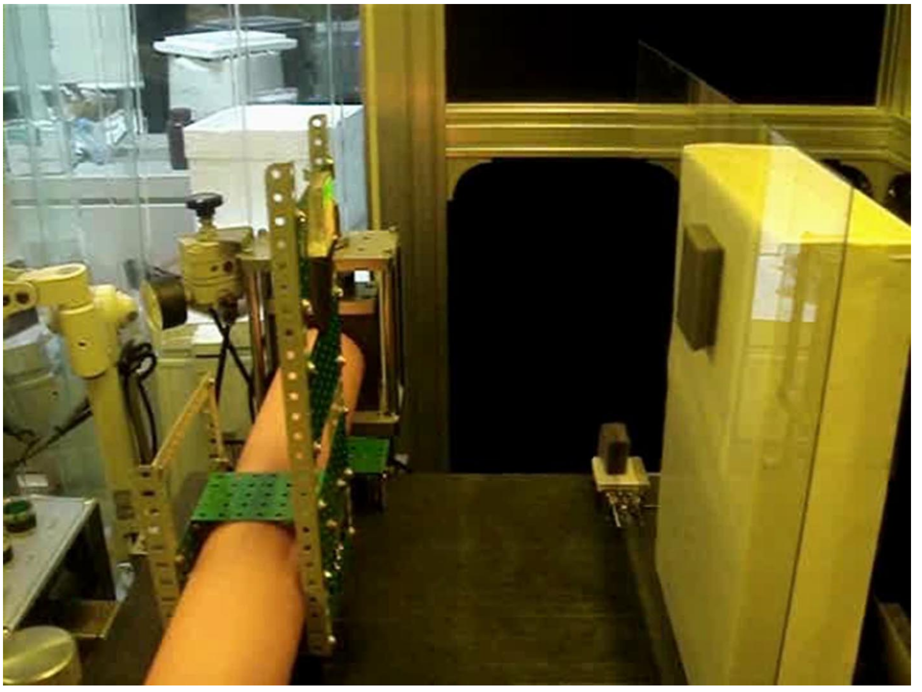


Fig. S7. (*A*) A titled SEM image of micro-/nanoscale combined hierarchical PUA hairs fabricated by a 2-step UV-assisted capillary molding. (*B*) A titled SEM image of real gecko foot hairs, which are very similar to the hierarchical PUA hairs in *A*.



Movie S1. This movie clip shows the transport of a large-area LCD glass by using a dry-adhesive patch with slanted nanohairs. The glass used in the movie is 0.9 mm in thickness, 47.5×37.5 cm^2 in area, and 470 g in weight.

Movie S1 (WMV)