Supporting Information

Simplified Fabrication of Hierarchical Structure for Flexible Pressure Sensor with Wide Linearity Range and Highly Sensitive

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 Surface Microstructures.

In the ABS mold fabrication process, we marked the acrylonitrile butadiene styrene (ABS) mold with smooth surface and rough surface at the laser current of 6A, respectively. After the laser marking, it could be observed that the hierarchical microstructures could be acquired by marking the ABS mold with smooth surface. When the surface of ABS mold possessed rough surface, the hemispherical microstructures were obtained.

To further verify the influence of ABS mold roughness, the ABS mold with smooth surface was polished by 800# sandpaper. From Figure S1a-c, it could be found that ABS mold became rougher after it was polished by sandpaper. The roughness of the ABS mold might influence how the ABS mold and the laser interacted on the surface, and therefore the morphologies of polydimethylsiloxane (PDMS) (peeled off from the polished ABS mold by 800# sandpaper, 800#-PDMS) were investigated. Firstly, the polished ABS mold was marked by laser marking machine. Then the PDMS mixture was drop-casted onto the prepared mold. After curing, the replicated surface structure of the ABS mold was peeled off. As displayed

in Figure S1d-f, it could be observed that the top view of peeled-off 800#-PDMS SEM images were similar to the hemispherical microstructure (Figure S1d). The results suggested that the hierarchical microstructure of the marked ABS mold was significantly associated with the surface roughness of ABS mold.

Furthermore, in the past reports, the researchers have focused on searching the interaction between the laser and the nonmetallic material.¹ They found that the laser marking intensity was closely related to the incident angles of laser. The roughness of the nonmetallic material surface was directly affected by the incident angles of laser. Hence, the roughness of the nonmetallic material surface might also influence the interaction between the laser and ABS mold.



Figure S1. The surface roughness of the rough ABS mold, b), and the smooth ABS mold c) the polished ABS mold by 800# sandpaper. Top-view SEM images of d) the hemispherical microstructure, e) the hierarchical microstructure, and f) the hemispherical microstructure (800#).



Figure S2. a) The surface roughness of the smooth ABS mold, and b) the rough ABS mold.



Figure S3. a) Raman spectra of PDMS, PDMS with GO layers and PDMS with rGO layers.



Figure S4. Top-view SEM images of microstructure with the diameter of, a) 220 μm b) 240 μm c) 260 μm d) 280 μm.



Figure S5 The 3D optical image of the hierarchical microstructure arrays with the pitch of a) 350 μ m, b) 300 μ m.



Figure S6. a) Optical images of hierarchical structure arrays under pressure varied from 1.23 kPa to 253.72 kPa. b) Optical images of hemispherical structure arrays under pressure varied from 0.78 kPa to 92.29 kPa.



Figure S7. Limit of detection of the pressure sensor detemined through loading with pressure of a) 16 Pa, b) 30 Pa, c) 40 Pa, d) 150 Pa.



Figure S8. The humidity stability of the pressure sensor under different humidity storage with 24 hours.

 Table S1. Sensitivity and linearity range of flexible pressure sensors with surface microstructures.

| Sensitivity | Linearity Range | Mold or Surface | Conductive | Dof |
|----------------------|-----------------|-------------------|---------------|------------|
| [kPa ⁻¹] | [kPa] | Microstructure | layer | Kel. |
| 0.0064 | 100 | Semi-sphere | water-based | S2 |
| | | | carbon | |
| 20.08 | 0.8 | Pillar | AgNWs | S 3 |
| 0.02 | 50 | Micro-dome | Carbon powder | S4 |
| 25.1 | 2.6 | Sand paper | rGO | S5 |
| 10 | 0.5 | Banana leaf | Silver | S6 |
| 15 | 5 | Mono-dispersed PS | Gold | S7 |
| 851 | 20 | Microhump | PEDOT:PSS | S 8 |

| 8.5 | 12 | Hierarchical structural | Graphene | S9 |
|------|------|-------------------------|----------------------------|-----|
| 6.8 | 0.25 | zinc oxide nanowire | Pt | S10 |
| 1.35 | 2 | Rose petal | Cu/Ag core-shell nanowires | S11 |

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