Irregular Micro-dome Structure-Based Sensitive Pressure Sensor Using Internal Popping of Microspheres

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Abstract

Modifying the surface morphology of an elastomer surface into three-dimensional (3D) microstructures is crucial for various soft sensor applications. However, processes based on typical mold microfabrication and elastomer casting remain the dominant methodologies. This study demonstrates a novel strategy for generating 3D and irregular micro-dome structures for flexible pressure sensors through the internal popping of microspheres. When thermal treatment is applied to a composite film composed of microspheres and an elastomer matrix, the microspheres expand, and irregular micro-dome structures are generated. This composite film with an irregular micro-dome structure is utilized as a piezoresistive pressure-sensing film. The sensor shows high sensitivity (–50.45 kPa⁻¹) owing to the heterogeneous contact change between the irregular micro-dome structure and a laser-induced graphene electrode; it also exhibits a short response time \((\tau_{10\%-90\%} \approx 39 \text{ ms})\), excellent repeatability, and high reliability. The sensor is applied in a fingertip-shaped pressure sensor to detect the blood pulse on the index finger and on electronic skin to recognize soft objects. Furthermore, a pressure sensor array that enables various functions such as drawing, multi-touch, and deep learning-based motion control, is demonstrated.

1. Introduction

The development of flexible pressure sensors has attracted significant attention owing to their various potential applications in wearable devices,\(^{[1]}\) human-machine interfaces,\(^{[2,3]}\) and health monitoring systems.\(^{[4-6]}\) For many of these applications involving low pressure detection such as wrist pulse or soft touch (0-10 kPa), high sensitivity in the low-pressure range are required. Pressure sensing mechanisms can be divided into piezoresistive,\(^{[7-12]}\) piezo-capacitive,\(^{[13-17]}\) piezoelectric,\(^{[18,19]}\) or piezo-transmittance\(^{[20-22]}\) types. Piezoresistive flexible pressure sensors
convert physical stimuli into a change in electrical resistance by changing the contact area and pressure between electrodes and sensor structures. This is the most widely used type of soft pressure sensor, since it has the advantages of easy operation, high sensitivity in the low-pressure range, simple structure, and fast response.

An effective approach for fabricating a piezoresistive soft pressure sensor structure with high sensitivity in the low-pressure range is to construct microstructures such as pyramids, domes, and pillars. When a physical force is applied to the sensor structure with micro/nano-scale 3D surface morphology, localized stress concentration occurs, inducing a large local strain. Therefore, the sensitivity of the sensor increases in the low-pressure range, and even subtle physical forces can be detected and distinguished. Sensor structures with 3D surface morphology exhibit desirable sensing characteristics, including sensitivity, response time, and limit of detection (LOD). These approaches can be used to manufacture various types of microstructures for high-sensitivity pressure sensor applications.

In general, an effective approach for fabricating a sensor structure with micro/nano-scale 3D surface morphology is the microfabrication of molds and elastomer casting processes. The mold fabrication methods can be divided into the following categories: i) top-down approaches, such as photolithography and silicon etching, and ii) bottom-up approaches using self-assembled particles. For example, Peng et al. fabricated a piezoresistive-based flexible pressure sensor based on conductive PDMS thin films generated by the soft lithography replication of 3D printed templates. The fabricated sensor exhibited a high sensitivity of approximately 3.6 kPa⁻¹. The authors showed that the sensor could detect a subtle pressure of approximately 1 Pa. Zhang et al. proposed microstructures using a self-assembled monolayer as a sacrificial template. The fabricated sensor showed a high sensitivity of approximately 15 kPa⁻¹ in a pressure range of 100 Pa or less and could detect a subtle pressure of approximately 4 Pa.
Although these researches showed that flexible pressure sensors with high sensing performance could be fabricated via conventional microfabrication of molds and elastomer casting, there is still a limitation in fabricating sensor structures with large-scale 3D surface morphology and sensitivity. First, geometrical parameters such as space and size of microstructures should be controlled to enhance the sensing performance. However, abovementioned methods require several iterative processes to fabricate molds such as photolithography and etching, which have low degrees of freedom in the fabrication perspectives. In addition, since regular 3D microstructures are simultaneouslly deformed together under the pressure and quickly saturated, there is a limitation to improve the sensor sensitivity. Therefore, a novel approach for controlling 3D surface morphology with high degrees of freedom in the fabrication process and well-designed irregular 3D microstructures is needed to achieve the fabrication of the highly sensitive pressure sensor.

In this paper, we propose a novel approach to fabricate a sensor structure with a microscale 3D surface morphology via the internal popping of thermally expandable microspheres. When a thermal treatment is applied to a composite film composed of microspheres and an elastomer matrix, the microspheres expand, and irregular micro-dome structures are generated. This method allows not only controllable microscale 3D surface morphology via high degrees of freedom in the fabrication process but also highly sensitive pressure sensor owing to the irregular micro-dome structures. This composite film with an irregular micro-dome structure coated with a conductive metal layer is used as the top active layer, and laser-induced graphene (LIG) acts as the bottom electrode for pressure sensing. Furthermore, finite-element method (FEM) simulations are employed to understand the compressive behavior of irregular micro-dome structures as a function of the pressure. We evaluate the sensing performance during static (I-V curves, limit of detection) and dynamic (reliability, response time, and repeatability) loading, including the sensitivity of the pressure sensors. To demonstrate the potential
applications in electronic skin, a fingertip-shaped pressure sensor was developed to detect the blood pulse on the index finger and recognize soft objects. Finally, we demonstrated that large-area and multi-pixel sensor array pads could realize various functions, such as drawing, multi-touch, and deep learning-based motion controllers.

2. Results and Discussion

2.1 Surface Morphology Control via Internal Popping of Thermally Expandable Microspheres

The mechanism of 3D surface morphology control using the internal popping of thermally expandable microspheres is illustrated in Figure 1. The microspheres consist of thermoplastic resins (acrylonitrile (AN) copolymers) at the outer shell and liquid hydrocarbon (isopentane) at the inside core (Figure 1a). When a specific temperature is applied to the microspheres, the AN copolymer shell becomes soft (state 1), and the hydrocarbon at the inside core undergoes a phase change from liquid to gas (state 2), gasified hydrocarbon starts to permeate through thin outer shell to diffuse into the composites. The volume expansion of the microspheres results by three to six times (Video S1, Supporting Information). Figure 1b shows the field-emission scanning electron microscopy (FE-SEM) images and particle diameter distribution before (noted as “initial state”) and after (noted as “expansion state”) the expansion of microspheres at 170°C. The FE-SEM images of the four steps of microsphere expansion are shown in Figure S1 in the Supporting Information: at room temperature (RT), at 150 °C where the expansion begins, at 170 °C where the maximum expansion occurs, and finally at 200 °C where the microspheres are shrunken. A histogram and average diameter were obtained by measuring 100 samples after expansion at different temperatures (Table S1 and Figure S2, Supporting Information). At the initial state, the average diameter of the microspheres was
measured as $8.49 \pm 0.53 \, \mu m$. There was no significant change in diameter at $140 \, ^\circ C$. Some microspheres began to expand at $150 \, ^\circ C$, and the average diameter rapidly increased with the heating temperature (e.g., average diameter of $41.35 \, \mu m$ at $170 \, ^\circ C$). The inside of microsphere exhibits a rough surface and thickness of shell was measured as $\sim 1 \, \mu m$ (Figure S3, Supporting Information). However, as the temperature increased above $170 \, ^\circ C$, the microspheres shrank due to polymer degradation occurring at the melting temperature, and the overall diameter gradually decreased.

Figure 1c shows a schematic of the 3D surface morphology formation on an elastomer surface using microsphere expansion. Commercial microspheres were mixed with liquid PDMS, in which the resin and hardener were mixed in a 10:1 weight ratio. The PDMS/microsphere precursor was spin-coated on the PI substrate at 1,000 rpm for 30 s and then cured at $80 \, ^\circ C$ for 4 h. The fabricated composite film of microspheres and elastomer exhibits a smooth surface before expansion. Then, irregular micro-dome structures are generated throughout the surface when heat is applied to the composite film (Video S2, Supporting Information). The formation mechanism of the 3D and irregular micro-dome structure is explained by the pressure difference between the microspheres and the composite matrix (Figure 1c). In the initial state (Figure 1c (i)), there is no significant change in the surface of the composite film. When a specific temperature is applied to the composite film, the expansion of the microspheres begins due to the increased pressure. The high pressure within the microspheres causes their expansion, forming irregular micro-dome structures on the surface (Figure 1c (ii)). The microspheres expand until their pressures are in equilibrium with that of the surrounding composite film, resulting in a final 3D surface morphology (Figure 1c (iii)). Figures 1d and e show the top and cross-sectional FE-SEM images of the fabricated composite film before and after microsphere expansion. Before microsphere expansion, the
composite film exhibited a smooth surface with no irregular micro-dome structures. When microspheres were expanded, irregular micro-dome structures were randomly and densely formed on the composite surface. The 3D profiles of irregular micro-dome structures were measured using a confocal laser scanning microscope to observe the changes in morphology by microsphere expansion (Figure 1f). The inset graphs show the cross-sectional profiles along specific lines (AA’ and BB’). Before microsphere expansion, the maximum height on the composite film was measured as only 0.41 μm. In contrast, the composite film after microsphere expansion exhibited a maximum height of 13.41 μm. To demonstrate that the microscale 3D surface morphology can be controlled, composite films with different microsphere concentrations (1-9 wt%) were compared (Figure S4, Supporting Information for details of area fraction). The area fraction of the composite film increased from 4.33% to 56.92% when the microsphere concentration raised from 1 wt% to 9wt%. The uniformity of the suggested composite film with irregular micro-dome structures was evaluated with respect to the arithmetic mean height at various points (Figure S5, Supporting Information for the details of the arithmetic mean height). From the results, the 3D surface morphology can be controlled with high scalability and uniformity according to the microsphere concentration.

2.2 Fabrication Process and Structure of Flexible Pressure Sensor

The fabrication process of the sensor structure is shown in Figure 2a. The top electrode was fabricated by coating a liquid mixture of microspheres and PDMS prepolymer, heating the cured composite film to form a micro-dome structure, and depositing a thin metal film as a conductive layer. The bottom electrode was fabricated by applying a CO2 laser on a cleaned polyimide (PI) substrate and connecting this electrode with a copper wire using silver epoxy. The surface of the PI film was irradiated by a CO2 laser and locally heated at ~2,500 °C via the
photothermal effect, which breaks the C-O, C=O, and N-C bonds, and generates C-C bonds in the chemical structures of the PI film.\[32\] The oxygen and nitrogen components are significantly reduced, and the sp\(^3\) bonds of carbon are converted into sp\(^2\) bonds to produce a high-purity graphene. Energy-dispersive X-ray spectroscopy (EDS) was used to compare the composition of the PI substrate and LIG layer (Figure S6, Supporting Information). The ratio of carbon and oxygen was changed by varying the CO\(_2\) laser irradiation treatment on the PI substrate, and the carbon content increased from 69.06 to 94.83 wt\%, while the oxygen content significantly decreased from 30.75 to 4.15 wt\%. In addition, X-ray photoelectron spectroscopy (XPS) was used to analyze the composition of LIG. The C1s profile of LIG exhibits distinct C-C peaks (Figure S7, Supporting Information). The Raman spectrum of LIG reveals three dominant peaks: the D peak at ~1,350 cm\(^{-1}\) induced by defects or bent sp\(^2\)-carbon bonds, the first-order G peak at ~1,580 cm\(^{-1}\), and the 2D peak at ~2,700 cm\(^{-1}\), originating from second-order zone-boundary phonons (Figure S8, Supporting Information). We also observed the surface of the fabricated LIG under various fabrication conditions (Figure S9 and S10, Supporting Information for details of the LIG surface observation). We also measured the sheet resistance of the LIG based on the laser irradiation power and scan steps to check the electrical conductivity characteristics (Figure S11, Supporting Information for the details of the measurements of electrical conductivity characteristics).

Figure 2b shows a schematic of the sensitive, flexible pressure sensor, where the conductive metal-deposited irregular micro-dome structure is used as an active layer for pressure sensing and an interdigitated electrode (IDE) based on LIG layer on a PI substrate acts as an electrode. In this study, we introduce an IDE design because its piezoresistance variation depends on the change in contact area between the micro-dome structure and LIG electrode. Furthermore, the irregular micro-dome structure enables stress concentration, leading to a heterogeneous
increase in the contact area, resulting in higher sensitivity. The detailed working principle of the pressure sensor is shown in Figure 2c. In the initial state (no pressure), only a few micro-dome structures are in contact with the bottom LIG electrode. Given the small contact area, the conductive pathway is narrow and little current flows between the top active layer and the bottom electrodes. When the pressure is increased, additional contacts are formed between other micro-dome structures and the LIG electrode, resulting in a rapid decrease in resistance. The micro-dome structures are deformed with increasing pressure, and the contact area with the LIG electrode increases. With this irregular micro-dome structure, the total resistance of this sensor \( R_t \) can be expressed as the sum of the resistance of the metal-deposited \( R_f \), the resistance of the LIG electrodes \( R_G \), and the contact resistance between them \( R_c \), which can be expressed as follows:

\[
R_t = R_G + R_c + R_f = R_G + \frac{1}{\frac{1}{R_{C1}} + \frac{1}{R_{C2}} + \cdots + \frac{1}{R_{Cn}}} + R_f \tag{1}
\]

where \( R_{Ci} \) and \( n \) correspond to the contact resistance at the contact point \( #i \) and total number of electrical contacts between the top and bottom layers, respectively. As the resistance of \( R_G \) and \( R_f \) are very low owing to the high conductivity of top and bottom layers, Equation 1 can be expressed as \( R_t \approx R_c \). Therefore, the change in \( R_c \) has a dominant effect on the sensing performance as the contact resistance between the micro-dome structures and the bottom LIG electrode changes according to the applied pressure (Figure S12, Supporting Information for details of the equivalent circuit model).

2.3 Sensitivity of the Flexible Pressure Sensor

Figure 3 depicts the sensing performance of the flexible pressure sensor with and without an irregular micro-dome structures in the pressure range of 0–400 Pa. The sensitivity \( S \) can be
calculated by $S = \frac{\delta(\Delta I/I_0)}{\delta p}$, where $\Delta I$, $I_0$, and $p$ represent the current change, initial current, and the applied pressure, respectively. The resistive-type pressure sensor based on the composite films with 5 wt% of microspheres exhibited two different linear sensitivities under applied pressure (Figure 3a). In the first linear section up to 50 Pa, the pressure response exhibited a linear behavior and high sensitivity of 50.45 kPa$^{-1}$. Notably, the sensitivity of the proposed sensor (w/microsphere expansion) is ~85.5 times higher than that of the sensor with a smooth surface (w/o microsphere expansion). The second linear section exhibited a sensitivity of 4.35 kPa$^{-1}$ at 50–400 Pa, which is ~7.37 times higher than that of the sensor with a smooth surface. Previously suggested flexible pressure sensors based on regularly arranged microstructures commonly have relatively low sensitivity in the low-pressure range owing to simultaneous deformation,$^{[11,12,23,27]}$ However, our sensor, based on the irregular micro-dome structures, demonstrated excellent sensing performance in terms of sensitivity, sensing range, response time, and reliability than previously developed sensors (Figure 3b and Table S2). We also checked the reproducibility by the following tests: i) Size distribution of micro-dome structures and ii) Responses of the fabricated flexible pressure sensors. The four areas of a composite film with irregular micro-dome structures exhibited approximately similar tendencies (Figure S13, Supporting Information). In addition, these 10 different sensors exhibited similar changes in the current responses, with negligible deviations (Figure S14, Supporting Information). These results indicate that the irregular micro-dome structure significantly improved the sensor sensitivity in the low-pressure range while maintaining high reproducibility. Figure 3c shows the response characteristics according to the microsphere concentration with the applied pressure. Because of the microstructures, the sensor with a microsphere concentration of 5 wt% showed the highest sensitivity. At microsphere concentrations higher than 5 wt%, the composite film was bent downward for releasing compressive residual stress caused by expansion (Figure S15, Supporting Information). In
addition, composite film with 5 wt% of microsphere concentration exhibited reduced mechanical properties (i.e., compressive stress and Young’s modulus) for the porous effect (Figure S16, Supporting Information). Therefore, a microsphere concentration of 5 wt% was used for the additional sensor evaluation and application.

2.4 Simulation of Compressive Behavior of the Irregular Micro-dome Structures

Finite element (FE) simulations were conducted using a representative volume element (RVE) to understand the compressive behavior of the irregular micro-dome structure. The RVE was generated using an FE-SEM image of the irregular micro-dome structure, using the following procedure. First, the center point locations and radii of each irregularly dispersed micro-domes in the FE-SEM image of 200 × 250 μm² size were characterized using the Image J program (NIH, USA). Next, a realistic RVE consisting of irregular micro-domes having the same locations and radii of the microstructure image was reconstructed into a 3D FE simulation model (Figure S17, Supporting Information). In total, 29 micro-domes were modeled in the FE simulation model. Finally, compressive deformation of the RVE via contact of the outer surface of the RVE to a flat plate was simulated using the FE method. The simulation assumed that the microspheres with a wall thickness of 0.5 μm were covered by a 2 μm thick PDMS film near the outer surface. The microspheres and PDMS were modeled as linear elastic materials, with an elastic modulus of 500 kPa and Poisson’s ratio of 0.49. We found that the simulated pressure-displacement curve from the RVE model under a pressure range of 0–400 Pa matched the experimental results very well (Figure 3d). Consistent with the experiments, the compressive displacement increased sharply with pressure until an applied pressure of 50 Pa and then moderately increased. This pressure-displacement behavior is directly linked to the sensitivity of the pressure sensor because the larger the compressive deformation, the more the
number of micro-domes that are in contact between the LIG electrode. The simulation results suggest that the sharp increase in the displacement (and therefore the high sensitivity of the pressure sensor) in the pressure range of 0-50 Pa originates from the irregular micro-dome structure. For such a relatively low applied pressure, the contact between the irregular micro-domes and the LIG electrode can occur only by a few largest micro-domes, as shown by the local stress distribution of the FE model for the compressive displacement of 4 μm in Figure 3e. In this situation, the contact area increases rapidly with the applied pressure owing to the locally concentrated deformation of the few largest micro-domes, thus leading to the high sensitivity of the pressure sensor. When the compressive displacement is higher than 6 μm, other smaller micro-domes also make electrical contacts (Figure 3e). In addition, the micro-dome structures are deformed with increased pressure, and the contact area between the micro-dome structures and the counter electrodes is enlarged accordingly (Figure 3f). These results suggest that the second linear section at 50–400 Pa are attributed to the wider the contact area between the irregular micro-domes and the LIG electrode.

2.5 Static and Dynamic Sensing Performance of the Flexible Pressure Sensor

The sensing performance of the proposed pressure sensor was evaluated under static and dynamic pressure loadings (Figure 4). The relative change in the current over five cycles of various pressure inputs (15, 30, 45, and 60 Pa) was measured, and the repeatability results are presented in Figure 4a. We observed that our sensor exhibited stable and repeatable signals at various pressure ranges. Moreover, we examined the dynamic response characteristics of the proposed sensor. The 10% to 90% rising time ($t_r$) and 90% to 10% falling time ($t_d$) were measured as 39 ms and 31 ms, respectively. We also measured the subtle-pressure, as shown in Figure 4b. The current between the two electrodes of the sensors was measured in real-time,
and water droplets (43 mg) were sequentially detected by placing the droplets on top of the sensor (Figure 4c). The sensor exhibited a continuous response after the droplets were placed four times (Video S3, Supporting Information). We also calculated that the limit of detection (LOD) of our sensor based on sequential detection of water droplets, as shown in Figure S18. The theoretical LOD of our sensor was calculated as low as 0.209 Pa. To compare the sensitivity of the sensors with a smooth surface and the irregular micro-dome structures, we obtained the current-voltage (I-V) curves under various applied pressures (Figure 4d). When pressure was applied to the sensor with a smooth surface, the resistance hardly changed because of low sensitivity; therefore, all the I-V curves almost overlapped. In contrast, the I-V curves of the flexible pressure sensor with an irregular micro-dome structures changed dramatically owing to the enhanced sensitivity. This result means that the proposed pressure sensor is sensitive enough to detect ultra-low pressures, enabling applications to various target fields.

For the fabricated sensors to be used in practical applications, they must maintain stable sensing characteristics under repeated pressure loading. We evaluated the reliability of our sensor by applying repeated compression/release cycles. A pressure of 1 kPa was applied to the sensor 4,000 times to investigate its long-term stability. Figure 4e presents the measurement results. Neither a shift in the sensing response nor a structural change in the sensor was observed during 4,000 cycles of compression. The inset values in the middle of the graph show the average responses and standard deviations for each cycle. In the initial 1–20 cycles, the average response was 6.834, and the standard deviation was 0.292%. Similar values were consistently measured throughout repeated compression cycles. For example, in cycles 3001-3020, the response and standard deviation were 6.951 and 0.269%, respectively. In addition, 3D profile results and FE-SEM images of composite film before and after reliability test exhibit that no significant structural distortion occurred in the micro-dome structure (Figure S19,
Supporting Information).

2.6 Demonstration of Fingertip-shaped Pressure Sensor

Given the excellent sensing characteristics such as high sensitivity, stability under static and dynamic applied loading, fast response, and high reliability, the proposed flexible pressure sensors are applicable for wearable devices. As one of the practical applications, we attached the sensor inside the customized thimble that perfectly fit the user’s finger (Figure 5a). Using this fingertip-shaped pressure sensor, we demonstrated the detection of the blood pulse on the index finger and electronic skin to recognize a soft object (Figure 5b). Our sensor exhibited a high sensitivity in the low-pressure range for blood pulse detection, which is appropriate for detecting a weak signal of the blood pulse at the fingertip compared to that at the wrist. The sensor clearly detected the pulse waves under normal conditions (60–66 beats per minute) with typical wave shapes, as shown in Figure 5c (Video S4. Supporting Information). A typical carotid pulse waveform with three clear peaks was observed: the percussion wave (P-wave), tidal wave (T-wave), and diastolic wave (D-wave), which are related to the systolic and diastolic blood pressures, late systolic augmentation, and heart rate, respectively. These results suggest that the sensor can detect subtle pressures, such as the pulse at the fingertip, which indicates that it can be applied to various health monitoring fields. For the electronic skin to recognize a soft object, it is necessary to detect subtle pressure changes on the electronic skin when handling fragile/soft objects. As shown in Figure 5d, our sensor could accurately detect small compressive forces from soft objects, such as marshmallows or sponges. Moreover, the sensor can detect and distinguish between gentle-, medium-, and high-pressure touch, as shown in Figure 5e.
2.6 Demonstration of Pressure Sensor Array Pad

We present the potential applications of the proposed technology by demonstrating the pressure sensor array pad (Figure 6). Because the pressure sensor array pad will become one of the most important electronic devices for human-computer interaction (HCI), high performance and easy fabrication of the pressure sensor array pad are essential. Our sensor exhibits excellent sensing performance in the low-pressure range and can be readily fabricated in large-scale and complex patterns. Thus, the fabricated pressure sensor array pad can detect small changes in tactile information from the user and transmit it to the computer for various applications. In this study, we demonstrated a sensor array pad to detect a pressure distribution with 10 × 16 pixels. Figure 6a shows a schematic of the sensor array pad. The sensor array pad was fabricated by assembling a metal-deposited film with micro-domes on the surface and LIG electrode patterns interconnected by a silver (Ag) electrode. For user convenience, the sensor array pad was covered with a soft sponge (ethylene-vinyl acetate foam). When pressure was applied to the sensor array pad, it measured the pressure applied to each pixel in real-time using an FPCB board and software. To increase the resolution and express visually from the sensor signals, we utilized a 2D/3D mapping and interpolation technique, as shown in Figure 6b. Compared to original sensor signals (Figure 6b(ii)), the interpolated sensor signals (Figure 6b(iii)) allow for the high resolution prediction of pressure levels at adjacent locations using a limited number of sensing points. For the drawing application (Figure 6c), we drew a wave curve using a fingertip on the sensor array pad. First, we slightly pressed the sensor array pad (Figure 6c (i, ii)) and then gradually increased the pressure level while drawing a curved trajectory (Figure 6c (iii, iv)). The 2D/3D pressure mapping results show that our sensor array pad can recognize the location and sensitive pressure change during fingertip drawing (Video S5 and S6, Supporting Information). For the multi-touch application (Figure 6d), we applied
pressure onto three different points of the sensor array pad with different pressure levels (gentle touch (Figure 6d(i)) and medium pressure touch (Figure 6d(ii)). From the 2D/3D mapping results, the sensor array pad successfully recognized the location and pressure levels (Video S7, Supporting Information).

After verifying the performance of the sensor array pad, we further developed a wrist-motion-based controller using deep learning (Figure 6e-h). Deep learning has been traditionally used for image processing; however, it recently has been successfully utilized in various tactile sensing applications.\cite{33} Owing to the high sensitivity of our sensor in the low-pressure range, the sensor array pad can measure small changes in the wrist posture when the wrist is on the sensor array pad. As shown in Figure 6f, the sensor signals exhibit different patterns by different wrist movements. We used a convolutional neural network (CNN) to classify four different functions: up/down/right/left movements from the sensor array pad (Figure 6g). First, the sensor signal was gathered with different wrist postures several times. Then, one hot encoding was used to provide the algorithm with the true labels. The signals (80 × 50 data) were put into a 1 × 4000 tensor. Then the output of the tensor was passed to two consecutive fully connected layers of sizes 40 and 20, respectively, with ReLU activation layers. Using this algorithm, we successfully demonstrated the control of the mouse cursor using our sensor array pad (Figure 6h and Video S8, Supporting Information).

3. Conclusion

The present study details a novel strategy to generate 3D and irregular micro-dome structures with microspheres that expand at a specific temperature. By mixing microspheres and elastomer and applying a heat expansion treatment, we could fabricate large-area, flexible, and
irregular micro-dome structures without the need for any micro-pattern mold. A composite film with an irregular micro-dome structure was applied to the sensing film of a high-performance pressure sensor, simultaneously featuring high sensitivity and flexibility. The proposed flexible pressure sensor based on an irregular micro-dome structure exhibited much higher sensitivity than previously suggested sensors based on regular microstructures. Through FEM simulations and experiments, we confirmed that the sensitivity enhancement of the pressure sensor originates from the heterogeneous contact change between the irregular micro-dome structures and LIG electrodes. In addition, the sensor showed a fast response time ($\tau_{10-90\%} \sim 39$ ms), excellent repeatability, and high reliability. The flexible pressure sensor was successfully utilized in a fingertip-shaped pressure sensor to monitor subtle pulse signals at the index finger and to recognize soft objects in real-time. Finally, to demonstrate the large-area application of the proposed pressure sensor, a multi-pixel sensor pad was fabricated and evaluated for HCI applications. As a result, we confirmed that the suggested 3D surface morphology via internal popping of microspheres could be applied in pressure sensors with high performance in terms of sensitivity, flexibility, scalability, and reliability. We believe that the present study opens a new avenue for the fabrication of 3D surface morphology for highly sensitive pressure sensors with excellent sensing performance. We expect that our study will provide a new route toward advanced pressure sensors for promising applications including human-machine interfaces, health-monitoring systems, interactive robot control, etc.

4. Experimental Section

4.1 Fabrication of the Composite Films with 3D Surface Morphology

First, thermally expandable microspheres (Microsphere, MSH,320, SDI Korea, Korea) were
added to the well-mixed liquid PDMS precursor (Sylgard 184, Dow Corning Corporation, USA), in which the resin and hardener were mixed in a 10:1 weight ratio. For uniform mixing and degassing, the PDMS/microsphere precursor was mixed for 90 s in a 7:6 revolution-rotation ratio using a planetary mixer (KK-V350W, KURABO, Japan). The thickness of the spin-coated composite film with microsphere affects the surface morphology, the PDMS/microsphere precursor was spin-coated on the PI substrate at 1,000 rpm for 30 s to control the thickness of the composite film as ~110 μm (Figure S20, Supporting Information). And then the composite film cured at 80 °C for 4 h. To maximize the microsphere expansion, the fully cured composite film was heated at 170 °C for 5 min. Metal deposition (Ti/Au 10/30 nm) was conducted by sputtering on the surface with irregular micro-domes to form a uniform conductive layer. Second, a commercial PI film (PI film, Young Woo Trading, Korea) with a thickness of 75 μm was cleaned and prepared. The LIG was grown directly on a PI substrate using CO₂ laser equipment (C30, Coryart, Korea). The scanning power and interval between the scan steps were 30% of the maximum power (40 W) and 0.075 mm, respectively. Sensing lines were connected to both pads on the IDE using a silver epoxy (CW-2400, ITW Chemtronics, USA).

4.2 Characterization of the Composite Films with Irregular Micro-dome Structure and Flexible Pressure Sensor

Real-time images of microspheres during expansion were obtained using an optical microscope (VHX-500F, KEYENCE, Japan). The morphologies of the composite film and LIG were characterized using FE-SEM (Supra 40VP, Carl Zeiss NTS GmbH, Oberkochen, Germany). A 3D laser scanning confocal microscope (VX-X1050, KETENCE, Japan) was used to measure the 3D surface morphology. The morphology of the composite film with 3D surface morphology was characterized using a 3D laser scanning confocal microscope (VX-X1050,
KEYENCE, Japan). Surface roughness measurements were performed using a portable surface roughness tester (SJ-210, MITUTOYO, Japan) with a scan length of 4 mm. The chemical composition of the composite structure was analyzed from the C1s spectrum obtained via XPS (Axis Supra, Kratos Analytical Limited (Shimadzu), Manchester, United Kingdom). A four-point probe (CMT-SR2000N, Advanced Instrument Technology, Korea) was used to measure the sheet resistance of the LIG, and the average and deviation were calculated by measuring 50 points for each condition. To verify the sensing performance, a high-precision universal testing machine (UTM) (AGS-X (1 kN), Shimadzu Corporation, Japan) equipped with a load cell (maximum load= 1,000 N) was used. A disk with a diameter of 50 mm was used to apply a uniform pressure to the sensor. The response of the sensor was measured using a parameter analyzer (Keithley 4200 SCS, Tektronix, USA). The UTM was employed to compress the composite film at 0.8 mm displacement (10% strain). The composite film was compressed at a speed of 1 mm/min, and had a diameter of 30 mm and thickness of 8 mm.

### 4.3 Fabrication of the Fingertip-based Pressure Sensor

A fingertip-based pressure sensor was fabricated using the following sequential steps. First, Alja Safe powder (SmoothOn, USA) was mixed with water to form a rubbery elastomer, which was subsequently poured into a container. Second, the target user’s finger was dipped into the container for 5 min until it was cured to form a mold for the customized thimble. Third, Ecoflex00-35 (Smooth-On, USA) was poured into the mold back and forth to form a customized thimble. Finally, the sensor was placed in the thimble and then covered with a thin Ecoflex00-35 (Figure S21, Supporting Information). The output from the sensor was measured using a source meter (Keithley 2400, Tektronix, USA). This experiment was approved by the institutional review board (IRB) of Korea Advanced Institute of Science and Technology (KAIST) (IRB no. KH2021-151).
4.4 Fabrication of the Pressure Sensor Array Pad

The pressure sensor array pad was fabricated by the following sequential steps. First, a shadow mask was mounted onto composite films with irregular micro-domes on the surface. Second, a metal layer (Ti=10 nm, Au= 30 nm) was deposited using sputtering process. Third, the LIG electrode was fabricated using patterned LIG with 10 × 16 pixels and screen printing with silver (Ag) ink (Figure S22 and S23, Supporting Information). Finally, the sensor array pad was fabricated by assembling the composite film and LIG arrays with a covering sponge (Figure S24, Supporting Information). Subsequently, this pressure sensor array was connected to the microcontroller (Teensy, PJRC, USA) to measure the resistance. Using software programs such as MALAB (MathWorks, USA) and Jupyter Notebook (Jupyter, USA), 2D/3D visualization and deep learning were conducted. Regarding the deep learning process, the sensor signal was gathered several times. Then, one hot encoding was used to provide the algorithm with the true labels. The signals (80 × 50 data) were put into a 1 × 4000 tensor. Then the output of the tensor was passed to two consecutive fully connected layers of sizes 40 and 20, respectively, with ReLU activation layers.

References


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**Author Contributions**

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.
Competing interests

All authors declare that they have no competing interests.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.
Figure 1. Concept of 3D surface morphology formation using thermal expansion of microspheres. a) Thermal expansion mechanism of microspheres. The AN copolymer shell becomes soft (state 1) and hydrocarbon at the inner core undergoes a phase change from liquid to gas (state 2) by applied temperature. b) FE-SEM images and particle diameter distribution.
before and after the expansion of microspheres. c) Schematic of 3D surface morphology on an elastomer surface using internal popping of microspheres. d-e) Top and cross-sectional FE-SEM images of the fabricated composite film before (d) and after (e) microsphere expansion. f) 3D profile measurement results of the composite films before (left) and after (right) microsphere expansion.
Figure 2. Fabrication procedure for the sensor structure and sensing mechanism. a) Fabrication method of the metal-deposited composite film with irregular micro-dome structure and interdigitated electrode using laser-induced graphene (LIG). b) Schematic illustration of the pressure sensor with metal-deposited composite film with irregular micro-dome structure on the top active layer and interdigitated LIG on the bottom sensing electrode. c) Sensing mechanism of the fabricated pressure sensor.
Figure 3. Experimental and numerical analysis of the flexible pressure sensor with irregular micro-dome structure. a) Measured sensitivity of the flexible pressure sensor under the pressure range of 0–400 Pa. The sensitivity of the sensor based on irregular micro-dome structure (w/ microsphere expansion) was ~85.5 times higher than that of the sensor with a smooth surface (w/o microsphere expansion). b) Comparison of flexible pressure sensor with previously developed pressure sensors with regular micro-structures. c) Response characteristics according to the microsphere concentration with applied pressure. d) Calculated and experimental pressure-displacement curve under the pressure range of 0–400 Pa. Experimental results were normalized for comparison of tendency. The simulation results were consistent with the experimental ones. e) and f) Finite-element simulation results of the local stress.
distribution and deformation of the irregular micro-dome structure under different applied pressure. The irregular micro-dome structures are deformed with increased pressure, and the contact area with the counter electrodes increases accordingly.
Figure 4. Analysis of the sensing performance of the pressure sensor based on irregular micro-dome structure under static and dynamic loading conditions. a) Repeatability test of the pressure sensor under different applied pressures. b) Response time of flexible pressure sensor to for a compressive loading of 100 Pa. The 10% to 90% rising time ($t_r$) and the 90% to 10% falling time ($t_d$) are 39 and 31 ms, respectively. c) Detection of lightweight object, i.e., water droplets of 43 mg. d) Current-voltage (I-V) characteristics of the pressure sensors with a smooth surface and irregular micro-dome structures. e) Reliability test by cyclic loading of 1 kPa over 4,000 cycles. The blue, red, green, and purple graphs show the results of each 20 cycles (1-20, 1001-1020, 2001-2020, and 3001-3020 cycles, respectively).
Figure 5. Demonstration of the fingertip-shaped pressure sensing with developed pressure sensor. a) Schematic illustration of the fingertip-shaped pressure sensor. b) Schematic illustration and photographs of demonstration of the blood pulse on index finger (i) and electronic skin for recognition of a soft object (ii). c) Sensor signal on the index finger. The response shows pulse waves under normal conditions (60–66 beats per minute) with typical wave shapes. d) Demonstration of the electronic skin for recognition of a soft object such as marshmallow. The sensor could accurately detect the small compressions of the soft object. e)
Sensor signals with the different pressure used to touch the objects. The sensor can detect and distinguish between gentle-, medium-, and high-pressure touches.
Figure 6. Demonstration of pressure sensor array pad with 10 × 16 pixels for human computer interaction. a) Photographs of demonstration of the pressure sensor array pad. The sensor array
pad was fabricated through assembling metal deposited irregular micro-dome structures, LIG patterned electrodes, and the soft sponge. b) Illustration of signal processing of the pressure sensor array pad. When pressure is applied to the sensor array pad, pressure at each pixel is measured in real time (i), using 2D/3D mapping and interpolation technique (ii, iii). c) Demonstration of the drawing application. At first, we slightly pressed the sensor array pad (i, ii), and then increased pressure level gradually (iii, iv) while drawing a curved trajectory. d) Demonstration of the multi touch application. Applying pressure onto three different points of the sensor array pad with different pressure levels (gentle touch (i) and medium pressure touch (ii)). e) Photographs of demonstration of wrist motion-based controller using deep learning. f) Sensor signals with different patterns with different wrist movements (up (i), down (ii), right (iii), and left (iv)). g) Deep learning process for real time classification of four different functions such as up/down/right/left movements from the sensor array pad. h) Result of deep learning process for classification of movements and control of the mouse cursor.
Irregular micro-dome structure with a design and manufacturing flexibility is generated by internal popping of microspheres. By integrating this structure with laser-induced graphene electrode, large scale pressure sensor with excellent sensing performances such as high sensitivity (−50.45 kPa⁻¹), short response time (τ₁₀−₉₀%=~39 ms), and high reliability (stable sensor response under 1 kPa and 4000 cycles) is realized.

Young Jung†, Jungrak Choi†, Wookjin Lee, Jong Soo Ko, Inkyu Park*, and Hanchul Cho*

Irregular Micro-dome Structure-Based Sensitive Pressure Sensor Using Internal Popping of Microspheres
Supporting Information

Irregular Micro-dome Structure-Based Sensitive Pressure Sensor Using Internal Popping of Microspheres

Young Jung†, Jungrak Choi†, Wookjin Lee, Jong Soo Ko, Inkyu Park*, and Hanchul Cho*

Table S1. Observation and comparison of microsphere size (average diameter and standard deviation) at different heating temperatures.

<table>
<thead>
<tr>
<th>Temp (°C)</th>
<th>RT</th>
<th>150</th>
<th>160</th>
<th>170</th>
<th>180</th>
<th>190</th>
<th>200</th>
</tr>
</thead>
<tbody>
<tr>
<td>Avg (μm)</td>
<td>8.49</td>
<td>11.85</td>
<td>28.33</td>
<td>41.35</td>
<td>37.36</td>
<td>34.59</td>
<td>22.36</td>
</tr>
<tr>
<td>SD (μm)</td>
<td>1.87</td>
<td>5.35</td>
<td>4.64</td>
<td>3.27</td>
<td>5.36</td>
<td>5.59</td>
<td>4.92</td>
</tr>
</tbody>
</table>
**Figure S1.** FE-SEM images of four steps of microsphere expansion. The size of the microsphere changed according to the applied temperature. a) Room temperature, b) 150 °C where the expansion begins, c) 170 °C where the maximum expansion occurs, and d) 200 °C where the microspheres are shrunken.
Figure S2. Histogram and measured diameter of microspheres after expansion at different heating temperatures. a) At room temperature, the microsphere diameter was measured as $8.79 \pm 0.53 \, \mu m$. b) Some microspheres began to expand at $150 \, ^\circ C$. c) The average diameter rapidly increased with the heating temperature, and the average diameter of microspheres was $\sim 41.35 \, \mu m$ at $170 \, ^\circ C$. d) As temperature increased above $170 \, ^\circ C$, microspheres shrunk due to the thinner outside shell thickness and increased external pressure, and the overall diameter gradually decreased.
Figure S3. FE-SEM images of cross-section of expanded microsphere. While the outside of the microsphere exhibits a smooth surface, the inside has a rough surface. In addition, thickness of shell was measured as ~1 μm.
Figure S4. Area fraction of 3D surface morphology according to the microsphere concentration.

We prepared the composite films with 1, 5, and 9 wt% of microsphere concentration and observed the surface morphologies using FE-SEM and threshold function of the Image J program. The composite film with 1 wt% has a slight 3D surface morphology on the surface, and the area fraction per unit area was calculated as $4.33 \pm 1.05\%$. The 3D surface morphology of the composite film with 5 wt% increased significantly as $28.08 \pm 0.64\%$. Finally, a maximum fabrication concentration of 9 wt% with a large number of microspheres was observed on the surface; most of the surface was covered with microspheres (Area fraction: $56.92 \pm 1.83\%$).
Figure S5. Uniformity of the 3D surface morphology by measuring the arithmetic mean height of various parts of the fabricated composite films with 5 wt% of microsphere concentration. The roughness of the top, bottom, left, right, and center parts was measured 10 times each, the average ($\sigma$) and standard deviation ($SD$) of arithmetic mean height were calculated. The arithmetic mean height ($R_a$) and maximum height ($R_z$), which are mainly used for surface roughness measurement, were measured. $R_a$ indicates the average of the absolute value along the scale length.

$$R_a = \frac{1}{l} \int_{0}^{l} |f(x)| \, dx$$  \hspace{1cm} (S1)

where $l$ is the scan length and $R_z$ is the sum of the largest height value within the defined scan length.

$$R_z = R_p + R_z$$  \hspace{1cm} (S2)

where $R_p$ is the highest profile peak and $R_z$ is the deepest profile valley.

The average roughness $R_a$ was $\sim 0.92$ $\mu$m. The error of the left ($R_a = 0.959$ $\mu$m) and bottom ($R_a = 0.882$ $\mu$m), where the largest error occurred, was $\sim 8.03\%$. These results indicate that each surface of the composite film made by this fabrication process exhibits high scalability and uniformity on a 4-inch wafer scale.
Figure S6. Composition analysis of the PI film and LIG using energy dispersive X-ray spectroscopy (EDS). a) PI film, b) LIG. The ratio of carbon and oxygen is changed by varying the CO$_2$ laser irradiation.
Figure S7. X-ray photoelectron spectroscopy (XPS) spectrum of PI film and LIG. The C1s XPS profile of LIG exhibits a distinct C-C peak with greatly suppressed C-N, C-O, and C=O peaks. After PI was converted to LIG, the nitrogen and oxygen peaks were remarkably decreased.
Figure S8. Raman spectrum analysis results of PI film and LIG. Three dominant peaks: the D peak at ~1,350 cm\(^{-1}\) induced by defects or bent sp\(^2\)-carbon bonds, the first-order G peak at ~1,580 cm\(^{-1}\), and the 2D peak at ~2,700 cm\(^{-1}\), originating from second-order zone-boundary phonons were observed.
Figure S9. FE-SEM images of the fabricated LIG under various laser irradiation power from 8 W to 20 W. At the power of 8 W, LIG grown on the PI substrate was not observed due to low temperature by low laser irradiation power. At the power of 12 W, LIG was fabricated to form vertically aligned fibers. From the power of 16 W, the density of LIG grown by high laser irradiation power was very high and a clustered form of LIG was manufactured on the surface.
Figure S10. FE-SEM images of the fabricated LIG under different laser scan steps. At a scan step of 0.025 mm, part of the LIG melted due to the narrow laser irradiation interval. As the scan steps increased, the density of LIG decreased.
Figure S11. Electrical characteristics of LIG according to the laser irradiation conditions. Four-point probe was used to measure the sheet resistance of fabricated LIG, the average and standard deviation were calculated by measuring 50 points for each condition. a) Effect of laser power: The sheet resistance decreased as the laser irradiation power increased. At the laser power of 12 W, the sheet resistance was 111.9 Ω/□. Conversely, at the laser power of 20 W, the sheet resistance decreased by ~20 Ω/□ to ~89.6 Ω/□. b) Effect of laser scan step: The fabricated LIG at a scan step of 0.025 mm was ~49.6 Ω/□; when the scan step increased to 0.125 mm, the sheet resistance increased significantly to ~294.1 Ω/□. To maximize the performance of the sensor, it is important to use the bottom electrode with low sheet resistance.
Figure S12. Sensing mechanism of the flexible pressure sensor based on irregular micro-dome structures. a) Equivalent circuit model of the flexible pressure sensor with irregular micro-dome structure. b) Contact mechanism between irregular micro-dome structure and interdigitated LIG electrode layer.

Table S2. Comparison of sensing performances of previous pressure sensors in terms of sensitivity,
sensing range, response time, and reliability.

<table>
<thead>
<tr>
<th>Structure (Sensing mechanism)</th>
<th>Sensitivity (kPa⁻¹)</th>
<th>Sensing range (Pa)</th>
<th>Response time (ms)</th>
<th>Reliability (Pressure/cycles)</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td><em>Irregular microstructure</em> (Piezoresistive)</td>
<td>50.45 and 4.35</td>
<td>0–50 and 50–400</td>
<td>39</td>
<td>1 kPa/4000 cycles</td>
<td><em>Our work</em></td>
</tr>
<tr>
<td>Regular microstructure (Piezoresistive)</td>
<td>15.14</td>
<td>0–500</td>
<td>40</td>
<td>-</td>
<td>[8]</td>
</tr>
<tr>
<td>Regular microstructure (Piezoresistive)</td>
<td>21.14, 1.39, and 0.08</td>
<td>0–25, 25–250, and 250–1200</td>
<td>52</td>
<td>250 Pa/10000 cycles</td>
<td>[10]</td>
</tr>
<tr>
<td>Regular microstructure (Piezoresistive)</td>
<td>1.80</td>
<td>0–300</td>
<td>10</td>
<td>1 kPa/67500 cycles</td>
<td>[11]</td>
</tr>
<tr>
<td>Regular microstructure (Piezoresistive)</td>
<td>9.95</td>
<td>0–100</td>
<td>200</td>
<td>-</td>
<td>[12]</td>
</tr>
<tr>
<td>Regular microstructure (Piezo-capacitive)</td>
<td>0.55 and 0.15</td>
<td>0–200 and 200–7000</td>
<td>300</td>
<td>1.5 kPa/10000 cycles</td>
<td>[14]</td>
</tr>
<tr>
<td>Regular microstructure (Piezoresistive)</td>
<td>5.53 and 0.1</td>
<td>0–100 and 100–1400</td>
<td>0.2</td>
<td>100 Pa/5000 cycles</td>
<td>[23]</td>
</tr>
<tr>
<td>Regular microstructure (Piezo-capacitive)</td>
<td>30.2</td>
<td>0–130</td>
<td>25</td>
<td>15 Pa/100000 cycles</td>
<td>[24]</td>
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<tr>
<td>Regular microstructure (Piezoresistive)</td>
<td>6.61 and 2.1</td>
<td>0–110 and 0–16000</td>
<td>100</td>
<td>22 Pa/3750 cycles</td>
<td>[25]</td>
</tr>
<tr>
<td>Regular microstructure (Piezoresistive)</td>
<td>1.80</td>
<td>0–350</td>
<td>-</td>
<td>-</td>
<td>[27]</td>
</tr>
<tr>
<td>Regular microstructure (Piezoresistive)</td>
<td>2.0</td>
<td>0–220</td>
<td>50</td>
<td>1.5 kPa/10000 cycles</td>
<td>[28]</td>
</tr>
<tr>
<td>Regular microstructure (Piezoresistive)</td>
<td>15</td>
<td>0–100</td>
<td>100</td>
<td>100 Pa/1000 cycles</td>
<td>[31]</td>
</tr>
</tbody>
</table>
**Figure S13.** Sizes distribution of micro-dome structures. The four areas of a composite film with irregular micro-dome structures exhibited approximately similar tendencies. The average and standard deviation of the micro-dome structures’ sizes were measured as $20.8 \pm 5.1 \, \mu m$, $21.4 \pm 5.5 \, \mu m$, $21.2 \pm 4.9 \, \mu m$, and $20.5 \pm 5.1 \, \mu m$, respectively.

**Figure S14.** Reproducibility of flexible pressure sensor with irregular micro-dome structure by
comparing 10 fabricated sensors. These 10 sensors exhibited similar responses with negligible deviations.

**Figure S15.** The fabricated composite film with irregular micro-dome structure. a) The composite film with a microsphere concentration of 5 wt% remained flat state after microsphere expansion. For the microsphere concentration of 7 wt%, the composite film was bent downward. b) Schematic of influence of residual stress on the substrate with microsphere
expansion. (i) Equilibrium state. (ii) With microsphere expansion, the closer distance between the microspheres generates the compressive residual stress. (iii) The residual stress is released by the downward bending of the composite film. Therefore, a microsphere concentration of 5 wt% was used for the sensor evaluation and further applications.

Figure S16. Mechanical evaluation of the composite film with irregular micro-dome structure. a) and b) Measured compressive stress-strain curves of composite films before and after expansion based on the microsphere concentration. c) Calculated Young’s modulus of composite films. The Young’s modulus dramatically decreased after the thermal expansion of microspheres, acting as micropores inside the elastomer.
Figure S17. Real microstructure image based representative volume element (RVE) model. a) Comparison of FE-SEM image that is used for reconstructing RVE model to 3D FEM model used for the computational analysis. b) Enlarged view of 3D FEM model.
Figure S18. The linear calibration curve of the pressure sensor responses for the calculation of limit of detection (LOD). The theoretical LOD of our pressure sensor was calculated based on the slope ($S$) of linear calibration curve and the signal-to-noise ratio. The noise level was estimated as a standard deviation of intercept ($SDI$), which are the relative change of current values without applying pressure. Therefore, the theoretical LOD value can be expressed as the following equation with a distinguishing factor of 3.3:

$$LOD = 3.3 \times \frac{SDI}{S} = 3.3 \times \frac{4.8122 \times 10^{-5}}{0.7584 \text{ kPa}^{-1}} = 2.0934 \times 10^{-4} = 0.209 \text{ Pa}$$ (S3)

We confirmed that the theoretical LOD of our pressure sensor is as low as 0.209 Pa. This result means that the proposed pressure sensor is sensitive enough to detect ultra-low pressures, enabling applications to various target fields.
Figure S19. a) 3D profile results of the composite film before and after reliability test of 1 kPa over 4,000 cycles. The size distribution exhibited approximately similar tendencies, the average and standard deviation of the micro-dome structures’ sizes were measured as 20.3 ± 4.8 μm and 21.2 ± 5.5 μm, respectively. b) FE-SEM images of the irregular micro-dome structures before and after 4,000 times of 1 kPa. Inset FE-SEM images showed the magnified surface of micro-dome. This exhibits that no significant structural distortion occurred in the micro-dome structure.
Figure S20. Effect of spin coating speed on the thickness of the spin-coated composite film and surface morphology after thermal expansion of microspheres. a) Measured thickness of composite film with 5 wt% of the microsphere concentration according to the spin coating speed. b) FE-SEM images of the surface of composite films. With increasing of spin coating speed, the microspheres are removed from the substrate due to a high centrifugal force, which significantly changes the density of the microspheres.
Figure S21. Fabrication process of the customized thimble. a, b) First, Alja Safe powder was mixed with water to form a rubbery elastomer, which was subsequently poured into a container. Second, the target user’s finger was dipped into the container for 5 min until it was cured to form a mold for the customized thimble. c) Ecoflex00-35 were poured into the mold back and forth to form the customized thimble.
Figure S22. CAD design of the sensor array. a) LIG electrode. b) Bottom electrode #1. c) Bottom electrode #2. d) Insulating layer. e) Top electrode. f) Assembly.
**Figure S23.** Schematic illustration of the LIG patterned electrode. First, the LIG electrode with 10 × 16 pixels was fabricated by applying a CO\textsubscript{2} laser treatment on polyimide (PI) substrate. Second, a sensing line #1 was manufactured using screen printing with silver (Ag) ink. Third, to prevent electrical connection between two sensing lines, insulating tape of 1.5 mm × 1.5 mm was formed. Finally, sensing line #2 was fabricated using screen printing with a silver ink.
Figure S24. Fabrication process of the pressure sensor array pad. a) LIG electrode. b) Putting the micro-dome structure film on the LIG electrode. c) Covering the soft sponge on the sensor.