A Flexible Piezoelectret Actuator/Sensor Patch for Mechanical Human-Machine Interfaces

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ABSTRACT

Flexible and wearable devices with the capabilities of both detecting and generating mechanical stimulations are critical for applications in human-machine interfaces, such as augmented reality (AR) and virtual reality (VR). Herein, a flexible patch based on a sandwiched piezoelectret structure is demonstrated to have a high equivalent piezoelectric coefficient of $d_{33}$ at 4050 pC/N to selectively perform either the actuating or sensing function. As an actuator, mechanical vibrations with peak output force of more than 20 mN have been produced, similar to those from the vibration mode of a modern cell phone and can be easily sensed by human skin. As a sensor, both pressure detection limit of 1.84 Pa for sensing resolution and excellent stability of less than 1% variations in 6000 cycles have been achieved. The design principle together with the sensing and driving characteristics can be further developed and extended to other soft matters and flexible devices.

KEY WORDS

human interactivity, wearable electronics, actuator/sensor, dual-functional, piezoelectret
A fully interconnected and intelligent living environment has been a grand challenge for future smart homes and cities and it is critically essential to development interactive sensing and actuating systems to bridge the human-machine interfaces.\textsuperscript{1-5} Previously, researchers have used various piezoelectric materials such as lead zirconate titanate (PZT),\textsuperscript{6-8} polyvinylidene fluoride (PVDF) or its co-polymers,\textsuperscript{9-12} and zinc oxide (ZnO)\textsuperscript{13-15} to sense or emulate a wide range of mechanical signals, including pressure, strain, and sound.\textsuperscript{16-18} An alternative is the piezoelectret (also referred as ferroelectret) material. For example, cellular polypropylene (PP) has piezoelectric-like properties with high equivalent piezoelectric coefficient, good flexibility, soft structure, and light weight.\textsuperscript{19-25} Devices based on either piezoelectric or piezoelectret materials can be designed to work as a sensor as well as an actuator. For instance, PVDF or cellular PP has been successfully constructed in various systems such as energy harvesters by converting mechanical movements into electrical outputs, as well as loudspeakers by converting electrical signals to mechanical motions.\textsuperscript{5,11,19,21} However, most research works have been focusing on sensing/energy harvesting applications without the actuator functions for wearable electronics. Specifically, the integration of perception and action in response to stimuli from environment/human is important for any human-machine interactive systems, where both functions of sensing and actuation are indispensably connected. As such, a flexible actuator/sensor array for interactive feedback communications with high-resolution, lightweight and low manufacturing cost would be desirable for various applications.

Herein, we design a flexible, sandwich-structured, and piezoelectret-based device to enable both the sensor and actuator functions, as shown in Figure 1, which is composed of the top and bottom fluorinated ethylene propylene (FEP) electret films, the middle Ecoflex spacer, with the
gold (Au) and aluminum (Al) electrodes at the top and bottom surfaces, respectively. With the application of an alternating voltage, the function of an actuator can be realized by means of electrostatic force to induce vibrational stimulations that can provide haptics feedback to human skin (Figure 1a). In a similar fashion, under the mechanical deformation like human body motions, the function of a sensor can be achieved to induce electrical outputs without any electrical power supply (Figure 1b). With the combination of actuators and sensors in the form of a wearable device, real-time sensing and actuation feedback and long-distance haptics communications are some possible immediate applications. Furthermore, the equivalent piezoelectric coefficient of our piezoelectret, $d_{33}$ value, is characterized as 4050 pC/N, which exceeds those of many traditional piezoelectric or piezoelectret materials or devices to achieve several important features: (i) a soft actuator using low driving voltages to produce a strong vibration force close to the vibration mode of a cell phone at 20 mN to be easily sensed by human skin; (ii) a flexible pressure sensor without the need of battery power with less than 1% readout variations for more than 6000 operations to reach the minimum pressure detection limit of 1.84 Pa from dandelion seed; (iii) assistance of functions between sensor and actuator for wearable scenario; (iv) an integrated actuator/sensor device with multiple pixels on the same flexible film with negligible cross-talks to act as a dual-function transducer array. It should be noted that current piezoelectret device is independently demonstrated sequentially as a sensor for human motions measurement and an actuator for the haptic feedback applications in this work.
Figure 1. Schematic diagram depicting the actuator/sensor structures as a wearable, flexible device for mechanical human-machine interfaces for applications such as real-time feedback and long-distance haptics communications. (a) The working principle of an actuator by means of electrostatic actuation to induce mechanical vibration under an alternating voltage. The measured force vs. time curves (bottom) for a prototype device under a driving voltage of 3.33 V/μm to generate 20 mN peak outputs. (b) The working mechanism of a sensor by generating electrical current outputs due to the mechanical deformation under external forces (bottom) for a prototype device under a stimulating pressure of 2.5 kPa.
RESULTS AND DISCUSSIONS

Sandwich-structured piezoelectret with high equivalent piezoelectricity

The sandwich-structured piezoelectret device in Figure 2a (detailed fabrication process in Figure S1) has an Ecoflex spacer layer chemically treated by 3-aminopropyltriethoxysilane (APTES) and mechanically sandwiched by two FEP films which are oxidized with the O₂ plasma. Ecoflex is very soft, while FEP is an excellent electret material to capture and retain surplus charges with good stability in the order of hundreds of years. The bonding mechanism of Ecoflex and FEP is assisted by the APTES and hydroxyl groups (-OH). The cross-sectional scanning electron microscope (SEM) image (Figure 2b) indicates that these films are tightly bonded, where the circular-shape cavities are designed in the Ecoflex spacer with 2 mm in diameter in the prototype devices. When the sandwich structure is forcibly separated (Figure S2), some Ecoflex debris are found on both the top and bottom surfaces of the FEP films as the results of strong bonding. A prototype sample is mechanically twisted between two human fingers to illustrate the flexibility (Figure 2c).

A corona charging process is used to generate megascopic electrical dipoles inside the air cavities (Figure S3a). The existence of the megascopic electrical dipoles is proved by a thermally stimulated discharge (TSD) method (Figure S3b). Polarization-electric field (PE) loops in Figure S4 also prove that FEP films are polarized to hole charges after corona charging and such charges are not mainly caused by triboelectric effect contribution. The aforementioned O₂ plasma treatment increases the surface roughness of the FEP electret film (Figure S5a and S5b) to improve its charge-retaining capability (Figure S5c). The equivalent piezoelectric coefficient of the device is characterized by the “weight moving” method (Figure S6), and found to be related to both the
density of the circular-shape cavity (hole ratio) (Figure S7a) and the thickness of the Ecoflex film which is controlled by the spin coating process (Figure S7b to S7d). Specifically, a thinner spacer layer and higher cavity to spacer area ratio can both improve the piezoelectricity, as indicated in Figure S7e. However, if the spacer layer is too thin or the cavity to spacer area ratio is too high, the top and bottom FEP layer may contact each other under a small mechanical deformation, thereby compromising the originally designed functions. It should be noted that piezoelectric coefficient of our sandwich-structured piezoelectret is mainly contributed by $d_{33}$.24

Among the tested structures, the piezoelectret film with a thickness of 150 μm and a cavity to spacer area ratio of 39.8% exhibits a stable equivalent $d_{33}$ coefficient of 4050 pC/N, which is much higher than those of previously reported piezoelectric and piezoelectret materials, such as cellular PP and PZT, etc (Figure 2d and Table S1).20,30-35 The high equivalent piezoelectric coefficient ($d_{33}$) of our sandwich-structured piezoelectret benefits by the rich stored charges in the FEP electret films and the soft property (low Young modulus) of the Ecoflex spacer. The stability of the piezoelectric property is further demonstrated by placing the sample in an indoor environment for 8 weeks and the equivalent $d_{33}$ is found to only drop in the first week but to approach a constant of 4050 pC/N afterwards as shown in Figure 2e. The equivalent $d_{33}$ values drop significantly during the first week as the charges stored by the FEP film after the Corona charging process will decrease rapidly because of the neutralization process with defects inside the FEP film, which is a typical phenomenon observed in all electret materials.27 As the defects are saturated overtime, the $d_{33}$ values become stable.
Figure 2. Sandwich-structured piezoelectret with high piezoelectric performance. (a) The key components of the sandwich-structured piezoelectret device are two FEP layers on the top and bottom and a soft Ecoflex layer at the centre having circular-shape holes. (b) Cross-sectional SEM image of a fabricated sample. (c) An optical photo of a fabricated structure being twisted by human fingers. (d) Comparison of the equivalent $d_{33}$ coefficients for the sandwich-structured piezoelectret device and other reported piezoelectric and piezoelectret materials or structures. (e) Measured equivalent $d_{33}$ coefficients of the sandwich-structured piezoelectret device under the indoor atmosphere condition for 8 weeks.
Working mechanisms

The two outer surfaces of the top and bottom piezoelectret films are coated with Au (50 nm in thickness) and Al (50 nm in thickness), respectively, as electrodes to form a flexible piezoelectret actuator/sensor device. The top FEP layer can bend downward slightly after the corona charging process because of the built-in electrostatic attraction force. Under alternating electrical voltages, the piezoelectret device can vibrate as an actuator. When a positive voltage is applied, for example, the electric field is superimposed onto the initial built-in electrical field generated by charges maintained by FEP electret films. The gap distance between the electrodes will decrease to cause the mechanical deformation of the structure Figure S8a and Video 1). If the driving voltage is removed, the structure can recover to its initial shape due to the elasticity of the film. The zero to positive alternating driving voltage thereby induces the vibration of the actuator and the detailed description for the actuator and the performances based on a cantilever structure are shown in Supplementary Explanation 1. It is found that the power consumption for a prototype actuator is 8.25 mW under a driving input of 3.33 V/μm (500 V for a 150 μm-thick piezoelectret device) (Figure S9).

As a pressure sensor, compressing and releasing the device will change the distance of the air gap and the moments of the megascopic electric dipoles, as indicated in Figure S8b. As a result, mechanical deformations are converted into electrical signals. For instance, as shown in Video 2, tapping the sensor with the fingertip can generate electricity to illuminate liquid crystal display (LCD) pixels showing the number of “1”. The detailed description for the sensor function is given in Supplementary Explanation 2.
**Soft piezoelectret actuator**

A force measurement setup is shown in Figure 3a and the prototype actuator is attached on top a soft PDMS film (thickness of 1 mm, simulating human skin). A 150 g preload is placed on top of the actuator and a force gauge is placed at the bottom of the PDMS film. The force vs. time curves generated by the actuator under different driving inputs at 60 Hz are shown in Figure 3b. As expected, a high driving electrical field can increase the vibration magnitude and the peak force values from about 3 mN at 0.67 V/μm to 20 mN at 3.33 V/μm. The Fast Fourier Transform (FFT) result for the force vs. time curves under a driving input of 3.33 V/μm is recorded (Figure S10), which shows that the input and output frequency spectrum match well with each other. Changing the external load on top of the actuator could alter the applied pre-stress on the soft actuator, resulting in the change of actuating output force (Figure S11). The force generated by a typical cell phone (weight of 165 g) is also measured in the same setup for comparison as shown in Figure S12. The peak cell phone force measured under the vibration mode is between 25 to 35 mN as shown in Figure 3c.

In the human subject sensing experiment, a referencing actuator and a testing actuator are placed side-by-side on a PDMS film (thickness of 1 cm) (Figure 3d). The reference actuator is constantly driven by an input of 1.33 V/μm (200 V) at 100 Hz to give a strong vibration feeling as the reference (the sensing threshold for the human skin is about 1 mN36). A total of 15 participating volunteers are asked to tap the two actuators (touching the top Au ground electrode) in turns with the same index finger, and describe their sensation feelings with the ranks of 0 (no sensation), 1 (weak), 2 (medium – same as the feeling of the reference), and 3 (strong). Each volunteer is asked to repeat the same test for 3 times and none-integer sensation feelings ranks are allowed. Different
driving inputs (0.33, 0.67, 1.33, 2 V/μm) and frequencies (25, 50, 100, 150, 200, 250, 300, 350, 400, 450, and 500 Hz) have been applied (Figure 3e). It is found that a higher driving input can produce stronger actuation force for higher sensation intensity. Furthermore, driving frequencies between 100 to 150 Hz can induce the strongest sensation intensity under variously tested driving inputs. With low driving frequency or high driving frequency above 500 Hz, the sensation feelings are lost under our current testing conditions. Simulation result shows that the resonant frequency of the piezoelectret actuator on human tissue is about 3 kHz (Figure S13). Although the actuator is found to vibrate up to 1700 Hz and act as a loud speaker to produce audible sounds (Video 3), human fingertip loses its sensation feeling at either low driving force or high vibration frequency. For example, the applied electrical field of 0.33 V/μm (50 V for a 150 μm-thick piezoelectret film) produces very weak sensation feelings for the fingertip in all tested frequencies as shown in Figure 3f. At 0.66 V/μm, an appreciable vibration sensation is detected with the sensation feelings, similar to that produced by a previously reported commercial PVDF actuator under 150-250 V (50-75 V/μm). The comparisons with other previously reported actuators are summarized in Table S2 and Figure 3g, and this work shows low electrical field requirement (driving voltage per thickness) and high piezoelectric coefficient (d33) as the result of large charges stored by the FEP electret films and soft property of Ecoflex spacer.
Figure 3. The performances of the soft piezoelectret actuator. (a) Schematic diagram for the setup of force characterizations using PDMS as the coupling material and a 150 g-load as the externally applied force. (b) Measured force vs. time curves for the actuator under increased driving voltage at a constant frequency of 60 Hz. (c) Measured force vs. time curves for the cell phone under its own vibration mode. (d) An optical photo depicting the finger sensation intensity test, where the reference actuator is driven under 1.33 V/μm at 100 Hz and the testing actuator is driven by randomly selected voltages and frequencies. The average sensation intensity collected from the
testing results of 15 subjects (e) under the driving inputs of 0.33, 0.67, 1.33 and 2 V/μm and frequencies from 25 to 500 Hz; and (f) under the driving frequencies of 50, 100, and 200 Hz and different voltage inputs from 0.33 to 2 V/μm. (g) Comparison of the typical driving inputs and piezoelectric coefficients (d_{33}) for different materials/structures based on previously reported piezoelectric and piezoelectret devices.

The contribution of piezoelectret effects

Finite element simulations (details in Supplementary Explanation 1) are used to analyse the strategy for performance improvements and the effectiveness of piezoelectret materials. The standard working unit has a circular air cavity with the diameter of 2 mm and the overall device thickness is 150 μm. The central deformation of the film under a driving voltage of 500 V is defined as D₀ without the Corona charging process for the FEP layer and the charge density (σ_{FEP}) is close to zero. Figure 4a shows that if the σ_{FEP} is increased such as by introducing extra charges using the Corona charging process, the displacement amplitude of the actuator increases (black line). The highest reported charge density of FEP electret film is about 0.5 mC/m²,⁴ and the relative deformation is about 10 times larger than D₀ if all other parameters remain the same. Meanwhile, the necessary applied voltage to produce the deformation of D₀ reduces from 500 to 55 V, as σ_{FEP} increases from zero to 0.5 mC/m² (red line). Figure 4b shows the effectiveness of decreasing the air gap distance for the performance of the actuator with the increasing relative displacement as the device thickness reduces under 500 V (black line) and decreasing voltage requirement as the device thickness decreases (red line). For example, if the σ_{FEP} value is 0.03 mC/m² (this value is close to
that of the real condition of our prototype), an applied voltage of 665 V and 185 V are required to induce the deformation of $D_0$ for the 300 $\mu$m- and 37 $\mu$m-thick devices, respectively.

![Figure 4](image)

**Figure 4.** Simulation results showing the piezoelectret effects using the example of a standard working unit having a circular air cavity with the diameter of 2 mm. (a) Relative displacement under a driving voltage of 500 V (black line) and driving voltage to induce deformation of $D_0$ (red line) vs. $\sigma_{FEP}$ from 0 to 0.5 mC/m$^2$. (b) Relative displacement under a driving voltage of 500 V (black line) and driving voltage to induce deformation $D_0$ (red line) vs. the device thickness from 300 to 37 $\mu$m under a constant $\sigma_{FEP}$ of 0.03 mC/m$^2$. $D_0$ means the displacement amplitude for the case with $\sigma_{FEP}$ of zero, thickness of device 150 $\mu$m and under driving voltage of 500 V.
**Flexible piezoelectret sensor**

A force gauge with a 3D moving stage for tunable mechanical stimulations is utilized to characterize the performances of a sensor (overall active size area of 4 cm$^2$). Under a fixed stimulation frequency of 3 Hz, the resulting peak currents and voltages with respect to the applied pressures are plotted (Figure 5a and Figure S14a to S14c). At the maximum tested pressure of 5 kPa, the corresponding peak current and voltage are 110.1 nA and 4.3 V, respectively. Furthermore, before the applied pressure of 1.75 kPa, the slopes of both the current and voltage are higher than those in the higher-pressure region (1.75-5 kPa). The current and voltage slopes for the lower-pressure regions are 41.53 nA/kPa and 1.45 V/kPa and those for the higher-pressure regions are 10.72 nA/kPa and 0.56 V/kPa. These observations closely resemble those of other sensors based on piezoelectret materials. For a constant applied pressure of 2.5 kPa under different frequencies (1-5 Hz) (Figure S14d), the peak currents increase approximately linearly with respect to the frequency (Figure 5b red curve and Figure S14e), while the peak voltages remain approximately constant at 3.2 V (Figure 5b blue curve and Figure S14f). As electrons can flow freely between the electrodes, the peak values of the short-circuit current depends on both the displacement magnitude and the rate of the displacement changes. Hence, the applied pressure and frequency can both affect the peak current. On the other hand, no charges will flow under the open-circuit condition and the peak voltage depends only upon the displacement magnitude changes.

The sensor has been stimulated under a constant applied pressure of 2.5 kPa and frequency of 3 Hz for over 6000 cycles as shown in Figure 5c. It is found that the output current remains stable with slight variation of less than 1% in the time waveforms (Figure 5d), demonstrating good stability. A 0.075-gram dandelion seed (Figure 5e) is put on a prototype sensor to investigate the
sensitivity (Figure 5f and Video 4) and the responding peak current is about -1.5 nA under pressure of 1.84 Pa, (Figure 5g). As illustrated in Figure S15, a pressure sensor is also assembled with a wristband to measure human arterial pulse signals at the wrist to demonstrate its high sensitivity and potential applications in wearable scenario, which can clearly reflect the dynamic (short-circuit current) and static details (open-circuit voltage) of pulse waves. These tests demonstrate the excellent sensitivity and stability for the sensing function of the device due to the high and stable piezoelectric coefficient ($d_{33}$).

**Figure 5.** The performances of the flexible piezoelectret sensor. (a) Peak short-circuit currents and open-circuit voltages under varying applied pressure magnitudes at a constant frequency of 3 Hz.
(b) Peak short-circuit currents and open-circuit voltages under an applied pressure of 2.5 kPa at different frequencies. (c) Short-circuit current vs. time of a prototype device under a constant applied pressure of 2.5 kPa at 3 Hz. (d) Expanded short-circuit current vs. time curves during the long-term stability test. (e) An optical photo showing the weight of a dandelion seed on a scale. (f) An optical photo for a dandelion seed on the prototype device. (g) The responding current signal for a dandelion seed on the prototype device.

A feedback control example using both the sensor and actuator

Intimate contact is critical for the operation of either the sensing or actuation function of wearable devices and some adjustments (tightening or loosening) are usually needed to have the proper pre-load between the device and skin for optimal operations (Figure 6a). An integrated instrument to measure the level of contact and an in-situ adjustment process are desirable to properly place the wearable devices. Here, a testing procedure is demonstrated to illustrate the possible application of the actuator/sensor patch. Specifically, a prototype patch is attached on a wristband and an air pump is utilized to apply the external load (Figure 6b) while the peak output voltage of the sensor is calibrated by a pressure gauge as shown in Figure 6c. As expected, the randomness of the placement of the patch results in large variations in the experimental results repeated for 5 times as shown.

The actuator/sensor patch is utilized to compensate these variations as the sensing function is switched to the actuator function under the same pre-load pressure levels. A total of 10 volunteers are tested for their skin sensations using a driving voltage of 2 V/μm at 100 Hz (Figure 6d) for 3 times. It’s found that the average sensation intensity increases as the external pressure increasing
and the maximum average sensation intensity of 2.6 is obtained for the pre-load pressure level 3 (average peak output voltage of 3.8 V from the sensor). As the pre-load pressure between the patch and skin increases, better intimate contact is established and the sensation is increased under the same actuation source. However, if the external load is further increased (pre-load pressure level 4 or 4.3 V from sensor readout), the average sensation intensity is reduced, probably due to the ultra-tight contact between the actuator and skin which limits the motions of the actuator. In order to adjust the human sensation to a similar level under different pre-load pressure levels, the driving voltage of the actuator is adjusted as shown in Figure S16. It is found that the cases of initially low average sensation can be adjusted to values close to the sensation value of 2.5 by using the actuator mode of 3.6 V/μm (pre-load pressure level 1), 3.2 V/μm (pre-load pressure level 2), and 2.6 V/μm (pre-load pressure level 4), as shown in Figure 6e. In other words, the sensation signal differences due to the initial installation tightness of wearable devices and/or the muscle movements of the arms could be compensated in real-time by using the sensing function of the device as the guidance to adjust the magnitude of the actuator for better sensing sensation uniformity.
Figure 6. The feedback control example using both the sensor and actuator functions. (a) Schematic diagram indicating the loose and tight contact between the actuator/sensor patch and skin when the arm stretches and bends. (b) Photo for the setup to apply pre-load pressure on the sensor/actuator patch, in which the loading pressure is adjusted by an air pump and the sensor is calibrated by a pressure gauge. (c) Applied pressure vs. sensor output voltage as a calibration process for the device using the sensor function. (d) The average sensation intensity averaged by 10 volunteers of about 1, 1.5, 2.6 and 1.7, respectively, under 4 different pre-load pressure levels.
of 1.25, 2.5, 3.75 and 5 kPa with the 2.2, 3.1, 3.8 and 4.3 V readouts from the sensor. (e) After adjusting the applied voltage for the device using the actuator function with the feedback of the pre-load pressure levels, similar sensation intensity under different pre-load pressure levels can be achieved.

**Integrated actuator/sensor array**

A single piezoelectret film is utilized to construct 5 sensors and 4 actuators with the size of $1 \times 1 \text{ cm}^2$ each and the gap between each element is 0.2 cm, as shown in Figure 7a, Figure 7b and Video 5 show the operation of the sensors connected to LCDs. When a specific sensor pixel (S3 in this demonstration) is pressed, the corresponding LCD is illuminated without crosstalk as the voltage output of S3 is much larger than that of any other pixel at over 1.5 V and the detailed testing results of various conditions are summarized in Figure S17 and Figure S18. On the other hand, the actuation characteristics of the actuator pixels are measured using a scanning LDV (Laser Doppler Velocimetry) as shown in Figure 7c. A PDMS film (thickness of 1 cm) substrate is utilized and actuators are driven under a constant electrical field and frequency of 1.33 V/μm and 55 Hz, respectively. The measured displacement distribution shows much larger displacements at the centers of the pixels with a peak displacement of over 300 nm, as indicated in Figure 7d. FFT analysis results in Figure S19 show an obvious frequency response at 55 Hz from the center of an actuator (A2 in this demonstration), implying that the measured signals are from the actuator with minimum interferences. The actuator pixels are also driven with frequencies of 50, 60, 65, and 70 Hz as summarized in Figure S20, further proving the anti-interface capability between pixels.
Figure 7. Integrated sensors and actuators. (a) An optical photo showing 5 sensor (S) pixels and 4 actuator (A) pixels on the same piezoelectret film. (b) An optical photo for touching a specific sensor pixel (S3) with a human finger. (c) An optical photo depicting the setup for measuring the vibration amplitudes of actuator pixels. (d) Measured displacement distributions for the actuator pixels under a driving voltage and frequency of 1.33 V/μm and 55 Hz, respectively.
Conclusion

Large equivalent piezoelectric coefficient is the key to enhance the performances of both the actuator and sensor functions, including the sensitivity of the sensor and to lower the driving voltage of the actuator. In this work, the equivalent piezoelectric coefficient of our sandwich-structured piezoelectret reaches 4050 pC/N, which provides the good foundation for both sensor and actuator functions with outstanding performances, such as the pressure sensing limitation as low as 1.84 Pa and a peak output force as high as 20 mN under an applied input of 3.33 V/μm. Experimental and simulation results further prove that increasing the charge density on the FEP electret film with geometric design and material optimizations of the device, such as thinner thickness and larger cavity to spacer area ratio are effective strategies to improve the piezoelectric coefficient and performances of the device. However, limited structural geometries (circular-shape cavity) and spacer sizes have been implemented in this work, while optimizing the geometry design (such as honeycomb structure) in the future should enhance the performance of the piezoelectret device. Furthermore, by replacing FEP with electret having larger maximum charge density value such as silicon dioxide at 34 mC/m² could further enhance the performances. These are some of the principles and directions to expand the current results toward actuator/sensor patches for practical mechanical human-machine interface applications.
Experimental section

**Fabrication of the Ecoflex film with circular-shape holes.** Two kinds of Ecoflex precursor (Ecoflex 00-35, Smooth-On) are uniformly mixed in a 1:1 ratio. Then, the mixture is spin-coated on a Teflon (PTFE) substrate (size of 7.5×5 cm²) using speeds (Laurell Model WS-650MZ) of 200, 300 and 400 rpm to form films with thickness of 350, 250 and 150 μm, respectively. After 5 min under a curing temperature of 50 °C, the films are cured. A tissue biopsy punch (Milex, Japan) with diameter of 2 mm is used to perforate the Ecoflex films to have different hole concentrations, with assorted hole to spacer area ratios of 4.4%, 17.3% and 39.8%, respectively. The prepared Ecoflex films can be easily peeled off from the PTFE substrate because of the low surface energy of PTFE.

**Fabrication of sandwich-structured piezoelectret.** Both sides of Ecoflex spacer are treated with O₂ plasma for 20 min with plasma power of 210 W and O₂ pressure of 350 mTorr (Plasma Equipment Technical Services Inc.). Then the spacer is soaked in a 10% wt APTES (99% purity, Sigma-Aldrich) aqueous solution for 24 h to fully modify the surface with APTES. Following the soaking process, the Ecoflex spacer is thoroughly dried with N₂ gas. At the same time, FEP films (thickness of 25 μm, Chemours Teflon) are treated with O₂ plasma for 10 min with the plasma power of 210 W and O₂ pressure of 350 mTorr to have -OH groups on the surface. Then, two pieces of -OH-modified FEP films are pressed to sandwich a treated Ecoflex spacer for 12 hours under 40 °C. The FEP films are permanently affixed to the Ecoflex film due to the strong chemical bonding between the APTES and -OH groups.
**Characterization.** The morphology of the samples is examined by a high-resolution Field Emission Scanning Electron Microscope (FEI Quanta 3D FEG) and Atomic Force Microscope (AFM, Dimension 3100 Veeco). A resonator (Electroforce 3200, Bose, MN) is utilized to supply the periodic compression force to a fabricated device attached to a force gauge (50 lb-f Load Cell). The output current signals from the sensor are recorded using a Stanford low-noise current preamplifier (Model SR570) and NI PCI-6259, and the output voltage signals are recorded by a Keithley 6514 electrometer. The actuator is driven by a TOS 5051 high voltage power source or a PI E-463 HVPZT AMPLIFIER. The vibrational signals of the actuator are measured by a Laser Doppler Velocimeter (LDV, Polytech OFV5000). The force signals generated by the actuator are detected by a sensitive force gauge (2 lb-f Load Cell).

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**Supporting Information Available.** More detailed information about additional characterizations of materials and devices, tables for comparison of the equivalent piezoelectric coefficients and different types of actuators, supplementary videos and supplementary Explanation 1 and 2. These materials are available of charge via the Internet at [http://pubs.acs.org](http://pubs.acs.org).
REFERENCES AND NOTES


A sandwich-structured piezoelectret-based actuator/sensor patch has action and perception in response to environmental and user’s stimuli. The high piezoelectric coefficient endows such devices with excellent performances in providing haptics feedback to human skin and sensitively detecting pressure/force signals from human motions, proving the potential wearable and human interactive applications in augmented reality (AR) and virtual reality (VR), etc.