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Gel-Based Artificial Photonic Skin to Sense a Gentle Touch by Reflection

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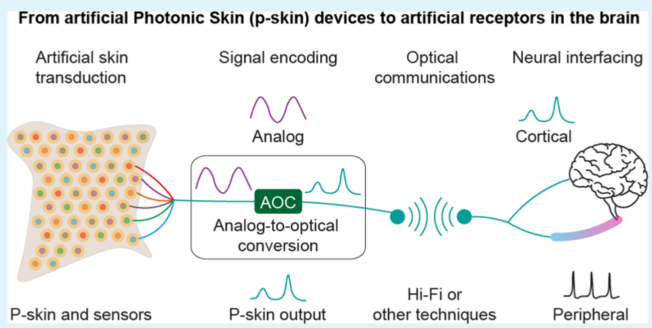
Supporting Information

ABSTRACT: This work demonstrates that engineering a three-dimensional photonic crystal (3DPC) structure in a highly flexible gel is a potential method to achieve flexible tactile artificial photonic skin (p-skin) for future visible-light communication (VLC). We investigated the photonic output modes of 3DPC-coated gel-based pressure sensors and explored their ability to sense low pressures (<10 kPa) through reflection. Such sensors with high sensitivity, fast response, and adjustable detection range can be fabricated in arrays of dots covering large, complex/uneven surfaces and are promising in the development of stimuli-responsive soft materials for future artificial intelligence, health monitoring, and photonic communication systems.



KEYWORDS: *artificial photonic skin (p-skin), gel, tactile sensing, reflection, photonic crystal*

The development of flexible, sensitive artificial skins that can respond to tactile stimuli in the environment is of paramount importance for future generations of smart robots, implantable artificial skins, and physiological monitoring systems. As an uncommon tactile sensing technology, artificial photonic skin (p-skin) devices have a number of advantages and are not only supported by rapidly developing techniques in applied spectroscopy but also fueled by the need for advanced photonic communications. In addition, p-skin devices offer a high degree of stability because they are far less susceptible to corruption by environmental influences such as electromagnetic fields than electrical/magnetic devices. Therefore, we focus on the p-skin technology in this article. However, the majority of reported photonic pressure sensors have shown low sensitivity, slow recovery (in minutes or more), large hysteresis, and poor flexibility and moldability in low-pressure regimes (<10 kPa).[1−6] In sharp contrast, natural skins are extremely flexible and moldable so as to continuously cover the whole body, safeguarding the internal organs from external environments, using countless pressure-sensitive pixels with high sensitivity in regimes of both low (<10 kPa, comparable to a gentle touch) and medium (10−100 kPa, suitable for object manipulation) pressure.[7] Some flexible soft materials, such as rubbers polydimethylsiloxane (PDMS) and poly(methyl methacrylate) (PMMA), have been widely applied as flexible pressure sensors to mimic natural skins.[1,2,6−10] Although the surfaces built from nonabsorbing soft materials can reflect light specularly or diffusely with great efficiency, poor photonic feedback has impeded the development of the flexible p-skin technology.

Recently, soft materials with three-dimensional photonic crystal (3DPC) structures have been widely studied and applied in various sensors.[11−15] Due to the structural periodicity, 3DPC materials exhibit remarkable reflection within a certain frequency range called photonic band gap (PBG), where electromagnetic waves are forbidden to propagate through the materials. Changes in the periodicity of 3DPC materials contribute to variations in the position and intensity of the reflection peaks.[16] With the unique photonic property, 3DPC-based p-skin devices can achieve remarkable photonic feedbacks in response to gentle touch, which are impossible to achieve with conventional photonic approaches. However, the fabrication of most 3DPC materials requires complex machinery or delicate operations, such as photo-

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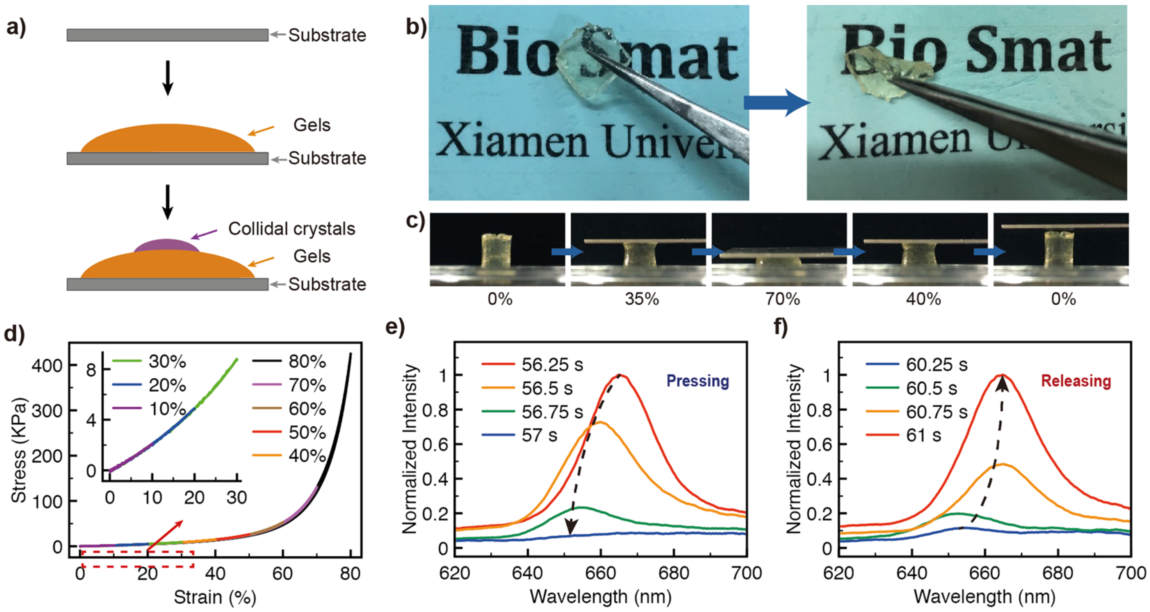

Figure 1. (a) Schematic of the process of sensor fabrication. (b) Photographs of the folding process of a keratin-based gel sample. (c) Photographs of a gel sample (cut into a circular cylinder 5 mm in diameter and 6 mm in height) under different pressure strains (as labeled below). (d) Mechanical properties of the keratin-based gel. (e) Reflection spectra of a gel-PC sample during the pressing process. (f) Reflection spectra of the gel-PC sample during the release process.

lithography, electron beam etching, direct laser writing, and inverse-opal techniques.[17−23] The above fabrication methods are suitable for the small-scale production of mostly inorganic materials, including metals and semiconductors, but are not applicable to microstructured soft materials due to their limited chemical/physical stability. Therefore, to overcome the barrier, there is a need to introduce a simple, mild, and cost-effective strategy for achieving 3DPC-based p-skin devices using soft materials.

Very recently, several groups, including ours,[24−28] have focused on soft material-enabled, flexible hybrid devices, which originate from wool, spider silks, and cocoon silks. On the basis of the above advanced soft materials, we report a simple, mild, and cost-effective strategy to achieve a flexible p-skin device for tactile sensing. With the unique photonic features of 3DPC materials and the remarkable mechanical properties of keratin gel, 3DPC-coated gel-based p-skin devices exhibit a simple, fast, and sensitive photonic response to various pressure regimes. In addition, keratin materials are easily coated over complex/uneven surfaces, and later behave as stable gel materials with large compression plasticity. Moreover, colloids are spontaneously assembled in close-packed arrays on the gel surface, which can rapidly respond to tiny deformations in the keratin gel as pressure is applied. Intelligent p-skin devices can be suitable platforms for the development of stimuli-responsive soft materials for future robotics, prosthetics, and artificial intelligence and health monitoring systems.

Figure 1a illustrates the fabrication process of flexible pressure-sensitive p-skin devices incorporating high-flexible gel with 3DPC material. The keratin-based gel was initially achieved via the Michael addition reaction (Figure S1). The original keratin solution was first drop-cast upon complex/uneven surfaces, and in the later stage, the solution turned to stable gel with high flexibility, large compression modulus and low stress relaxation (Figure 1b−d, Figure S2). A monodisperse colloidal emulsion was then drop-cast upon the gel to obtain the 3DPC-coated gel material.

Such fabricated keratin-based gel is good candidate for tactile detection applications. The self-assembled colloidal crystals were originally in close-packed arrays with small randomness and exhibited bright coloration (Figure S3). Because of the mechanical and adhesion features, the gel can quickly respond to pressure stimuli by undergoing deformation, which simultaneously affects the arrangement of the colloid crystals. To prove the pressure-response ability, we recorded the dynamic reflection spectra of polystyrene (PS) colloids for further analysis (Figure S4). The detailed spectra of a 3DPC-coated gel sample with a diameter of 1 cm indicate the fast (approximately 0.75 s) and sensitive response to pressure cycles of 2.42 ± 0.05 kPa (Figure 1e, f). When pressure was applied upon the 3DPC-coated gel sample, the reflection intensity descended from 100% to approximately 10%, and the reflection peak was blueshifted by approximately 10 nm. When the pressure was released from the sample, the reflection intensity rose back to the original value of 100%, and the reflection peak was red-shifted back to the original peak position at approximately 665 nm (Text S1). The 3DPS-coated gel sensors are suitable for reproducible detection of low pressure.

To further demonstrate the working principle of the pressure-sensitive p-skin devices, the photonic output will be revolved by two independent cases. The first one was to stretch the 3DPC-coated gel samples by approximately 20% (without pressing), resulting in a blueshifting of the reflection peak (λ_{max}) by approximately 30 nm and a reduction in the reflection intensity by 50% (Figure 2a). The simulated results in Figure 2b, c show that in the stretching process, with the transition of the state from ordered to disordered, the increasing randomness of the colloidal arrays resulted in the blueshifting of the reflection peaks and the decrease in the reflection intensity. The randomness is one of vital structural parameters, i.e., not the only cause of reflection shifting (Text S2).

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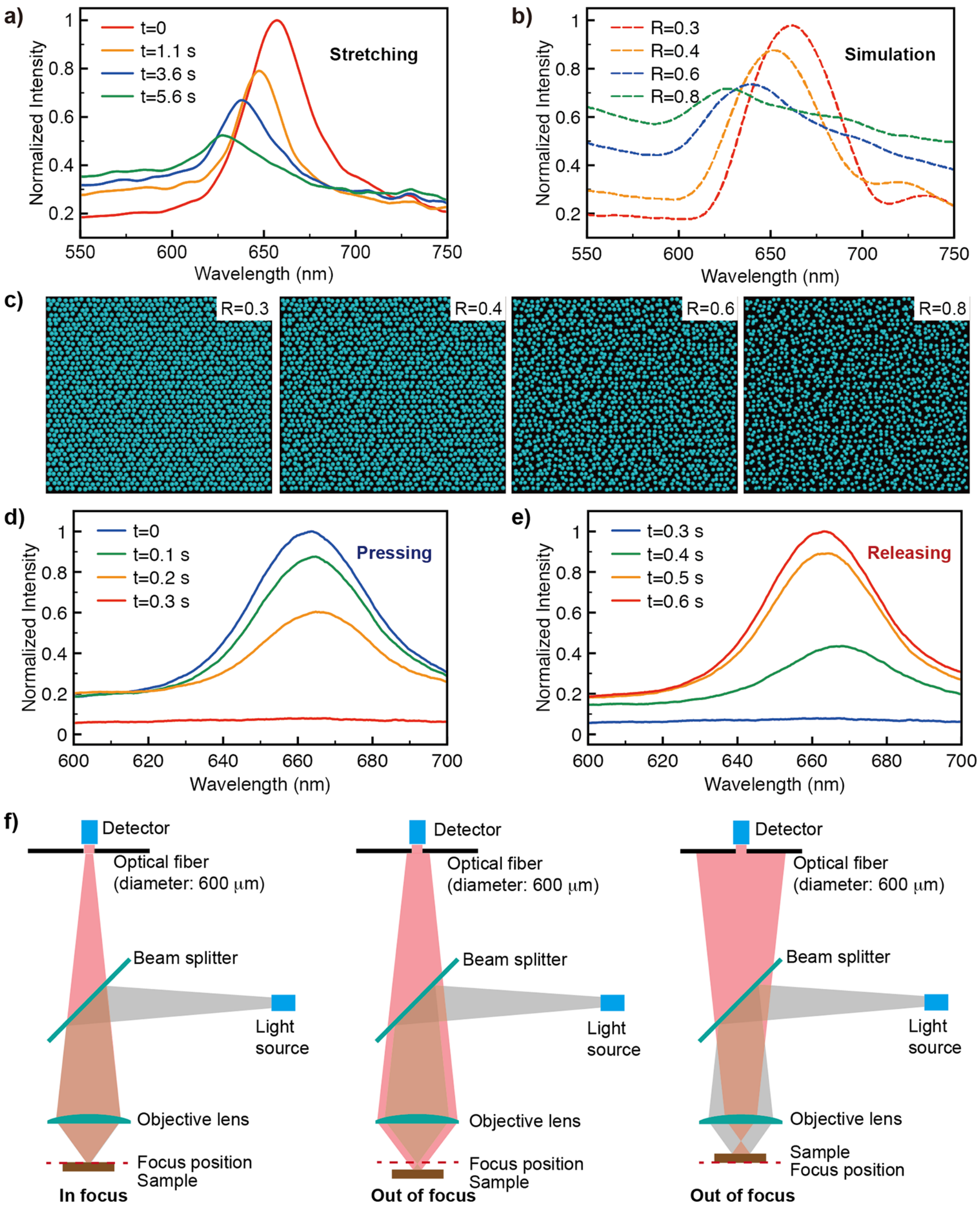

Figure 2. (a) The reflection spectra changed when a gel-PC sample was stretched. (b) Reflection spectra and (c) images of a simulation of PS colloidal arrays with different randomness. The reflection spectra of the gel-PC samples changed when pressure was (d) applied and (e) released at high speed. (f) Schematics of the in-focus and out-of-focus detection modes.

In contrast, the second case was to quickly apply pressure cycles upon the PC-coated gel samples (without stretching). The colloidal arrays quickly and remarkably responded to the external pressure stimuli, as shown by the measured reflection intensity, whereas the reflected peaks were positioned at approximately 665 nm (Figure 2d, e). The progress is briefly illustrated in Figure 2f. When pressure is applied at the in-focus stage, a hand-held spectrometer (detector) can harvest the focused reflected light through an optical fiber with a diameter of 600 μm. By contrast, when pressure is applied at the out-of-focus stages, the detector can collect less reflected light than that collected at the in-focus stage; therefore, the reflection intensity is decreased. Here, the optical fiber functioned as a valid light entrance. Thus, the 3DPC-coated gel sensors can provide a multisignal photonic output for pressure stimuli.

To simplify the response analysis process, the reflection intensity of the 3DPC-coated gel-based sensors was optimized as the main evaluation parameter in further investigations. During the pressure cycles, the 3DPC-coated samples with a diameter of 1.0 cm exhibited reproducible and reliable photonic responses to pressure ranging from 0.24 to 2.42 kPa (Figure 3a–d). Notably, after 300 pressure cycles of approximately 2.42 kPa with an approximately 85% reduction in the reflection intensity, the gel can still be stretched repeatedly without macroscopic breakdown and is thus suitable for tactile sensing applications.

Because the deformation of the gel is dependent on its dimensions, the photonic outputs of the colloid coating will change as the gel size varies (Text S3). 3DPC-coated gel samples with different diameters (1.0 and 0.7 cm) were fabricated to investigate the pressure–response ability. The

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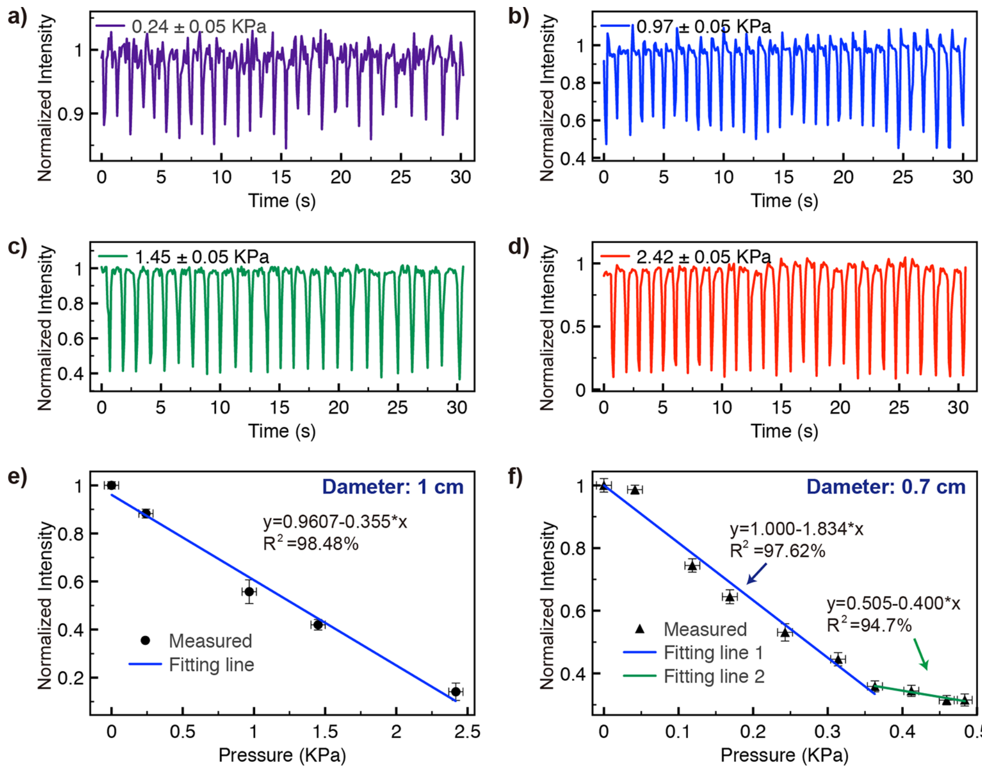

Figure 3. Normalized reflection intensity changes of a gel-PC sample with a diameter of 1 cm when pressed at (a) 0.24 ± 0.05 kPa, (b) 0.97 ± 0.05 kPa, (c) 1.45 ± 0.05 kPa, (d) 2.42 ± 0.05 kPa and (e) different levels (0−2.50 kPa), respectively. (f) Normalized reflection intensity changes in a gel-PC sample with a diameter of 0.7 cm when pressed at different levels (0−0.50 kPa).

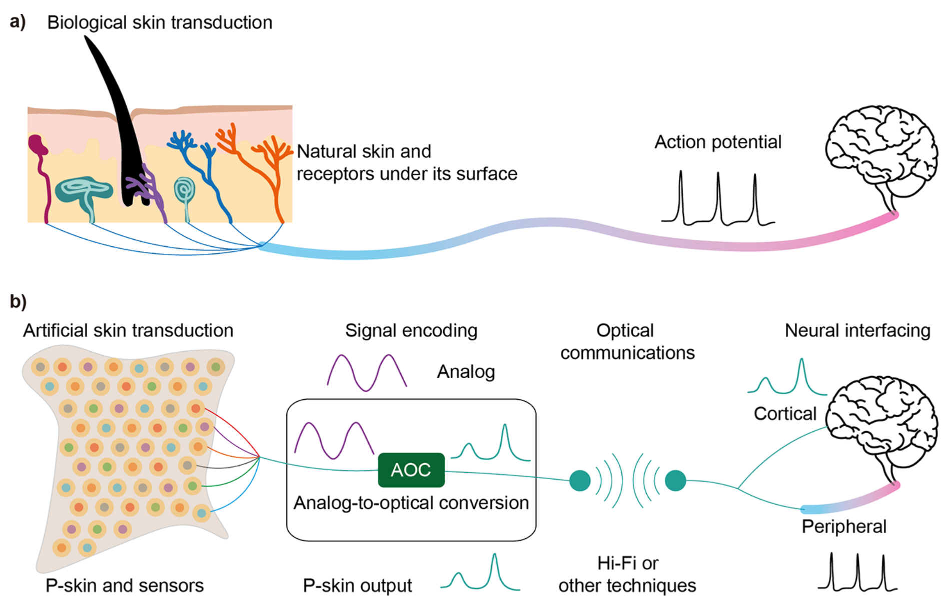

Figure 4. Schematics of the steps required for the transduction of sensory stimuli from (a) natural or (b) artificial receptors in the brain. Signal collection, encoding in photonic signals mimicking action potentials, photonic communications and neural interfacing are all key issues that need to be addressed to add sensing capabilities to p-skin devices.[29]

3DPC-coated samples with a diameter of 1.0 cm exhibited a sensitivity of 0.355 kPa^{-1} and a linearity of 98.48% in the pressure-detection range of 0−2.5 kPa (Figure 3e). The 3DPC-coated samples with a diameter of 0.7 cm exhibited a nearly linear response in the pressure range of 0−0.35 kPa, with a sensitivity of 1.834 kPa^{-1}, a linearity of 97.62%, and a steady trend in the pressure range of 0.35−0.50 kPa (Figure 3f). As expected, the gels with smaller sizes achieved smaller pressure-detection ranges and higher sensitivity.

Moreover, to illustrate the coloration ability of 3DPC materials, we coated a keratin gel sample with the Allura Red dye as the control, and the pressure−response behavior was characterized by reflection spectroscopy. Figure S5 shows broad spectra and fairly low output for the sample coated with the Allura Red dye, while the spectra for the sample coated

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with PS colloids were sharp with a striking contrast. The results indicate that the p-skin design of 3DPC-coated gel can provide remarkable, sensitive, and reproducible photonic output with a controllable detection range in response to an external gentle touch.

Importantly, the sensitivity of the PC-coated gel-based sensors can be further improved by decreasing the gel size. Therefore, the pressure sensors could be fabricated on large, complex/uneven surfaces in arrays of dots with excellent uniformity in size, height, shape, and period using 3D printing or other advanced techniques. The extremely small variance in the size of the dots can ensure a clearly defined and reproducible photonic output for pressure sensing. If the resolution of the dot-shaped pressure sensors with different sizes was tuned at the resolution of natural skins, the pressure sensing would cover the entire body with greatly improved detection sensitivity and broad detection range. Our hypothesis demonstrates that PC-coated gel-based sensors are promising mechanical sensing p-skin designs with potential applications.

Furthermore, multifunctional p-skin devices will be developed to offer more applicability and better practicality with rapidly developing new materials and new processing approaches. Our initial hypothesis of a future multifunctional p-skin system is that the main components will comprise a photonic sensor, a signal encoder, photonic communications, and an approach to convey the photonic output to the nervous system (Figure 4).[29] Although some of the components are currently limited, especially in photonic communications and neural interfacing, several technologies, such as flexible photonic devices, light fidelity (Li-Fi) technology, and artificial nervous systems, have achieved remarkable results for the further development and production of p-skin devices. Meanwhile, the development of p-skin technology is promising for rapid and continuous advancements in multiple fields, including material science and engineering, photonic communication, and neural interfacing and associated materials and devices. Therefore, the 3DPC-coated gel-based pressure sensors we have developed may prove valuable in further multifunctional p-skin devices. Both challenges and potentials exist in the applications of comprehensive and programmable p-skin devices for future artificial intelligence, health monitoring, and photonic communication systems.

In conclusion, we report here a simple yet efficient coating approach to fabricate an artificial p-skin device by incorporating highly flexible gel with a 3DPC material. The entire fabrication process is accessible, controllable and cost-effective. Our p-skin devices could rapidly detect low pressures (0−2.5 kPa) with a sensitivity of 0.355 kPa^{-1} and a linearity of 98.48% through reflection in the low-pressure regime (with a diameter of 1 cm). We also demonstrated that the sensitivity and measurement range of the p-skin devices could be tuned simply by adjusting the dimensions of the gel. Moreover, the p-skin devices, which possess the excellent flexibility of the gel and the extraordinary photonic output of the colloidal crystals, are uniform in size, shape and period, and the devices can cover and adhere to large-scale, complex/uneven surfaces. The properties will contribute to the real-time, ultrasensitive, and reproducible photonic communications for an external touch such as that of natural skin. Furthermore, we demonstrate an artificial skin transduction system through photonic communications. We believe that p-skin devices can be developed with more functions and may prove valuable in future artificial intelligence, health monitoring, and light communication systems.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsami.9b01649.

Materials, experimental details, reflection peak positions, definition of randomness, measurement strategy and dimensions of gel, characterizations of keratin-based gel (SEM, photographs), mechanical property of silicone rubber and PDMS, characterizations of PS colloidal crystals (SEM, microphotograph), pressure response of a gel-PC sample, pressure sensing of a dye-coated gel sample, schematic of measurement setup, dimensions of gel (PDF)

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

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Notes

The authors declare no competing financial interest.

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ABBREVIATIONS

3DPC, three-dimensional photonic crystal;; p-skin, photonic skin; VLC, visible-light communication; PDMS, polydimethylsiloxane; PMMA, poly(methyl methacrylate); PBG, photonic band gap; PS, polystyrene; Li-Fi, light fidelity

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