

Integrated Soft Ionotronic Skin with Stretchable and Transparent Hydrogel–Elastomer Ionic Sensors for Hand-Motion Monitoring

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Abstract

Skin-like stretchable sensors with the flexible and soft inorganic/organic electronics have many promising potentials in wearable devices, soft robotics, prosthetics, and health monitoring equipment. Hydrogels with ionic conduction, akin to the biological skin, provide an alternative for soft and stretchable sensor design. However, fully integrated and wearable sensing skin with ionically conductive hydrogel for hand-motion monitoring has not been achieved. In this article, we report a wearable soft ionotronic skin (iSkin) system integrating multichannel stretchable and transparent hydrogel–elastomer hybrid ionic sensors and a wireless electronic control module. The ionic sensor is of resistive type and fabricated by curing ionic hydrogel precursor on a benzophenone-treated preshaped elastomer to form a hydrogel–elastomer hybrid structure. The hydrogel–elastomer hybrid iSkin is highly stretchable ($\sim 300\%$ strain), transparent ($\sim 95\%$ transmittance in the visible light range), and lightweight (<22 g). Experiments demonstrate that the fully integrated iSkin system can conformably attach onto the dexterous hands for recognizing the joint proprioception and hand gesture, and understanding the sign language. Our iSkin system would also provide a test bed for customized material selection and construction in a variety of applications.

Keywords: ionotronic skin, ionic sensor, hydrogel–elastomer hybrid, soft and stretchable sensor, hand-motion monitoring

Introduction

SKIN PLAYS AN IMPORTANT ROLE for humans to feel the world.¹ Achieving skin's sensing capability in wearable devices and robotics is a long-lasting challenge. Although many skin-inspired wearable or implantable sensors have been developed for motion detection and health monitoring, they are generally made of rigid and brittle electronic materials without high enough stretchability, resulting in fundamental mismatch and uncomfortable in mechanics with human bodies.^{2–5} Human skin not only has powerful sensory organs but also is soft and stretchable to adapt the body motions. Development of soft and stretchable sensors is of significance in the realm of wearable sensing skins and has

the advantages of higher flexibility, better adaptability, and conformal integration over their rigid counterparts.⁶

Over the years, intensive efforts have been made in skin-inspired stretchable sensors. One clever concept is to exploit inorganic semiconductor materials in the unconventional deterministic thin architectures (such as micropatterned, wavy, or buckled geometries).^{7–10} However, such semiconductor sensors are flexible but not intrinsic stretchable, and complicated structures and fabrication techniques are required for stretchable sensing system design. To support soft and stretchable functionality, the majority of stretchable electronics have been proposed by using the soft conductive electrodes with intrinsic stretchable substrates (such as silicone rubbers).^{11–15} The choice of conductive electrodes

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includes conductive fabrics and polymers,^{16–19} organic semiconductor nanocomposites,^{20–22} carbon nanotube films,^{3,23–26} and liquid metals.^{27–30} These conductive electrodes transmit the sensing signals using electrons, whereas sensors in our skin transmit signals using ions.³¹ Alternatively, stretchable ionotronics using the soft ionic conductors have been becoming another promising technology for wearable sensing skins.^{32,33} Hydrogel is one attractive selection as ionic conductors owing to its distinctive attributes, including inherent softness (with the modulus of ~ 8 kPa), high stretchability (beyond ~ 20 times the initial length), transparency, biocompatibility, and/or biodegradability.^{34–40} Currently, various ionic hydrogel conductors have been reported, for instance, ionic cables,^{41,42} strain/pressure sensors,^{43–46} touch panels,⁴⁷ self-healing sensors,^{48,49} and hybrid circuits.^{50,51} These studies have well demonstrated the capability of ionic hydrogel conductors to sense body motions like human fingers. However, efforts to incorporate the multichannel ionic hydrogel sensors into fully integrated and untethered wearable sensing skins for hand joint proprioception and gesture recognition have not been achieved.

In this study we report a highly stretchable transparent wearable soft ionotronic skin (iSkin) system integrating 10-channel hydrogel–elastomer hybrid ionic sensors and a wireless electronic control module (Fig. 1), which conformably attaches onto our hand and monitors the real-time hand motions. The ionic sensor is of resistive type and fabricated by curing hydrogels on a benzophenone-treated preshaped elastomer to form the hydrogel–elastomer hybrid structure. The integrated iSkin system with 10 ionic sensors covers the metacarpophalangeal joint and proximal-interphalangeal joint of each finger for joint proprioception and hand gesture monitoring, which is highly stretchable ($\sim 300\%$ strain),

transparent ($\sim 95\%$ transmittance in the visible light range), and lightweight (<22 g). For ease of usability, we further integrate the iSkin system with a wireless electronic control module consisting of digitizers, a microcontroller, a Bluetooth transceiver, and a cell battery. Flexibly wavy circuit interconnects are used to connect the ionic sensors to the control module. In this sense, the real-time hand motions can be detected by our integrated wireless iSkin system and displayed in distal devices (such as a mobile phone). By experiments, we demonstrate that the integrated wireless iSkin system exhibits the ability to identify various hand motions, including the joint bending, hand posture, gesture, and sign language. The stretchability, transparency, wearability, and soft interaction make our integrated wireless iSkin system promising in various applications such as soft robotics, prosthetics, and wearable medical devices.

Materials and Methods

Synthesis of the ionic hydrogel

The ionic hydrogels are the polyacrylamide (PAAm) hydrogel containing lithium chloride (LiCl). The PAAm–LiCl hydrogels are synthesized by using the acrylamide (AAM; J&K) as the monomer, *N,N*-methylenebisacrylamide (MBAA; Molbase) as the crosslinker, LiCl monohydrate ($\text{LiCl} \cdot \text{H}_2\text{O}$; Sinopharm Chemical Reagent) as the ionic conductive medium, and 2-ketoglutaric acid (Adamas) as the photoinitiator. The synthesis steps are described as follows. The monomer solution is prepared by mixing AAM, $\text{LiCl} \cdot \text{H}_2\text{O}$, and deionized water at the weight ratio of 9.98%:16.16%:73.86%. MBAA solution is dissolved in deionized water with a weight ratio of 1.2%. Then, the monomer

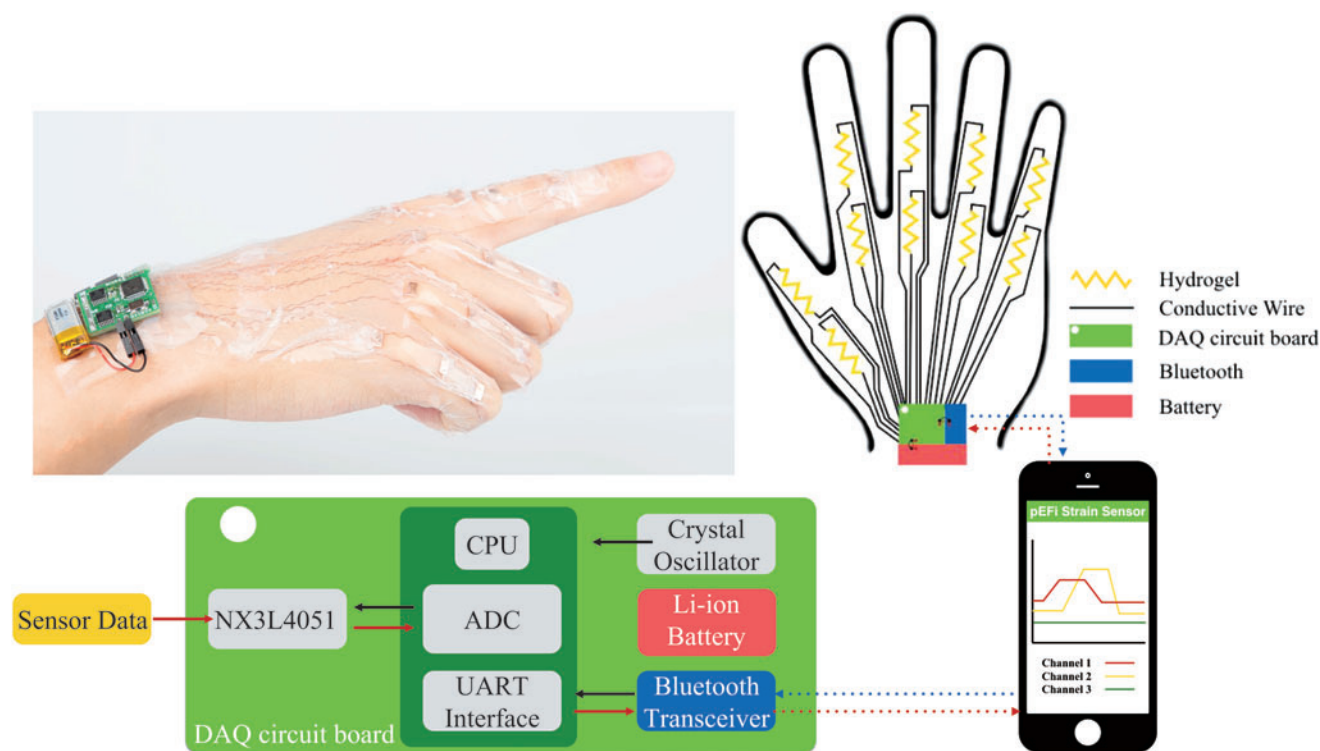


FIG. 1. Overview of the soft iSkin system integrating 10-channel hydrogel–elastomer hybrid ionic sensors and a wireless electronic control module. iSkin, ionotronic skin. Color images are available online.

solution, MBAA solution, and 2-ketoglutaric acid are mixed with the weight ratio of 96.67%:1.13%:2.20% to form the hydrogel precursor ink for further fabrication. We may mention that LiCl is contained into the hydrogels, not only serving as conductive medium but also as one kind of hygroscopic salts.⁵² To further avoid the dehydration problem of ionic hydrogels, we will encapsulate them with the elastomer layers, which will be detailed in the following development.

Fabrication of hydrogel–elastomer hybrid iSkin

To prevent the dehydration of ionic hydrogels and achieve the robust strain sensing, we fabricate the iSkin with the hydrogel–elastomer hybrid structure. The used elastomer is made of polydimethylsiloxane (PDMS) elastomer (Silpuran film; Wacker), which has good breathability and biocompatibility.⁵³ As shown in Figure 2A, the ionic hydrogel is sandwiched by two PDMS elastomer films to form anti-dehydration structure. To alleviate oxygen inhibition effect and activate elastomer surfaces for the robust bond with ionic hydrogels, benzophenone is used to modify the PDMS elastomer surface.⁴² As shown in Figure 2B to D, we can see that the fabricated hydrogel–elastomer hybrid ionic sensor is highly stretchable ($\sim 300\%$ strain) and transparent ($\sim 95\%$ transmittance in the visible light range).

In Figure 2E, we provide a schematic illustration of the fabrication process for the iSkin system with 10 ionic sensors and the wireless control module. (i) A thin PDMS elastomer film ($100\ \mu\text{m}$) is used as the sensor substrate, and a laser cut mold array for the ionic hydrogel sensors is arranged onto the

substrate according to the distribution of metacarpophalangeal joints and proximal-interphalangeal joints of human hands. After the surface treatment with the benzophenone, the hydrogel precursor ink is injected into the mold through a needle tubing. (ii) Ultraviolet light with 500 W and 365 nm wavelength is applied to initiate the crosslinking for about 20 min. (iii) The mold array is peeled off and electrical circuits including the flexible interconnects and portable control module are integrated onto the elastomer substrate. Notably, the flexible interconnects are made of serpentine shape copper wires for a better stretchability.⁴⁰ (iv) The integrated system is cut into a hand-like shape and packaged by assembling another thin PDMS elastomer film ($100\ \mu\text{m}$), where the pre-elastomer resin (DowCorning Sylgard 184) is used as the adhesive between the two thin PDMS films. Notably, the integrated iSkin system will be further cured for about 1 h under room temperature.

Description of the control module

The wireless electronic control module used in the iSkin system consists of an analog interface with two 8-channel analog switches (NX3L4051; NXP) for multiplexing, a 32-bit microcontroller (STM32L152RC; ST Microelectronics) integrating a 12-bit analog-to-digital converter (ADC), and a Bluetooth transceiver (BC-04-B; Cambridge Silicon Radio). The multiplexing analog interface can read 16 channel inputs in maximum. The 12-bit ADC in the microcontroller converts the analog inputs to digital signals by a series connected load

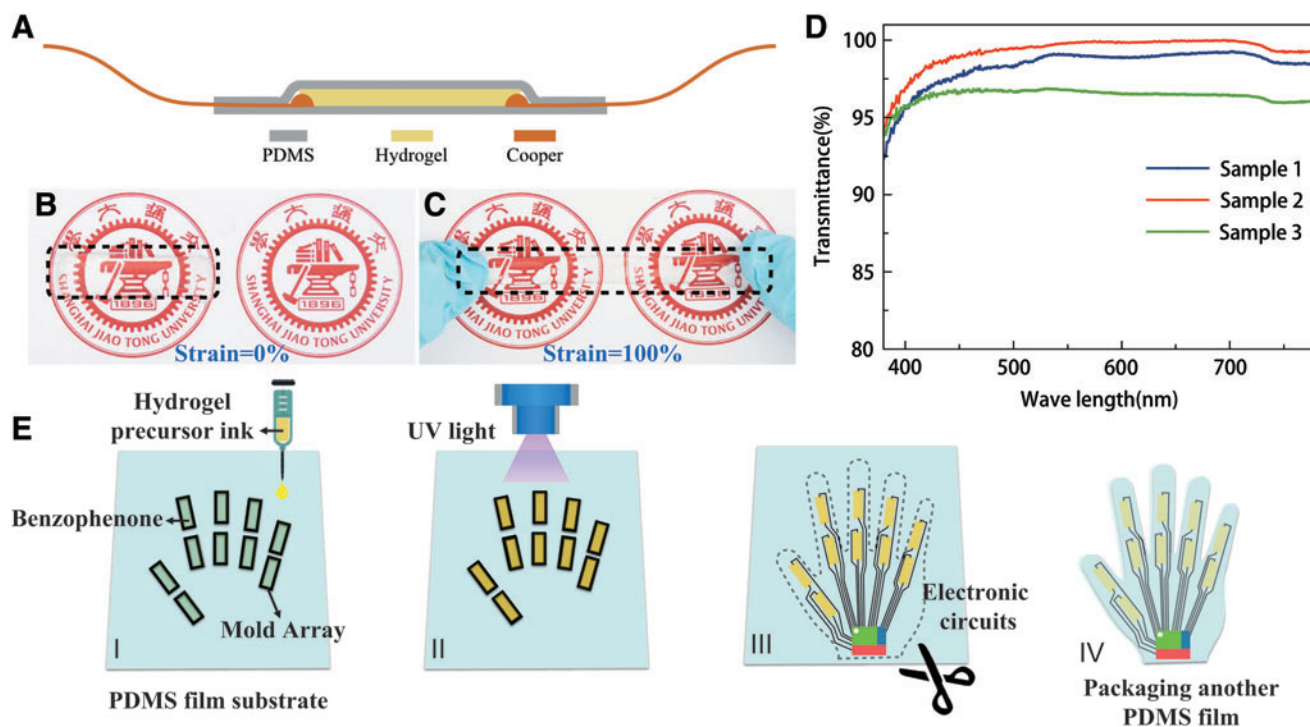


FIG. 2. Fabrication of the soft ionic sensor and iSkin system. (A) Schematic illustration of the ionic sensor in the sandwiched hydrogel–elastomer hybrid structure. The ionic hydrogel layer is strongly bonded to the PDMS films through the benzophenone treatment. (B) The ionic sensor with 0% strain. (C) The ionic sensor with 100% strain. Since the hydrogel ionic sensor is transparent, the black dotted box indicates the real size of the ionic sensors. (D) The transmittance of ionic sensor. (E) Schematic illustration of the fabrication process of our integrated iSkin system as listed from I to IV. PDMS, polydimethylsiloxane. Color images are available online.

resistor with a sampling time of 3 ms for further processing. The Bluetooth module is used to build wireless communications with the mobile phone that can transmit a robust signal up to 10 m. The whole transceiver is powered by a 3.3 V rechargeable Li-ion battery. A software developed in Android is run in the mobile phone to process, store, and display the data.

Results and Discussions

Sensor performance characterization

To thoroughly characterize the performance of our hydrogel–elastomer hybrid ionic sensor, a series of performance indicators are introduced and characterized, including the sensitivity, hysteresis, repeatability, durable stability, and detection limit.¹³

The gauge factor (GF) is used to define the sensitivity

$$GF = \frac{\Delta R/R_0}{\varepsilon},$$

where ΔR is the relative resistance change with respect to the nominal resistance R_0 at zero strain and ε denotes the strain of the sensor. Figure 3A plots $\Delta R/R_0$ as a function of ε by performing cyclic loading up to 100% strain at a frequency of 30 Hz. The GF is ~ 0.7762 , with linearity of $R^2 = 0.9944$ in both loading and unloading directions. The results also show that the hysteresis of our ionic sensor is very low, by comparing with the carbon–grease-based strain sensor.¹⁹ This may be attributed to the robust hydrogel–elastomer hybrid structure for a more uniform resistive distribution during stretching.

Extended use of the sensors is to characterize the repeatability by performing cyclic loading up to 100% strain for 50 cycles. The results indicate that the sensor signals are stable and repeatable (Fig. 3B). Further cyclic tests are performed to show the durable stability of our sensor up to 7 days (Fig. 3C). The experimental data demonstrate that the sensor responses agree well among days and the sensor is still sensitive at the end of the experiment, showing the capability for long time use.

To identify its segment retentivity, we stretch and then hold the sensor for 10 s at increasing strains of 20%, 40%, 60%, 80%, and 100%. The real-time data $\Delta R/R_0$ are plotted as a function of time in Figure 3D. We can see that the segment strains are well distinguished and there is slight signal creep when holding it at the strained state. By putting the sensor on metacarpophalangeal joint of an index finger, we can clearly identify the bending angle of the joint at the increasing angles of 0°, 30°, 60°, and 90° (Fig. 3E). From the results in Figure 3D and E, we can see some fluctuation of the ionic sensors. This may be caused by the following: (i) the motion of the finger is generally dynamic, which cannot be fully steady and (ii) the soft materials, including the PDMS and hydrogels, have the inherent viscoelasticity, which needs certain time to stabilize the responses. To further demonstrate its static stability, we used a linear-guide platform to hold the ionic sensor with a certain strain (i.e., 50%), and the measured results are shown in Figure 3F. It can be seen that the resistance of the sensor is relatively static after the viscoelastic response, which will not influence its sensitivity.

We have also performed tests to show the temperature effects on the sensor in Figure 3G and H. The results dem-

onstrate that around the room temperature (i.e., 27°C in our test), the resistance of the hydrogel sensor is almost steady. However, with the increase of the temperature, the resistance of the sensor slightly changes (for instance, the change will be <0.06 when the temperature increases from 17°C to 30°C).

Finally, we demonstrate that the strain limit of the ionic sensor is $\sim 300\%$ (Fig. 3I), which is higher than stretchability of our skin.⁹ By the experimental characterization, our ionic sensor with the hydrogel–elastomer hybrid structure demonstrates the robust performance over large strains and cyclic loadings, which can satisfy the requirements of a variety of potential applications.

Integrated iSkin system

Next, we fabricate the soft iSkin to adhere to the hand using a kind of water-borne adhesive (Romantic Angel RL-7030) for commercial cosmetics and monitor the hand motions. Since the moduli of the PDMS (~ 10 kPa) and the hydrogel sensors (~ 8 kPa) are much smaller than our skin (between 0.42 and 0.85 MPa⁵⁴), our iSkin system can provide conformal contact with human hands. As shown in Figure 2E, the iSkin system contains 10 embedded hydrogel ionic sensors that cover the metacarpophalangeal joint and proximal interphalangeal joint of each finger. When a finger joint bends, the located sensor accommodates the bending strain, causing correspondingly change in the electrical signal; when the finger straightens, the sensor also recovers to its initial resistance (Fig. 3E). To be a wearable device, we integrate our iSkin with the portable electronic control module using the flexible wavy circuit interconnects.⁴⁰ The control module captures the sensor signals and wirelessly transmits the signals to a mobile phone through the Bluetooth transceiver protocol. The signals can then be real-time displayed in the virtual interface of the remote mobile phone.

The functionality of our iSkin system is demonstrated by a series of tests. In these tests, the fingers sequentially move through multiple finger gestures. Figure 4A shows the real-time responses recorded from the corresponding 10 sensors of our iSkin system. As can be seen, every slight movement of the metacarpophalangeal joint and proximal interphalangeal joint on each finger can be fast and accurately detected, showing the electrical and mechanical robustness of the integrated iSkin system. By the combination of different sensor signals, we can monitor the hand gesture to recognize the digits 0–9 of the American Sign Language and display them in the virtual interface of the mobile phone (Supplementary Movie S1). Figure 4B also shows that our iSkin system can automatically recognize the gestures of the letters S, J, T, and U of the international sign language to composite into SJTU (abbreviation of Shanghai Jiao Tong University) (Supplementary Movie S2). Since the iSkin system is soft, stretchable, wearable, and biocompatible, it is flexible for multiple users.

Applications of the integrated iSkin system

We next demonstrate the potential applications with our integrated iSkin system in daily life. Figure 5A shows the example to control the brake of a bike using the index finger and middle finger of a hand. It can be seen that the iSkin system can monitor the isochronous motions of two fingers while cyclically opening and closing the index and middle fingers to brake. In contrast, when using a mouse, the index

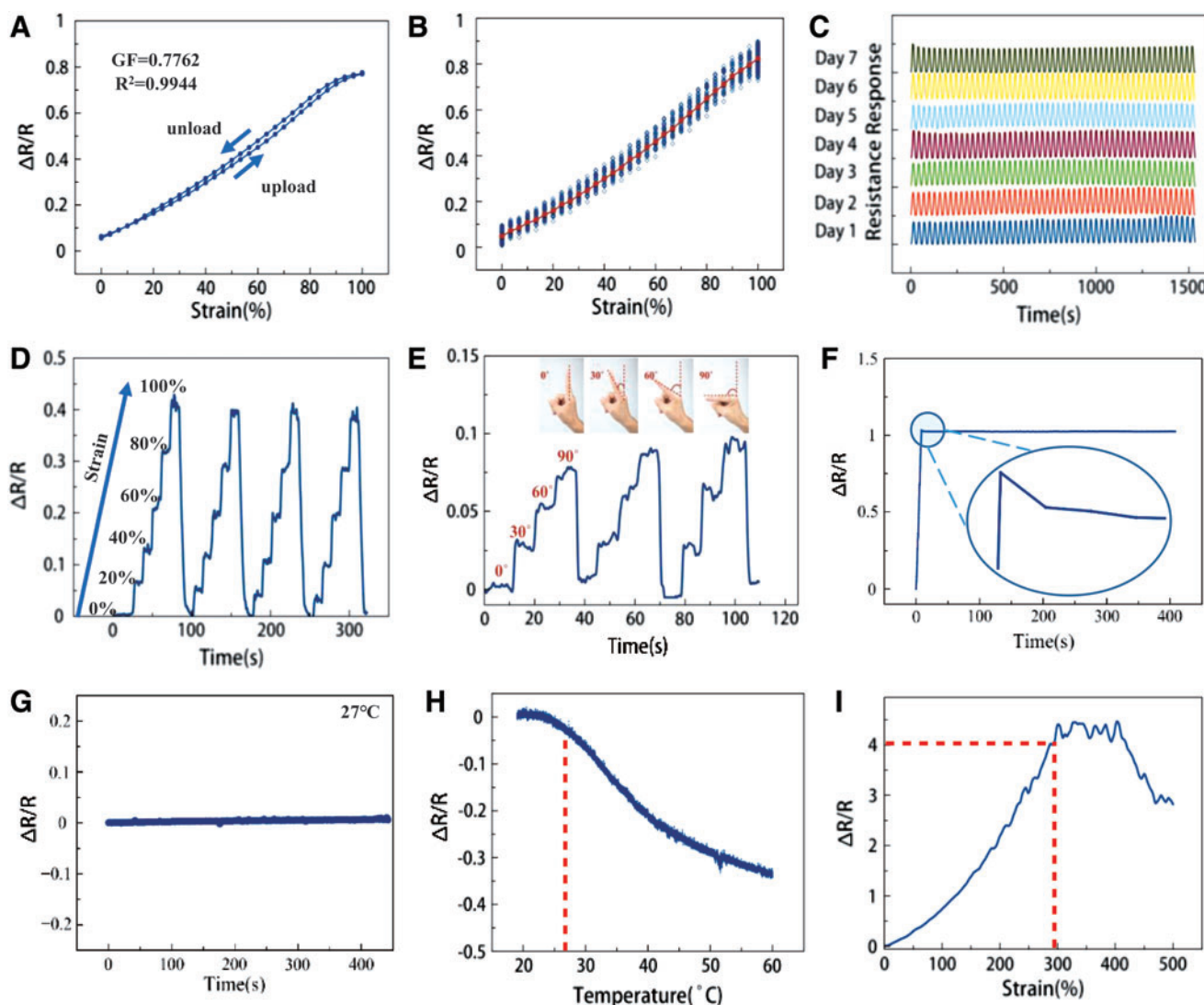


FIG. 3. The performance of a single hydrogel sensor. (A) $\Delta R/R_0$ is plotted as a function of ε by performing cyclic loading up to 100% strain. (B) $\Delta R/R_0$ is plotted as a function of ε by performing cyclic loading up to 100% strain with 50 cycles. (C) Measured resistances of an ionic sensor are plotted as a function of time at 7 days, respectively. (D) $\Delta R/R_0$ is plotted as a function of time with increasing strains of 20%, 40%, 60%, 80%, and 100%. (E) $\Delta R/R_0$ is plotted as a function of time with increasing bending angles of 0°, 30°, 60°, and 90°. (F) $\Delta R/R_0$ is plotted as a function of time when the sensor is kept with a specific strain. (G) $\Delta R/R_0$ is plotted as a function of time maintained around room temperature (i.e., 27°C in our test). (H) $\Delta R/R_0$ is plotted as a function of temperature from 17°C to 60°C. (I) The strain limit test of the ionic sensor. Color images are available online.

finger frequently slid the roller while middle finger generally keeps motionless, leading to different motion states of two fingers, which can also be differentiated by our iSkin system (Fig. 5B). The ability of a spatially resolved strain-sensitive wearable iSkin system would be used to provide proprioception feedback for potential applications in soft robotics, prosthetics, and virtual/augmented reality interaction.

As a proof of concept, we can also use the iSkin system to communicate with the sign language. Bending/straightening of each finger is captured and wirelessly transmitted from the iSkin system to a virtual device (i.e., a mobile phone). Then, the remote virtual device can translate the information into words. Figure 6 and Supplementary Movie S3 provide an example that one gesticulates “HELLO SJTUER” and the mobile phone understands the information. Therefore, with

our iSkin system, a deaf-mute person may successfully communicate with the persons who do not understand the sign language. Our integrated iSkin system thus provides the potential to be a smart “glove” and has advantages over conventional glove systems based on the optical fibers and metal-strain gauges in terms of high stretchability, transparency, fabrication cost, and simplicity.¹³

Conclusions

In this article, we present a fully integrated highly stretchable transparent wearable untethered iSkin system for hand-motion monitoring, which is composed of multiple hydrogel-elastomer hybrid ionic sensors and a wireless electronic control module. We provide an efficient and

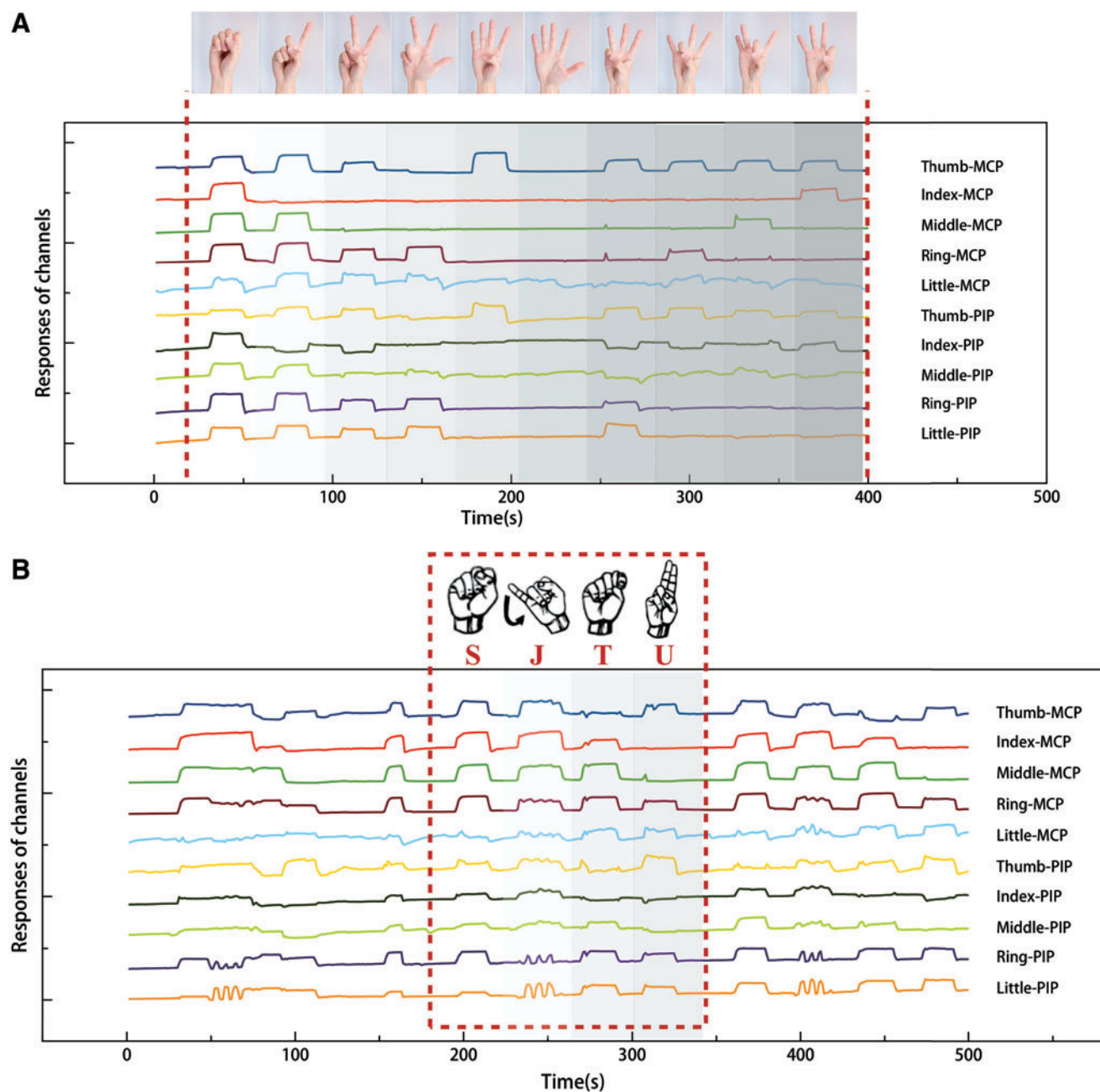


FIG. 4. Demonstrations of hand gesture recognition and sign language identifying with the integrated iSkin system. **(A)** The real-time responses recorded from the corresponding 10 sensors of our iSkin system when the subject gesticulates the digits 0–9 of the American Sign Language. **(B)** The real-time responses recorded from the corresponding 10 sensors of our iSkin system when the subject gesticulates the letters S, J, T, and U in international sign language. Color images are available online.

practical method to fabricate these robust hydrogel–elastomer hybrid ionic sensors. The adopted materials are all based on commonly available commercial products. The hydrogel–elastomer hybrid structure enables robust interface and durable sensitivity of the iSkin system. We characterize the performance of the ionic sensor with high stretchability, good sensitivity, small hysteresis, high repeatability, and durable stability. After being tested for 7 days, no significant change is observed in the sensing performance. We also demonstrate the capability of the integrated iSkin system to

monitor the joint bending and hand gesture, and communicate with the sign language. This work paves the way for practical design and integrates hydrogel–elastomer hybrid ionic sensors for highly stretchable wearable sensing skins in various applications, such as prosthetics, robotics, and human–robot interfaces. In future work, we will explore fabricating a fully transparent multifunctional iSkin system that can monitor the hand movement and detect the pressure and temperature with a better performance of precision, flexibility, adaptability, and comfortability.

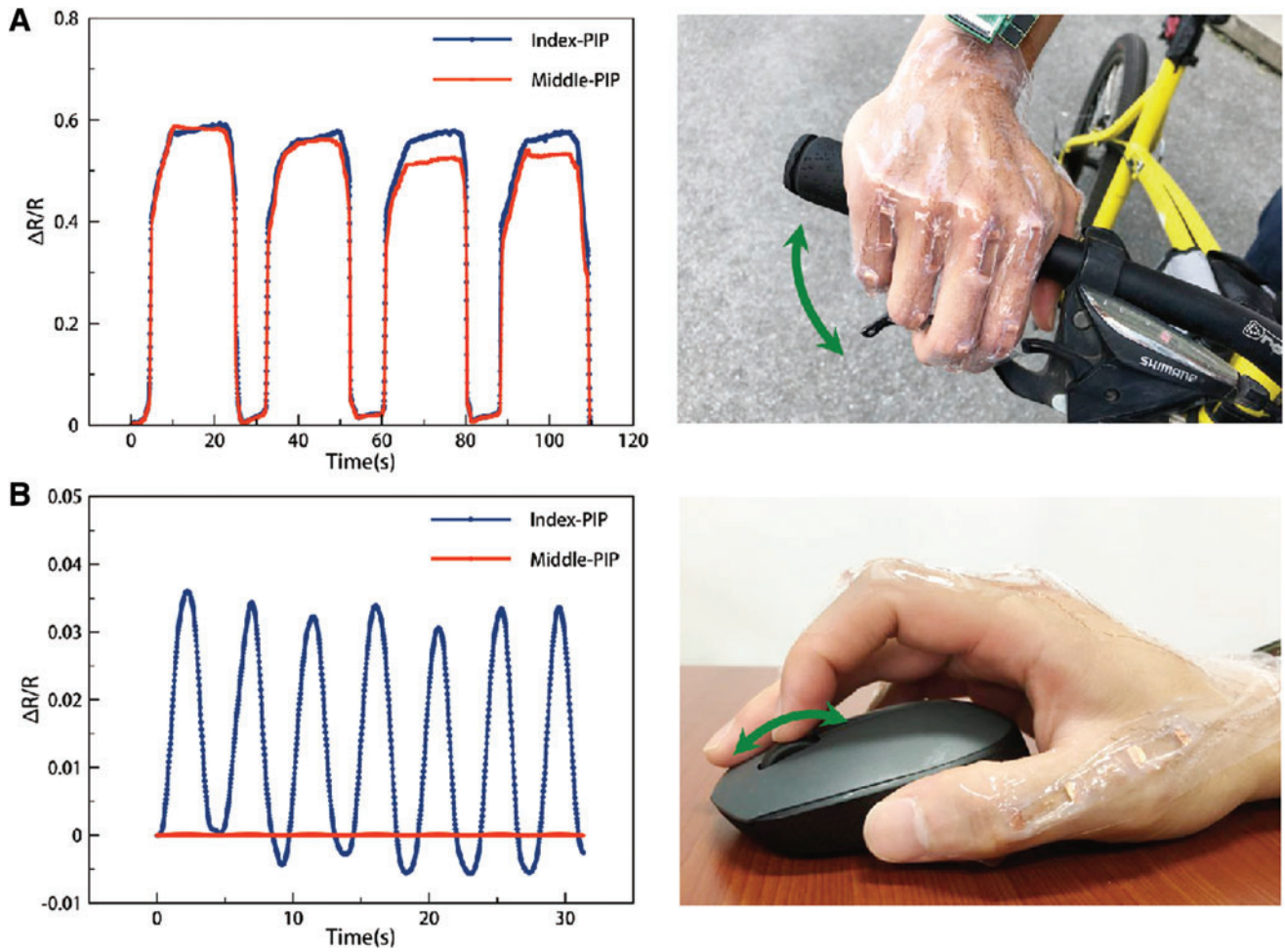


FIG. 5. (A) Application of the integrated iSkin system when braking a bike. (B) Application of the integrated iSkin system when sliding a mouse. Color images are available online.

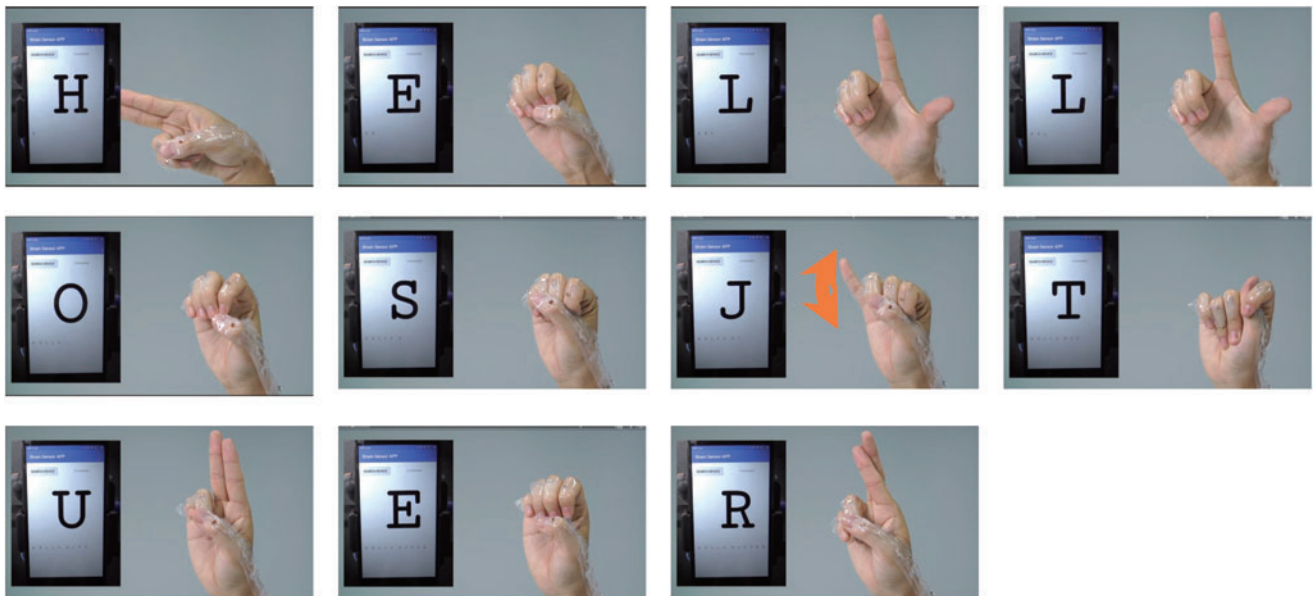


FIG. 6. A subject wearing the integrated wireless iSkin system gesticulates the sign language (i.e., “HELLO SJTUER”), which is understood by the remote virtual device (i.e., a mobile phone). Color images are available online.

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Author Disclosure Statement

No competing financial interests exist.

Supplementary Material

Supplementary Movie S1
Supplementary Movie S2
Supplementary Movie S3

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