Waterproof Mechanically Robust Multifunctional Conformal Sensors for Underwater Interactive Human–Machine Interfaces

Shengshun Duan, Binghao Wang, Yucheng Lin, Yinghui Li, Di Zhu, Jun Wu,* Jun Xia, Wei Lei, and Baoping Wang

Wearable sensors with water resistance and mechanical durability are of great value in dealing with long-term movement and remote control in harsh environments. However, achieving high sensitivity with long-term stability and real-time remote control in a watery environment is still a challenge. Herein, the waterproof wearable sensors with good mechanical robustness composed of laser-induced graphene and in situ-coated protective silicone layers are reported. By being integrated with high-capacitance ion-gel dielectrics, the conformal sensors can detect multiple stimuli, including strain, temperature, and pressure. The long-term water resistance of strain sensors is evaluated by continuously monitoring the resistance in underwater, sweat, and saline environment for up to 5.5 h. Underwater wireless remote control of a robotic hand is further demonstrated by mounting five sensor arrays. Moreover, different finger gestures are well recognized, making these sensor devices promising candidates for versatile waterproof wearable electronics and robotics technology.

1. Introduction

Given that wearable electronics need continuous operation for long time in various environments, excellent robustness, environment-stimulus tolerance, and performance stability are highly required for these wearable electronics to achieve interactive human–machine interfaces (iHMI).[11–13] Moisture is a huge issue that can damage electronic devices even in tiny amount. For wearable electronic devices, it is hard to avoid being exposed to watery environment in their daily activities, such as sweating, diving operations, underwater sports, and falling into the river accidentally. Thus, waterproof wearable sensors are supposed to allow for remote operation, underwater motion monitoring, and alarm sending capabilities.[14,15] In addition, the mechanical flexibility and long-term durability of waterproof conformal sensors can provide accurate monitoring and improve their lifetime during thousands of cycles of elastic deformation caused by the body movements.[16–19]

To achieve real-time health monitoring in watery environment, waterproof and long-lasting flexible hybrid electronic systems that assemble conformal physical/chemical sensors with miniaturized silicon-based chips can provide consumer-grade performance and reliability.[17,20–24] In general, commercial sensors are generally encapsulated by rigid shells in case of water intruding into the internal electronic components, but the rigid and bulky encapsulation layer not only weakens the sensitivity of sensors owing to the indirect contact between sensing components and measured objects, but also increases the difficulty in carrying out measurements on objects with irregular surfaces.[14] In recent years, various waterproof designs and encapsulation strategies have been explored for flexible and wearable sensors. For instance, the non-swellable graphene-assisted hydrogel by introducing a hydrophobic and hydrophilic network exhibits excellent nonswelling and wet-adhesion performance in organic aqueous solution and has been successfully applied as wearable sensors in aquatic environments.[25] On the other hand, encapsulation of wearable sensors with a hydrophobic soft layer is a facile but effective method for excellent water resistance.[14,15,25–30] However, achieving good sensitivity after encapsulation and long-term underwater durability is still challenging. Also, the identification and remote control in aquatic environment require waterproof sensor devices to unaffectedly work well underwater, which has been seldom reported.

Here, we report the waterproof conformal sensor with mechanical robustness and long-term operation stability.
(≈5.5 h) in underwater, sweat, and saline environment. The laser-induced graphene (LIG) from polyimide (PI) film is used as the sensing layer, which is encapsulated with silicone through in situ casting and curing. The process enables strong adhesion between LIG and silicone that can be detached from PI substrate with ease. Then, the mechanical robustness and electrical tolerance to water are demonstrated by cyclic mechanical/electrical tests under various harsh conditions (i.e., sandpaper rubbing, tape sticking, and underwater). In addition, owing to the unique surface morphology and high conductivity of the LIG, the e-skin offers high sensitivity to strain (a gauge factor (GF) of 132.5 at strain from 16% to 36%) and excellent stretchability (over 1000 cycles). Moreover, when mounted on human skin, the e-skins exhibit multifunctional sensing abilities involving pulse wave monitoring, muscle movements, and external temperature/pressure stimuli. We further demonstrate the underwater hand gesture recognition in a real-time wireless remote-control mode by mounting five e-skins on hands.

2. Results and Discussion

Figure 1a shows the fabrication process of multifunctional e-skins for the remote-control system. First, the PI substrate was selectively etched by laser engraving to form conducting graphene layer with predesigned patterns (Figure S1, Supporting Information). The laser reduction processing not only removes most of the oxygen-containing groups, but also effectively guarantees the smoothness and completeness of graphene patterns. The weak adhesion of LIG to the bottom PI substrates ensures easy transferring. Here, the silicone substrate is chosen due to its durability, biocompatibility, and stretchability. The silicone solution (1:1 weight ratio between the curing agent and the polymer) was poured onto LIG and then solidified at room temperature for 4 h with the thickness of 0.3 mm. Owing to the porous feature of LIG, silicone solution would partially penetrate the LIG layer and firmly bond it after solidification. Then, the PI layer was peeled off, finished by facile encapsulation with an additional silicone layer (0.3 mm thick). We further mounted...
the device on human skin as e-skins, which exhibits multifunctional sensory characterizations. The e-skin can sense strain (16–36%) with a high GF of 132.5. It is also temperature-sensitive with a work temperature ranging from 25 to 43 °C and exhibits good sensitivity (0.3% °C⁻¹) and linearity.

When pressure is exerted onto the e-skin, its capacitance increases dramatically (about five times higher under 1.9 kPa). Most importantly, e-skin devices are water-resistant with minimal conductivity change after being immersed into water for 5.5 h. To achieve underwater remote control of the robotic hand, e-skins are mounted on joints of each finger and connected with home-designed electronic systems that have the capability of analog signal acquisition, conditioning, processing, encoding, wirelessly transmitting, receiving, decoding, and controlling (Figure 1b,c, and Figure S2, Supporting Information). Results show that the robotic hand can be well remotely controlled by human fingers, even in underwater condition. In comparison with previous reports about LIG/silicone-based sensors and other waterproof sensors (Table S1 and S2, Supporting Information),[40–45] our conformal sensors feature long-term water resistance, high GF, mechanical robustness, multifunctionality, and underwater iHMI properties.

2.1. Mechanical and Electrical Characterization

Figure S3, Supporting Information, shows the peeled LIG with an interdigital pattern on silicone substrates, and the peeling process has a minimal effect on the LIG conductivity. The sheet resistance increases from 200 to 420 Ω cm⁻² (Figure S4, Supporting Information). Thus, the LIG on silicone is used as both the electrodes and the sensing layer. The scanning electron microscope (SEM) images in Figure S5, Supporting Information, show that unencapsulated LIG/silicone has rough surface with porous and hierarchical microstructures, which endow them with excellent conductivity and strain sensitivity.[37,46,47] In contrast, the MXene/silicone sensors fabricated through a drop-casting method exhibit the weak bonding ability, and cracks are easily formed during manipulation (Figure S6–S9, Supporting Information).

The electrical durability of devices based on LIG/silicone and MXene/silicone is first evaluated under strains. The devices are gradually stretched by a stretching tester with the real-time resistance being continuously monitored. When the resistances reach infinite, the strains on the devices are released, and then, the resistances at the initial stage are recorded. Figure 2a shows the initial resistance of both devices after being stretched/released for ten times. The initial resistance of the MXene/silicone device increases from 7.0 to 90.0 kΩ in the first six times of being stretched/released (∼32% strain); then, it dramatically increases to 2476.3 kΩ after ten times. In contrast, the initial resistance of LIG/silicone device keeps relatively low values and is very stable (from 280.7 to 535.4 Ω) during the ten-time tests (∼150% strain). The optical images (Figure S10, Supporting Information) show that obvious cracks form on the MXene/silicone device after only ten stretching–releasing cycles under 30% strain, whereas the LIG on the silicone substrate is still uniform and continuous with negligible cracks. Accordingly, during the ten-time test cycles, the initial resistance of LIG/silicone device is much more stable than that of the MXene/silicone device (Figure S11, Supporting Information). Moreover, the

Figure 2. Mechanical and electrical properties. a) The cyclic initial resistance after the two devices was stretched by a stretching tester until the resistance reaches infinite. b) Strain sensing property at a strain from 0% to 75%. c) GF measurement at a strain from 16% to 36%. d) The photograph shows that a 2 cm-long sensing unit can be stretched to 3 cm. Scale bar: 1 cm. e) Durability test for up to 1000 continuous stretching–releasing cycles. f) Generated resistance signals of the resistive e-skin, with various tensile strains at a fixed strain rate.
unencapsulated LIG/silicone device exhibits much better mechanical and electrical durability under mechanical loads, such as sandpaper rubbing and tape sticking (Figure S12–S14, Supporting Information). The resistance of MXene/silicone device is larger than 100 Gohm at ≈30% strain, whereas for the LIG/silicone device, the resistance is at the level of 10 MΩ at ≈95% strain (Figure S15, Supporting Information).

Before quantifying the strain-electrical properties of the LIG/silicone devices, we first encapsulate LIG/silicone devices with a silicon rubber layer. The stress–strain–resistance measurements are conducted at a constant strain rate of 15 mm min⁻¹, and their relationship is plotted in Figure 2b. The relative change of resistance (ΔR/R₀) increases slowly at first, and then increases significantly when the strain is larger than 40%. Also, a linear region is observed for the strain between 16% and 36% (Figure 2c), and the GF, defined as GF = ΔR/ΔεR₀,[48] is as high as 132.5. Figure 2d,e shows the cyclic test setup and ΔR/R₀ evolution during 16 000 stretching/releasing cycles (0%–50% strain). The value of ΔR/R₀ increases to ≈115 times when the LIG/silicone device is stretched to 50% strain, but it can fully recover after release. No electrical degradation is observed after 10 000 stretching/releasing cycles, demonstrating excellent durabilities. Note that most parts of human skin during motion would not be stretched more than 30%; thus, our devices are qualified for motion monitoring when mounted on human skin.[49] In addition, Figure 2f shows cyclic variations of resistance for different peak strains of 2%, 5%, 10%, 20%, and 30%, respectively. The test is taken at a constant strain rate of 24 mm min⁻¹. The relative resistance change (ΔR/R₀) profiles are stable and proportional to the tensile strain in terms of the signal amplitude.

2.2. Waterproof Multifunctional Conformal Sensors

To mimic mimic human skins with multifunctions, we mount the robust and conductive LIG-based e-skins on human skin, which are used as the temperature, strain, and pressure sensors (see Table 1, Figure S16–S18, Supporting Information, and Experimental Section for details). Figure 3a shows the resistance response of LIG-based e-skin to temperature variation. The resistance (at 2 V) decreases from 0.62 to 0.37 kΩ, as the temperature increases from 25 to 42 °C. The resistance exhibits a linear temperature dependence, and the resistance sensitivity (S_T), defined as S_T = δ(ΔR/R₀)/δT × 100%,[50] is estimated to be 0.3%/°C via a linear least-squares fitting method, which is ten times higher than that (0.04%/°C) of commercial Pt-based thermistor.[51] As shown in Figure 3b, warm water (50 °C) is poured into an empty beaker, and the temperature of the attached sensor increases from room temperature (≈23 °C) to 50 °C owing to the heat conduction. The response time, defined as the time when resistance rises from 10% to 90% of the final resistance,[52] is about 8 s due to poor heat conduction of the silicone encapsulation layer. Then, a few pieces of ice are added into the beaker to accelerate the cooling process of hot water. Accordingly, the relative change of resistance recovers to the original resistance. Moreover, we test the temperature-discrimination capacity of our sensors by periodically putting it on the 60 °C hot plate and the 0 °C ice bag. The relative change of resistance of our sensor exhibits repeatable temperature response, demonstrating stable and reliable sensing capacity (Figure 3c). The encapsulated resistive e-skin is also attached to the ring finger to characterize the dependence of ΔR/R₀ on bending speed and angle, where large strains (up to 55%) occur near one’s finger joint. The device responses rapidly with high sensitivity during continuous bending (about 120°) and exhibits excellent operational stability (Figure 3d). Next, ΔR/R₀ under different degrees of finger movements, which correspond to different extents of strain, is evaluated. The resistive e-skin exhibits excellent electrical stability with high resolution at different bending angles due to the excellent sensitivity. As shown in Figure 3e, excellent synchronization of the relative change of resistance and the corresponding stretching force is observed, which indicates a response time of ≈130 ms and a recovery time of ≈240 ms, thereby guaranteeing an accurate expression of finger movements. More information about response and recovery is shown in Figure S19, Supporting Information. Also, the devices are also explored for monitoring throat movement and minimal facial expression, corresponding to small strains (Figure S20, Supporting Information). Results show that characteristic signals are generated in response to repeatable swallowing, the opening of the mouth, and the smile, respectively.

Regarding the pressure sensor, the interdigital LIGs on silicone substrates are used as electrodes, and the ion gel with ultrahigh capacitance (on the orders of tens to hundreds of nF) is used as a sensing part, which exhibits perfect immunity to the

Table 1. Structure, function, and representative device figure of merit reported in this work.

| Device       | Device structure (up-down) | Dimension | Application               | Figure of merit                        |
|--------------|---------------------------|-----------|---------------------------|----------------------------------------|
| Resistor     | Silicone–LIG–silicone     | W = 0.5 cm| Strain sensor             | GF ≈ 132.5                             |
|              |                           | L = 1 cm  |                           | Range = 0%–80%                         |
|              |                           | H = 0.65 mm| Underwater hand gesture recognition|
| Resistor     | LIG–silicone              | W = 0.3 cm| Temperature sensor         | Sensitivity = 0.3%/°C                  |
|              |                           | L = 1 cm  |                           | Range = 25–43 °C                       |
|              |                           | H = 0.35 mm|                                           |
| Supercapacitor| Silicone–hydrogel–LIG–silicone| W = 1.1 cm| Pressure sensor            | High capacitance ≈212 nF              |
|              |                           | L = 2.4 cm|                           | 500% increase in C under 1.9 kPa       |
|              |                           | H = 1 cm  |                           |                                           |
| Supercapacitor| Silicone–LIG–hydrogel–LIG–silicone| W = 0.35 cm| Pulse monitoring          | Clear demonstration of pulse waves     |
|              |                           | L = 1 cm  |                           |                                           |
|              |                           | H = 0.25 mm|                                           |
parasitic noises (on the orders of several pF). Specifically, the capacitance between interdigital LIG electrodes increases from 3 pF to 212 nF after mounting the ion gel (Figure S21, Supporting Information). Thus, the relative capacitance change ($\Delta R/R_0$) of supercapacitive e-skin reaches 5 when a small pressure ($\approx$1.9 kPa) is exerted by 50 g weight (Figure 3f). Next, the supercapacitive e-skin with a plane parallel structure is attached to the wrist of a volunteer for pulse monitoring. A clear pulse waveform is observed (Figure S22, Supporting Information). The long-term waterproofness and robustness of the e-skin are investigated in underwater conditions. Figure 3g compares the resistance change of the e-skins with/without encapsulation when tested in tap water for up to 1 h. The value of $\Delta R/R_0$ of e-skin without encapsulation keeps constant in the first 90 s; then, it gradually increases and reaches 10% after immersing in water for 15 min due to the water penetration. After that, the value of $\Delta R/R_0$ almost keeps unchanged when tested underwater for up to 1 h. The waterproof property is further improved after encapsulated with silicone. No obvious change in resistance is observed during the 1 h test in tap water (Figure 3d, and Video S1 and S2, Supporting Information). Furthermore, given that wearable sensors are usually confronted with saline environments when attached on skin, we test the electrical robustness and stability for about 5.5 h in three different solutions involving tap water, saline environment (0.9% wt NaCl in deionized water), and sweat (pH $\approx$ 5.1). Owing to excellent encapsulation, all the sensors exhibit remarkable electrical stability in all the three solutions, as shown in Figure S23, Supporting Information. Moreover, the stable strain-sensing capability of e-skins for the same bending angle is observed when the device is tested in ambient or underwater conditions (Figure 3e, and Figure S24 and Video S3, Supporting Information).
Supporting Information), clearly demonstrating the excellent waterproof performance.

### 2.3. Underwater Remote Control

To demonstrate the capability of resistive e-skins for robotic remote control, the analog signals of current are first converted into analog signals of voltage, followed by analog-to-digital signals conversion. Then, the signals can be wirelessly transmitted to a terminal display and/or applied for remote control (Figure 4a). As discussed earlier, the resistance of the resistive e-skin increases from hundreds of ohms to hundreds of kilo-ohms during finger bending, which can lead to computational errors during signal processing. Thus, a resistance/voltage conversion circuit is adopted to convert resistive signals into voltage signals and normalize the voltage signals between 0 and 5 V (Figure 4b, and Figure S25, Supporting Information). Figure 4c shows the real-time voltage signals from a resistive e-skin under six different motion statuses. The output reliability of the resistive e-skin is confirmed by continuous repetition of each finger motion (three times for each status), resulting in three stable and uniform voltage peaks at each status. Next, the direct correlation between the relative voltage changes of the e-skins with specific gestures is monitored by mounting five strain sensors on the finger joints of a hand (Figure 4d). For example, the e-skins can accurately recognize letter C, number 8, and phrase “I love you,” without any interchannel interference.

Finally, we use the waterproof and long-term stable resistive e-skins for underwater remote control of a robotic hand. The flow diagram of the human–machine remote control system is shown in Figure 5a, which includes a hand gesture recognition process and a robot hand control process. Voltage signals through each path are the terminal voltage across resistive e-skins. The data processing and transmission path for each channel are implanted, concerning the corresponding transduced signals with an analog circuit. The major function of this analog circuit is to remove interference signals and environmental noises using low-pass filters, which ensures that the final analog output of each resistive e-skin can precisely express the movement information and be suitable for subsequent processing by an analog-to-digital converter (ADC). Microcontroller 1 would receive digital signals from ADC and encode different hand gestures using a multithreshold code algorithm (Figure S26, Supporting Information). Then, codes are wirelessly transmitted to another microcontroller 2 that controls the robotic palm. Microcontroller 2 would decode the codes into different commands, which are then sent to the executive module, thereby controlling the robotic hand movement with the same gestures as the human hand (Figure 1b and 5a).

To demonstrate real-time gesture recognition, eight hand gestures (A, I, L, Y, 3, 8, and “I love you”) are selected, which are mainly fundamental elements of communication—numbers, letters, and phrases (Figure 5b). The corresponding voltage profiles of these hand gestures are shown in Figure 5c. The different colors of lines represent different fingers (thumb, index, middle, ring, and little), and the height of lines represents the voltage across the e-skin adhered to fingers under different bending statuses. The latter four gestures are tested in a watery environment, and output voltages reveal the corresponding finger gestures. Demonstrations of robot remote control in ambient and underwater conditions are shown in Figure 5e, and Video S4.
Supporting Information. Various gestures are wirelessly synchronized between a human hand and a robotic hand without obvious time delay.

3. Conclusion

In summary, we demonstrate waterproof e-skins for underwater hand gesture monitoring and remote control. The LIG technique, combining with a one-step infiltration process, is developed for robust multifunctional sensors. The as-fabricated e-skin exhibits an excellent conductivity of $420 \, \Omega \cdot \text{cm}$ and a high $GF$ of 132.5 at a strain from 16% to 36%. The e-skin shows good electrical stability during 1000 stretching/releasing test cycles at a strain of 50%. In addition, pulse monitoring, human posture recognition, and external stimulus detection toward temperature and pressure are successfully demonstrated. Most importantly, the outstanding mechanical robustness and electrical tolerance to an aquatic environment of the e-skins are validated under different test conditions, including underwater finger bending test and real-time remote control. We believe that our e-skins promote advances in waterproof wearable electronics for a wide range of applications.

4. Experimental Section

Fabrication of LIG: Laser induction was conducted on PI substrates using a laser cutter (Universal Laser System, VLS3.50) equipped with 10.6 $\mu$m CO$_2$ laser. The image resolution varied from 100 to 1000 pixel per inch (PPI), and the scanning speed was fixed at 10 cm s$^{-1}$. Laser average power was changed by tuning the duty. In a typical experiment, the LIG on 100 $\mu$m-thick PI substrate was formed at a 1000 PPI with a 64% duty.
cycle. The various graphene patterns on PI substrate, for example, interdigital and square patterns, were designed through AutoCAD 2020. The PI would be etched according to the predesigned pattern to form the same graphene pattern.

**Infiltration of LIG for E-Skin:** Silicone (Hong Ye Jie Technology Co., Ltd.) was mixed in a weight ratio of 1:1 of components A and B. The LIG on PI substrates was placed in a mold (10 cm in diameter) and wetted with deionized water. The silicone was poured on top of the LIG/PI and de-bubbled in a vacuum oven, followed by solidification in ambient.

After that, the PI layer was peeled away, and the LIG/silicone composite was formed.

**Fabrication of the Ionic Hydrogel:** Acrylamide (3.15 g) (98%, Aldrich) and NaCl (3 g) (99.5%, Shanghai Lingfeng Chemical Reagent Co., Ltd.) were dissolved in 20 mL of deionized water. Then, 0.00189 g of N,N'-methylene-bisacrylamide (99%, Shanghai Aladdin Biochemical Technology Co., Ltd.) and 0.005355 g of ammonium persulfate (98%, Sigma) were added to the solution. After degassing in a vacuum chamber, 10 μL of N,N',N'-(tetramethylene)enediamine (99%, Shanghai Taitan Scientific Co., Ltd.) was added. The solution was poured into a polytetrafluoroethylene (PTFE) mold and covered with a 3 mm-thick glass plate. After being cured using a UV light (254 nm wavelength) for 20 min under a power of 8 W, ion gels were formed.

**Fabrication of the Resistive E-Skin for Human Posture and Temperature Detection:** The LIG/silicone composite with a square LIG pattern was first fabricated. Cu wires as electrodes were fixed at the two ends of the LIG pattern using silver paste. Then, some amount of liquid silicone was dropped on the top of the LIG/silicone composite to seal the LIG/silicone composite to make waterproof resistive e-skins.

**Fabrication of the Supercapacitive E-Skin for Pulse Detection:** The LIG/silicone composite with a square LIG pattern was fabricated as electrodes. The ion gel was sandwiched by two LIG/silicone layers. Two Cu wires were fixed at the LIG pattern of two LIG/silicone layers using the silver paste, separately. Two pieces of polyurethane films as protective layers were attached to the top and bottom sides of the LIG/silicone composite.

**Fabrication of the Supercapacitive E-Skin for External Stimulus Detection:** The LIG/silicone composite with an interdigital LIG pattern was fabricated as electrodes. The ion gel was placed on the top side of the LIG/silicone composite. Cu wires as electrodes were fixed at the interdigital LIG pattern. One piece of polyurethane film as the protective and adhesive layer was then attached to the top side of the ion gel.

**Silicone Encapsulation Layer:** After preparing LIG/silicone sensor and fixing Cu electrodes, we drop-cast the silicone mixture (Hong Ye Jie Technology Co., Ltd.), mixed in a weight ratio of 1:1 of components A and B, onto the upper surface of LIG/silicone sensor, followed by solidification in ambient about 1 h, to protect LIG from external mechanical damage and water intrusion.

**Underwater Stability Test:** The capsulated devices were immersed into the solutions for up to 5.5 h with their real-time resistances were recorded. The saline solution was prepared by adding 27 mg of NaCl into 30 g of DI water. The sweat was directly collected from the human body (Male, Age 24) after 60 min running.

**Device Characterization:** The structure and morphology of the e-skin and MXene-based e-skin were observed by a scanning electron microscope (Quanta 200 FEI). The electrical characterization was measured with a source meter (Keithley 2400) under the signal of 2 V. The capacitance of the supercapacitive e-skin was measured by a precision LCR meter (Tonghui, TH2829C). A force gauge (Pubtester, TST-01H) was used to apply the external force.

The experiments involving human subject have been performed with the full, informed consent of the author.

**Supporting Information**

Supporting Information is available from the Wiley Online Library or from the author.

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**Conflict of Interest**

The authors declare no conflict of interest.

**Data Availability Statement**

Research data are not shared.

**Keywords**

electronic skin, human–robot remote control, laser-induced graphene, strain sensors, water resistance

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