Hydrogel-elastomer-based stretchable strain sensor fabricated by a simple projection lithography method

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ABSTRACT

Stretchable strain sensor detects a wide range of strain variation and is therefore a key component in various applications. Unlike traditional ones made of elastomers doped with conductive components or fabricated with liquid conductors, ioni-
cally conductive hydrogel-based strain sensors remain conductive under large deformations and are biocompatible. However, dehydration is a challenging issue for the latter. Researchers have developed hydrogel-elastomer-based strain sensors where an elastomer matrix encapsulates a hydrogel circuit to prevent its dehydration. However, the reported multi-step approaches are generally time-consuming. Our group recently reported a multimaterial 3D printing approach that enables fast fabrication of such sensors, yet requires a self-built digital-light-processing-based multimaterial 3D printer. Here, we report a simple projection lithography method to fabricate hydrogel-elastomer-based stretchable strain sensors within 5 minutes. This method only requires a UV projector/lamp with photomasks; the chemicals are commercially available; the protocols for preparing the polymer precursors are friendly to users without chemistry background. Moreover, the man-
ufacturing flexibility allows users to readily pattern the sensor circuit and attach the sensor to a 3D printed soft pneumatic actuator to enable strain sensing on the latter. The proposed approach paves a simple and versatile way to fabricate hydrogel-elastomer-based stretchable strain sensors and flexible electronic devices.

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1. Introduction

Stretchable strain sensor is a type of sensor that is highly stretchable and capable of detecting a wide range of strain variations. Therefore, it is a key component in various emerging fields including soft robots [1–4], wearable devices [5–7], smart prosthetics [8,9], and others [10–12]. In the past years, many efforts have been made to develop stretchable strain sensors, and the approaches mainly rely on two types: elastomer-based strain sensors and liquid-based strain sensors. The elastomer-based strain sensors are made of elastomers doped with conductive components such as silver nanoparticles/nanowires, carbon nanotubes, and graphene. However, the conductivity of such strain sensors degrades remarkably as a result of the separation of doped wires or particles [13,14]. Therefore, they are not ideal for sensing large deformations. Liquid-based strain sensors are fabricated with liquid conductors such as liquid metal and grease. While such type of strain sensors can sustain large deformations, the accumulated localized plastic deformation resulting from cyclic loading deteriorates the conductivity and leads to signal drifts [15,16]. More importantly, many of those liquid conductors are not biocompatible, and the efforts to prevent them from leakage result in additional design complexity and manufacturing costs [14].

Unlike electronic conductors, ionically conductive hydrogels are highly stretchable, conductive under large deformation and biocompatible. Therefore, they have found various applications in strain sensors [17–22], touch panels [23], and hybrid circuits [24]. However, despite the recent great progress in the applications of ionically conductive hydrogels, dehydration is a challenging issue that significantly affects the performance and functionality of ionically conductive hydrogel-based flexible electronic devices [25]. To address this issue, researchers have developed a hydrogel-elastomer hybrid flexible electronic device where hydrogel forms an electronic circuit to transfer signals and an elastomer matrix encapsulates the hydrogel circuit to protect it from dehydration. However, in general, hydrogels and elastomers are dissimilar materials, and the adhesion between the two materials is inherently low. To address this issue, Yuk et al. reported a simple yet versatile method to create synthetic hydrogel–elastomer hybrids by using benzophenone to modify the cured chemical bonding with hydrogels [25]. This approach successfully fabricated hydrogel–elastomer hybrids with interfacial bonding tougher than epidermis–dermis. However, this approach is restricted to specific sequences to form the
hybrids: only grafting a hydrogel onto an elastomer. To address this challenge, Liu et al. proposed an approach that is realized by adding silane coupling agents into the precursors of both hydrogels and elastomers, and is independent of the sequence of bonding process [26]. The above methods enable the fabrication of the hybrid structures that are used to protect, reinforce, or add new functionalities to hydrogel structures including elastomeric biomedical devices with hydrogel-based hydrophilic and lubricant skin [27–29], hydrogel-based flexible electronics with elastomeric anti-dehydration coating [25,26], hydrogel composites reinforced by elastomeric fibers [30,31], and others [32–34]. Despite those rapid progresses, the above approaches usually include multiple steps, and are time-consuming. Recently, our group reported a multimaterial 3D printing approach that enables fast fabrication of hydrogel–elastomer hybrids in complex 3D forms [4]. However, such a 3D printing approach requires a self-built digital light processing-based multimaterial 3D printer that is not commercially available. Therefore, a more general, simple, and low-cost fabrication method is desired to manufacture hydrogel–elastomer-based stretchable strain sensors.

Here, we report a simple projection lithography method to fabricate hydrogel–elastomer-based stretchable strain sensors. This method only requires an ultraviolet (UV) projector or even a UV lamp with photomasks, and therefore is accessible and affordable for most of the research groups. The entire fabrication process is simple, and only takes about 5 minutes. The chemicals introduced in this manuscript are all commercially available, and the preparation protocols of the UV curable polymer precursors are simple and friendly to the users who do not have chemistry background. By taking advantage of the manufacturing flexibility of this projection lithography, we are able to readily pattern the circuit of the stretchable strain sensor. After attaching the stretchable strain sensor fabricated by this method to the soft pneumatic actuator (SPA) made by 3D printing, we are able to create an SPA with the integrated strain sensing capability. The approach proposed in this work paves a simple and versatile way to fabricate hydrogel-elastomer-based stretchable strain sensors and flexible electronic devices.

**Figure 1.** Design and manual fabrication procedures of stretchable strain sensor. (a) Illustration of the sensor. (b) Details of the sandwiched structure of the strain. (c) Snapshot of the fabricated sensor. (d)–(f) Manual fabrication of the stretchable strain sensor.
2. Fabrication procedures

Figure 1(a,b) illustrates the sandwiched structure-based design of the hydrogel-elastomer strain sensor. In this design, the ionic hydrogel circuit is embedded into the elastomer matrix that works as the substrate to provide elastic support and protect the ionic hydrogel circuit from dehydration. The low resistance silver conductive tapes are embedded into the sensor and directly contacted with the hydrogel circuit. Figure 1(c) presents a snapshot of a fabricated hydrogel-elastomer strain sensor that can be made by a manual but simple manufacturing process described in Figure 1(d–f). As shown in Figure 1(d), the fabrication process starts by photo-curing a layer of elastomer substrate. The elastomer precursor is first injected into the cavity that is sandwiched by two glass slides. The thickness of the cavity is controlled by the spacers between the two glass slides. Then, a rectangular-shaped UV pattern is projected to the elastomer precursor, which transforms the liquid precursor into a solid rectangular pattern. As shown in Figure 1(e), after using ethanol to remove the uncured elastomer precursor, the ionic hydrogel precursor is injected into the cavity between the glass slide and the UV cured rectangular substrate. The ionic hydrogel circuit pattern is then formed by projecting the corresponding UV pattern to the hydrogel precursor. As shown in Figure 1(f), the middle

![Figure 2](image.png)

Figure 2. Chemicals used to form stretchable hydrogel and elastomer. (a) Chemicals used to prepare acrylamide-PEGDA hydrogel. (b) Illustration of the ionically conductive hydrogel network. (c) The corresponding chemical structure at crosslinking point of the acrylamide-PEGDA hydrogel. (d) Chemicals used to prepare BA-PEGDA elastomer. (e) Illustration of the elastomer network. (c) The corresponding chemical structure at crosslinking of the BA-PEGDA elastomer.
and upper layers of elastomer encapsulating the ionic hydrogel circuit are fabricated by following the similar procedures. It should be noted that the silver tapers are embedded after the hydrogel circuit is fabricated; the modulation of UV irradiation can be realized by using a commercial UV projector such as Wintech Digital PRO4500 (Wintech Digital System Technology Corp., San Marcos, CA, USA), or using a traditional UV lamp plus photomask.

3. Materials and characterization

3.1. Material preparation

Figure 2(a) presents the chemicals that are used to prepare the ionic conductive, highly stretchable, and UV-curable hydrogel precursor solution. We prepare the hydrogel precursor solution by mixing the acrylamide powders, poly(ethylene glycol) diacrylate (PEGDA, $M_w = 575$), and the self-prepared water-soluble photoinitiator 2,4,6-trimethylbenzoyl diphenylphosphine oxide (TPO), and lithium chloride into water. Figure 2(b) illustrates the ionic hydrogel network where acrylamide works as linear chain builder, PEGDA works as a crosslinker. The hydrogel network contains water as solvent where the salt–lithium chloride is dissolved, and the salt ions are able to freely travel so that the hydrogel is ionic conductive. In the acrylamide-PEGDA system, the PEGDA/acrylamide mixing ratio is 0.625 wt.%, and the water content of the AP hydrogel is 80 wt.%. The mechanical behavior is highly affected by the PEGDA/acrylamide mixing ratio, the molecular weight of PEGDA, as well as the water content [35]. It should be noted that the water-soluble TPO as photoinitiator is the key to making the acrylamide-PEGDA hydrogel precursor highly UV curable. The details about preparing the water-soluble TPO can be found in previous reports [36–38]. The water-soluble TPO is now also commercially available and can be directly purchased from Sigma Aldrich (Product Nos. 906808, 906816).

Figure 2(c) presents the chemicals that are used to photopolymerize the highly stretchable and UV curable elastomer where benzyl acrylate (BA) works as linear chain builder, PEGDA ($M_w = 700$) works as crosslinker, and TPO with 1 wt.% is added as a photoinitiator. Figure 2(d) illustrates the corresponding elastomer network. Compared with previously reported butyl acrylate [37] or 2-hydroxyethyl acrylate [38], the higher molecular weight makes benzyl acrylate less odoriferous, and therefore more user-friendly. The mixing ratio of PEGDA/BA also affects the mechanical behavior of elastomer system, and the relationship will be discussed in Section 3.2. We emphasize here that all the chemicals that we used to prepare the hydrogel and BA elastomer are commercially available, and users can purchase them from Sigma Aldrich or other local chemical providers; the protocols to prepare the hydrogel and elastomer precursors are simple and friendly to the users who do not have chemistry background.

3.2. Mechanical property characterization

As demonstrated in Figure 3(a), the photopolymerized acrylamide-PEGDA hydrogel exhibits excellent stretchability. As previously studied, the mechanical property of the
photopolymerized acrylamide-PEGDA hydrogel is highly tailor able by tuning the PEGDA/ acrylamide mixing ratio, the molecular weight of PEGDA, as well as the water content [35]. In this work, we prepare a hydrogel where the PEGDA/acrylamide mixing ratio is 0.625 wt. %, molecular weight of PEGDA is 575 wt.%, and water content is 80 wt.%. We examined the stress-strain behavior of the hydrogel on a commercial uniaxial tensile testing machine (Guangzhou Precision Control Testing Instrument Co. Ltd, XLD-100E, China) with a strain rate of 1.33 min⁻¹. As shown in Figure 3(b), the hydrogel can be stretched by ~750% with the Young’s modulus of ~4 kPa. In order to achieve ionic conductivity, we add lithium chloride (LiCl) into the acrylamide-PEGDA hydrogel system, which also affects its mechanical behavior. As shown in Figure 3(b), adding more LiCl leads to an increase in modulus as well as strength of the hydrogel, but a decrease in fracture strain. As summarized in Figure 3(c), the increase in the LiCl concentration from 0 to 5 mol/L
leads to the rise in Young’s modulus from 4 kPa to 12 kPa, but the decrease in the elongation-at-break from 750% to 560%.

Figure 3(d) shows the great stretchability of the BA-PEGDA elastomer. We further performed uniaxial tensile tests to investigate the effect of the crosslinker PEGDA on the mechanical behavior of the elastomer. Again, the test was performed on a commercial uniaxial tensile testing machine (Guangzhou Precision Control Testing Instrument Co. Ltd., XLD-100E, China) with strain rate of 1.33 min\(^{-1}\). As presented in Figure 3(e), the increase in PEGDA content results in higher Young’s modulus but lower stretchability of the elastomer. As summarized in Figure 3(f), the increase in PEGDA content from 1 wt.% to 3 wt.% leads to a slight raise in Young’s modulus from 0.7 MPa to 1.05 MPa but a significant decrease in the elongation-at-break from 900% to 375%. In this work, we prepared the BA-PEGDA elastomer with 1 wt.% of PEGDA.

Since the stretchable strain sensor is fabricated in the hydrogel-elastomer hybrid form, the interfacial bonding between the hydrogel and elastomer becomes critical. As shown in Figure 3(g), we conducted the 180°-peeling tests to investigate the hydrogel-elastomer interfacial toughness. Both hydrogel and elastomer were attached to polymeric backing layer to constrain deformation. The hydrogel and the elastomer are bonded in an area of 20 mm × 50 mm, and the thickness of hydrogel and elastomer is 1 mm. The peeling tests were conducted on a commercial uniaxial testing machine (Guangzhou Precision Control Testing Instrument Co. Ltd., XLD-100E, China). The loading rate was 1.33 min\(^{-1}\). Figure 3(h) shows the interface image during the peeling test where the hydrogel undergoes a cohesive failure that leaves the hydrogel residue on the elastomer (insets in Figure 3(i)). Figure 3(i) presents the measured interfacial toughness (150 N/m) which is in fact stronger than the previously reported results (less than 100 N/m) summarized in List 1. The bonding mechanism can be explained as follows: after one layer of hydrogel (or elastomer) is UV cured, there are still some unreacted monomers or crosslinkers left in the cured material; when we start to cure the next layer of elastomer (or hydrogel), the unreacted monomers or crosslinkers penetrate the elastomer–hydrogel interface and form the covalent bonds between hydrogel-elastomer to achieve high interface adhesion [4].

| Hybrid materials                          | Interfacial toughness | Refs. |
|-------------------------------------------|-----------------------|-------|
| Acrylamide Hydrogel-Tango/Agilus elastomer| ~20 N/m               | [4]   |
| Acrylamide Hydrogel-PDMS/Ecoflex          | 60–100 N/m            | [14,26,40] |
| Acrylamide Hydrogel-Benzyl acrylate Elastomer | 150 N/m               |       |
| (this work)                               |                       |       |

### List 1. Comparison on the interfacial toughness of hydrogel-elastomer hybrids.

**3.3. Electrical property characterization**

We characterize the electrical property of the acrylamide-PEGDA hydrogel using a precision LCR meter (TH2838H, Changzhou Tonghui Electronic Co., Ltd., China). In order to avoid dehydration during tests, as illustrated in Figure 4(a), we fabricated a hydrogel dumbbell sample that was encapsulated in elastomeric protection layers. The silver conductive tapes were attached to the two ends of the hydrogel sample. To
investigate the effect of the testing frequency on the electrical performance of the ionic conductive hydrogel, we carried out impedance-frequency tests. As shown in Figure 4(b), the impedance is a function of testing frequency: the impedance is 5–8 kΩ at low-frequency range; the impedance drops significantly by increasing the testing frequency, and it becomes stable in the range of 5–10 Ω when the testing frequency is greater than 10 kHz. This phenomenon has been previously studied \[14,41\], and is attributed by the existence of an electrical double layer (EDL) between the ionic hydrogel and the silver conductive tapes. The impedance of EDL $X_C$ equals to $(2\pi fC)^{-1}$ where $f$ is the testing frequency and $C$ is the capacitance of EDL. When the testing frequency is low, the contribution of $X_C$ to the whole impedance is significant, while the testing frequency is high, the contribution of $X_C$ is negligible. The negative-phase angle in Figure 4(b) supports the above explanation: when the testing frequency is low, the negative-phase angle maintains at 80°; while the testing frequency is greater than 1 kHz, the obvious drop of the negative-phase angle can be seen. Figure 4(b) also demonstrates the effect of salt
content on the conductivity of the hydrogel, which is summarized in Figure 4(c): the conductivity increases from 2.8 S/m to 9 S/m by adding the LiCl from 1 mol/L to 5 mol/L.

We further measure the resistance change of the ionic hydrogel under uniaxial tension. As shown in Figure 4(d), the increase in tensile strain leads to a higher relative resistance change $\Delta R/R_0$. Moreover, Figure 4(d) shows that $\Delta R/R_0$ is independent of the salt content when the tensile strain is less than 100%; when the tensile strain is greater than 100%, the hydrogel with 5 mol/L LiCl is slightly more sensitive than the hydrogels with less LiCl. We further investigate the sensing performance of the hydrogel under different strains. As shown in Figure 4(e), the $\Delta R/R_0$ value increases correspondingly with different tensile strains applied. The resistance returns to the initial value after unloading in each cycle. Furthermore, as presented in Figure 4(f), the tensile strain gradually increases from 0% to 5%, 10%, 15%, 25%, 50%, and 100%, the $\Delta R/R_0$ responses synchronously. Both Figure 4(e,f) indicate that the ionic hydrogel has great strain sensing stability and reversibility. The elastomeric encapsulating layer prevents the hydrogel from dehydration. To prove this, we carried out dehydration comparison tests to examine the effect of the elastomeric layer on the anti-dehydration. Through the comparison (Figure 4(g)), we find that despite the existence of salt that slowed the dehydration process [42], the hydrogel with no encapsulating layer lost more than 30 wt.% water within 7 days; in contrast, the protected hydrogel lost less than 5 wt. % water, indicating the effective anti-dehydration of the elastomeric encapsulation layer. Moreover, as shown in Figure 4(h), the electric performance of the hydrogel that was encapsulated in the elastomeric layer was more stable than that of the unencapsulated hydrogel.

4. Hydrogel-elastomer strain sensor

4.1. Characterization of a single hydrogel-elastomer strain sensor

As shown in the inset of Figure 5(a), by taking advantage of the manufacturing flexibility of this projection lithography, we are able to readily pattern the circuit of the hydrogel-elastomer-based stress–strain sensor. We then measure the resistance change of a hydrogel-elastomer-based strain sensor under uniaxial tension. As shown in Figure 5(a), we deform the sensor by the stretch $\lambda$ ($\lambda = 1 + \varepsilon$, $\varepsilon$ is the strain of the material) from 1 to 3, and measure the current resistance $R$ to be about 5.5 times of the original resistance $R_0$. The $R/R_0 - \lambda$ relation exhibits a nonlinearity that can be precisely captured by the previous-derived equation [4]: $R = a\lambda^2 + b\lambda^{-1}$, where $R_0 = a + b$, $R_0 = 174$ k$\Omega$, $a = 110$ k$\Omega$, $b = 64$ k$\Omega$. It should be noted that the parameter $a$ is the resistance of the part along the length direction of the sensor, and the parameter $b$ is the resistance of the part perpendicular to the length direction of the sensor. For the bulk hydrogel sample in Figure 4(e), $a = 1.46$ k$\Omega$; $b = 0$ which causes the difference in the $R - \lambda$ relation between Figure 4(e) and Figure 5(b).

To further investigate the dynamic response of the hydrogel-elastomer-based strain, we performed cyclic loading-unloading tests with different strain amplitudes (50%, 100%, 150%, 200%, respectively). As shown in Figure 5(b), the strain sensor responds precisely by following the $R/R_0 - \lambda$ relation in Figure 5(a). Moreover, we perform a stepwise loading by gradually increasing the strain from 0% to 25%, 50%, 75%, and 100%, and then gradually decreasing the strain back 0%. Figure 5(c) shows that the strain sensor responds...
4.2. Strain sensor on a soft pneumatic actuator

The hydrogel-elastomer-based strain sensor can be readily attached and bonded to structures and devices made of acrylate-based polymers. To demonstrate this advantage, as illustrated in Figure 5(d), we attach the hydrogel-elastomer-based strain sensor to the top surface of a fully 3D printed soft pneumatic actuator (SPA) made of a BA-PEGDA elastomer. Since the sensor is attached to the top of SPA, it stretches when the SPA bends under pressure (Figure 5(e)). The SPA can be printed on a commercial digital light processing-based desktop 3D printer such as Asiga Pico, Envision TEC, or other similar products. The sensor and the SPA can be firmly bonded by simply applying a layer of BA-
PEGDA precursor between the sensor and the SPA followed by UV curing. Figure 5(f) presents the snapshot of the fabricated SPA with a hydrogel-elastomer-based stretchable strain sensor. As shown in Figure 5(g), the attached sensor is capable of measuring tensile strain and bending angle of the SPA by mapping the relation between strain/bending angle and resistance change. Based on this, as presented in Figure 5(f), we can further confirm that the tensile strain and bending angle have a linear relationship.

5. Conclusion

In this work, we report a simple projection lithography method to fabricate hydrogel-elastomer-based stretchable strain sensors. This is a general and affordable method: it only requires a UV projector or even a UV lamp with photomasks; the chemicals used in this method are commercially available; the protocols for preparing the UV-curable polymer precursors are friendly to the users who do not have chemistry background. This manuscript introduced the general fabrication procedures, presented details on the preparations of hydrogel and elastomer precursors, and mechanical and electrical characterizations of the prepared materials, provided an application example where the manually manufactured stretchable strain sensor is attached to a 3D-printed pneumatic soft actuator to measure the strain and bending angle during actuation.

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Disclosure statement

No potential conflict of interest was reported by the author(s).

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