

Performance characterization of ionic-hydrogel based strain sensors

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Ionic hydrogels, owing to the advantages of stretchability, conductivity and transparency, have attracted more attention for developing new soft sensors and artificial skins. Existing works on ionic-hydrogel based sensors mostly focus on material synthesis, structure design and functional integration, while few studies investigate the characterization of their sensing performances. In this paper, we present a method to characterize the performance (e.g., sensitivity, linearity and repeatability) of a kind of ionic-hydrogel based strain sensors by varying the testing frequencies and the sensors' geometry (e.g., length-width (L/W) ratio). To this end, we first develop an experimental testing platform and fabricate a series of strain sensors made of the polyacrylamide (PAAm) hydrogel containing ionic conductive medium. We establish an equivalent electrical model to represent the ionic-hydrogel based strain sensors, indicating the influence of the testing frequencies and L/W ratio of the ionic hydrogels on their sensing performances. These theoretical predictions are in agreement with results obtained through experimental measurements. We further demonstrate that (1) the sensitivity of the strain sensors, characterized by the gauge factor (GF), increases with the rise of testing frequencies but tends to be stable over the frequency of 5 kHz; (2) the sensitivity GF has a nonlinear relation with the L/W ratio of the strain sensors, but with a certain maximum value under the same testing frequency when the L/W ratio equals to 4.80. We verify the above experimental observations with two commonly used electrolytes, including lithium chloride and sodium chloride. With the optimum testing frequency and L/W ratio, we finally conduct various experiments to demonstrate the low hysteresis and good repeatability of our ionic-hydrogel based strain sensors. This work provides an approach to characterize the performance of the ionic-hydrogel based strain sensors, which may be an important step forward in further applications of ionic hydrogels in soft robotics.

ionic hydrogels, sensor characterization, large-strain sensor, stretchable electronics

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1 Introduction

Soft sensors are essential in various emerging fields such as soft robotics [1], wearable devices [2–4], human-machine interfaces [5,6], and prosthetics [7,8]. The inherent stretchable ability makes them suitable for detecting large de-

formation while keeping the conformal contact with measured objects, which is not applicable for traditional rigid sensors without stretchability [9].

During the last decade, many soft sensors with different measuring principles like resistive [10,11], capacitive [12,13], or piezoresistive [14,15] have been developed. In these designs, the key technology is the use of a stretchable electrode, such as liquid metal [11] and ionic gel [16]. Alternatively, ionic hydrogels are promising as a candidate due

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to the inherent softness, high stretchability, transparency, and good biocompatibility [17]. By adding ionic conductive medium, many kinds of ionic-hydrogel based soft sensors such as strain sensors [18,19], touch panels [20], and hybrid circuits [21] have been recently developed. In these works, the main contributions lie in synthesizing materials, designing structures, and integrating multi-functional sensors, while few studies investigate their sensing performances with different testing conditions. In addition, due to the feature of the ionic conduction, an electrical double layer is generated between the electrical conductor and the ionic hydrogels [22,23], which makes the characterization of the ionic-hydrogel based sensors difficult.

In this paper, we provide a method to characterize the sensing performance of a kind of ionic-hydrogel based strain sensors by varying the testing frequencies and length-width (L/W) ratio of the ionic-hydrogel electrodes. To this end, we first establish an experimental testing platform and fabricate a series of strain sensors made of the polyacrylamide (PAAm) hydrogels containing ionic conductive medium. An equivalent electrical model is then developed to represent the ionic-hydrogel based strain sensors, indicating the influence of testing frequencies and the L/W ratio of the ionic hydrogels on their sensing performances. The theoretical predictions are in agreement with results obtained through experimental measurements. From the experimental results, we further demonstrate that with the increase of the testing frequencies, the sensitivity of the strain sensors, characterized by the gauge factor (GF), rises as well, which tends to be stable over the frequency of 5 kHz. In addition, we notice that the sensitivity GF has a nonlinear relation with the L/W ratio of the ionic-hydrogel electrodes, but with a certain maximum value under the same testing frequency when the L/W ratio equals to 4.80. Furthermore, with the optimum testing frequency and L/W ratio, several experiments are conducted to investigate the linearity and repeatability of the ionic-hydrogel based strain sensors, which demonstrates the low hysteresis and good reliability.

The remainder of this paper is organized as follows. Section 2 presents the experimental setup, the synthesis process of the ionic hydrogels and the fabrication of ionic-hydrogel based strain sensors. The equivalent circuit model and the experimental results are reported in Section 3. Section 4 discusses the optimum testing frequency and L/W ratio and also shows the low hysteresis and good repeatability of our development. Finally, conclusions and an outlook for future work are drawn in Section 5.

2 Description of the experimental platform

2.1 Experimental setup

Figure 1(a) shows the experimental setup to test the ionic-

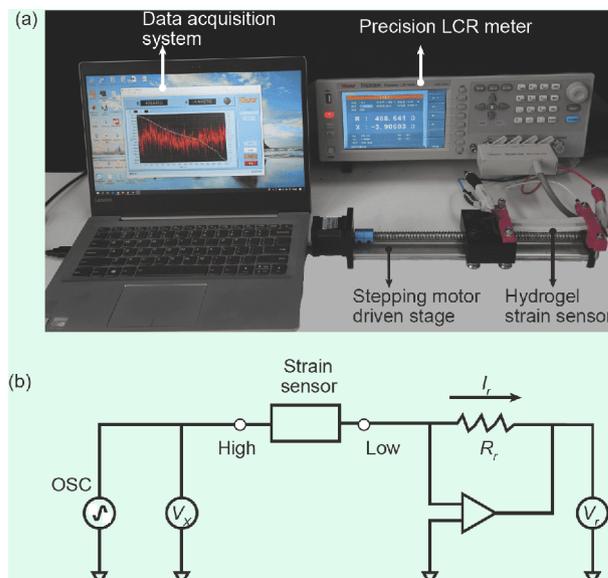


Figure 1 (Color online) Experimental description for characterization of the ionic-hydrogel based strain sensors. (a) Experimental setup; (b) schematic diagram of the testing circuit.

hydrogel based strain sensors, and the diagram of the testing circuit is illustrated in Figure 1(b). The experimental setup consists of a stepping motor driven stage (ST42H4809, Changzhou Sutai Electrical Appliance Co., Ltd., China), a precision LCR meter (TH2838H, Changzhou Tonghui Electronic Co., Ltd., China) and a commercial data acquisition system. Since the LCR generally requires hundreds of milliseconds to complete a measurement in our setup, we set the sampling frequency of 2.5 Hz in this work to investigate the influence of the wide-range testing frequencies (i.e., 20 Hz–2 MHz). During the testing, the strain sensors are mounted on the driven stage with a self-designed fixture and the stage is controlled by the Arduino board to move at a speed of 1.25 mm/s with a maximum strain of 100%. We may mention that the strain limit of our ionic-hydrogel based strain sensors is about 300%. Focusing on the influences of the testing frequency (f) and geometry on sensors' GF , we restrict the maximum strain of 100% in this work, which is available in most applications such as human-motion detections and electrical skins [18,24,25]. The output resistances of sensors are measured and collected by the precision LCR meter automatically.

In this work, we focus on investigating the influences of testing frequencies and L/W ratio of the ionic-hydrogel electrodes on the sensing performance. Therefore, we first fabricate the strain sensors with the same electrode's geometry and conduct experiments by varying testing frequencies from 20 Hz to 2 MHz. Then, we fabricate the strain sensors with different L/W ratios of ionic-hydrogel electrodes from 1.76 to 11.2 and perform experiments with the same testing frequency.

2.2 Synthesis of the ionic hydrogels

The ionic hydrogels are synthesized by mixing the acrylamide (AAM; J&K) monomer solution, *N,N'*-methylenebisacrylamide (MBAA; Molbase) crosslinker solution and 2-ketoglutaric acid (Adamas) as the photoinitiator with the mass ratio of 96.67:1.13:2.20 to form the precursor ink. The monomer solution is prepared by mixing AAM, electrolytes (LiCl·H₂O; Sinopharm Chemical Reagent Co., Ltd., or NaCl; Shanghai Macklin Biochemical Technology Co., Ltd.), and deionized water with the mass ratio of 9.98:16.16:73.86. MBAA solution is prepared by dissolving MBAA into deionized water with a mass ratio of 1.2%. In each mixing process, the solution should be agitated with a magnetic stirrer for 1 h to form a homogeneous solution.

Here, we choose two commonly used electrolytes (NaCl and LiCl) as conductive medium in the hydrogel precursor to fabricate two types of strain sensors with mass ratio of 10% based on our previous work [26], to verify the generality of the effect of testing frequencies and L/W ratio on sensitivity.

2.3 Fabrication of ionic-hydrogel based strain sensors

The designed ionic-hydrogel based strain sensors are composed of three layers made of stretchable materials (Figure 2(a)). The intermediate layer is the ionic-hydrogel electrode (2 mm). Both the top and bottom layers are VHB

elastomers (4910, 3M) as the encapsulation to effectively prevent the dehydration of the ionic-hydrogel electrodes and enduringly achieve the robust sensing performance [27]. Then, two strips of carbon fabric are laid at two ends in contact with the ionic-hydrogel electrode for connecting the external measuring equipment.

To fabricate the ionic-hydrogel based strain sensors, we utilize the molding method, which is shown in Figure 2(b). First, a piece of VHB elastomer (1 mm) as the substrate of the strain sensor is pretreated by being exposed to oxygen plasma to introduce hydroxyl-activated surface oxides on the surface (30 W at a pressure of 27 Pa; Atto, Diener Co., Ltd., Germany) for 5 min. Then, the substrate is treated with the benzophenone alcohol solution (benzophenone monomer and the alcohol solution mixed in a mass ratio of 1:9) to alleviate oxygen inhibition effect and activate elastomer surface for the robust bonding with ionic hydrogels [28,29]. After pretreatment, a laser cut mold (VLS3.50, Universal Laser Systems, Inc., USA) is arranged on the substrate and the hydrogel precursor ink is injected into the mold through a needle. Afterwards, the ultraviolet light is applied to initiate the crosslinking for about 20 min with the power of 500 W and the wavelength of 365 nm. After crosslinking process, the mold is peeled off from the cured hydrogel and wires are arranged at both ends of the sensor for a better interconnection with the readout machine. Finally, another layer of VHB elastomer (1 mm) is laminated on the top for en-

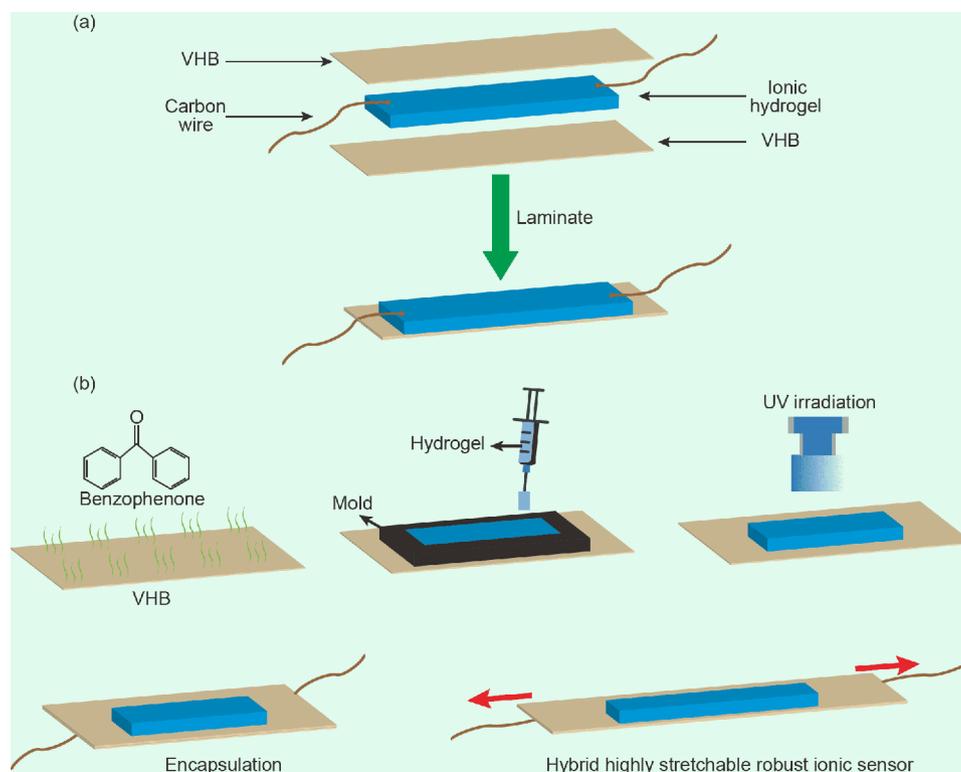


Figure 2 (Color online) Schematic of the designed ionic-hydrogel based strain sensor and its fabrication process. (a) Exploded view of the strain sensor and the assembled view of a strain sensor; (b) fabrication steps.

capsulation. In order to investigate the effect of the geometry size, a series of strain sensors with different ionic-hydrogel electrodes (L/W ratio varying from 1.76 to 11.2) are fabricated.

3 Experimental results

3.1 Electrical model and performance characterization

To evaluate the performance of a strain sensors [26,30–32], the sensitivity, characterized by the GF , is usually employed, which can be expressed as

$$GF = \frac{\Delta R/R_0}{\varepsilon}, \quad (1)$$

where $\Delta R=R-R_0$ is the resistance change of the strain sensor with R_0 as the original resistance of the unstretched state and R as the current resistance of the stretching state when the strain is ε .

For the ionic-hydrogel based strain sensors, modeling the resistance is generally complex due to the ionic conduction and the existence of the electrical double layer (EDL) [22,23,30]. In this work, we present an equivalent circuit model for our ionic-hydrogel based strain sensors as shown in Figure 3. In this model, the main part of the ionic hydrogel is described as a pure resistance, R_g , which is related to electrical resistivity (ρ), length (l), cross-sectional height (h) and cross-sectional width (w) according to the Ohm's law, $R_g=\rho l/(hw)$. At the interface between ionic hydrogel and electronic connection, the electrolyte-electrode interface is considered as a parallel circuit of the EDL capacitance C_e and the leakage resistance R_e . R_{ic} represents the resistance of the carbon fabric. The impedance of the C_e is calculated by

$$X_C = \frac{1}{2\pi f C_e}, \quad (2)$$

where f is the frequency, relating with the testing circuit as shown in Figure 1(b).

According to the equivalent circuit model in Figure 3, we can calculate the original resistance R_0 of the ionic-hydrogel based strain sensor as

$$R_0 = 2 \frac{R_e}{\sqrt{(2\pi f C_e R_e)^2 + 1}} + R_g + 2R_{ic}. \quad (3)$$

For the ionic hydrogels, only configuration of hydrogel's polymer network and water molecules are changed, which

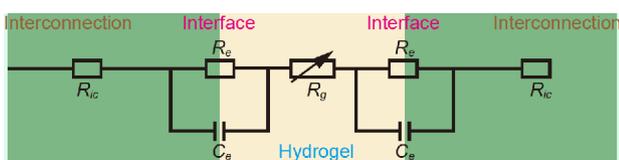


Figure 3 (Color online) Equivalent circuit model of hydrogel-interconnection interface for the ionic-hydrogel based sensors.

has a negligible influence on the ionic conductivity [30]. Therefore, we suppose that when the ionic-hydrogel based strain sensor is stretched, only R_g increases due to the change of l , h and w , while other resistance and capacitance components (i.e., R_e , R_{ic} and X_C) keep constant. Thus, we can conclude that although R and R_0 depend on testing frequency f , the change of the resistance $\Delta R=R-R_0=\Delta R_g$ is independent on f as shown in Figure 4.

Based on this assumption, we can further express the GF of the ionic-hydrogel based strain sensor as

$$GF = \frac{\rho(\varepsilon+2)}{\frac{w}{l} \cdot \frac{2hR_e}{\sqrt{(2\pi f C_e R_e)^2 + 1}} + \rho + \frac{w}{l} \cdot 2hR_{ic}}. \quad (4)$$

From eq. (4), we can demonstrate that the GF of the ionic-hydrogel based strain sensor is mainly influenced by f (relating to the testing frequency) and R_g (relating to the length and width of the ionic hydrogels). However, we may note that it is generally difficult to exactly obtain the value based on eq. (4) due to the unknown of R_e , C_e , R_g , and R_{ic} . In the following, we will present the experimental method to verify our model and investigate the influence of the testing frequency and geometry of the ionic hydrogels on the GF of the strain sensors.

3.2 Effect of the testing frequencies

To sufficiently verify the influence of testing frequencies, we first carry out impedance-frequency tests on various ionic-hydrogel based sensors. We measure the impedance of the sensor as a function of testing frequencies. The results show that the impedance is high at the low-frequency range due to a large capacitive impedance that arises from the hydrogel-carbon fabric interface [33]. When the testing frequency is higher than 5 kHz, the impedance tends to be stable. This may be explained that the capacitive effects at the interface

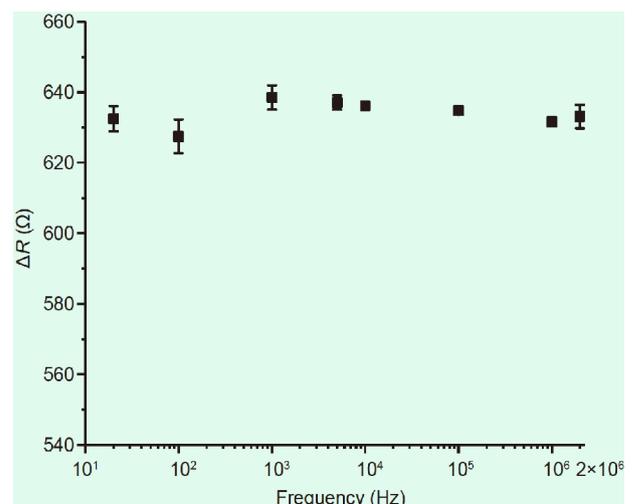


Figure 4 Effect of the testing frequency on the resistance change.

are inversely proportional to frequency and the impedance decreases with the increasing high frequencies. We can see from Figure 5 that this result is consistent regardless of the type of electrolyte.

Then we characterize the response of the ionic-hydrogel strain sensor with different testing frequencies (i.e., 20 Hz, 100 Hz, 1 kHz, 5 kHz, 10 kHz, 100 kHz, 1 MHz, and 2 MHz). We may mention that the frequency range of 20 Hz–2 MHz depends on the used LCR meter, which also covers commonly used frequencies for ionic-based hydrogel strain sensors in refs. [30,34]. Figure 6(a) and (c) show the relative changes of the resistance $\Delta R/R_0$ versus strain under different testing frequencies with the electrolyte of LiCl and NaCl, respectively. The experimental results show that the curves of the sensor under 5 kHz, 10 kHz, 100 kHz, 1 MHz, and 2 MHz almost overlap, while there are noticeable differences in the range between 20 Hz and 5 kHz. Furthermore, Figure 6(b) and (d) indicate that the GF dramatically rises with the increase of the testing frequency until the value of the frequency reaches approximately 5 kHz. After that the GF tends to be constant. These results demonstrate that using a higher testing frequency (i.e., 5 kHz) for ionic-hydrogel based strain sensor can get a better sensing performance in sensitivity, which is also consistent with the theoretical

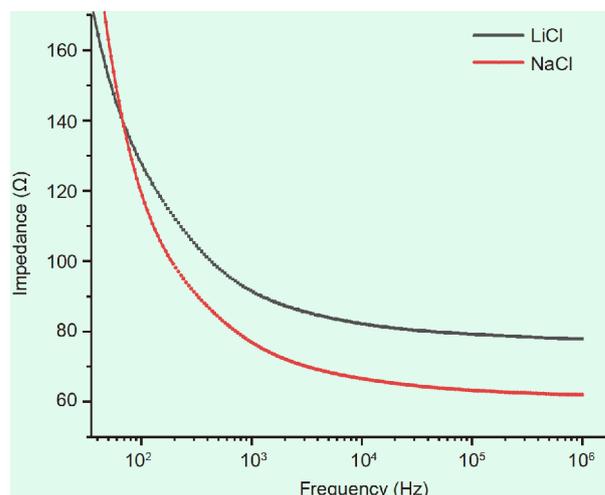


Figure 5 (Color online) Impedance spectra of PAAm ionic hydrogels with the same geometry size of varying electrolytes.

analysis. Another interesting phenomenon we find is that $\Delta R/R_0$ sharply increases when the strain (ϵ) is beyond 20%, which demonstrates that pre-stretching the strain sensors for certain times (i.e., 20%) may contribute to better sensing performance and the result is similar to the phenomenon in

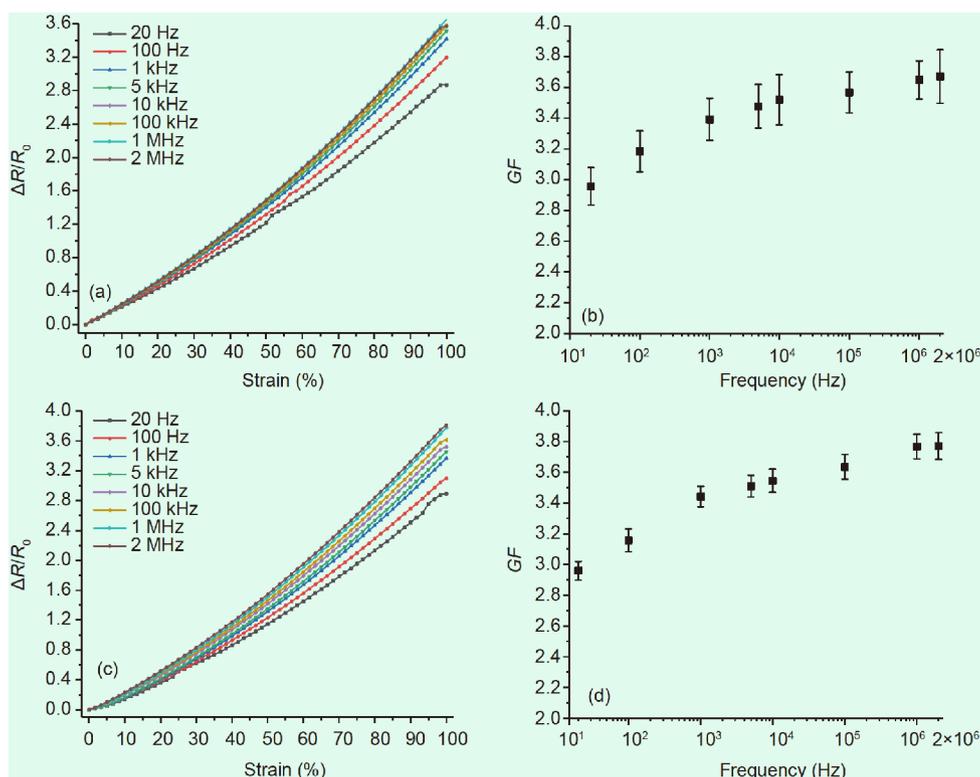


Figure 6 (Color online) Effect of the testing frequencies on the sensing performances. (a) The relative changes of the resistance are plotted as a function of strain under different testing frequencies with the electrolyte of LiCl (we only select one load process of four cycle tests in the picture); (b) GF of the sensor is plotted as a function of the testing frequency with the electrolyte of LiCl (GF is plotted with the mean value of four cycle tests); (c) the relative changes of the resistance are plotted as a function of strain under different testing frequencies with the electrolyte of NaCl (we only select one load process of four cycle tests in the picture); (d) GF of the sensor is plotted as a function of the testing frequencies with the electrolyte of NaCl (GF is plotted with the mean value of four cycle tests).

capacitive strain sensor we have found in our previous work [35].

3.3 Effect of the length-width ratio

Next, we conduct experiments to investigate the effect of the geometry (i.e., the length and width) of the ionic hydrogels on the sensitivity of the strain sensors. For ease of investigation, we use the dimensionless ratio, i.e., L/W ratio. To this end, a series of ionic-hydrogel based strain sensors are fabricated with different L/W ratios of 1.76, 2.31, 2.67, 4.80, 6.93, 9.07, and 11.2, while all other fabrication parameters are kept consistent. Figure 7(a) and (c) show the relative change of resistance $\Delta R/R_0$ versus the strain for different L/W ratios with the electrolyte of LiCl and NaCl respectively. As shown in Figure 7(b) and (d), we can see that the GF first dramatically increases and thereafter decreases when the L/W ratio overtakes 4.80. This result is interesting for designing the ionic-hydrogel based strain sensors with better performance. We note that this coupling effect has not been reported in the literature. We may explain it by the combination influence of the pure resistance (R_g) and the complex interface-effect between the electronic and ionic conductors (i.e., C_e and R_e) according to eq. (4). Firstly, the

change of L/W ratio occupies the dominant status, which will result the increase of GF . However, the effect of L/W ratio gradually weakens and the complex interface-effect becomes the main factor affecting sensitivity as the L/W ratio continues to increase, which will cause a decrease in GF . As a result, there is an optimum value in the L/W ratio.

Therefore, based on above observations, the L/W ratio should be considered when designing and fabricating the strain sensor to obtain a better performance in sensitivity. In this work, we select the relatively optimum value of 4.80 for ionic-hydrogel based strain sensor.

4 Discussion

The developed equivalent circuit model of the ionic-hydrogel based strain sensors reveals the qualitative effect of testing frequencies and geometry of the ionic hydrogels on the GF , which have been further verified in our experiments. The results show that the sensitivity GF steadily levels up with the increase of the testing frequencies and reaches a plateau when the frequency is relatively high. It should be notable that the high-frequency testing certainly requires high cost and also makes the design of testing equipment

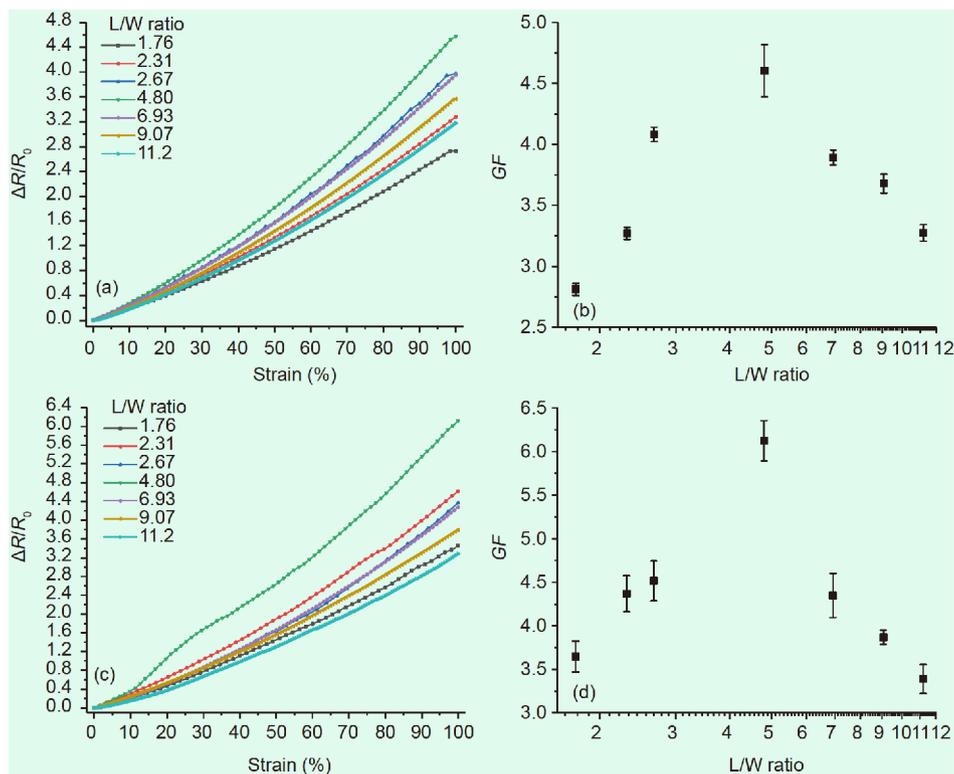


Figure 7 (Color online) Effect of the L/W ratio on the sensing performances. (a) The relative changes of the resistance are plotted as a function of strain under different L/W ratio with the electrolyte of LiCl (we only select one load process of four cycle tests in the picture); (b) GF of the sensor is plotted as a function of the L/W ratio with the electrolyte of LiCl (GF is plotted with the mean value of four cycle tests); (c) the relative changes of the resistance are plotted as a function of strain under different L/W ratio with the electrolyte of NaCl (we only select one load process of four cycle tests in the picture); (d) GF of the sensor is plotted as a function of the L/W ratio with the electrolyte of NaCl (GF is plotted with the mean value of four cycle tests).

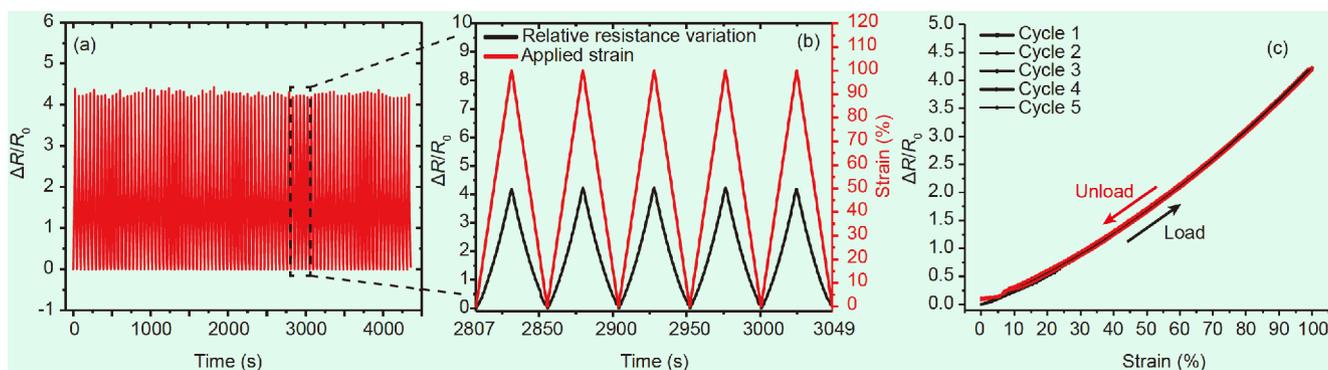


Figure 8 (Color online) Measured performance of the fabricated sensor under multiple cycles. (a) The relative change of resistance $\Delta R/R_0$ of the ionic-hydrogel based strain sensor upon cyclic strain up to 100% for 4356 s; (b) $\Delta R/R_0$ and strain of the ionic-hydrogel based strain sensor vs. time from 2807 to 3049 s based on the results in (a). (c) $\Delta R/R_0$ is plotted as a function of strain based on the results in (b).

difficult. Therefore, we select 5 kHz as the optimum testing frequency, which ensures good sensing performance as well as relatively low testing cost. According to the results presented in Figure 7, there is also an optimal range (about 4.80) for the geometry size of the ionic-hydrogel electrode for better sensitivity of strain sensors. Besides, there are still some other factors influencing the performance of the ionic-hydrogel electrode such as the dehydration [34,36,37]. However, it is generally difficult to consider this effect in the development model, which will be investigated in our future work. To reduce the dehydration effect in this work, we have encapsulated the ionic-hydrogel electrode in two pieces of elastomer.

We also use two kinds of ionic conductive medium to conduct the same experiments, and show that the ionic conductive medium has little effect on the results of performance characterization. However, for different conditions we suggest different kinds of electrolyte. For example, in most applications, lithium chloride is added into hydrogel because LiCl is proved to be the most effective electrolyte for water retaining [38]. While due to its slightly corrosive characteristic, it is inappropriate to be applied in wearable devices which may directly contact with human skins. On the contrary, hydrogel mixed with NaCl represents better biocompatibility which is suitable for wearable devices and the human-machine interface.

With the above discussion, we also conduct various experiments to show the low hysteresis and robustness of our ionic-hydrogel based strain sensors when the testing frequency is 5 kHz and the L/W ratio is 4.80. Figure 8(a) and (b) show the cyclic response of the relative change of resistance $\Delta R/R_0$. We can see from the results that the ionic-hydrogel based strain sensor has good repeatability during long-time cyclic applications. In addition, based on results in Figure 8(c), we plot $\Delta R/R_0$ as a function of the strain of the ionic-hydrogel based strain sensor, which demonstrates the low hysteresis (about 0.0129, the mean value of five sets of

cyclic data in Figure 8(c)).

5 Conclusions

In this paper, we present a method to characterize the performance of the ionic-hydrogel based strain sensors by varying the testing frequencies and L/W ratio of the sensors' geometry. An experimental testing platform is established and a series of ionic-hydrogel based strain sensors are fabricated based on the polyacrylamide (PAAm) hydrogel containing two commonly used types of ionic conductive medium. We develop an equivalent electrical model to represent the ionic-hydrogel based strain sensors, which clearly reflects the influences of the geometry of the ionic hydrogels and testing frequency on sensing performance. We then present experimental results to verify the theoretical predictions. Finally, we also conduct a series of experiments to show the low hysteresis and good repeatability of our strain sensors with the selected testing frequency and L/W ratio of the ionic hydrogels. In future work, we will extend our work for exploring other influence factors (e.g., dehydration and ionic concentration) on the performance of the ionic-hydrogel based strain sensors, and integrating them in the applications of soft wearable robotics.

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