Flexible Highly Sensitive Pressure Sensor Based on Ionic Liquid Gel Film

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Supporting Information

ABSTRACT: Flexible, semitransparent ionic liquid gel (ionogels) film was first fabricated by in situ polymerization. The optimized ionogels exhibited excellent mechanical properties, high conductivity, and force sensing characteristics. The multifunctional sensor based on the ionogel film was constructed and provided the high sensitivity of 15.4 kPa−1 and wide detection range sensing from 5 Pa to 5 kPa. Moreover, the aforementioned sensor demonstrated excellent mechanical stability against repeated external deformations (for 3000 cycles under 90° bending). Importantly, the sensor showed advantages in detection of environmental changes to the external stimulus of subtle signals, including a rubber blower blowing the sensor, gently touching, torsion, and bending.

INTRODUCTION

Pressure sensors have been of great interest in Internet of things applications,1−5 including smart windows, displays, security system, mobile phones, and prospective electronic skin.1−6 Among various aspects of pressure sensors, it is necessary to develop excellent flexibility, high sensitivity, and a wide-range sensing,7−9 which are significantly important to detect human activities.10−12 Hence, enormous efforts are being paid to fabricate the sensors with high sensitivity and excellent flexibility. The piezoresistive sensors inserting the micro/nanopatterns were used to improve the sensitivity, and fabricating the nanostructure-based hybrid films in sensing layers to mend the flexibility has been developed. Despite the great progress achieved so far, the aforementioned methods are still limited in extensibility, high cost, and poor durability, thus restricting their application in wearable sensors.

Ionic conductors13 are a class of functional materials with key roles in energy storage, solar energy conversion sensors,14−17 and electronic devices. One important kind of ionic conductor is hydrogels18 containing electrolytes, which offer a unique combination of high stretchability, transparency, and bio-compatibility. They have showed great promise as stretchable ionic devices, such as electrically activated, soft, transparent actuators19 and ionic skins sensing the location and pressure of touch. However, the poor electromechanical stability of aqueous electrolytes limits their further application in wearable electronics. Comparably, the ionogels20 represent an important sensing material including the polymer composites with ionic liquids (ILs), which possess the attributes of the solid and the liquid: the polymer network provides an elastic solid form that avoided the drawbacks of IL leakage, and the ILs enable electrical conduction. Thus, the ionogels have been developed intensely in wearable electrical fields, in particular in E-skin devices.21,22 It is worth noting that the nonconductive polymer in the ionogels sometimes affects the conductivity of the ionogels, thus limiting the sensitivity.23 Despite the great progress in the ionogel-based E-skin achieved so far, the development of a highly sensitive ionogel-based pressure sensor with multifunction remains a considerable challenge due to the limitation of materials and engineering.

Herein, air-stable, highly conductive, semitransparent ionogels were fabricated by in situ synthesizing 1-vinyl-3-ethylimidazolium dicynamide ([VEIm][DCA]) ILs containing a carbon–carbon double bond.23,24 The effects of the components of ionogels on its properties such as flexibility, conductivity, and transparency were investigated. The optimized ionogel-based sensors exhibited excellent mechanical properties, high conductivity, and force sensing characteristics. Importantly, the aforementioned pressure sensor provides high sensitivity of 15.4 kPa−1, little hysteresis, and a wide detection range, which had potential application in monitoring the external stimulus of subtle signals and human activities.

RESULTS AND DISCUSSION

The chemical structures of the materials and preparation process of the ionogels were schematically illustrated in Figure 1a and 1b, respectively. The detailed descriptions of the ionogel
fabrication process were presented in the Experimental Section. In brief, the N,N'-methylene bis(acrylamide) (NNMBA) and ammonium persulfate (APS) were first dissolved in 1-vinyl-3-ethylimidazolium dicyanamide ([VEIm][DCA]) ILs to form the prepolymer solution, and then the solution was heated at 80 °C in a vacuum oven. After heating for 2 h, the colorless solution would change into the ionogels with the orange color, as shown in Figure 1c.

The effects of the ionogel components on its properties, especially for the film morphology, color, transmittance, and conductivity, were investigated in detail. As shown in Figure 2 and Figure S1, the proportions of NNMBA and APS on ionogels had significant effects on the morphology and the color. When the ratio of NNMBA/APS was more than 1:1 in the mixture of 2 mL of ILs with 25 mg of APS, significant cracks occurred, which might be attributed to the coexistence of multiple phases due to nonreactive NNMBA.25−27 With the decrease of the ratio of NNMBA, the surface of the gel film became smooth, and the color changed from light black to brown. At 1:1 ratio of NNMBA/APS, the orange gel film with smooth surface could be obtained (Figure 2a and 2b). With the further decrease of the ratio, the ionogels cannot be formed due to lack of polymerization. In addition, the content of the APS had obvious effects on the film morphology and transparency. As shown in Figure 2c, some cracks occurred again, and the gel color becomes deeper from brown to black, which reduced the transmittance of the gel film when there was an increase of APS in the mixed solution with 2 mL of ILs and 25 mg of NNMBA. Figure 2d presented a photograph of the outside view of the laboratory through the ionogel film. It was obvious that a semitransparent film was formed. The composites of the ionogels were investigated by the EDX and FT-IR spectra, and the results were shown in Figure S2 and Figure S3. It was obviously seen that the new peak was observed at the 1750 cm⁻¹ in the FT-IR after reaction. Moreover, the EDX element analysis spectrum exhibited only C, N, and O peaks without the S peak. These results further confirmed that the ionogels were formed, including of the ILs and the NNMBA without the APS.
In addition, the ionogels also obtained good thermostability, which also depended on the composite of the ionogels. As shown in Figure S4, there was a two-step weight loss process as the temperature rose from room temperature at 10 °C/min. The first region of the ionic gel polymer appeared from 120 to 230 °C, where the heat loss of the ionogels with NNMBA/APS of 1:1 was minimum corresponding to 6−12% weight loss only. With the further increase of the temperature, the heat loss increased dramatically which had reached 44−47% at about 450 °C. Moreover, the sharp endothermic peaks appeared in the DSC thermogram of the ionogels with varied ratios of NNMBA/APS corresponding to melting temperature \( T_m \) of \( \sim 260 \) °C, together with a crystallization \( T_c \) of \( \sim 250 \) °C and a decomposition temperature \( T_d \) of \( \sim 330 \) °C (shown in Figure S5). Moreover, the intensity of the exothermic peak gradually decreased with increasing of the ratio of NNMBA/APS in the ionogels. Therefore, under the above optimized condition, the NNMBA, APS, and ILs were completely reacted, avoiding the phase separation, resulting in higher transparency, smooth surface, and better stability.

Moreover, it was found that the composite of gel had an obvious effect on the flexibility and conductivity except the transparency and morphology. It was seen that the device based on ionogels fabricated on the 1:1 showed the better flexibility. Furthermore, the aforementioned film also showed the high conductivity as described from Figure 3a and 3b, which might provide a promise in the high sensitive pressure sensor. Based on the above description, the ionogels with 1:1 ratio of NNMBA/APS showed the higher transparency and conductivity and better morphology and flexibility. Therefore, the composition comprised of 25 mg of NNMBA, 25 mg of APS,
and 2 mL of ILs was used as the optimal composite for the following sensing investigation. High sensitivity was necessary to detect the subtle pressure signal, which was important for both fundamental research and practical applications. To investigate the sensitivity, the typical sensor was constructed on the optimized ionogels. The device structure was schematically illustrated in Figure 4a, where the size of the ionogels was 15 × 7 × 0.4 mm, and a detailed fabrication process was shown in the Experimental Section. The sensitivity of the sensor could be defined as $S = \delta(\Delta I/I_0)/\delta P$, where $\Delta I$, that is $I - I_0$, was the relative current change when a certain pressure was applied on the devices; $I_0$ was the pristine current of the device under no pressure; and $P$ was the applied pressure. Figure 4b showed the sensitivity of a pressure based on ionogel film. Similar to most of the reported pressure sensors, the plot for sensitivity was composed of different regions. Generally, in the low-pressure regime, the sensor obtained the higher sensitivity, while in the high-pressure regime, the sensor had lower sensitivity. The sensitivity was approximately 0.45 kPa⁻¹ in the region of more than 30 Pa and was 6.21 kPa⁻¹ in the range of 4 Pa ~ 25 Pa, at the bias voltage of 4 V. In addition, the sensitivity increased with the bias voltage, which could arrive to 15.4 kPa⁻¹ at the bias voltage of 10 V as shown in Figure S4. The dependence of the sensitivity on the voltage might be attributed to the Schottky barrier between ionogels and the electrodes.

The hysteresis behavior of the sensor was also investigated during loading and unloading pressure cycles and shown in Figure 4b. The loading and the unloading curves were almost overlapped, indicating little hysteresis existed. Moreover, the degree of hysteresis was calculated as follows:

$$DH = \frac{A_{\text{Loading}} - A_{\text{Unloading}}}{A_{\text{Loading}}} \times 100\%$$

where $A_{\text{Loading}}$ and $A_{\text{Unloading}}$ were the area of loading and unloading curves, respectively. A lower DH value indicated lesser hysteresis in the electrical response. The DH value of the obtained sensor was only 5.36%. The above result further confirmed that the sensor based on ionogels was capable of detecting pressure quantitatively and independently of the pressure history. Despite the facile fabrication process, the sensitivity based on the ionogel film can be comparable to or higher than most of the reported results in the literature as shown in Figure 4c. Compared to these sensors based on the ionogels, the aforementioned sensor had higher sensitivity than that of the capacitive pressure sensor based on ionogel film with the flat structure. In addition, the sensor possessed the detection limit of 5 Pa, which could be comparable to the most reported microstructure ionogel film. The above excellent performances may be attributed to the adhesion between ionogels and the Eco-Flex elastomeric soft matrix, which caused surface self-wrinkling of ion gels, resulting in the excellent force sensitivity of the sensor. As we know, the response current signals would be larger for high conductive pressure sensors, and thus very small applied pressure would be able to produce the detectable signals. Therefore, the high conductivity of ionogels and good contact between the conductive layer and electrode should be the main factors resulting in the high sensitivity.

Importantly, the sensor showed fast response when the 5 Pa pressure was loaded on the surface, demonstrating a similar response with the fast response and high sensitivity (shown in Figure 4d). Moreover, the sensor also showed obvious response to a small leaf (5.5 Pa) and light flower flake (4.6 Pa). The ionogel-based sensor possessed a high sensitivity and a low detection limit simultaneously, exhibiting potential applications for the protection of some precision instruments and a potential Hazard Alarm System. The pressure sensing mechanism of the sensor based on the ionogels was similar to the hydrogel and ionic nanofabric materials. Under subtle external pressure, the ionogel structure experienced compression with an increase of the high-density mobile ions, resulting in higher electrical conductivity.

A highly sensitive pressure sensor based on ionogel film allows us to efficiently detect a wide range of pressures in a single-device platform. Here, we monitored the relative resistance or current changes arising from a variety of pressure sources including minute pressure of leaf and light flower (∼5 Pa), low pressure of rubber blowing, and generally touching (<10 kPa). The results in Figure 5 showed that the sensor was capable of accurately detecting the various pressures describe above. Specially, the sensor showed high sensitivity and fast response to the touching. A gentle touch by a human hand (9 mN) was bigger than natural perturbation of breeze by swinging a feather (0.009 mN). When a finger gently touched the sensor surface, the resistance of ionogels immediately fluctuated with external loading and unloading. Moreover, the response signal had a sharp peak instead of a state curve, which illustrated a fast response and no hysteresis with the fast switching (Figure 5a). The touching and releasing...
time were 0.072 s and 0.753 s, respectively, which was far better than the reported results. The excellent features in touching sensing would enrich the application of the sensor in a robot touching sensor. Furthermore, the sensors could detect the noncontact actions such as air movement generated by human respiration. As shown in Figure 5b and Figure S5, notable peaks were seen when a man breathed toward the device and a rubber blower blew the surface of the device. During detection, the absence of direct contact action acted as a medium for transmitting the action triggers to the pressure sensor. The intensity of the signal peaks corresponded to weak and strong air movement, where the air movement was similar to loading to about 60 Pa pressure on the surface. The noncontact pressure sensors based on ionogels were expected to react to potential applications in many new fields such as turbulent flow detection, vibration monitoring, and an acoustic transducer and so on.

To explore the potential application in the flexible electronics, the bending property of the obtained pressure sensor was investigated. First, the ionogel-based sensor was integrated on the PET substrate. The sensor inherited superb flexibility of the PET upholder, and the bending angle was defined in the inset of Figure 6a. Even with the minute bending of 1°, an obvious relative resistance change was obtained, which corresponded to 0.05% bending strain. We harnessed the bending-induced strain on the upper surface of the PET slab as a stimulus to the sensor, which in turn reflected the bending extent. Figure 6a showed the relative resistance change from 0° to 15°, which was enough to illustrate that the sensor had a very keen response to the smaller deformation and, under 90° bending, also showed a stable and lasting response (Figure 6b). Even with the minute bending of 1°, an obvious relative resistance change was obtained. As we know, the electrical stability was an important factor in application of the pressure sensor. The response and restoration curves of the sensor measured for 3000 cycles under 90° bending were shown in Figure 6c. The output signals of the relative resistance change were stable, indicating the remarkable electrical stability. As aforementioned, this robust response was associated with the stable ionogels and good contact between the ionogel film and the electrodes. In addition, the sensor was sensitive to torsion stimulus and showed relatively stable resistance changes under the torsion loading and unloading (Figure 6d), where twisting degree was defined similar to the reported literature. The outstanding properties in bending or torsion sensing enabled the sensor to the application of monitoring joint bending of humans and robots. Importantly, this work would have a profound influence on the synthesis and device assembly of the next new ionogels, especially the design and construction of an ultrasensitive ionogel-based pressure sensor with various functions (bending, twisting, pressure, friction, stretching, and pulse). Most importantly, it would be used to monitor or capture the human body real-time signal in the smart wearable equipment and perceive a slight change in the surrounding environment.

**CONCLUSIONS**

Flexible and semitransparent ionogels were constructed by in situ synthesizing [VEIm][DCA] containing a carbon–carbon double bond. The ionogels exhibited highly conductive and force-sensing characteristics. The optimized ionogel-based sensor demonstrated high sensitivity of 15.4 kPa⁻¹, little hysteresis of 5.36%, and a low detection limit of 5 Pa and thus was extremely sensitive to ambient environmental changes including of weak gas flow, small leaf, flake flower, and gentle...
touched. Moreover, the sensor exhibited excellent mechanical stability against repeated external deformations (3000 cycles under 90° bending). Importantly, the optimized sensor showed multifunctional sensing characteristics including the pressure, bending, and twisting. The excellent performance and facile process suggested the potential application of the ionogel-based sensor in the detection of the interaction of human and robot and detection of the environment change.

**EXPERIMENTAL SECTION**

**Materials.** Polydimethylsiloxane (PDMS-184) was purchased from Dow Corning Corporation Midland-Michigan USA. Eco-flex 0030 was purchased from SMOOTH-ON. N,N'-Methylene bis(acrylamide) (NNMBA), ammonium persulfate (APS), and 1-vinyl-3-ethylimidazolium dicyanamide ([VEIm]-[DCA]) were purchased from Sigma-Aldrich Corporation, Shanghai, China. This IL with 99% purity was used directly without further purification.

**Ionogel Preparation.** NNMBA and APS were dissolved in 2 mL of [VEIm][DCA] ILs, and then the mixture was vigorously stirred for 30 min to form the solution. The mixed solution was heated at 80 °C in a vacuumed oven. After heating for 2 h, the colorless solution became the ionogels.

**Sensor Fabrication.** First the PDMS prepolymer mixed solution was first spin-coated on the clean silicon wafer at 350 rpm and heated at 80 °C for 2 h. The solidified PDMS film was cut into slices as a mold in the following process. Second, the mixture of Eco-flex was poured on the PDMS mold surface and solidified in a Petri dish. The Eco-flex film was peeled off, and PDMS mold was removed, forming the small channel with the height of 0.4 mm. Then the mixed solution was filled in the small channel with copper foils attached as electrodes at both ends and polymerized at 80 °C for 2 h. To enhance the contact of the ionogel film and the Eco-flex substrate, another two Eco-flex films were sealed on both ends of the devices, and the mixture solution of Eco-flex was poured on the surface of the covered film. After solidification of the Eco-flex, the sensor based on the ionogels was obtained.

**Characterization.** Thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) were performed in air using a TG/DTA6000 (Seiko Inc., Japan). The samples were heated from room temperature to 500 °C at 10 °C/min. The morphology of the ionogels was analyzed with an environment scanning electron microscope (ESEM, Quanta FEG250) and the reflection detection microscope (CMM-90AE). The composition of the ionogels was measured by Fourier transform infrared (FTIR, Thermo Fisher Scientific FTIR 6700) and the energy-dispersive X-ray spectroscopy (EDX). Electrical properties were measured using semiconductor meter Keithley4200.

**ASSOCIATED CONTENT**

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsomega.7b01575.

The photographs of ionogels; the EDX spectrum; the FT-IR spectrum; TG curves; DSC thermograms; the relative current change under different pressures; the relative current variations when breathing toward the device at the bias voltage of 4 V (PDF)

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**Notes**

The authors declare no competing financial interest.

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**ABBREVIATIONS**

NNMBA, N,N-methylene bis(acrylamide); APS, ammonium persulfate; ionogels, ionic liquid gel; ESEM, environmental scanning electron microscope; EDX, energy-dispersive X-ray spectroscopy; FT-IR, Fourier transform infrared spectroscopy; TG, thermogravimetry; DSC, differential scanning calorimetry

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