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Novel silicone-ionic liquid composite with keratin utilized as pressure sensor

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ABSTRACT

Dielectric elastomer (DE) sensors have great potential for applications in soft robotics, wearable devices and medical diagnostic. A novel pressure sensor with remarkably improved force sensing characteristics was obtained through combined usage of polydimethylsiloxane (PDMS) and ionic liquid (IL). The regenerated keratin from wool was added and dispersed homogeneously in the PDMS matrix acting as reinforcing fillers. The influence of the amount of IL on the electro-mechanical properties of the composites was investigated. One obvious result was that the permittivity of the IL-containing elastomers increased dramatically with the increased amount of IL loaded. Furthermore, the sensitivity of the composite elastomers as pressure sensors was investigated by recording the response of the voltage when a small force is applied to the top surface of the pressure sensor. The elastomers with IL loaded exhibit excellent response of the voltage and the maximum sensitivity of the composite elastomer is 2.64 mV/N.

Keywords: Polydimethylsiloxane, ionic liquid, keratin, pressure sensor, high sensitivity

1. INTRODUCTION

Pressure sensors have attracted much attention over the past few years due to their various applications in electronic skin^[1-3], soft robotics^[4-5] and energy harvesting^[6-8]. Various types of sensing technologies have been explored based on force-induced changes in piezoresistivity, capacitance and piezoelectricity^[9-10]. In recent years, enormous efforts have been made towards improving the performance of pressure sensors. A lot of nanomaterials, including carbon nanotubes^[11-14], gold/silver nanowires^[15-18], graphene nanosheets^[19-21], metal nanoparticles^[22-23], have been chosen for incorporation with polymers to fabricate novel, more sensitive pressure sensors. Tao et al.^[19] proposed a graphene-paper pressure sensor that shows excellent performance in the range of 0-20 kPa, and a sensitivity up to 17.2 kPa⁻¹ with a response time of 120 ms. Cho et al.^[24] presented a flexible capacitive pressure sensor that shows superior pressure sensing performance with a broad sensing range and fast response time. A flexible, highly sensitive pressure sensor based on an ionic liquid gel film was reported by Zhang et al.^[25] The reported pressure sensor has a high sensitivity of 15.4 kPa⁻¹ and a wide detection range sensing from 5 Pa to 5 kPa. However, a problem reveals from the above pressure sensors that a pressure sensor fabricated by incorporating polymers with nanomaterials has high sensitivity, but also exhibits limited stretchability, complex preparation process and high cost, which limit its application to a large extent. Therefore, it is necessary to develop a flexible sensor with high sensitivity and facile fabrication process as smart sensors for further application.

Ionic liquids (ILs), which have high permittivity and chemical stability, are a class of functional materials for novel pressure sensors for improvement of their performance^[26]. However, the poor electromechanical stability of ILs limits their further application as pressure sensors. Therefore, an ionic pressure sensor based on polymer composites with ILs, possessing both the elasticity of the polymer and the electrical conduction of the ILs, has received a lot of attention from researchers^[25, 27-28]. For example, a highly sensitive ionic pressure sensor with a microstructured dielectric layer with a sensitivity of 35.96 kPa⁻¹ has been prepared by Su et al.^[29]. Despite the great progress in ionic pressure sensors achieved so far, the development of ionic pressure sensor are still limited due to complicated fabrication processes and high cost.

Silicone elastomers represent an excellent matrix for sensors^[30-31] due to their inherent stability^[32-33], also upon elevated temperatures^[34]. However, silicone polymers are usually difficult to mix with other components and therefore tedious mixing schemes are usually required to ensure homogeneous composites.

Herein, a novel silicone-based composite consisting of silicone elastomer with ionic liquid and keratin was developed. Moreover, keratin, which has excellent biocompatibility and stability, was for the first time used in silicone elastomers as a reinforcing filler. This method not only improves the performance of the material as pressure sensors but it is also simple and facile, and thereby demonstrates a great potential for large-scale production of ionic materials. The effect of the amount of IL on the electro-mechanical properties of the composites was investigated. The presence of IL not only increased the dielectric permittivity of the resulting elastomer, but also benefitted to an increased pressure sensitivity. Especially, a maximum sensitivity of the composite elastomer up to 2.64 mV/N was demonstrated. Therefore, this novel pressure sensor will have great potential in the fields of smart skins, healthcare, and wearable devices, amongst others.

2. EXPERIMENTAL

2.1 Materials

Vinyl-terminated PDMS, DMS-V31 ($M_n = 28000 \text{ g mol}^{-1}$) and a cross-linker HMS-301 ($M_n = 1950 \text{ g mol}^{-1}$) were obtained from Gelest Inc, USA. The regenerated keratin from wool was purchased from Tokyo Chemical Industry Co., Ltd, and the platinum cyclovinyldimethyl siloxane complex catalyst (511) was purchased from Hanse Chemie, Germany. 1-butyl-3-methylimidazolium hexafluoroantimonate (BmimSbF₆) was purchased from Chengjie Chemicals Ltd, China. The structures of the chemicals can be seen in Figure 1.

Figure 1 Structures of telechelic vinyl functionalized PDMS (V31), hydride functional crosslinker (HMS301), and ionic liquid BmimSbF₆ used in this work.

2.2 Fabrication of BmimSbF₆ loaded pressure sensor

The fabrication of the BmimSbF₆ loaded pressure sensor is simple and facile, as mentioned previously. First, vinyl-terminated PDMS, DMS-V31 (5g, 0.18 mmol), cross-linker HMS-301 (0.1045g, 0.0536 mmol) and the catalyst (Pt 511) (15 ppm) were mixed using a speed mixer (DAC 150.1 FVZ-K, Synergy Devices Ltd, UK) at 3500 revolutions per minute (rpm) for 2 min. on a FlackTek Inc. The proportion of chemicals was based on previous research^[35]. Thereafter, keratin (25 wt%) and BmimSbF₆ were added in the mixture as solid filler and liquid filler, respectively, and the mixture was speedmixed once more. The homogenous mixture was coated on a glass substrate using a film applicator (3540 bird, Elcometer, Germany). Then, the film was cured in the oven at 80 °C for 2 h, after which, individual sensors were cut from the film with 5mm*5mm. The copper sheets were attached to the film as electrodes.

2.3 Fourier transform infrared spectroscopy (FT-IR) determination

Fourier transform infrared spectroscopy (FT-IR) spectra of the samples were conducted using a Nicolet iS50 FT-IR fitted with a diamond crystal attenuated total reflection accessory (ATR), which operated in the range of 4000-400cm⁻¹ at a resolution of 4 cm⁻¹ and 32 scans per measurement. All the spectra were baseline-corrected.

2.4 Dielectric properties determination

Dielectric relaxation spectroscopy (DRS) was carried out using a Novocontrol Alpha-A high-performance frequency analyser (Novocontrol Technologies GmbH & Co, Germany). The dielectric properties were tested in the frequency

range 10^{-1} to 10^6 Hz at room temperature and with a low electrical field (about 1 V mm^{-1}). The thickness of the tested samples was 1 mm, and the diameter was 20 mm.

2.5 Scanning electron microscopy (SEM) observation

The morphology of the samples was investigated with an FEI Quanta 200 ESEM (Thermo Fisher Scientific, USA) scanning electron microscope. This system was equipped with a field emission gun. The samples were coated with an approximately 5 nm thick gold layer by means of a sputter coater (Q150R ES, Quorum Technologies, UK) under vacuum condition and a current of 40 mA for 10 seconds before test. The sample's cross-section was detected with a secondary electron detector (SE) for an incident electron beam of spot 3.5 accelerated to 10 k eV.

2.6 Voltage response test platform

A voltage response test platform was designed and set up to study the sensing performance of the fabricated BmimSbF₆ loaded pressure sensor. This platform consists of a personal computer (PC)-controlled wave-maker and a signal acquisition system. A control signal was generated via National Instruments' LabVIEW software, and then the signal was recorded by an Asahi Seisakusyo APD-050FCA controller. A small force stimulus was generated by the wave-maker (SL-0505, Asahi Seisakusyo) and applied to the top surface of the pressure sensor via a shaker rod. The force that was applied to the pressure sensor was measured by a load cell (ZNLBM-1KG Micro Load cell, Bengbu Zongnuo Sensor Inc.). The voltage data generated by the pressure sensor was recorded using a National Instruments PCIe-6351 data acquisition board. The sensitivity (S) was calculated based on the following equation:

$$S = \frac{d(\Delta V)}{dF} \quad (1)$$

where ΔV is the relative voltage change, F is the applied force^[27].

3. RESULTS AND DISCUSSION

3.1 Morphology of ionic liquid in elastomer films

In order to verify that the BmimSbF₆ is loaded successfully into the silicone elastomer, the FT-IR spectra are investigated. As illustrated in Figure 2, all films loaded with BmimSbF₆ show distinctive C-H and N-H stretching peaks at approximately 3200 cm^{-1} and 3180 cm^{-1} , respectively, and all of the films display the characteristic peaks of the imidazole ring at 1580 cm^{-1} . The imidazole peaks of the material with 70 phr BmimSbF₆ are sharper than those of the materials with low amount of BmimSbF₆, thereby confirming the higher amount of BmimSbF₆ presents in this material.

Figure 2 FT-IR spectra of films with different amounts of BmimSbF₆ loaded.

The morphology of the samples was investigated by using scanning electron microscope (SEM). As shown in Figure 3, regenerated keratin powders (white dots) are evenly dispersed on the surface of the elastomer films. It can be clearly observed that BmimSbF₆ droplets (black dots) are present in the films with BmimSbF₆ loadings, and the higher loadings of the BmimSbF₆, the more BmimSbF₆ droplets, as would also be expected. Furthermore, the SEM images show that the BmimSbF₆ is well dispersed in the silicone network.

Figure 3 SEM images of elastomers, (a) pure PDMS, (b) PDMS with 10 phr BmimSbF₆ loaded, and (c) PDMS with 70 phr BmimSbF₆ loaded.

3.2 Effect of the BmimSbF₆ amount on the dielectric properties

The dielectric properties of the films with different BmimSbF₆ loadings were investigated as a function of frequency. Unlike conventional PDMS elastomers, these films with BmimSbF₆ exhibit high permittivity, as shown in Figure 4. The dielectric permittivity of elastomers with BmimSbF₆ increased in line with the increased amount of BmimSbF₆, while the dielectric permittivity of the elastomer with 70 phr BmimSbF₆ has the highest permittivity. The dielectric permittivity at 10⁶ Hz increased from 2.24 for the pure film up to 10.3 for the sample with 70 phr BmimSbF₆. In other words, the dielectric permittivity of an elastomer with 70 phr BmimSbF₆ has a 4.6x increase compared to the pure PDMS elastomer. The storage permittivity at 10⁶ Hz and 10⁻¹ Hz are also presented in Table 1.

Figure 4 Storage permittivity of films with different amounts of BmimSbF₆ at room temperature.

Table 1 Dielectric properties of the investigated elastomer composites at room temperature.

Samples	Pure	1 phr BmimSbF ₆	5 phr BmimSbF ₆	10 phr BmimSbF ₆	30 phr BmimSbF ₆	50 phr BmimSbF ₆	70 phr BmimSbF ₆
Permittivity at 10 ⁶ Hz	2.24	2.87	3.15	3.90	6.05	8.10	10.34
Permittivity at 10 ⁻¹ Hz	2.56	3.57	4.00	5.61	8.24	13.70	20.73

3.3 Effect of the BmimSbF₆ amount on the response of the voltage

To evaluate the performance of the pressure sensors, a dedicated test bench consisting of a personal computer (PC)-controlled wave-maker and a signal acquisition system was designed. To describe the effect of the amount of BmimSbF₆ on the performance of the films as pressure sensors, the voltage change curves upon a constant force 1.1 N for the different elastomers are compared in Figure 5. The developed sensors showed tunable voltage change depending on the loading of BmimSbF₆. The voltage change of the films increased with increasing loadings of the BmimSbF₆. The pure PDMS does not respond to force changes (Fig. 5b), and the film with 70 phr BmimSbF₆ exhibited the highest voltage change (Fig. 5d). This behavior indicated that the addition of BmimSbF₆ has a positive effect on the response of elastomer film to force.

Figure 5 Voltage changes of films with different amounts of BmimSbF₆ upon a constant force 1.1 N, (a) Applied force, (b) Voltage changes of film without BmimSbF₆, (c) Voltage changes of film with 50 phr BmimSbF₆, and (d) Voltage changes of film with 70 phr BmimSbF₆.

3.4 Sensitivity of the film with BmimSbF₆ loaded

The sensitivity of the pressure sensor with 70 phr BmimSbF₆ loaded was calculated based on equation (1). As shown in Figure 6, the pressure sensor with 70 phr BmimSbF₆ has an obvious voltage change when a small force is applied. It also can be seen that the sensitivity of the pressure sensor with the thickness of 800 μm is 1.88 mV/N. Based on previous report, the thickness of the elastomer film has effect on the sensitivity of the pressure sensor^[36]. Therefore, the effect of the thickness of the pressure sensor on the sensitivity was also investigated by calculating the sensitivity of the materials with different thicknesses. In Figure 6, the sensitivity of the pressure sensor with a thickness of 160 μm is 2.64 mV/N, which is much higher than that of the pressure sensor with a thickness of 800 μm . This result indicated, as expected, that the thickness of the pressure sensor plays an important role on the sensitivity of the pressure sensor, which is consistent with the conclusions drawn from the literature^[36].

Figure 6 Sensitivity of film with 70 phr BmimSbF₆ upon varying forces and thicknesses.

4. CONCLUSION

In summary, a novel high-performance of pressure sensor was fabricated by incorporating the ionic liquid BmimSbF₆ into a silicone elastomer, and the regenerated keratin from wool was added as solid fillers to strengthen the elastomer. The effect of the loading of BmimSbF₆ on the performance of the elastomer composites as pressure sensors was also investigated. Dielectric permittivity increased in line with increased amount of BmimSbF₆, while the dielectric permittivity of the film with 70 phr BmimSbF₆ was 4.6x higher than that of the pristine silicone elastomer. The developed sensor showed tunable voltage change, depending on the loaded amount of BmimSbF₆, and the optimized pressure sensor exhibited excellent force sensing characteristics. Especially, the sensitivity of the pressure sensor with 70 phr BmimSbF₆ is 2.64 mV/N. Moreover, the thickness of the pressure sensor also effects the sensitivity, and thinner film showing higher sensitivity for the pressure sensor. The excellent performance and facile process suggested the potential application of the novel pressure sensor in the healthcare and smart wearable devices.

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CONFLICT OF INTEREST

The authors declare no conflict of interest.

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