Highly Sensitive and Optically Transparent Resistive Pressure Sensors Based on a Graphene/Polyaniline-Embedded PVB Film

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***Abstract*— The development of a facile, low-temperature, and low-cost method to fabricate highly sensitive and optically transparent resistive pressure sensors remains a challenge. In this paper, a graphene/polyaniline-embedded polyvinyl butyral (GPANI-PVB) composite film is employed as the active layer in a resistive pressure sensor; this film is ultrathin, highly sensitive, optically transparent, anisotropically conductive, highly durable, highly flexible, and bending insensitive. A flexible touch panel based on the GPANI-PVB composite film is fabricated using a facile process, which supports multitouch and multilevel pressure detection. The GPANI-PVB composite film offers great potential for applications in flexible and transparent interactive electronic devices.**

***Index Terms*— Flexible touch panel, graphene/polyaniline-embedded polyvinyl butyral (GPANI-PVB) composite, high sensitivity, high transparency, resistive pressure sensor.**

I. Introduction

PRESSURE sensors, including capacitive, piezoelectric, and resistive types, are widely used in displays, robotics, and prosthetics [1]–[3]. For application in displays, pressure-sensitive touch panels serve as an intuitive interactive interface and provide users with touch depth perception as well as conventional contact positioning on the screen surface. Current pressure-sensitive touch technologies applied in smart phones mainly make the use of capacitive pressure sensors, such as 3-D touch proposed by some suppliers. With a pressure applied, such touch panels can detect tiny increase in capacitance due to the reduction in distance between sensor electrodes [4]. However, they have issues of low sensitivity due to large parasitic capacitance, high susceptibility to electronic noises [3], and possible sensitivity variation due to bending for flexible display applications. Pressure-sensitive touch panels based on self-powered piezoelectric pressure sensors [5] were also reported. However, they usually cannot support the detection of static pressure and multipress, and need extra pressure sensors in addition to conventional capacitive touch sensors. Therefore, capacitive and piezoelectric pressure sensors are possibly not the best candidates for flexible pressure-sensitive touch panels.

Compared with capacitive [4], [6]–[9] and piezoelectric types [5], [10]–[12], resistive pressure sensors respond to applied pressure with a change in the resistance of the active layer. Composites consisting of insulating elastomers and conductive fillers, such as carbon nanotubes [13], [14] and silver nanowires [15], have been developed to fabricate resistive pressure sensors. Recently, microstructures, such as porous [13], pyramidal [16], and triangular [17] morphologies, have also been introduced into composites to improve the sensitivity of pressure sensors. Basically, these sensors are based on two mechanisms [3]. One uses piezoresistive elastomers whose resistivity changes with the applied pressure due to the rearrangement of conductive fillers in elastomers [13], [18], [19]. Such pressure sensors usually suffer from low sensitivity, high hysteresis, and temperature dependence of pressure sensitivity. The second mechanism achieves high sensitivity by fabricating microstructures on elastomers or electrodes to increase the relative change in contact resistance with a pressure applied [16], [17]. However, because of the abundance of conductive fillers, these two kinds of sensors usually have low transparency, hindering their application in touch panels. Recently, a transparent bending-insensitive resistive pressure sensor fabricated using an electrospinning process was proposed [20]. Although high transmittance (more than 90% in the visible-to-infrared wavelength region) can be obtained, the pressure sensor exhibits high sensitivity only in the pressure regime of less than 1 kPa, which is not applicable for pressure-sensitive touch panels (sustaining

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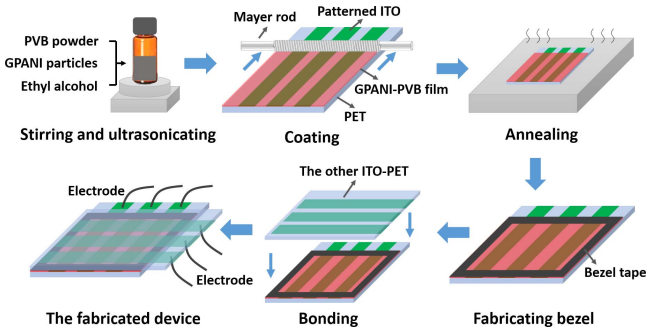

Fig. 1. Fabrication process of the GPANI-PVB composite film and the flexible touch panel.

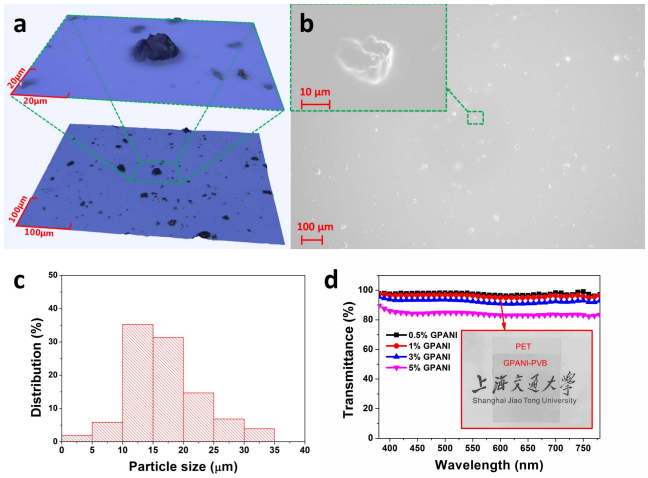

Fig. 2. Fabricated GPANI-PVB composite film. (a) and (b) Surface morphology of the composite film characterized by a 3-D optical profile and an FE-SEM, respectively. (c) Size distribution of the GPANI particles. Film thickness: 3 μm; concentration of GPANI in PVB: 1 wt%. (d) Transmittance of the composite film with different concentrations of GPANI in PVB. Film thickness: 3 μm. Inset: composite film coated on a PET substrate with a GPANI concentration of 1 wt%.

pressures of 0–100 kPa). Moreover, the fabrication process and required equipment are complicated and challenging. Recently, a flexible and transparent resistive pressure sensor array based on percolative graphene films was reported for touch panels [21]. However, because its pressure-sensitive feature relies on the stretchability of the graphene films, the sensor array may suffer from sensitivity variation due to bending. Moreover, its fabrication process is complicated and time consuming.

Therefore, we developed a resistive pressure sensor based on a graphene/polyaniline-embedded polyvinyl butyral (GPANI-PVB) film, which is ultrathin, highly sensitive, optically transparent, anisotropically conductive, highly durable, highly flexible, and bending insensitive. The fabrication process is facile, low temperature, and all solution based. Therefore, the composite film is suitable for manufacturing low-cost and large-area touch panels. The range of variation in contact resistance and pressure sensitivity can be readily controlled by the concentration of GPANI in PVB and the thickness of the composite film. Based on the composite film, a flexible pressure-sensitive touch panel was fabricated, which supports multitouch and multilevel pressure detection.

II. Fabrication Process

Fig. 1 shows the fabrication process of the GPANI-PVB composite film. A mixture of PVB powder, GPANI microparticles, and absolute ethyl alcohol was stirred for 4 h and subsequently ultrasonicated for another 30 min at room temperature. Because PVB is soluble in ethyl alcohol and GPANI particles can be well dispersed in a PVB-ethyl alcohol solution [22], the mixture finally became a homogenous solution. The resulting solution was coated on a patterned indium tin oxide-coated polyethylene terephthalate (ITO-PET) layer by a Mayer rod. ITO and PET served as the electrodes and substrate of pressure sensors, respectively, which can be replaced with other suitable materials. The thickness of the wet composite film (with ethyl alcohol) was approximately 68.6 μm. After the composite film was annealed at 80 °C for 15 min, the ethyl alcohol evaporated and the GPANI-embedded PVB film was fabricated. The thickness of the dry composite film (without ethyl alcohol) was determined by the concentration of PVB in ethyl alcohol. The thicknesses measured by a surface profiler were 1.8, 2.7, and 3 μm for PVB concentrations of 2.5, 5, and 10 wt%, respectively. The surface morphology of the composite film was characterized by a 3-D optical profiler [Fig. 2(a)] and a field-emission scanning electron microscope [FE-SEM, Fig. 2(b)]. The size of the GPANI particles was mainly in the range of 10–20 μm [Fig. 2(c)], and at different positions of the PVB film, the GPANI particles showed high uniformity.

Because the size of the GPANI particles was several times larger than the thickness of the PVB film, the majority of the GPANI particles were embedded and even penetrated into the PVB film. Therefore, the composite film was conductive in the direction perpendicular to the film surface. Due to the discrete distribution of GPANI microparticles in the PVB film, the composite film was nonconductive in the direction parallel to the film surface. The anisotropic conductivity of the composite film makes it a good candidate for application in touch panels because it will not electrically connect adjacent electrodes of the same layer.

Because of the high intrinsic transmittance of the PVB film and the discrete distribution of GPANI microparticles in the PVB film, the GPANI-PVB composite film displayed high transparency [Fig. 2(d)], which can be controlled by the concentration of GPANI in the PVB film. For a GPANI concentration of less than 1 wt% in the PVB film, the composite film exhibited a uniform transmittance of more than 96% throughout the visible wavelength region (380–780 nm). This high optical transparency is a useful feature for integration of the composite film in displays. It should be noted that the size of the GPANI particles must be well controlled to be invisible to human eyes. According to Rayleigh criterion, with a viewing distance of larger than 18 cm, which is always the case for a touch panel, human eyes cannot make out particles with a size of less than 25 μm (assuming that the minimum resolution angle of human eyes is 0.0077°), which implies that the GPANI-PVB composite film is adequate for application in touch panels.

By bonding a second ITO-PET layer onto the GPANI-PVB film-covered ITO-PET by bezel tape with the ITO sides face to face, a flexible pressure-sensitive touch panel based on the composite film was fabricated. The ITO layers had been previously patterned with parallel strip electrodes with a width of 4.5 mm and a gap of 0.5 mm. The two layers of strip electrodes were arranged perpendicularly, forming a 10×10 resistive pressure sensor array. As a comparison, a touch panel with the same two ITO-PET layers bonded together but without a GPANI-PVB composite film in between was also fabricated.

III. Performance of the Resistive Pressure Sensors

The bezel tape induced an ultrathin air gap between the two layers of ITO-PET. With no pressure applied, the two layers of strip electrodes were electrically insulated because of the air gap. If a finger-like rubber tip touched the panel, the top ITO layer began to contact the GPANI-PVB film on the bottom ITO layer, and conductive paths were built up. Thus, resistance could be detected at the position where the touch occurred. As the applied pressure increased from a gentle touch to a hard press, the detected resistance decreased due to the increase in contact area and the decrease in contact resistance between the GPANI particles and the two ITO layers.

Fig. 3(a) illustrates the relative change in resistance ($\Delta R/R$) as a function of the applied pressure for different concentrations of GPANI in PVB. To initialize the contact between the GPANI particles and the ITO layers, the resistance detected at a pressure of 3 kPa was defined as the initial resistance. Although increasing the GPANI concentration in PVB resulted in decreased resistance, the relative variation in resistance (corresponding to the sensitivity) remained stable.

We propose a simplified model to explain the influence of GPANI concentration on sensitivity [Fig. 3(b)]. Assuming that the GPANI particles are balls of the same size, the resistance of the pressure sensor (R) under pressure can be given by the following equation:

$$R = \frac{R_c}{A\sigma} \tag{1}$$

where R_c denotes the equivalent contact resistance resulting between one GPANI particle and the ITO layer, A denotes the contact area, and σ denotes the areal density of GPANI particles in the PVB film. As the applied pressure increases, R_c decreases and A increases, reducing the resistance of the pressure sensor. The relative variation in resistance can be expressed as

$$\frac{\Delta R}{R_0} = \frac{R_0 - R}{R_0} = \frac{\frac{R_{c0}}{A_0\sigma} - \frac{R_c}{A\sigma}}{\frac{R_{c0}}{A_0\sigma}} = 1 - \frac{R_c}{R_{c0}} \cdot \frac{A_0}{A} \tag{2}$$

where R_{c0} and A_0 denote the equivalent contact resistance and the contact area for the initial state. The sensitivity of the pressure sensor (S) can be defined as

$$S = \frac{\Delta R}{R_0} \cdot \frac{1}{\Delta P} = \left(1 - \frac{R_c}{R_{c0}} \cdot \frac{A_0}{A}\right) \cdot \frac{1}{\Delta P} \tag{3}$$

where ΔP denotes the change in applied pressure. Equation (3) implies that the concentration of GPANI in PVB (σ) has no effect on sensitivity. Therefore, we can achieve low power consumption for resistance measurement and meanwhile retain the high sensitivity by fabricating the pressure sensors with a low concentration of GPANI in PVB.

Based on (2), in the pressure range of 0–20 kPa, the resistance of the pressure sensor decreases sharply due to the rapid increase in contact area. In the pressure range of 20–100 kPa, the contact area remains constant (4.5 mm $\times$ 4.5 mm), and the resistance of the pressure sensor decreases more gradually because of the slow decrease in equivalent contact resistance. For a given change in contact area, the main factor affecting the sensitivity is the equivalent contact resistance between the GPANI particle and the ITO layer, which can be tuned by the size of the GPANI particles and the thickness of the composite film [Fig. 3(c)]. As the thickness of the composite film increases, the resistance of the pressure sensor increases and the sensitivity decreases, which is attributed to the fact that a thinner film has a larger relative change in equivalent contact resistance.

The pressure sensors also exhibit high durability [Fig. 3(d)]. After 7000 cycles of applying and releasing a hard press (corresponding to a pressure of approximately 100 kPa) by a finger, the resistance of the pressure sensor as a function of the applied pressure (0–100 kPa) remained stable.

Due to the influence of the bezel tape and our laboratory fabrication process, the initial resistance at different locations of the sensor array was not strictly constant and had a maximum deviation of 78.5% from the average value at a pressure of 3 kPa [Fig. 3(e)]. This will not be an issue if we employ an appropriate driving circuit which enables the measurement of large resistance variation of more than 10 times and the recording of initial resistance of the whole sensor array (see more details in Section V). Moreover, the relative change in resistance as a function of the applied pressure, which defines pressure sensitivity, was nearly the same (maximum deviation: 7.6%) [Fig. 3(f)], implying high uniformity. By well controlling the size of the GPANI particles, the variations in resistance and sensitivity may be further reduced. The high uniformity of pressure sensitivity implies that even for a large-area touch panel, the GPANI-PVB composite film is also applicable.

The pressure sensors also show a low hysteresis of less than 8.5% [Fig. 3(g)], which is lower than the hysteresis reported in [16] and [17]. The hysteresis was defined as the maximum discrepancy between the forward and backward curves divided by the full-scale resistance change. Resistive pressure sensors in the previous work usually employ elastomeric materials with microstructures as active layers and electrodes, which may suffer from viscoelastic creep due to irreversible entanglement of polymer chains with a pressure applied [9], resulting in high hysteresis. However, the pressure sensor based on the GPANI-PVB composite film relies on the change in contact area and contact resistance resulting between the more incompressible ITO-PET film and composite film [Fig. 3(b)], which yields a more reversible contact process, resulting in lower hysteresis.

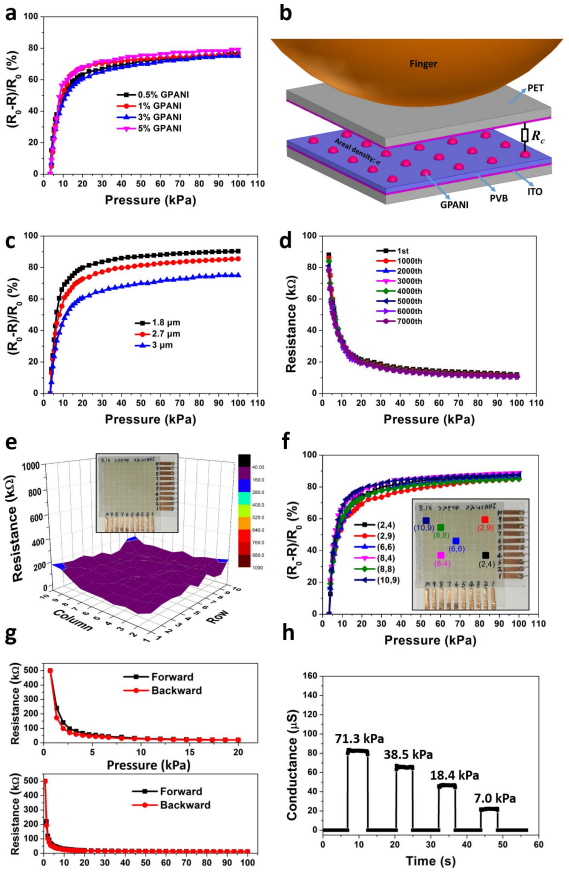

Fig. 3. Performance of the fabricated pressure sensors as a function of applied pressure. (a) Influence of GPANI concentration in PVB on sensitivity. Sensor position: (6, 6) in the array; film thickness: 3 μm; initial resistances: 660, 200, 140, and 110 kΩ for GPANI concentrations of 0.5, 1, 3, and 5 wt%, respectively. (b) Simplified model to explain the working mechanism of the pressure sensors. R_C denotes the equivalent contact resistance resulting between one GPANI particle and the ITO layer. (c) Influence of film thickness on sensitivity. Sensor position: (6, 6) in the array; concentration of GPANI in PVB: 3 wt%; initial resistances: 88, 90, and 140 kΩ for film thicknesses of 1.8, 2.7, and 3 μm, respectively. (d) Stability and durability of the pressure sensors. A test was performed after every 1000 cycles of applying and releasing a hard press by a finger. (e) Resistance distribution of the sensor array at a pressure of 3 kPa. (f) Uniformity of the sensor array. Initial resistances: 150, 140, 90, 140, 94, and 180 kΩ for pressure sensor positions of (2, 4), (2, 9), (6, 6), (8, 4), (8, 8), and (10, 9) in the array, respectively, which represent key positions over the array. (g) Hysteresis of the pressure sensors. Two different loops (0–20–0 kPa and 0–100–0 kPa) were measured. (h) Response of the pressure sensors to different pressure levels. To include the extremely high resistance values with no pressure applied, the y-coordinate represents the conductance of the pressure sensors. For (d)–(h), film thickness: 2.7 μm; concentration of GPANI in PVB: 3 wt%. For (d)(g)(h), sensor position: (6, 6) in the array.

The response of the pressure sensors to different pressure levels was shown in Fig. 3(h). The test was performed by quickly applying and releasing a finger press with different pressure levels on the pressure sensors. The response and recovery time of the pressure sensors were both less than 57 ms, which was already beyond the limit of our measurement system.

Compared with resistive pressure sensors in [16], [17], and [20], the proposed pressure sensor may not have such a high sensitivity in ultralow pressure regime (less than 1 kPa), but its high sensitivity in the pressure regime of 0–100 kPa (gentle touch to hard press), high transparency,

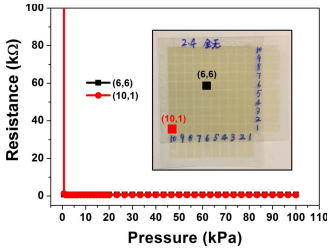

Fig. 4. Resistance between the two layers of strip electrodes as a function of applied pressure for the touch panel without the GPANI-PVB composite film in between.

ultrathin thickness, high stability and durability, low hysteresis, fast response (not comparable to previous work under current measurement conditions), and simplified fabrication process due to the absence of microstructures makes the pressure sensor a good candidate for application in touch panels.

IV. Performance of the Flexible Touch Panel

By directly bonding two layers of patterned ITO-PET films together without the GPANI-PVB composite film in between, a conventional resistive touch panel supporting multitouch detection was fabricated (Fig. 4) [23]. With no pressure applied, the ultrathin air gap induced by the bezel tape also results in electrical insulation between the two layers of trip electrodes. However, once a very gentle touch (less than 1 kPa) occurs on the touch panel, the resistance between the two layers of strip electrodes will sharply reduce to a very low value (several hundreds of ohms), which is mainly the resistance of ITO electrode; even if the applied pressure increases, the resistance remains nearly a constant. Therefore, the resistive touch panel without the composite film can only function as a switch and cannot detect pressure levels. Moreover, due to the ultralow ON-state resistance, the touch panel has the issue of high power consumption.

Sensor array in the touch panel with the GPANI-PVB composite film in between enables the detection of both touch position and applied pressure at the same time. Due to its high stability, the resistance of each pressure sensor in the array at a pressure of 3 kPa was recorded and set as the threshold. When a finger gently touched a certain pressure sensor, only the resistance of the touched pressure sensor was below the threshold, and the touch position was precisely identified [Fig. 5(a)]. When the finger pressed the pressure sensor hard, the resistances of the surrounding eight pressure sensors were also below the threshold because of the increase in contact area, but the relative variations in resistance did not exceed that of the central pressure sensor [Fig. 5(b)], which can precisely derive the applied pressure. Therefore, the touch panel supported multilevel pressure detection [Fig. 5(e)]. The test was performed by manually manipulating a pressure meter to successively apply multilevel pressure on the touch panel via a finger-like rubber tip. The pressure-sensitive touch panel also supported multitouch detection [Fig. 5(c) and (d)] using the positions where the peaks of the relative variation in resistance were lying. Fig. 5(a)–(d) also demonstrates from another perspective that the variation in initial resistance is not an issue due to the high uniformity of pressure sensitivity.

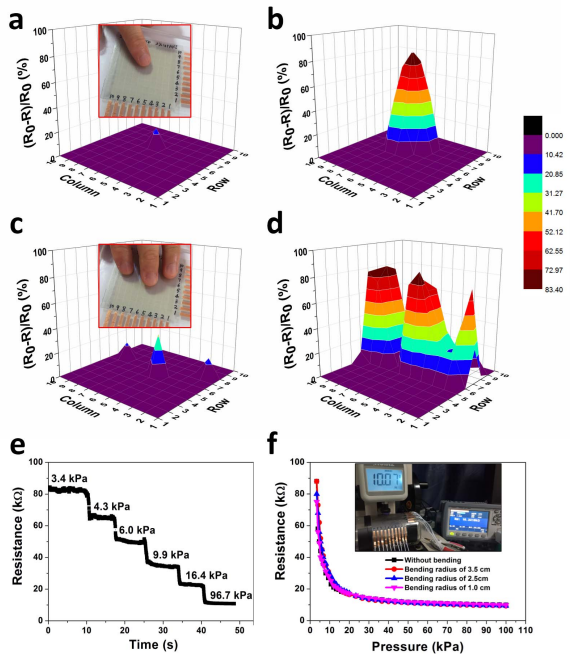

Fig. 5. Performance of the pressure-sensitive touch panel. (a) Gentle single touch. (b) Hard single press. (c) Gentle multitouch. (d) Hard multipress. (e) Touch panel supports multilevel pressure detection. (f) Bending tests. Film thickness: 2.7 μm. Concentration of GPANI in PVB: 3 wt%.

Bending tests with the touch panel attached to a curved rigid substrate were also conducted [Fig. 5(f)]. Even at a bending radius of 1 cm, the sensitivity of the touch panel remained stable due to the identical bending deformation of the two ITO-PET layers, which implies that the touch panel is bending insensitive and of high flexibility. However, because of the brittleness of ITO, the resistance of ITO-PET film has a dramatical increase with a bending radius of less than 7.5 mm, reducing the sensitivity of the touch panel. Using an ITO-replacement material of higher flexibility, such as silver nanowires and metal mesh [24]–[27], the bending insensitivity, and flexibility of the touch panel may be improved.

Compared with conventional resistive touch panel without the composite film, the proposed touch panel can not only support multilevel pressure detection, but also reduce the power consumption due to the controllable contact resistance (in the level of tens to hundreds of kilohms). Compared with current pressure-sensitive touch technologies [4], [5], [21], the proposed touch panel combines the detection of both touch position and pressure, and has high noise immunity due to resistance measurement, enabling its application in harsh circumstances, such as under water or in intense electromagnetic fields; it is also bending insensitive and of high flexibility, and can support multitouch and multilevel pressure detection.

V. Discussion

For practical use, the influence of temperature and humidity on the performance of the touch panel should be considered. Although humidity has a negligible effect on the performance [Fig. 6(a)], an increase in temperature results in a nearly linear reduction in the resistance of the pressure

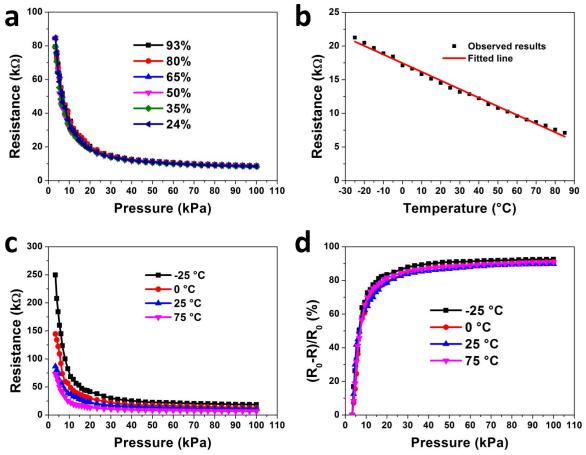

Fig. 6. Influence of humidity and temperature on the performance of the touch panel. (a) Change in sensor resistance as a function of applied pressure with different relative humidities. Temperature: 22 °C. (b) Change in sensor resistance as a function of temperature with an applied pressure of 50 kPa. (c) Change in sensor resistance as a function of applied pressure with different temperatures. (d) Relative change in sensor resistance as a function of applied pressure with different temperatures. Relative humidity: 45%. Sensor position: (6, 6) in the array; film thickness: 2.7 μm; concentration of GPANI in PVB: 3 wt%.

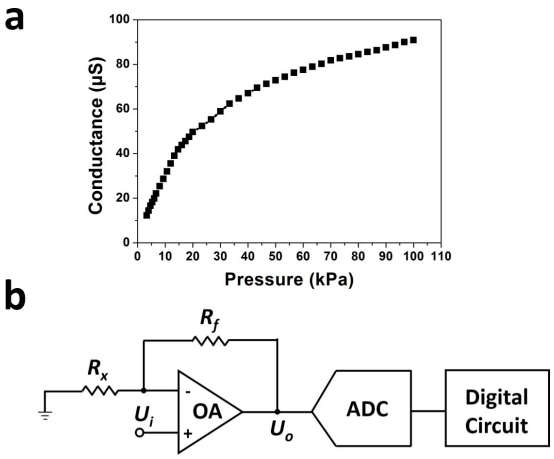

Fig. 7. Solution to the relatively low sensitivity of the pressure sensors in the pressure range of 20–100 kPa. (a) Conductance change of the pressure sensors as a function of applied pressure. (b) Driving circuit transforming the measurement of resistance change into output voltage change.

sensors [Fig. 6(b) and (c)]. The resistance variation may be attributed to the fact that an increase in temperature results in a reduction in the equivalent contact resistance between the GPANI particle and the ITO layer, considering that only a slight increase (2.1%) in the resistance of ITO-PET film is induced by an increase in temperature from −25 °C to 85 °C. However, the relative change in resistance (representing the sensitivity) remains stable [Fig. 6(d)]. Therefore, the temperature-induced resistance variation can be calibrated if we fabricate a similar sensor based on the GPANI-PVB film with a given pressure (such as 3 kPa) in the touch panel and measure its resistance as the threshold value.

Fig. 3 shows that in the pressure range of 20–100 kPa, the pressure sensors have a relatively low sensitivity due to the nearly constant contact area and the slow decrease

in equivalent contact resistance. However, situation can be much improved if we use the change in conductance, which is the reciprocal of resistance, to identify the applied pressure [Fig. 7(a)]. The improvement is due to the approximately inverse proportional relationship between the resistance of pressure sensors and the applied pressure (20–100 kPa) (Fig. 3). Fig. 7(b) illustrates a measurement circuit making use of the conductance change. According to the principle of noninverting operational amplifier (OA) with a negative feedback, the output voltage (U_o) can be expressed as

$$U_o = U_i \left(1 + \frac{R_f}{R_x}\right) \tag{4}$$

where U_i denotes the input voltage, R_f denotes the feedback resistance, and R_x denotes the resistance of pressure sensors to be measured. With no pressure applied, R_x is infinite and $U_o = U_i$ and as the applied pressure increases, R_x decreases and U_o increases. $R_x = R_f$ yields $U_o = 2U_i$ and $R_x = R_f/N$ under a high pressure yields $U_o = (N + 1)U_i$. Therefore, the driving circuit transforms the measurement of resistance change into output voltage change which is a linear function of the conductance of pressure sensors, improving the pressure sensitivity in the pressure range of 20–100 kPa. Moreover, the change in output voltage can be conveniently processed by back-end digital circuits via an analog-to-digital converter.

VI. Conclusion

The high sensitivity, high transparency, high flexibility, high uniformity, low hysteresis, fast response time, anisotropic conductivity, and bending insensitivity of the GPANI-PVB composite film make it a good candidate for fabricating flexible interactive electronic devices, such as flexible displays, transparent electronic skins, and health monitors. These devices can provide users or robotics with information regarding both the contact position and the applied pressure. Because these devices work by detecting resistance instead of capacitance, the influence of electromagnetic interference on measurement results is negligible, implying that these devices will show high reliability and applicability under harsh circumstances, such as under water or in intense electromagnetic fields. The facile, low-temperature, and all-solution-based method also enables the fabrication of low-cost, large-area interactive devices.

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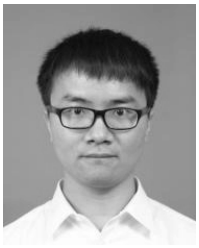

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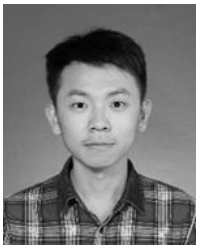

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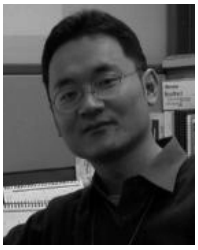

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