

Received September 10, 2019, accepted September 23, 2019, date of publication September 25, 2019, date of current version October 8, 2019.

Digital Object Identifier 10.1109/ACCESS.2019.2943736

Electrospun Ionic Nanofiber Membrane-Based Fast and Highly Sensitive Capacitive Pressure Sensor

XIAOFENG YANG¹, YISHOU WANG, AND XINLIN QING¹

School of Aerospace Engineering, Xiamen University, Xiamen 361005, China

Corresponding author: Xinlin Qing (xinlinqing@xmu.edu.cn)

This work was supported in part by the National Natural Science Foundation of China under Grant 11772279 and Grant 51975494, and in part by the Fundamental Research Funds for the Central Universities under Grant 20720180120.

ABSTRACT Flexible pressure sensor has been widely used in different industries, but there are still some challenges for the fabrication of sensor with a superb sensitivity and large sensing range. In this paper, a flexible capacitive pressure sensor based on electrospun ionic nanofiber membrane is developed. The sensor contains an ionic nanofiber dielectric layer and two indium tin oxide polyethylene terephthalate (ITO-PET) films coated on the top and bottom of the dielectric layer, respectively. The electrical double capacitors are formed at the interface between the ionic nanofiber membrane and the electrodes. When external pressure is applied on the sensor, the distance between two electrodes decreases, which causes the increase of the ionization degree of ionic liquid as well as the effective contact area between the two electrodes and ionic nanofiber dielectric layer. Above changes lead to a rapid increase of the capacitance of the sensor. The sensing principles and the theoretical models of the sensor are systematically studied. Comparing to traditional capacitive pressure sensors, the developed sensor provides a high sensitivity up to 78.54 nF/kPa and a rapid dynamic response (16 ms) for pressure measurement. Experiments are also conducted to investigate the influence of the thickness and the bending radius of ionic nanofiber membrane as well as temperature and humidity of the environment.

INDEX TERMS Flexible, sensor, pressure measurement, ionic membrane, capacitor.

I. INTRODUCTION

Recently, flexible, bendable and stretchable electronic devices have gained exponential attentions for potential applications in wearable and foldable devices [1]–[3]. Among the various types of electronic sensors, pressure sensors are one of the most studied elements due to their potential in variety of applications including foldable structures [4], [5], soft robots [6] electronic skins [7], [8] and biological sensing [9], [10]. Various sensing mechanisms have demonstrated the possibility of fabricated highly sensitive pressure sensors, including organic field effect transistor (OFET) [11], [12], piezoresistance [13], [14], piezoelectricity [15], [16] and capacitance [17], [18]. Among them, the high sensitivity and low cost are desired in this field. Piezoelectric sensors can achieve high sensitivity and self-power based on the piezoelectric effect, but they fail to detect static

pressures [19], [20]. Piezoresistive sensors, which transduce the pressure applied on the sensor to resistance signal, have been widely used due to their feasible production, low cost and easy signal analysis [21], [22]. However, the sensing materials of piezoresistive sensor are sensitive and vulnerable to environment factors [23], [24]. Owing to the high electrical sensitivity, low power consumption, and simple construction, capacitive pressure sensor has been more widely applied [17].

Conventionally, capacitive pressure sensor is formed by a pair of parallel plates. When pressure is applied on the upper plate, it deforms and results in the change of the distance between two plates of the capacitive sensor [25], [26]. However, the traditional capacitive pressure sensors are rigid, and thus, their low sensitivity is unsuitable for flexible devices. Recently, several flexible capacitive pressure sensors based on the flexible dielectric layer have been proposed. To improve the sensitivity, several microstructures, such as Pyramid [27], [28] micropillar [29], [30] and hemisphere [31], [32] shaped dielectric layer, have been integrated

The associate editor coordinating the review of this manuscript and approving it for publication was Yasar Amin¹.

to capacitive sensor to enhance their sensitivity. Although a high sensitivity can be obtained, it decreases rapidly as the applied pressure increases and the fabrication progress of the microstructure is very complicated. After that, porous elastomers have also been used for improving the sensitivity of the capacitive pressure sensor [33], [34], nevertheless, the sensitivity is still low. Ionic hydrogel with high ionic conductivity and outstanding mechanical properties has been used as the dielectric layer of capacitive sensor, which can form an electrical double capacitor in the surface of the electrode [35]–[37]. However, the hydrogel will lose water under compression and it is hard to deform under low external pressure.

Electrospinning is widely used for electrostatic fiber production which makes polymer solution to fibers by electrical force [38], [39]. During the electrospinning process, the as-prepared solution is sprayed through the syringe spinneret, and a Taylor cone is formed at the tip of the pendant droplets due to the high electrostatic forces between the tip and the collector. Under the critical voltage, electrostatic forces overcome the surface tension of the solution, and cause an electrified solution jet. The nanofibers are randomly entangled and stacked on the collector to form a porous membrane [40]–[43]. This technology has been broadly used to produce natural nanofibers and fabrics with controllable porous structure. Owing to the high specific area and low Young's modulus of the nanofiber structure, the electrostatic fiber membrane has been broadly used in sensing device [44]. To improve conductivity of the fiber membrane, some nano-conductive materials including the carbon nanotube, graphene and metallic nanowires have been added into the polymer solution to electrospun composite nanofiber film. However, because of a strong van der Waals, it is hard to disperse the conductive additions uniformly. Besides, the high contact resistance between conductive additions critically suppresses the high conductivity of nanofiber membrane. Recently, an ionic membrane has been used as the dielectric layer of capacitive sensor with high sensitivity [45]–[47]. However, previous works mainly focus on wearable electronics for monitoring physiological information. Moreover, the sensing mechanism and the influence of the environment factors as well as geometrical parameters have not been reported yet.

In this work, a flexible capacitive pressure sensor with high sensitivity and rapid dynamic response is developed. The sensor consists of an electrospun 3D nanofiber membrane and two indium tin oxide polyethylene terephthalate (ITO-PET) films, which are coated on the top and bottom of the nanofiber membrane, respectively. The sensing mechanism which generates electric double layer capacitors in the surface of electrodes is systematically discussed. The performances of the sensors with different ionic liquid additions, different thicknesses of ionic nanofiber membrane are studied. In addition, the effect of temperature and humidity are systematically investigated. The application of a sensor

network used for measuring the spatial distribution and magnitude of the noncontact air pressure is also studied.

II. EXPERIMENT

A. MATERIAL AND APPARATUS

Polyvinylidene Fluoride (PVDF) with $M_w \sim 1,000,000$ g/mol were purchased from the Aladdin Reagent Co. Ltd. Ionic liquid 1-butyl-3-methylimidazolium bis-(trifluoromethyl)imide (99%, Shanghai Chengjie Chemical Co. Ltd.) was selected as the sensing material. Acetone (99.5%) and N, N-Dimethylformamide (DMF, 99.5%) was supplied by the Sinopharm Chemical Reagent Co., Ltd. Polyethylene terephthalate (PET) coated with 100 nm thick layer indium tin oxide (ITO) was purchased from the Shenzhen South China Xiangcheng Technology Co., Ltd. Copper wire (20P) was supplied by Lida optoelectronic hardware factory, china. The conductive epoxy (3706) was supplied by Shenzhen xinwei electronic material co. LTD.

Scanning electron microscopes (SEM, SUPRA 55, ZEISS, Germany) was used to observe the microstructure of ionic nanofibers. Fourier transform infrared spectrometer (Nicolet IS10 FTIR Spectrometer, Thermo Fisher Scientific Inc, USA) was used to determine the molecular structure of the ionic nanofiber. Capacitance measurements were performed by a WK6500B impedance analyzer (Shenzhen Wenke Electronics Co., Ltd.). A vibration exciter (Wuxi Shiao Technology Co., Ltd.) was used to provide a dynamic load with different frequencies applied on the sensor. A homemade electrospinning system is used to spin the composite nanofiber membrane. A computer-controlled step motor (VELMEX, INC.) was used to apply load on the sensor. A force gauge with resolution of 0.01N (HP-100, Dongguan Zhiqiu Precision Instrument Co., Ltd) was used to measure the load applied on the sensor. A constant temperature and humidity incubator (Shanghai Baixin Instrument and Equipment Factory) was used to control the testing temperature and humidity.

B. SENSOR DESIGN

The ionic nanofiber membrane was fabricated using electrospinning method. Firstly, the acetone was added in the DMF with the weight ratio at 2:3 to forming a mixed-solvent. Secondly, the PVDF powder was dissolved in a mixed-solvent (The concentration of the PVDF powder in the solution is 10%), and then the mixture was mechanically stirred for 10 hours at 150 r/min with magnetic stirring at room temperature to obtain a homogeneous solution. Thirdly, the ionic liquid was added to five divided mixture solution with weight ratios to PVDF at 0:1, 1:4, 1:2, 1:1 and 2:1 and then stir continuously for 2 hours at 150 r/min to make ionic liquid disperse uniformly. After mechanically stirring, the mixed solution is transparent, and no stratification and precipitation occur after 10 hours of standing, which can be confirmed that the ionic liquid is dispersed uniformly in the mixed solution. Fourthly, the nanofibers were deposited from

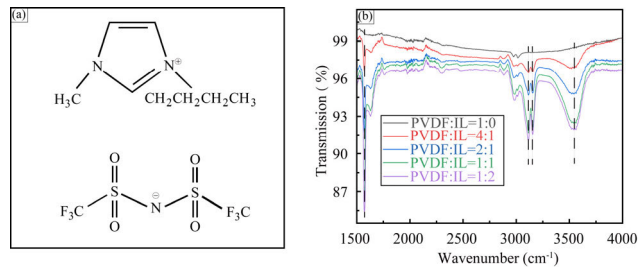


FIGURE 1. Characterization of morphologies of ionic nanofiber. (a) Chemical structure of the ionic liquid. (b) FTIR spectra of ionic nanofiber with different ionic liquid addition.

the as-prepared solution using electrospinning and further randomly entangled and stacked to form a porous membrane. Note that, owing to the high boiling point and low vapor pressure of ionic liquid, when organic solvent (DMF and acetone) evaporated in the electrospinning progress, the ionic liquid would be left in the nanofibers. In this work, the feed rate was 0.1 ml/h, the voltage was 15 KV and the distance between the collector which used for depositing nanofibers and syringe was 10 cm. In order to ensure the consistency of the experiments, the same experimental parameters were used in the electrospinning progress with different weight ratio of ionic liquid and PVDF. Finally, the ionic nanofiber membrane was dried for 2 hours at room temperature to volatilize the residual organic solvent and peeled off from the collector. The collector used in this work is the smooth aluminum foil. To prove the above theory, some molecular structures of the nanofibers with different ionic liquid addition have been tested by Fourier transform infrared spectrums (FTIR). The chemical structure of the ionic liquid is shown in Figure 1 (a). The FTIR spectra of the ionic nanofiber membrane in Figure 1(b) shows characteristic bands at 1580 cm^{-1} (C=N), 3120 cm^{-1} (N-H), 3160 cm^{-1} (N-H) and 3560 cm^{-1} (O-H), which does not appear in the spectra of the neat PVDF nanofiber membrane. The intensity ratio of the spectrums shows a significant increase with high level of ionic liquid addition. As shown in Figure 1(b), the 1580 cm^{-1} (C=N), 3120 cm^{-1} (N-H) and 3160 cm^{-1} (N-H) are the characteristic bands of the ionic liquid, and it demonstrates that the ionic liquid is absorbed in the nanofiber. The existence of 3560 cm^{-1} (O-H) is demonstrated as follows.

Since the oxygen in the air can dissolve in the ionic liquid and reduce via a one-electron transfer process to form the superoxide (O_2^-), the reverse reaction of oxygen being reduced to O_2^- is given by: [48]



Furthermore, the O_2^- is not stable in a humid atmosphere where reaction with water would suppress the reverse reaction because of its high capability for reduction. The reaction is expressed as: [48]

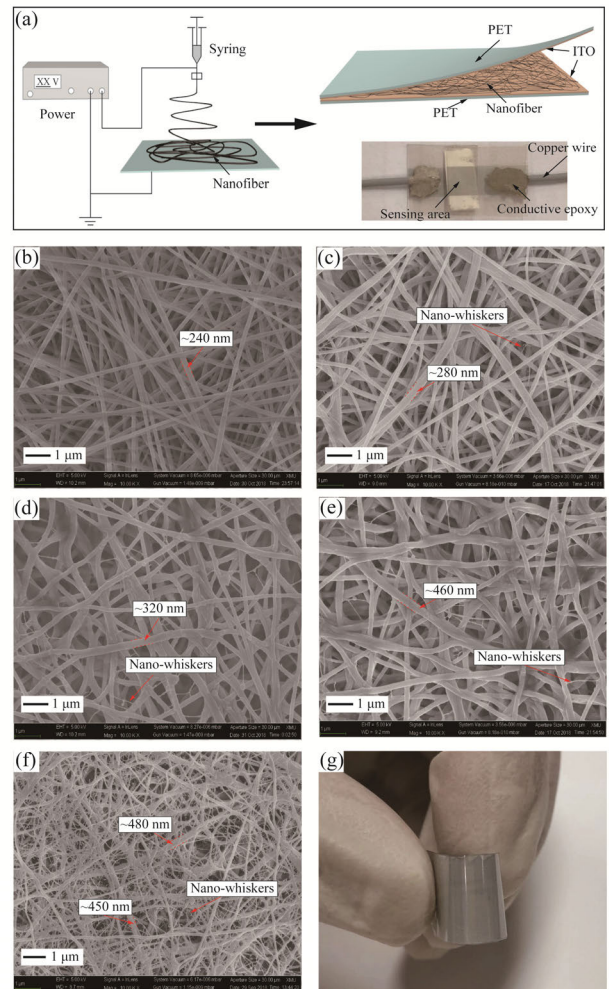


FIGURE 2. Flexible capacitive pressure sensor. (a) Schematic illustration of the fabrication of the sensing device. (b)-(f) Microstructure of the ionic nanofiber membrane with different ionic liquid additive (from 0:1 to 2:1). (g) Diagram of single sensor.

These theoretical analyses are in consistent with the experimental design and the FTIR spectrums results. As described in Equations (3) and (4), the ionic liquid will not react with water, but the oxygen in the air can dissolve in the ionic liquid and react with water to produce some charged ions (HO_2^- and OH^-). However, only a small amount of oxygen can dissolve in the ionic liquid and react with water. Therefore, the charged ions in the nanofibers will reach equilibrium soon when the nanofibers are pouring in the air. After reaching the equilibrium, the charged ions are stable in any humidity environment, and thus the sensing performance of the design sensor will not be influenced by the humidity.

After previous preparations, a single sensor and a sensor array can be fabricated on the nanofiber membranes and they are used as dielectric layer. The fabrication process of the sensing device and the microstructure of ionic nanofiber membrane with different ratios of ionic liquid

additive are shown in Figure 2 (a)-(f). Because of the existence of the ionic liquid, the nanofibers can be adhered together by the ionic liquid on their surfaces during the electrospinning progress, and thus, lead to an increase of the average size of nanofibers. As shown in the microstructure images, more nano-whiskers (The finer ionic PVDF nanofiber) can be seen in the nanofibers with more ionic liquid addition. Since the conductivity increases with more ionic liquid additive, the electrostatic force between the collector which used for depositing nanofibers and the syringe spinneret will increase. Because of the strong applied electrostatic force, the nano-whiskers were formed and connected with the main fibers though strong hydrogen bonds [49]. It will increase the sensitivity of the sensor. When the external pressure is applied on the sensor, the nanofiber membrane will be compressed, and the nano-whiskers contacted with the electrode will cause more contact area between the electrode and the nanofiber. Because of the larger contact area between the nanofiber with nano-whiskers and the electrode, the capacitance of the sensor will increase faster than the one without nano-whiskers under the same external pressure. Therefore, the developed sensor shows much higher sensitivity.

For the single sensor, as shown in Figure 2 (g), two pieces of the ITO-PET films connected with copper wires by conductive epoxy were fixed onto the top and bottom surface of composite nanofiber membrane as electrodes to record the capacitance variation under external pressure. For the flexible sensor array, line patterns of the ITO were prepared on two PET substrates. These two ITO-PET electrodes were assembled such that the directions of ITO electrodes were perpendicular to each other. Both the width of each ITO electrode and the spacing between two adjacent ITO electrodes were 5mm. Each elemental sensor was defined by the region covered with the opposite electrodes.

C. SENSING PRINCIPLE

The schematic of the proposed flexible capacitive sensor based on nanofiber membrane is shown in Figure 3. In order to improve the sensitivity of the sensor, an ionic nanofiber membrane used as the dielectric layer of the capacitive sensor, is developed by electrospinning. Comparing to the conventional thin film, there are two advantages in the electrospun nanofiber membrane. One is low Young's modulus of the nanofiber membrane because of the uniform pores distribution. The other is the increase of contact area between the nanofibers and the electrodes when the nanofiber membrane is compressed.

The sensing principle of the proposed sensor is based on the electrical double capacitors between ITO-PET electrodes and ionic nanofiber dielectric layer. When the sensor is energized, an electron field force will form between two electrodes. Under the electron field force, the charged ions in the ionic nanofiber membrane will move to the surface of the opposite-charged electrodes to form electrical

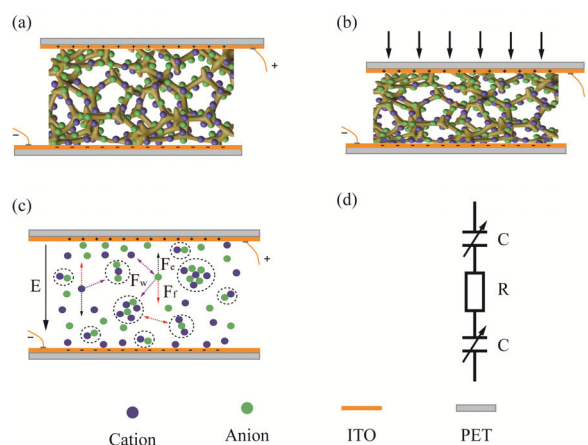


FIGURE 3. Sensing principle of the capacitive pressure sensor. (a) No external pressure applied on the sensor. (b) Pressure applied on the sensor. (c) Schematic image of three forces exerted to ions in nanofibers. (d) Equivalent circuit model of the sensor.

double capacitors which are ultrahigh unit-area capacitors. The capacitance depends on the amount of the charged ions and the contact area between the electrodes and the ionic nanofiber membrane. The sectional view of the sensor without external pressure applied on the sensor is shown in Figure 3(a). Under an external pressure, as shown in Figure 3(b), the distance between two electrodes decreases and the effective contact area between electrodes and ionic membrane increases. In addition, as shown in Figure 3(c), three forces are exerted to the ions in the nanofiber, F_e is the electrostatic force caused by electric field between two electrodes, F_w is the van der Waals force between nearby ions and F_f is the frictional retarding force caused by the nanofibers [50]. According to the equation $F_e = qE = qU/d$, the electrostatic force (F_e) will increase as electric field intensity (E) between two electrodes increases which can be achieved by compressing the nanofiber membrane. As the increase of electrostatic force, the original mechanical equilibrium of ions will be destroyed and the number of charged ions increases. According to the discussion above, more charged ions will move to the surface of the opposite-charged electrodes to form electrical double capacitors. Based on the superposition of the three parts discussed above, the capacitance of the sensor will increase rapidly. As shown in Figure 3 (d), the equivalent circuit model of the sensor can be simplified to two capacitors connected in series.

According to the relationship between the nominal stress and the engineering strain of the porous structure, the stress-strain relationship of the porous nanofiber membrane can be written as: [51]

$$\varepsilon = \frac{2P}{E_0 d(1 - \sqrt{\eta})} \quad (3)$$

where ε is the strain of porous structure, d is the cell size of the porous structure (center-to-center), E_0, η are the Young's

modulus and porosity of the structure. P is the external pressure applied on the sensor.

According to the binary system theory [52], the permittivity of the porous structure can be expressed as:

$$\epsilon_s = \epsilon_p \left(1 + \frac{n\eta(\epsilon_a - \epsilon_p)}{n\epsilon_s + (\epsilon_a - \epsilon_p)(1 - \eta)} \right) \quad (4)$$

where ϵ_s , ϵ_p and ϵ_a are the permittivity of porous structure, the nanofiber and the air, respectively, n denotes the parameter associated with the shape of pores. η is the porosity of the structure.

The capacitance of the capacitor is calculated from:

$$C = \frac{\epsilon_p \epsilon_a A}{h} \quad (5)$$

where C is the capacitance, A is the overlapping area of the two plates, h is the distance between two electrodes. ϵ_p and ϵ_a are the permittivity of the nanofiber and air.

Based on the three equations above, the relationship between the external pressure and the capacitance of the sensor can be described as follows:

$$C = \frac{\alpha A \epsilon_a \epsilon_p}{h(\alpha - 2P)} \left[1 + \frac{n(\alpha\eta - 2Ph)(\epsilon_a - \epsilon_p)}{n\alpha\epsilon_p + (\epsilon_a - \epsilon_p)(\alpha - \alpha\eta + 2Ph)} \right] + \beta C_0 \quad (6)$$

where α is a constant of $E_0 d(1 - \sqrt{\eta})$. C_0 is the unit capacitance of electrical double layer. βg is the constant of contact area. The meanings of other symbols are the same as those in previous equations.

According to the above equations, the capacitance of the sensor will be influenced by both material and the structure parameters, such as the Young's modulus, the permittivity and the thickness of the nanofiber membrane.

III. RESULTS AND DISCUSSIONS

A. RELATIONSHIP BETWEEN PRESSURE AND CHANGE OF CAPACITANCE

The experimental setup and the schematic illustration of load applied on the sensor are shown in Figure 4(a), (b). The load was supplied by a force gauge and the capacitance of the sensor was recorded by an impedance analyzer. Figure 4(c), (d) illustrate the change of capacitance of sensor with different pressures applied on the sensor, giving the weight ratio of ionic liquid and PVDF varies from 0:1 to 2:1. The area and the thickness of nanofiber between two electrodes are given at 10 mm × 10 mm and 50.2 μm, respectively. When the pressure was applied on the sensor, the capacitance of the sensor was measured by the impedance analyzer in real-time. The slope rate of the curve is defined as the sensing sensitivity. And various sensitivities are obtained by ionic liquid with different weight ratios addition. It is clear that the change of capacitance depends on the pressure on the sensor. It was observed that once the pressure was applied on the sensor, the ionic nanofiber dielectric layer was compressed and the contact area between electrodes and nanofiber dielectric layer, as well as capacitance was increased

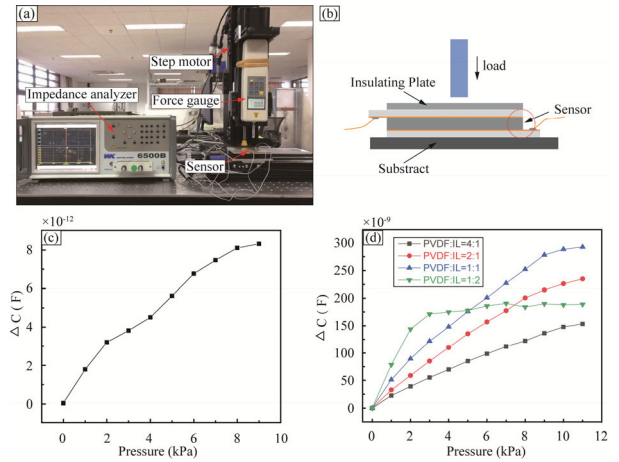


FIGURE 4. Characterization of the pressure sensing performance of the capacitive sensor. (a) Experimental systems for applying pressure and measuring capacitance. (b) The schematic illustration of load applied on the sensor. (c) The relative change in capacitance of the sensor without ionic liquid additive under different pressure applied. (d) The relative change in capacitance of the sensor with different weight ratios ionic liquid addition under different pressure applied.

immediately. As shown in Figure 4(c), the average sensitivity of sensor without ionic liquid addition is 0.76 pF/kPa below 9 kPa.

Since the electrical double capacitors are formed by electrons on the electrodes and the counter ions from the nanofiber membrane at the surface of electrodes, the sensitivity of the sensor with ionic liquid additive can be improved greatly. According to the experimental results, the capacitance of the electrical double capacitor is more than 1000 times larger than that of conventional capacitive sensor. Owing to the high capacitance of the electrical double capacitor, the signals from the capacitive sensor show a very high signal to noise ratio which do not need any denoising process or signal amplification. In order to improve the accuracy of the measurement, five sensors have been measure at the same condition, the final values were the average of the five sensors and the standard deviations (SD) was investigated. The ionic nanofiber membrane with weight ratio at 1:4 of ionic liquid and PVDF had an average pressure sensitivity of 22.31 nF/kPa below 1.6 kPa, which reduced to 12.52 nF/kPa from 1.6 kPa to 11.4 kPa. The ionic nanofiber membrane with a weight ratio at 1:2 of ionic liquid and PVDF had an average sensitivity of 32.60 nF/kPa below 1.3 kPa, which reduced to 19.43 nF/kPa from 1.3 kPa to 10.6 kPa. Moreover, the ionic nanofiber membrane with higher ionic liquid contents (1:1 of ionic liquid and PVDF) exhibited a high sensitivity of 51.28 nF/kPa below 1.5 kPa, but decreased to 26.41 nF/kPa for a higher pressure range from 1.5 kPa to 12.3 kPa. The ionic nanofiber membrane with a weight ratio at 2:1 of ionic liquid and PVDF had a highest sensitivity of 78.54 nF/kPa below 1.0 kPa, which reduced to 32.10 nF/kPa from 1.2 kPa to 3.6 kPa. The sensitivity and the sensing range of the sensors with different weight ratio of ionic liquid and PVDF were summarized in Table 1.

TABLE 1. Comparison of the sensing properties of the sensors with different weight ratio of ionic liquid and PVDF.

IL:PVDF	Sensitivity -1 (SD)	Sensing Range-1	Sensitivity -2(SD)	Sensing Range-1
0:1	0.76 pF/kPa (0.23)	0-9 kPa	-	-
1:4	22.31nF/kPa (2.36)	0-1.6 kPa	12.52nF/kPa (1.38)	1.6-11.4kPa
1:2	32.60nF/kPa (3.12)	0-1.3 kPa	19.43nF/kPa (2.57)	1.3-10.6kPa
1:1	51.28nF/kPa (2.18)	0-1.5 kPa	26.41nF/kPa (1.95)	1.5-12.3kPa
2:1	78.54nF/kPa (4.29)	0-1.0 kPa	32.10nF/kPa (3.89)	1.0-3.6kPa

Since the high concentration of ionic liquid addition leads to an increase of nano-whiskers, the amount of the main fibers of nanofiber membrane decreases in the same injection speed. The nanofiber membrane can be compressed easily due to the decrease of the main fiber; therefore, the sensing range of the sensor decrease as well. The sensitivity of the sensor increases with the increase of ionic liquid addition, which is inconsistent with the theoretical analyses before. Note that, when the pressure is continuously increased, the sensitivity decreases due to the increased Young’s modulus of the ionic nanofiber membrane. There are two relevant factors: (3) the “spring constant” of nanofibers increases under press, (4) more and more nanofibers get into contact with the electrodes under press.

In summary, the 1:1 weight ratio of ionic liquid and PVDF results in a higher sensitivity (about 67.5 times higher than the sensor without ionic liquid addition) and sensing range of the sensor, which has been proven by experiments.

In addition, a dynamic pressure applied on the sensor was provided by a vibration exciter driven by sine wave voltages with different driving frequencies at 5, 10, 25 and 50 Hz. The sensors were fabricated by ionic nanofiber membrane with weight ratio of 1:1 between ionic liquid and PVDF and the thickness was given at 50.2 μm. The dynamic experimental setup is shown in Figure 5(a), the dynamic load was supplied by vibration exciter and the capacitance of the sensor was recorded by impedance analyzer. To evaluate the reversibility and reproducibility of the pressure sensor, 1000 repetition cycle tests were conducted by applying and releasing pressure of 5 kPa with different driving frequencies. The relevant change in capacitance of the sensor with 1000 cycles under 5 Hz is shown in Figure 5(b). The capacitance curves of both initial fifteen and final fifteen cycles of the 1000 cycles with 5 Hz, 10 Hz, 25 Hz and 50 Hz are shown in Figure 5(c)-(f). The capacitance is stable and matching well with external pressure. It was observed that once the pressure was applied on the sensor, the ionic nanofiber membrane was compressed and the contact area between electrode and ionic nanofiber membrane as well as capacitance both increased

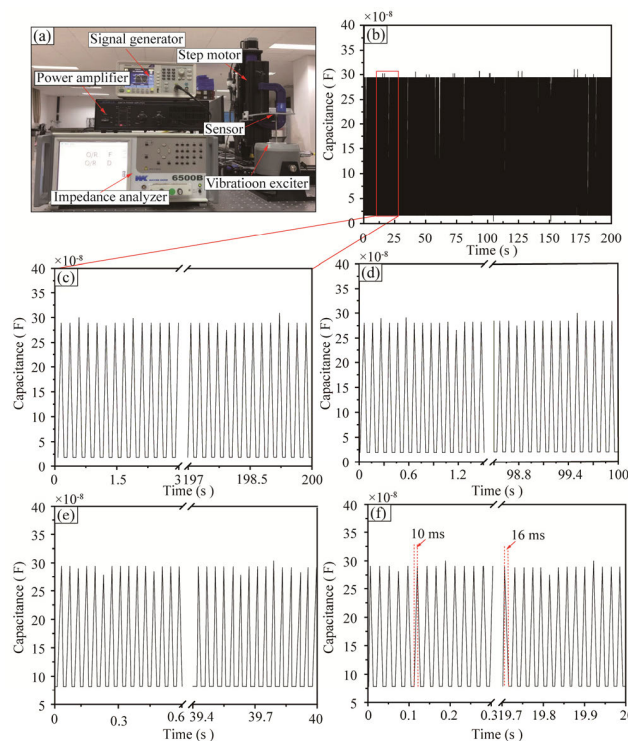


FIGURE 5. Characterization of the pressure dynamic sensing performance of the flexible capacitive sensor. (a) Experimental system for dynamic pressure applying and capacitance measuring. (b) Relative change in capacitance of sensor with 1000 cycles of repeated loading and unloading under 5 kPa at 5 Hz. The relationship of pressure and capacitance of the sensor compressed by a periodic pressure at c) 5 Hz, (d) 10 Hz, (e) 25 Hz, (f) 50 Hz.

immediately. The response and relaxation time of the sensor were 10 ms and 16 ms, which can be extracted from the upstroke response of the device readout signal in Figure 5(f). Moreover, the capacitance is back to the original value after unloading the pressure. Overall, the sensitivity of the pressure sensors is greatly enhanced by using the porous nanofibers structure compared with sensors fabricated by a conventional thin-film structure. In addition, the sensors were able to respond well to both static and dynamic pressure applied on them. Note that, because of the viscoelasticity of the PVDF, the capacitance change of the sensor cannot catch up with dynamic pressure applied on it when the frequency is over 50 Hz.

Table 2 summarizes the sensitivity and sensing range of the capacitive sensor as well as some similar capacitive pressure sensors. Compared to other capacitive pressure sensors, the developed sensors not only exhibit higher sensitivities but also demonstrate better linearity. In addition, the fabrication process and materials of the developed sensors was very simple and inexpensive.

B. SENSOR NETWORK FOR AIR PRESSURE SENSING

In order to verify the feasibility of sensor for a non-contact air pressure measurement, a homemade air pressure testing system was used to provide a controllable air pressure for the

TABLE 2. Comparison of capacitive sensor developed in this work with previous reported capacitive pressure sensors.

Dielectric layer	Sensitivity	Sensing Range	Sensing area
Pyramid PDMS ^[53]	18.6 fF/kPa	0-2.0 kPa	Point
Microlillar ^[54]	30.08 pF/kPa	0-0.4 kPa	15*15 mm ²
AgNW PDMS ^[26]	0.89 nF/kPa	0-0.12 kPa	5*5 mm ²
Porous PDMS ^[34]	2.08 pF/kPa	0-0.33 kPa	15*15 mm ²
Nanofiber (this work)	78.54 nF/kPa	0-1.0 kPa	10*10 mm ²

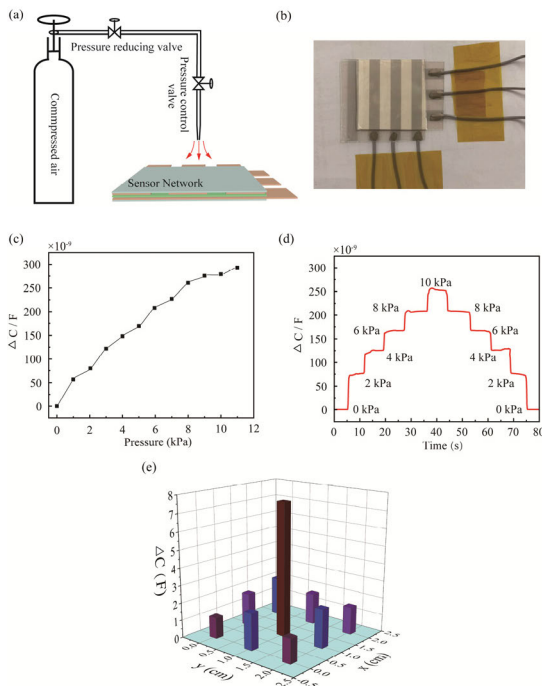


FIGURE 6. Performance of sensor network for air pressure measurement. (a) Experimental systems for applying controllable air pressure. (b) The photograph of the sensor array. (c) The relative change in capacitance of the sensor under different air pressure applied. (d) The relative change of capacitance in response to a step increased and decreased air pressure. (e) The measured capacitance changes of the sensor network with an air pressure applied on the center.

sensor, as shown in Figure 6(a). The photograph of the sensor array was shown in Figure 6(b). A controllable air flow was applied on the sensor vertically. The pressure was calculated by velocity of the airflow and the area of the sensing area. As shown in Figure 6(c), the capacitance of the sensor is increasing with the increment of the air pressure. The sensitivity is 53.27 nF/kPa below 1.1 kPa, while is reduced to 28.14 nF/kPa between 1.1 kPa to 11.8 kPa. Note that, because of the instability of the air pressure, the linearity of the signal was not as good as that in the static pressure test.

To verify the stability of the sensor used in air pressure measurement, a step change in air pressure was applied on the sensor. As shown in Figure 6(d), the capacitance of the sensor is stable during the measurement of the step increase

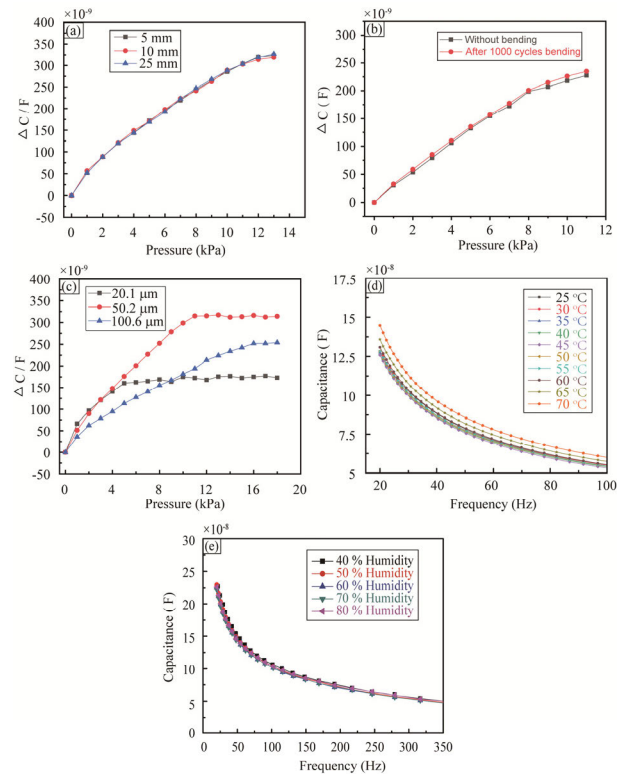


FIGURE 7. Influence of some factors on the sensing performance. (a) Relationship of the change in capacitance of the sensor attached on circular tubes with radius at 5, 10, 25 and pressure applied. (b) The capacitance changes with external load before and after 1000 cycles bending. (c) Different thickness of the nanofiber membrane, 20.1, 50.2, 100.6. (d) Capacitance change with frequency at temperature range from 25 °C to 70 °C. (e) Capacitance follows the frequency at humidity range from 40% to 80%.

and decrease air pressure. In order to investigate the ability of the sensors to measure the spatial distribution and magnitude of an air pressure, a 3×3 sensor array was fabricated based on the ionic nanofiber membrane and attached on a plate to measure the surface pressure. The pressure distribution can be easily obtained by measuring the capacitance change of the sensor array. As shown in Figure 6(e), the sensor network was attached on an arc structure. When the air pressure was applied on the surface of the sensor network, the area closer to where the pressure was applied would exhibit more deformation, leading to a larger capacitance variation. According to the results, this innovative sensor can be adapted to detect a variety of non-contact actions, such as the aerodynamic pressure measurements and impact position measurements.

C. EFFECT OF THE BENDING RADIUS AND THICKNESS OF THE DIELECTRIC LAYER

The influence of bending radius is important to the sensing performance of the flexible sensor. The sensors prepared with about 50.2 μm nanofiber membrane with 1:1 weight ratio of ionic liquid and PVDF were attached on the cylinders with radius of 5 mm, 10 mm and 25 mm, respectively. As shown in Figure 7(a), the capacitance changes of the

sensors attached on cylinders with different radii were negligible. Moreover, the influence of the sensing performance before and after repeated bending has been investigated. As shown in Figure 7(b), the influence of the sensing performance after repeated bending 1000 cycles was negligible. These features are of great significance for the pressure measurement of complex structure.

The thickness of the dielectric layer also plays a crucial role in the sensing performance of the sensor. Figure 7(c) shows the relationship between the change of capacitance and the actual pressure on the sensor, with the thickness of the ionic nanofiber dielectric layer varying from 20.1 μm to 100.6 μm . It is clear that as the thickness increased from 20.1 μm to 100.6 μm , the sensitivity decreased from 96.48 nF/kPa to 36.14 nF/kPa and the sensing range increased from 5 kPa to 16 kPa. Because the thin film can be compressed easily, the distance between two electrodes of the sensor fabricated by a thinner nanofiber membrane will be reduced more when the same external pressures is applied on the sensor. Therefore, the sensitivity of the sensor increases with decreasing the thickness of nanofiber membrane, but the sensing range decreases. In order to balance the sensitivity and the sensing range of sensor, 50.2 μm is the optimized thickness of nanofiber membrane in the experiments. Based on the study, it is clear that the sensitivity and the sensing range of the sensor can be adjusted by changing the thickness of the ionic nanofiber dielectric layer to meet the actual measurement needs.

D. EFFECT OF THE TEMPERATURE AND HUMIDITY

Importantly, the service environment of the sensor is very complex, humidity and temperature vary in time. Thanks to the stable characteristic of physical and chemical properties of ionic nanofiber dielectric layer, the environmental temperature fluctuation only causes minor influence on capacitance. As shown in Figure 7(d), the capacitance of the sensor was measured at the temperature range from 25 $^{\circ}\text{C}$ to 70 $^{\circ}\text{C}$. The sensor was set under the corresponding temperature for 0.5 hour to make sure that the signal is measured at the right temperature. It is clear that the capacitance increases slowly and with a sharp at 60 $^{\circ}\text{C}$, and then increases fast. Under the temperature between 25 $^{\circ}\text{C}$ to 60 $^{\circ}\text{C}$, the capacitance was stable with less than 2.3 % of the variation. This is most likely due to the stabilization of physical and chemical properties and the ionization of ionic liquid in this temperature range. When continuously increases the temperature, the capacitance rises fast, which may cause by the phase transformation of the ionic nanofiber dielectric layer and the increasing ionization of ionic liquid with the temperature above 60 $^{\circ}\text{C}$.

Furthermore, the performance of the sensor at different humidity levels has been investigated. The sensor is set at corresponding humidity for 0.5 hour to make sure that the signal is measured at right humidity. As shown in Figure 7(e), the capacitance is very stable at different humidity levels ranging from 40 % to 80 %. This result is in consistent with the theoretical analyses before. The ionic liquid will

not react with water. Although, the oxygen will dissolve in the ionic liquid and react with the water to produce the HO_2^- and OH^- , the solubility of the oxygen in the ionic liquid is low, the charged ions in the nanofibers will reach equilibrium soon after pouring in the air. Therefore, the performance of the sensor will be affected by the concentration of oxygen slightly, but not by humidity.

IV. CONCLUSION

A flexible capacitive sensor with a simple architecture and high sensitivity has been developed. Investigation shows that 1:1 weight ratio of ionic liquid and PVDF with thickness of 50.2 μm were optimal process parameters. From the experimental results, it is clear that the sensor responds well at both static and dynamic pressure with high sensitivity, good linearity and repeatability. Comparing to traditional pressure sensors, this sensor provides a high sensitivity up to 78.54 nF/kPa. Meanwhile, the sensors exhibit negligible capacitance change under different bending radii; moreover, the sensitivity and sense range of the sensor can be adjusted by modulating the weight ratios of ionic liquid and PVDF and the thickness of the ionic nanofiber membrane. Furthermore, the effects of temperature and humidity on the performance of sensor have been also investigated. The capacitance of sensor with selected materials is stable from 25 $^{\circ}\text{C}$ to 60 $^{\circ}\text{C}$, and not sensitive to humidity. Overall, the developed innovative sensor has significant potential for the low cost and reliable non-contact pressure measurement.

ACKNOWLEDGMENT

(Xiaofeng Yang and Yishou Wang contributed equally to this work.)

REFERENCES

- [1] S. Miyashita, L. Meeker, M. T. Tolley, R. J. Wood, and D. Rus, "Self-folding miniature elastic electric devices," *Smart Mater. Struct.*, vol. 23, no. 9, p. 094005, 2014.
- [2] T. Li, L. Li, H. Sun, Y. Xu, X. Wang, and H. Luo, "Porous ionic membrane based flexible humidity sensor and its multifunctional applications," *Adv. Sci.*, vol. 4, no. 5, p. 1600404, May 2017.
- [3] Y. Ma, J. Choi, A. Hourlier-Fargette, Y. Xue, H. U. Chung, and J. Y. Lee, "Relation between blood pressure and pulse wave velocity for human arteries," *Proc. Nat. Acad. Sci. USA*, vol. 115, no. 44, pp. 11144–11149, 2018.
- [4] A. Castrichini, V. H. Siddaramaiah, D. Calderon, J. E. Cooper, T. Wilson, and Y. Lemmens, "Nonlinear folding wing tips for gust loads alleviation," *J. Aircr.*, vol. 53, pp. 1391–1399, Feb. 2016.
- [5] D. Mueller, J. W. Gerdes, and S. K. Gupta, "Incorporation of passive wing folding in flapping wing miniature air vehicles," in *Proc. DETC*, San Diego, CA, USA, 2009, pp. 797–805.
- [6] D. Trivedi, C. D. Rahn, W. M. Kier, and I. D. Walker, "Soft robotics: Biological inspiration, state of the art, and future research," *Appl. Bionics Biomech.*, vol. 5, no. 3, pp. 99–117, 2008.
- [7] J. Kim, M. Lee, H. J. Shim, R. Ghaffari, H. R. Cho, D. Son, Y. H. Jung, M. Soh, C. Choi, S. Jung, K. Chu, D. Jeon, S.-T. Lee, J. H. Kim, S. H. Choi, T. Hyeon, and D.-H. Kim, "Stretchable silicon nanoribbon electronics for skin prosthesis," *Nature Commun.*, vol. 5, p. 5747, Dec. 2014.
- [8] J. Park, Y. Lee, J. Hong, Y. Lee, M. Ha, Y. Jung, H. Lim, S. Y. Kim, and H. Ko, "Tactile-direction-sensitive and stretchable electronic skins based on human-skin-inspired interlocked microstructures," *ACS Nano*, vol. 8, no. 12, pp. 12020–12029, 2014.

- [9] Y. L. Bunimovich, Y. S. Shin, W.-S. Yeo, M. Amori, G. Kwong, and J. R. Heath, "Quantitative real-time measurements of DNA hybridization with alkylated nonoxidized silicon nanowires in electrolyte solution," *J. Amer. Chem. Soc.*, vol. 128, no. 50, pp. 16323–16331, 2006.
- [10] J. H. Chua, R.-E. Chee, A. Agarwal, S. M. Wong, and G.-J. Zhang, "Label-free electrical detection of cardiac biomarker with complementary metal-oxide semiconductor-compatible silicon nanowire sensor arrays," *Anal. Chem.*, vol. 81, no. 15, pp. 6266–6271, 2009.
- [11] T. Someya, Y. Kato, T. Sekitani, S. Iba, Y. Noguchi, Y. Murase, H. Kawaguchi, and T. Sakurai, "Conformable, flexible, large-area networks of pressure and thermal sensors with organic transistor active matrices," *Proc. Nat. Acad. Sci. USA*, vol. 102, no. 35, pp. 12321–12325, 2005.
- [12] T. Someya, T. Sekitani, S. Iba, Y. Kato, H. Kawaguchi, and T. Sakurai, "A large-area, flexible pressure sensor matrix with organic field-effect transistors for artificial skin applications," *Proc. Nat. Acad. Sci. USA*, vol. 101, no. 27, pp. 9966–9970, 2004.
- [13] Y. Wang, L. Wang, T. Yang, X. Li, X. Zang, M. Zhu, K. Wang, D. Wu, and H. Zhu, "Wearable and highly sensitive graphene strain sensors for human motion monitoring," *Adv. Funct. Mater.*, vol. 24, pp. 4666–4670, Apr. 2014.
- [14] Y. Cheng, R. Wang, J. Sun, and L. Gao, "A stretchable and highly sensitive graphene-based fiber for sensing tensile strain, bending, and torsion," *Adv. Mater.*, vol. 27, pp. 7365–7371, Dec. 2015.
- [15] A. V. Shirinov and W. K. Schomburg, "Pressure sensor from a PVDF film," *Sens. Actuators A, Phys.*, vol. 142, no. 1, pp. 48–55, 2008.
- [16] I. Lee and H. Sung, "Development of an array of pressure sensors with PVDF film," *Exp. Fluids*, vol. 26, nos. 1–2, pp. 27–35, 1999.
- [17] S. Kohli, A. Saini, and A. J. Pillai, "MEMS based capacitive pressure sensor simulation for healthcare and biomedical applications," *Int. J. Sci. Eng. Res.*, vol. 4, no. 12, pp. 1855–1862, 2013.
- [18] G. Meng and W. H. Ko, "Modeling of circular diaphragm and spreadsheet solution programming for touch mode capacitive sensors," *Sens. Actuators A, Phys.*, vol. 75, no. 1, pp. 45–52, 1999.
- [19] A. Chortos, J. Liu, and Z. Bao, "Pursuing prosthetic electronic skin," *Nature Mater.*, vol. 15, p. 937, Jul. 2016.
- [20] J. H. Lee, H. J. Yoon, T. Y. Kim, M. K. Gupta, J. H. Lee, and W. Seung, "Micropatterned P(VDF-TrFE) film-based piezoelectric nanogenerators for highly sensitive self-powered pressure sensors," *Adv. Funct. Mater.*, vol. 25, pp. 3203–3209, 2015.
- [21] H. Li, K. Wu, Z. Xu, Z. Wang, Y. Meng, and L. Li, "Ultra-high-sensitivity piezoresistive pressure sensors for detection of tiny pressure," *ACS Appl. Mater. Interfaces*, vol. 10, pp. 20826–20834, Jun. 2018.
- [22] H. B. Yao, J. Ge, C. F. Wang, X. Wang, W. Hu, and Z. J. Zheng, "A flexible and highly pressure-sensitive graphene-polyurethane sponge based on fractured microstructure design," *Adv. Mater.*, vol. 25, pp. 6692–6698, Dec. 2013.
- [23] F. Motoo, X. Zhang, H. Q. Xie, H. A. Takahashi, T. Ikuta, and H. Abe, "Measuring the thermal conductivity of a single carbon nanotube," *Phys. Rev. Lett.*, vol. 95, no. 6, p. 065502, Aug. 2005.
- [24] S. Luo and T. Liu, "SWCNT/graphite nanoplatelet hybrid thin films for self-temperature-compensated, highly sensitive, and extensible piezoresistive sensors," *Adv. Mater.*, vol. 25, pp. 5650–5657, Aug. 2013.
- [25] D. P. J. Cotton, I. M. Graz, and S. P. Lacour, "A multifunctional capacitive sensor for stretchable electronic skins," *IEEE Sensors J.*, vol. 9, no. 12, pp. 2008–2009, Dec. 2009.
- [26] J. Wang, J. Jiu, M. Nogi, T. Sugahara, S. Nagao, and H. Koga, "A highly sensitive and flexible pressure sensor with electrodes and elastomeric interlayer containing silver nanowires," *Nanoscale*, vol. 7, pp. 2926–2932, Feb. 2015.
- [27] S. Mannsfeld, R. S. Tee, C. H. Chen, S. Barman, B. Muir, and A. Sokolov, "Highly sensitive flexible pressure sensors with microstructured rubber dielectric layers," *Nat. Mater.*, vol. 9, pp. 859–864, Sep. 2010.
- [28] C. M. Boutry, A. Nguyen, Q. O. Lawal, A. Chortos, S. Rondeau-Gagné, and Z. Bao, "A sensitive and biodegradable pressure sensor array for cardiovascular monitoring," *Adv. Mater.*, vol. 27, no. 43, pp. 6954–6961, 2015.
- [29] T. Li, H. Luo, L. Qin, X. Wang, Z. Xiong, H. Ding, Y. Gu, Z. Liu, and T. Zhang, "Flexible capacitive tactile sensor based on micropatterned dielectric layer," *Small*, vol. 12, no. 36, pp. 5042–5048, 2016.
- [30] W. He, G. Li, S. Zhang, Y. Wei, J. Wang, Q. Li, and X. Zhang, "Polypyrrole/silver coaxial nanowire aero-sponges for temperature-independent stress sensing and stress-triggered joule heating," *ACS Nano*, vol. 9, no. 4, pp. 4244–4251, 2015.
- [31] J. Park, Y. Lee, J. Hong, M. Ha, Y.-D. Jung, H. Lim, S. Y. Kim, and H. Ko, "Giant tunneling piezoresistance of composite elastomers with interlocked microdome arrays for ultrasensitive and multimodal electronic skins," *ACS Nano*, vol. 8, no. 5, pp. 4689–4697, 2014.
- [32] J. Park, M. Kim, Y. Lee, H. S. Lee, and H. Ko, "Fingertip skin-inspired microstructured ferroelectric skins discriminate static/dynamic pressure and temperature stimuli," *Sci. Adv.*, vol. 1, no. 9, p. e1500661, 2015.
- [33] S.-Y. Liu, J.-G. Lu, and H.-P. D. Shieh, "Influence of permittivity on the sensitivity of porous elastomer-based capacitive pressure sensors," *IEEE Sensors J.*, vol. 18, no. 5, pp. 1870–1876, Mar. 2018.
- [34] S. Chen, B. Zhuo, and X. Guo, "Large area one-step facile processing of microstructured elastomeric dielectric film for high sensitivity and durable sensing over wide pressure range," *ACS Appl. Mater. Interfaces*, vol. 8, no. 31, pp. 20364–20370, 2016.
- [35] J. Y. Sun, C. Keplinger, G. M. Whitesides, and Z. Suo, "Ionic skin," *Adv. Mater.*, vol. 26, no. 45, pp. 7608–7614, Dec. 2014.
- [36] B. Nie, R. Li, J. Cao, J. D. Brandt, and T. Pan, "Flexible transparent iontronic film for interfacial capacitive pressure sensing," *Adv. Mater.*, vol. 27, pp. 6055–6062, Oct. 2015.
- [37] X. Yang, Y. Wang, and X. Qing, "A flexible capacitive pressure sensor based on ionic liquid," *Sensors*, vol. 18, p. 2395, Jul. 2018.
- [38] S. Lee, A. Reuveny, J. Reeder, S. Lee, H. Jin, and Q. Liu, "A transparent bending-insensitive pressure sensor," *Nat. Nanotechnol.*, vol. 11, pp. 472–478, May 2016.
- [39] N. Bhardwaj and S. C. Kundu, "Electrospinning: A fascinating fiber fabrication technique," *Biotechnol. Adv.*, vol. 28, no. 3, pp. 325–347, May/Jun. 2010.
- [40] T. Xu, Y. Ding, Z. Wang, Y. Zhao, W. Wu, and H. Fong, "Three-dimensional and ultralight sponges with tunable conductivity assembled from electrospun nanofibers for a highly sensitive tactile pressure sensor," *J. Mater. Chem. C*, vol. 5, no. 39, pp. 10288–10294, 2017.
- [41] S. Yun, S. Park, B. Park, Y. Kim, S. K. Park, and S. Nam, "Polymer-waveguide-based flexible tactile sensor array for dynamic response," *Adv. Mater.*, vol. 26, no. 26, pp. 4474–4480, 2014.
- [42] B. Zhu, Z. Niu, H. Wang, W. R. Leow, H. Wang, Y. Li, L. Zheng, J. Wei, F. Huo, and X. Chen, "Microstructured graphene arrays for highly sensitive flexible tactile sensors," *Small*, vol. 10, no. 18, pp. 3625–3631, Sep. 2014.
- [43] A. P. Gerratt, H. O. Michaud, and S. P. Lacour, "Elastomeric electronic skin for prosthetic tactile sensation," *Adv. Funct. Mater.*, vol. 25, no. 15, pp. 2287–2295, 2015.
- [44] D. Ye, Y. Ding, Y. Duan, J. Su, Z. Yin, and Y. A. Huang, "Large-scale direct-writing of aligned nanofibers for flexible electronics," *Small*, vol. 14, p. e1703521, May 2018.
- [45] X. Yang, Y. Wang, H. Sun, and X. Qing, "A flexible ionic liquid-polyurethane sponge capacitive pressure sensor," *Sens. Actuators A, Phys.*, vol. 285, pp. 67–72, Jan. 2019.
- [46] R. Li, Y. Si, Z. Zhu, Y. Guo, Y. Zhang, and N. Pan, "Supercapacitive iontronic nanofabric sensing," *Adv. Mater.*, vol. 29, no. 26, p. 1700253, 2017.
- [47] M. F. Lin, J. Xiong, J. Wang, K. Parida, and P. S. Lee, "Core-shell nanofiber mats for tactile pressure sensor and nanogenerator application," *Nano Energy*, vol. 44, pp. 248–255, Feb. 2018.
- [48] H. Wan, H. Yin, and A. J. Mason, "Rapid measurement of room temperature ionic liquid electrochemical gas sensor using transient double potential amperometry," *Sens. Actuators B, Chem.*, vol. 242, pp. 658–666, Apr. 2017.
- [49] A. Walther and A. H. E. Müller, "Janus particles: Synthesis, self-assembly, physical properties, and applications," *Chem. Rev.*, vol. 113, no. 7, pp. 5194–5261, 2013.
- [50] S. G. Yoon, B. J. Park, and S. T. Chang, "Highly sensitive microfluidic strain sensors with low hysteresis using a binary mixture of ionic liquid and ethylene glycol," *Sens. Actuators A, Phys.*, vol. 254, pp. 1–8, Feb. 2017.
- [51] H. Chen, F. Zhu, K. I. Jang, X. Feng, J. A. Rogers, and Y. Zhang, "The equivalent medium of cellular substrate under large stretching, with applications to stretchable electronics," *J. Mech. Phys. Solids*, vol. 120, pp. 199–207, Nov. 2018.
- [52] T. Yamada, T. Ueda, and T. Kitayama, "Piezoelectricity of a high-content lead zirconate titanate/polymer composite," *J. Appl. Phys.*, vol. 53, no. 6, pp. 4328–4332, 1982.

- [53] S. C. Mannsfeld, B. C. Tee, R. M. Stoltenberg, C. V. H. Chen, S. Barman, and B. V. Muir, "Highly sensitive flexible pressure sensors with microstructured rubber dielectric layers," *Nature Mater.*, vol. 9, p. 859, Sep. 2010.
- [54] J. Yang, S. Luo, X. Zhou, J. Li, J. Fu, and W. Yang, "Flexible, tunable, and ultrasensitive capacitive pressure sensor with microconformal graphene electrodes," *ACS Appl Mater Interfaces*, vol. 11, no. 16, pp. 14997–15006, Mar. 2019.



YISHOU WANG received the B.Sc. and Ph.D. degrees from the Dalian University of Technology, Dalian, China, in 2002 and 2008, respectively.

He is currently an Associate Professor with Xiamen University, Xiamen, China. His main research interests include structural health monitoring and advanced sensing technology.



XIAOFENG YANG received the B.Sc. degree from the Jiangsu University of Science and Technology, Zhenjiang, China, in 2013, and the M.Sc. degree from Xiamen University, Xiamen, China, in 2016, where he is currently pursuing the Ph.D. degree. His main research interest includes advanced sensing technology.



XINLIN QING received the M.Sc. degree from Tianjin University, Tianjin, China, in 1991, and the Ph.D. degree from Tsinghua University, Beijing, China, in 1993.

He is currently a Distinguished Professor with Xiamen University, Xiamen, China. His main research interests include structural health monitoring and advanced sensing technology.

...