Sensors and Actuators A 324 (2021) 112629

Contents lists available at ScienceDirect

Sensors and Actuators A: Physical

journal homepage: www.elsevier.com/locate/sna

A flexible highly sensitive capacitive pressure sensor

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ARTICLE INFO

Article history:
Received 19 December 2020
Received in revised form 31 January 2021
Accepted 14 February 2021
Available online 19 February 2021

Keywords:
Flexible
Sensor
Pressure measurement
Ionic membrane
Capacitor

ABSTRACT

Flexible pressure sensor has been increasingly recognized over the past several decades, but there is still a challenge to fabricate it with a superb sensitivity and large sensing range. Herein, a highly flexible, sensitive and capacitive pressure sensor based on a porous ionic membrane is developed. The innovative sensor contains a polyvinyl alcohol/potassium hydroxide (PVA/KOH) porous ionic dielectric layer and two indium tin oxide polyethylene terephthalate (ITO-PET) films, which are coated on the top and bottom of the dielectric layer, respectively. The sensing mechanism of the sensor is based on the electric double layer capacitors between ITO-PET electrodes and PVA/KOH membrane. By improving the deformability of such elastomer dielectric layer with micro-pores, the sensitivity is greatly enhanced and the detectable range of pressure is significantly widened. Comparing to traditional pressure sensor, the developed sensor provides a high sensitivity up to 20.83/kPa and rapid dynamic responses (50 ms) for pressure measurement. More importantly, the sensor is insensitive to the bending deformation and stable in a low temperature range. Furthermore, a capacitive sensor network is fabricated to measure the spatial pressure distribution and magnitude.

1. Introduction

Recently with the increasing demand for the diversification of electronic devices, flexible electronics have gotten increasing attentions for their broad potential applications in portable and foldable devices [1–4]. According to different sensing mechanisms, electronic sensors can be classified into four categories: triboelectricity [5], piezoelectricity [6,7], piezoresistance [8,9] and capacitance [10,11]. Because of their low power consumption, fast response and low drift, capacitive sensors have been widely used in many important areas [12,13]. Despite that, the sensitivity still needs to be improved. To deal with these issues, small amount of conductive materials (Carbon nanotubes, Graphene, Metal nanowires etc.) have been added to the flexible polymers [14–16]. However, because of the low compressive strain and high Young's modulus of the polymers, the sensitivity of the sensor is still low. After that, some microstructures (Pyramid, Micropillar, Hemisphere etc.) have been used to improve the sensitivity [10,17,18]. Although a higher sensitivity can be obtained in those cases, it would drop dramatically when the pressure applied on the sensor exceeds certain range. Furthermore, micro-porous elastomers have been proposed as a kind of sensing materials for flexible sensors with high sensitivity [19,20]. Recently, a super-capacitive sensor based on the ionic nanofiber membrane has been developed, which can form an electrical double layer with ultrahigh unit area capacitance. This sensor has a relatively high sensitivity of 5.3 nF/kPa [21,22]. However, this fibrous sensor can be broken easily under the tensile force, which greatly limits its application. In addition, some ionic elastomers (ionic-liquid-loaded ionic thermoplastic polyurethane, elastic polyacrylamide composite hydrogel etc.) with microstructures (pillar, pyramid etc.) have been used for the dielectric materials of sensors [23,24]. Nevertheless, the sensors have the same drawbacks as the sensor with microstructures dielectric layer. Thus, it is urgent to develop a new flexible pressure sensor with higher sensitivity, lower drift and broader sensing range.

It is well known that, polyvinyl alcohol/ potassium hydroxide (PVA/KOH) film is commonly used as the solid electrolyte in battery. However, to our best knowledge, there is no reference reported about PVA/KOH capacitive pressure sensor. In this work, a flexible capacitive pressure sensor with high sensitivity and rapid dynamic response is developed. The fabrication procedure is facile and low cost, as it involves adding KOH to PVA solvent and cast molding. The sensor consists of a flexible and 3D porous polyvinyl alcohol (PVA) ionic liquid membrane, and is coated with two indium tin oxide polyethylene terephthalate (ITO-PET) films

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https://doi.org/10.1016/j.sna.2021.112629
0924-4247/

Table 1
Comparison of present capacitive pressure sensor with previous reported capacitive pressure sensors.

Types	Advantage	Disadvantage
Silicon based [25]	High stability	Rigid; Low sensitivity
Elastomer based		
Microstructure elastomer [10,17,18]	High sensitivity in low pressure	Low Sensitivity in high pressure
Composite Elastomer [19,20,26]	High flexibility Simple fabricate	Low sensitivity
Fibrous structures [27,28,29,30,31,32,33]	High sensitivity High stability	Unable to measure shear force
Ionic material based		
Ionic liquid Pu-sponge [22]	High sensitivity	Unable to measure shear force
PVA/KOH [This work]	High sensitivity High flexibility	–

on both top and bottom. The response mechanism is carefully discussed to illustrate the decreased of Young's modulus caused by the uniform pore distribution and the formed electrical double layer capacitors between electrodes and the PVA/KOH ionic membrane. Thus, the pressure measurement performance of the sensors along with various PVA and KOH ratios, different thickness of sensing membrane, temperature and humidity were systematically investigated. The developed sensors can overcome the limitations of the expensive materials, low sensitivity and complex fabrication methods and shows higher sensing range and fast dynamic response.

In conclusion, the advantage and disadvantage of the previous reported and the developed capacitive pressure sensors are concluded in Table 1.

2. Experiment

2.1. Material and apparatus

Polyvinyl alcohol 1788 was purchased from aladdin reagent Co. Ltd. KOH was supplied by Sinopharm Chemical Reagent Co., Ltd. Polyethylene terephthalate (PET, 50 μm) coated with 100 nm thick layer indium tin oxide (ITO) was purchased from Shenzhen South China Xiangcheng Technology Co., Ltd. Copper wire (20 P) 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 membrane. Fourier transform infrared spectrometer (Nicolet IS10 FTIR Spectrometer, Thermo Fisher Scientific Inc, USA) was used to determine the molecular structure of the ionic nanofiber. The crystallographic characteristics of the ionic membrane were demonstrated by X-ray Diffraction (XRD-7000, SHIMADZU, Japan). The capacitance of the senor was measured by the impedance analyzer (WK6500B, Wayne Kerr, USA). A computer-controlled step motor (VELMEX, USA) was used to apply load on the sensor. A force gauge with resolution of 0.01 N (HP-100, Dongguan Zhiqu, China) was used to measure the load applied on the sensor. A vibration exciter (SA-JZ002, Wuxi Shiao, China) was used to make a dynamic load with different frequency applied on the sensor. An incubator (DZF-6050, Shanghai Baixin, China) was used to control the testing temperature.

2.2. Sensor design

The PVA/KOH porous ionic membrane was fabricated using solution casting process at ambient conditions. The fabrication progress is shown in Fig. 1(a). A PVA solution was produced by dissolving the PVA particles in deionized water and stirring continuously at 80 °C for 2 h. KOH was then added into the solution in weight rations of 1:5, 1:2 and 1:1 of PVA for the following experiments. Specifically, monomers of PVA were crosslink by KOH via complexation reaction, which leads to forming 3D porous structures through coordinate bonds. The porous PVA/KOH membrane can be produced on a large scale and at low cost with homogeneous PVA/KOH mixed solution. In order to achieve a uniform pore distribution, the surface silicon model is modified to enhance the hydrophobicity. And then, the prepared PVA/KOH solution was poured into a silicon model and dried at room temperature. The porous PVA/KOH membrane was finally peeled off from the silicon model without any mechanical damage owing to the hydrophobic surface of silicon model. As shown in Fig. 1(b), the porous PVA/KOH membrane was very flexible and elastic. In this work, the porous PVA/KOH ionic membrane was used as the dielectric layer of the capacitive pressure sensor. The ITO-PET electrodes were cleaned by ultrasonic cleaner for 30 min in an acetone and IPA solution, respectively. For the single sensor, two pieces ITO-PET electrodes connected with cooper wires by conductive epoxy were fixed onto the top and bottom surface of PVA/KOH membrane as electrodes to record the capacitance variation under external pressure. The microstructures of PVA/KOH membranes with different KOH weight ratio of PVA were shown in Fig. 1(c)–(e).

2.3. Sensor principle

The schematic of proposed flexible capacitive sensor based on ionic sensing membrane is shown in Fig. 2. The conductive mechanism of the PVA/KOH solid electrolyte follows two steps: 1), the migrating ions are bounded with polar groups in polymer chains; 2), under the action of electric field, with the thermal movement of molecular chains in high elastic region, migrating ions and polar groups are complexing-decoupling constant which realizing the migration of ions [29]. The sensing principle of proposed sensor is based on the electric double layer capacitors between ITO-PET electrodes and PVA/KOH ionic membrane, which is an ultrahigh capacitor formed at the surface of the electrodes by electrons on the electrodes and the counter ions from the ionic membrane. The sectional view of the sensor without applied external pressure is shown in Fig. 2(a). As shown in Fig. 2(b), the PVA/KOH membrane deforms under an external pressure, the contact area between ITO-PET electrodes and PVA/KOH membrane increases. As a result, the capacitance of the sensor will increase due to the decrease of the distance between two electrodes and more changes will be accumulated at the interface between electrodes and PVA/KOH membrane. The equivalent circuit of the sensor is shown in Fig. 2(c) [21,34]. Notably, because of the capacitance of the electrical double layer capacitors are very large (about 1000 times higher than conventional capacitor), the signals from the capacitive sensor do not need any denoising processing or signal amplification that significantly reduces the cost of circuit design and makes it superior to other sensors.

3. Results and discussions

3.1. XRD and FTIR analysis of PVA/KOH membrane

In order to investigate the structure of porous PVA/KOH ionic membrane, XRD and FTIR were carried out to obtain the phase and chemical bond. Fig. 3(a) illustrates the XRD patterns of PVA with different KOH addition. By comparison to the neat PVA mem-

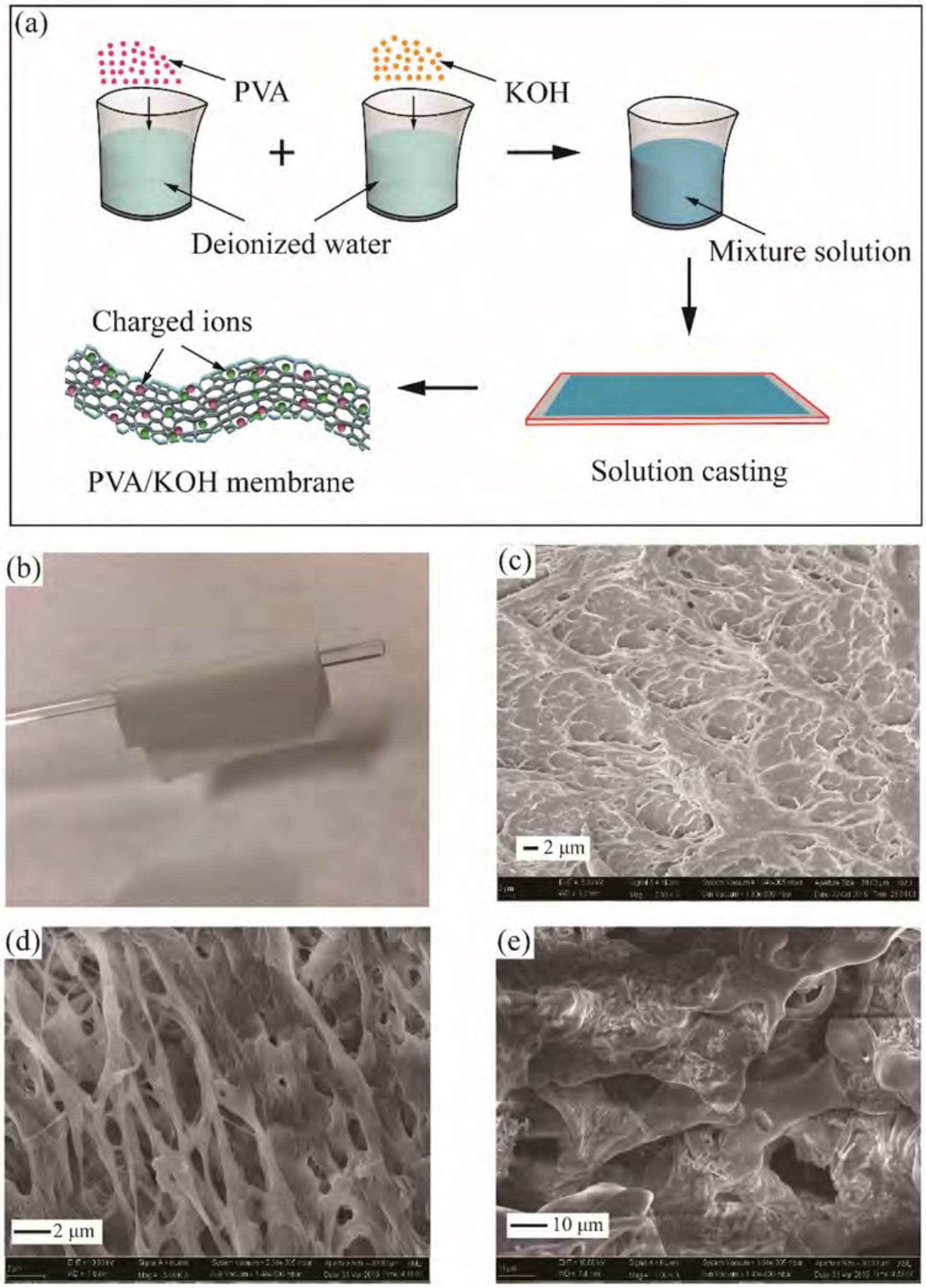

Fig. 1. Flexible ionic capacitive pressure sensor. (a) Schematic illustration of the fabrication of the sensing device. (b) The photograph of PVA/KOH ionic membrane. (c)-(e) Microstructures of the PVA/KOH ionic membrane with weight ratio in 1:5, 1:2, 1:1 between KOH and PVA.

brane, the membrane with KOH addition led to a lower diffraction intensity at about 19.4 (2θ) because of a reduction of the crystallinity, and thus the ionic conductivity is increased. There is an obvious broad peak for PVA/KOH amorphous structure with the KOH added to more than 50 % of PVA, and no KOH crystal peaks is observed.

The FTIR spectra of the PVA membranes with different KOH addition are shown in Fig. 3(b). The peaks located at 3206 cm-1 and 2940 cm-1 are corresponding to the O—H and C–H vibrations, respectively. The peaks at 1240 cm-1 (C—O) drop as the increase of KOH addition due to the decline in crystallinity of PVA. This finding has also been proved by XRD. In addition, the peaks of C═O is observed at 1650 cm-1 since the PVA molecules are dehydrated.

3.2. Relation between pressure and change of capacitance

The experimental setup and load application system were shown in Fig. 4(a), the load was supplied by a force gauge and the capacitance of the sensor was measured by impedance analyzer. The schematic illustration of the load applied on the sensor is shown in Fig. 4(b). As the pressure was applied on the sensor, the capacitance of sensor was measured by the impedance analyzer in real-time. The sensitivity of the sensor S can be described as:

$$S = \frac{\partial(\Delta C/C_0)}{\partial P} \tag{1}$$

Where $\triangle C$ is the capacitance change of the sensor with pressure applied on the sensor. C_0 is the capacitance of the sensor without

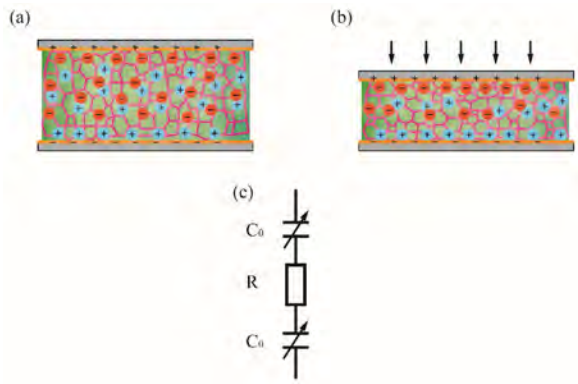

Fig. 2. Sensing principle of the flexible capacitive pressure sensor. (a) No external pressure applied on the sensor. (b) Pressure applied on the sensor. (c) The equivalent circuit of the sensor.

external pressure applied. *P* is the external pressure applied on the sensor.

Fig. 4(c), (d) illustrates the sensitivity of three capacitive sensors with different pressure applied on the sensor, giving the thickness of sensing membrane at about 50 μm and sensing area at 5 mm * 5 mm. It is clear that the change of capacitance depends on the pressure and the performances of three sensors are stable. And various sensitivities are obtained by KOH with different weight ratios addition. It is observed that once the pressure was applied on the sensor, the PVA/KOH dielectric layer was compressed and the contact area of PVA/KOH membrane and ITO-PET electrode as well as the capacitance of the sensor increases. The PVA/KOH membrane with weight ratio at 1:5 of KOH and PVA had an average pressure sensitivity of 10.18/kPa below 2.1 kPa, which reduced to 3.32/kPa from 2.1 kPa to 38.4 kPa. The PVA.KOH membrane with a weight ratio at 1:2 of KOH and PVA had an average sensitivity of 20.83/kPa below 5.0 kPa, which reduced to 11.70/kPa from 5.0 kPa to 32.0 kPa. Moreover, the PVA/KOH membrane with higher ionic liquid contents (1:1 of KOH and PVA) exhibited a high sensitivity of 46.28/kPa below 1.5 kPa, but decreased to 38.41/kPa for a higher pressure range from 1.5 kPa to 9.3 kPa. When the pressure was continuously increased, the sensitivity was decreased due to the reduced mechanical deformation of the PVA/KOH dielectric layer which the small deformation limit has been exceeded, and the contact area of PVA/KOH dielectric layer and ITO-PET electrode increase barely. Different levels of external pressure can be clearly distinguished and well recovered. The capacitance-pressure curves were stable over 1000 cycles, as shown in Fig. 4(e). A small reduction in pressure over the cycles due to the viscoelasticity of the ionic membrane.

In addition, a dynamic pressure of 5 kPa provided by a piezoelectric actuator was measured by the sensor (PVA:KOH = 1:2). As shown in Fig. 4(f), the durability and repeatability of the sensor was tested with repeated loading/unloading at a pressure of 5 kPa for 1600 cycles. It is obvious that the sensing performance of the sensor was stable during the thousands of test cycles. The response and relaxation time of the sensor were 50 ms and 41 ms, which can be extracted from the upstroke response of the device readout signal in Fig. 4(g). Moreover, the capacitance was back to the original value after unloading the pressure. Remarkably, this developed sensor shows a low hysteresis. In addition, the average of the amplitude, the response and relaxation of the 1600 cycles loading/unloading is 651.31 nF, 41.30 ms and 50.12 ms, which indicates that the sensor is stable. Overall, the sensor is able to respond well to the static and dynamic pressure applied on it.

In order to investigate the resolution and the minimum measurable capacitance of the sensor, a step increased and decreased pressures are applied on the sensor for 10 h. The capacitance curve of both the initial increase and the last decrease signals of the 10 h are shown in Fig. 4(h). It is obvious that, the capacitance of the sensor is stable during the measurement of the step shape pressure. The resolution and the minimum measurable capacitance of the sensor are 10 Pa and 1.26 pF.

As shown in Fig. 4(i), the capacitance of the sensor increases with the tensile strain. The sensor can withstand the maximum deformation of 30 %, which is a big improvement to our previous work [22,35,36].

Table 2 summarizes the sensitivity and sensing range of the capacitive sensors as well as some similar capacitive pressure sensors. Compared to other capacitive pressure sensors, the pressure sensors in this work not only exhibit higher sensitivities but also demonstrate a better linearity. In addition, the fabrication progress and materials of the sensors are very simple and inexpensive.

3.3. Effect of the curvature of the sensor, temperature and humidity

As for the flexible pressure sensor, one of the most important performance factors is that it can be attached on complex structure to measure the surface pressure. The sensors prepared with 50 μm PVA/KOH membrane was attached on the cylinder with radius of 0, 10 and 25 mm. As shown in Fig. 5(a), the relationships between the capacitance of the sensors with different curvature were stable and the external pressure was stable. The maximal signal variation

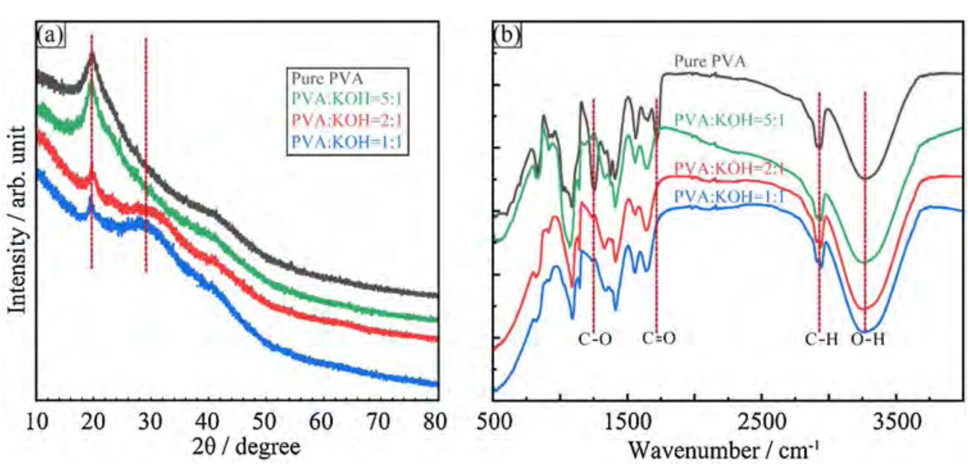

Fig. 3. The XRD patterns and FTIR spectra of the PVA membrane with different KOH addition.

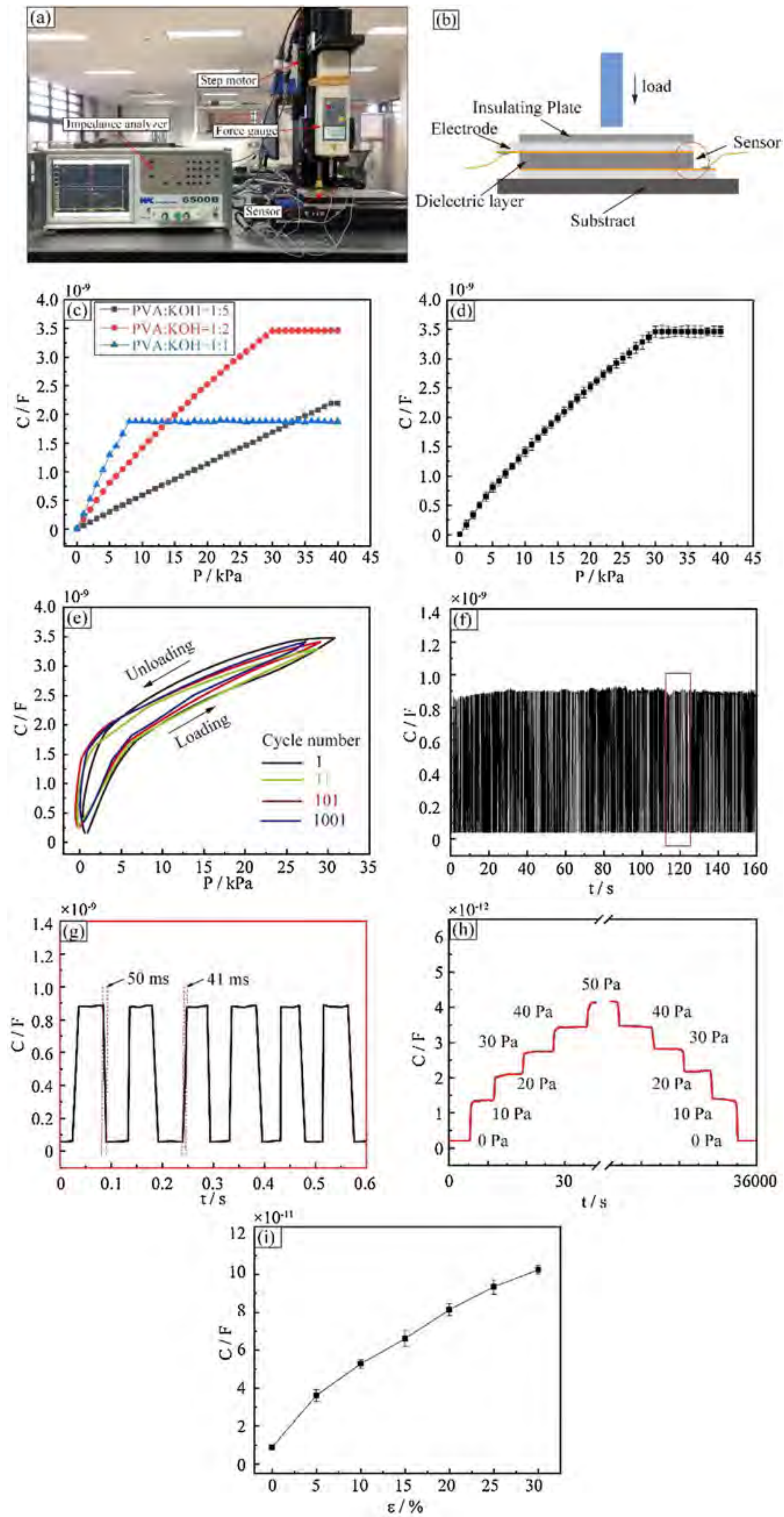

Fig. 4. Characterization of the pressure sensing performance of the flexible capacitive sensor. (a) Experimental system for applying pressure and measuring system. (b) The schematic illustration of the load applied on the sensor. (c) The relative change in capacitance of the sensor with different weight ratios KOH addition under different pressure applied. (d) Relative rate of change in capacitance of the sensor as a function of different pressure. (e) Pressure-capacitance curves of the sensor were measured for more than 1000 cycles. (f) Relative change in capacitance of the sensor with 1600 cycles of fast loading/unloading. (g) Response time of the sensor under external pressure of 5 kPa. (h) The capacitance in response to a step increased and decreased pressure. (i) The relation between the capacitance and the tensile strain.

Table 2
Comparison of present capacitive pressure sensor with previous reported capacitive pressure sensors.

Types	Dielectric layer	Sensitivity /kPa^{-1}	Sensing Range/ kPa
Elastic material-based	Ecoflex(porous) [37]	0.60	0–5
	CNT/PDMS [38]	5.54	0–0.03
	Pyramid PDMS [39]	0.55	0–2
	Porous PDMS [40]	0.26	0–0.33
	Hemisphere PAAm [23]	2.33	0–3
	Fabric structure [41]	0.012	0–100
	PVA/KOH (porous) this work	20.83	0–5

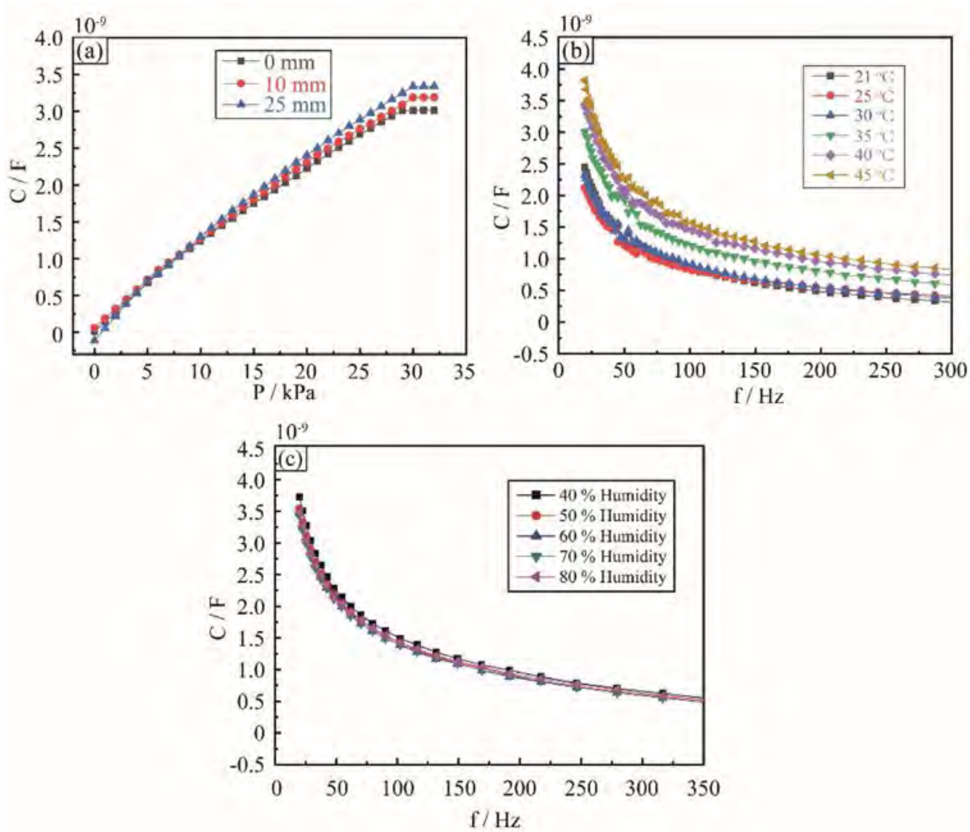

Fig. 5. Effect of two important factors. (a) Relationship between the capacitance of the sensors attached on different circular tubes and the external pressure. (b) Capacitance change with frequency at temperature range from 21 °C to 45 °C. (c) Capacitance change with frequency at humidity range from 40 % to 80 %.

of the three situations was less than 5%. The results indicate that the sensor is potential for tactile pressure measurement of complex structure.

Importantly, the environment of the sensor in service is very complex, for example, the environment temperature varies in time. As previous reports, the conductivity of the PVA/KOH membrane increases with the increase of the temperature which follows the Arrhenius equation [42]. Therefore, it is important to investigate the influence of environment temperature on the sensor. The sensor was set at corresponding temperature for half an hour to make sure that the signal is measured at the right temperature. As shown in Fig. 5(b), the capacitances of sensor were measured at the temperature range from 21 °C (room temperature) to 45 °C. It is clear that the capacitance increases slowly, and with a sharp peak at 30 °C, and then increases fast. At the temperature range of 21 °C to 30 °C, the capacitance was stable with less than 2 % variation. This is due to the stabilization of physical and chemical properties and the conductivity of PVA/KOH varying slightly in the low temperature range. When continuously increasing the temperature, more charged ions will move the surface of the opposite-charged electrode to form the electrical double capacitors resulting in the increase the capacitance, which can be contributed to the conductivity of the PVA/KOH dielectric layer increasing fast with the temperature above 30 °C.

Moreover, the influence of the humidity on the sensing performance also has been investigated. The sensor was set at corresponding humidity for half an hour to make sure that the signal is measured at the right humidity. As shown in Fig. 5(c), the capacitance is very stable at different humidity.

3.4. Application for electronic skin

An essential function of electronic skin is to sense the haptic perception, such as surface pressure. To investigate the ability of the sensors for measuring the spatial distribution and magnitude of external pressure, a 3 × 3 sensor network was fabricated and attached on the skin to measure the surface pressure. As shown in Fig. 6(a), (c), the sensor network was attached on the surface of the hand with one and two tactile pressure applied on it. Three experiments have been done to verify the functionality of the sensor network: 1) single tactile pressure was applied on the center of the sensor network; 2) double tactile pressures were applied on the diagonal of the sensor network; 3) some fast dynamic tactile

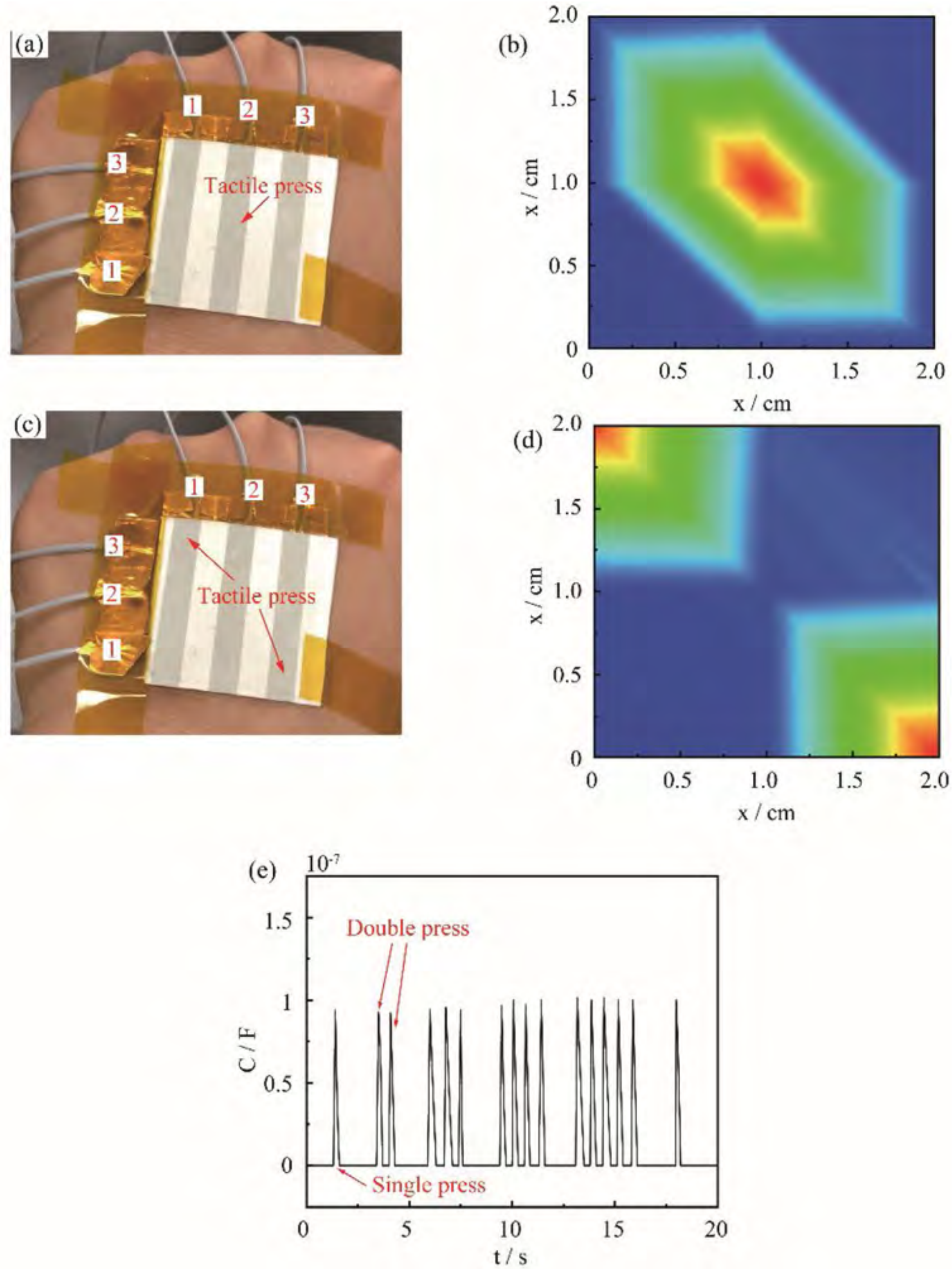

Fig. 6. Sensor network for tactile pressure measurement. (a) A tactile pressure applied on the 3 × 3 sensor network. (b) The capacitance changes of the sensor network with a single tactile pressure applied on the center. (c) Double tactile pressure applied on the 3 × 3 sensor network. (d) The capacitance changes of the sensor network with two tactile pressure applied on diagonal line. (e) The capacitance changes of the sensor network with a dynamic tactile pressure applied.

pressures were applied on one sensor of the sensor network. The capacitance of the sensor network was measured by the impedance analyzer one by one on each individual sensor. The contact areas under tactile pressure were compressed since the local pressure, which resulted in the increasing of capacitance in those areas, different from the uncompressed areas. The capacitance changes of the sensor network under both situations are shown in Fig. 6(b), (d), one with small tactile pressure applied on the center and the other with double tactile pressure applied on the diagonal of the sensor network. It is obvious that the sensor near the location of the tactile increase dramatically, which can be used to locate the impact on the structure. Based on the measurements from 9 discrete sensors, the pressure distribution can be obtained using the interpolation of the measurements over the area and the error is within 1 mm. In addition, the sensor network was also used to detect dynamic pressure variation, which was important for real-time monitoring of tactile pressure changes. As shown in Fig. 6(e), the real-time variation in capacitance of a sensor from the sensor network was press by a dynamic tactile pressure, when the pressure applied on the sensor the capacitance increases immediately and the capacitance variation was positively correlated with the pressure applied on the sensor. The capacitance of the sensor returned to its initial state rapidly when pressure released. Importantly, by repeating the tactile pressure for numerous times, we also demonstrated the low drift of the sensor. Comparing to our previous work, this sensor can withstand some tensile deformation, which has the potential value in the application of soft robotics and electronic skin. This is one of the potential applications of the sensor, it can be used at many important fields for pressure measurement. In addition, the other flexible sensors can also be used to measure the tactile pressure of the complex structure, but additionally need signal amplification or denoising processing.

4. Conclusions

A flexible capacitive sensor with a simple architecture and high sensitivity has been developed, and its pressure sensing properties show that a PVA/KOH mass ratio at 2:1 and thickness of 50 μm are optimal process. Experimental results show that the sensor responds well at both static and dynamic pressure with high sensitivity, good linearity and repeatability. Comparing to traditional pressure sensor, this sensor provides a high sensitivity up to 20.83/kPa, and good dynamic response (50 ms) for pressure measurement. Meanwhile, the sensitivity and sense range of the sensor can be adjusted by modulating the mass ratios of PVA-KOH. Moreover, the influence of bending deformation is negligible and the capacitance of sensor is stable from 21 °C to 30 °C. In addition, the signals from the capacitive sensor show a very high signal to noise ratio which do not need any denoising processing or signal amplification since the capacitance of the electrical double layer is very large. The results show in this work are based on the raw data of the experimental. Overall, the developed innovative sensor has significant potential for the low cost and reliable pressure measurement.

Declaration of Competing Interest

The authors report no declarations of interest.

Acknowledgement

The authors are grateful for the support by the National Natural Science Foundation of China (No. 11772279).

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