Smart textiles integrated with the function of recording tactile stimulation attract much attention due to its great potential for health monitoring and wearable electronics. However, the highly uneven surface and poor thermal stability of textiles become main challenges for precision measurement and device fabrications. To solve these issues, an ion-assisted pressure-sensitive textile using a novel photo-curing process is demonstrated in this work. Using a photo-cured fluorinated polymer poly(vinylidene fluoride-co-hexafluoropropene) (PVDF-HFP) and a transparent polydimethylsiloxane (PDMS) mold, a highly sensitive pyramid array of PVDF-HFP is successfully fabricated on a textile. Furthermore, after doping the ionic additive, tetrabutylammonium perchlorate (TBAP), the sensors show improved sensitivity due to the enhanced dielectric constant. The dielectric constant has a significant increase in the value over 37 after doping a small amount of TBAP (2 wt%). Due to the ion doping, the sensitivity of the textile pressure sensors show significant improvement (0.73 kPa⁻¹) higher than that of the PDMS-based one (0.19 kPa⁻¹). This work not only opens an efficient approach for the fabrication processes of a high-k elastomer pyramid array on a textile but also provide a design strategy to improve performance of capacitive pressure sensor for smart textiles.

1. Introduction

Smart textiles integrated with pressure-sensing capability has attracted much attention due to its great potential for health monitoring and wearable electronics, e.g., light touch and heartbeats.[1–7] Pressure sensing is an essential function of human skin to understand the stimulation from the surrounding. For example, light touch from human can generate pressure of 1–10 kPa (≈10–100 g cm⁻²); the pressure of carotid pulse is around 1.3 kPa[8,9]; the pressure generated from the weight of a butterfly is only 0.003 kPa.[2,3] Therefore, to sense various pressures from human gentle touch to the light weight of insect, a highly sensitive pressure sensor is one of the most important requirements for wearable tactile textiles. To date, three technologies have been developed for pressure sensors, including piezoelectric, piezo-resistive, and capacitive sensors. Piezoelectric sensors based on ferroelectric polymer, polyvinylidenefluoride (PVDF) are generally used for high pressure measurement (≥100 kPa).[10,11] However, they are not suitable for health monitoring or wearable electronics, because of their less sensitivity to gentle touch (≈2 kPa). In contrast, piezo-resistive sensors have attracted increasing attention due to their facile fabrication and high sensitivity.[12,13] By converting mechanical deformation or contact area to electrical resistive changes, piezo-resistive sensors can be used to measure both low (<10 kPa) and high pressure (10–100 kPa) regimes.[7,14] Nevertheless, many factors possibly affect electrical resistance changes, e.g., humidity, impurity, temperature, and electrode brittleness. These factors make piezo-resistive sensors less reliable and susceptible to electrical resistance hysteresis.[15,16] In addition to piezoelectric and piezo-resistive devices, capacitive sensors are an alternative promising route toward high-sensitivity pressure sensors.

Compared with piezoelectric and piezo-resistive devices, pressure-sensing capability of the capacitive sensors is mainly based on the distance between two parallel electrodes. Capacitance is defined by the equation, \( C = \varepsilon_0 A/d \), where \( \varepsilon_0 \) is the vacuum permittivity, \( A \) is the area of the electrodes, and \( d \) is the distance between the two electrodes.
is the vacuum permittivity, \( k \) is the relative dielectric constant, \( A \) is the overlap area between two electrodes, and \( d \) is the thickness of the dielectric layer.\(^{[17]}\) According to this equation, capacitance can be manipulated by three factors, including dielectric constant, thickness, and overlap area. Among these three factors, the change in the thickness is the most common way for pressure sensing. To further improve sensitivity of the capacitive sensors, Mannsfeld and Tee et al. designed a polydimethylsiloxane (PDMS)-based microscale elastic pyramid array.\(^{[19]}\) The shape of the pyramid allows more space for compression, leading to increased pressure sensitivity and reduced viscoelastic behavior and relaxation time after compression. The pressure sensitivity of the structured film is five times higher (0.55 kPa\(^{-1}\)) than an unstructured PDMS film (0.1 kPa\(^{-1}\)).\(^{[5,8]}\) Compared with other types of pressure sensors, the capacitive sensor can save more power, because the current passing through a capacitor is near zero ideally. Furthermore, the capacitor is able to be coupled with field-effect transistors.\(^{[27]}\) Transistors integrated with pressure sensors not only save power but also amplify signals and sensitivity. Schwartz et al. integrated polymer transistors with a PDMS pyramid pressure sensor.\(^{[28]}\) The capacitance change in PDMS pyramid film under compression is significantly amplified when integrating with transistors. The pressure sensitivity of 8.4 kPa\(^{-1}\) was achieved in the pressure-sensitive active matrix, which is more than ten times higher than the original capacitive sensor without coupling with a transistor. Recently, Kim and co-workers developed capacitive pressure sensors using a polyurethane-based materials with the ionic liquid [EMIM][TFSI]. The ionic pairs can generate strong capacitive responses from a few nF to \( \mu \)F when receiving mechanical deformation.\(^{[18–20]}\)

Although the capacitive sensor exhibited great potential for thin-film pressure-sensing applications, integrating capacitive sensor with textiles is rarely investigated. There are two main issues for the fabrication of pressure sensors on textiles: 1) limited thermal-curing temperature and 2) rough surface of textiles. The first issue originated from poor thermal stability of textiles. Typically, PDMS is commonly used for pressure sensor due to its commercial availability. This material requires relatively high temperatures (80–100°C) for solidifying the pyramid array for pressure sensors. However, the process temperature is close to (or higher than) the glass-transition temperature of polyester polyethylene terephthalate (PET)-based textiles (Tg: 75–80°C). Therefore, the thermal process easily causes the unintentional deformation of the polyester textiles. On the other hand, a lower temperature could be an option for thermal curing of the elastic polymer array. However, the lower temperature curing usually suffers low cross-linking degree of PDMS, which may reduce the pressure sensitivity and stability of devices. Therefore, the limited thermal-curing temperature causes a large obstacle for realization of pressure-sensing textiles. In addition to the limited curing temperature, the rough surface of textiles is another obstacle for the realization of pressure-sensing textiles. The internal and surface voids of the textile generate an extremely uneven surface. Because the capacitance of capacitive sensors is inversely proportional to the thickness of the dielectric layer, the uneven surface causes a large deviation of the read-out signals. This uneven surface is also a main factor for low sensitivity of pressure-sensing textiles. Lee et al. demonstrated fiber-shaped pressure sensors based on PDMS-coated conductive fibers with a moderate sensitivity of 0.21 kPa\(^{-1}\).\(^{[16]}\) This sensitivity of PDMS-based fibers is much lower than the thin-film devices made by PDMS (\( \approx 0.5 \text{ kPa}^{-1} \)). Limited contact area between the PDMS-coated fibers led to a small capacitance response under an external pressure. The small capacitance response and moderate sensitivity limit its applications. Therefore, according to the aforementioned literature, limited thermal-process temperatures and relatively low sensitivity remains as challenging issues for pressure-sensing textiles.

To improve the performance of capacitive sensors, we demonstrate a facile approach to fabricate tactile textiles using a photo-curable fluorinated elastomeric material. The fluorinated elastomer, poly(vinylidene fluoride-co-hexafluoropropene) (PVDF-HFP), has been used for organic field-effect transistors,\(^{[21]}\) light-emitting diodes, and solar cells,\(^{[22]}\) due to its high polarity and great stretchability. The dielectric constant of PVDF-HFP is much higher than PDMS. To further improve the capacitance response, we introduce the quaternary ammonium salt, tetrahydroammonium perchlorate (TBAP), into the PVDF-HFP film. It is expected that the addition of the ionic dopant may further increase capacitive response of the dielectric polymer. To avoid the thermal deformation of the polyester textiles, we developed a new photo-curable approach for the solidification of the fluorinated elastomer using a transparent PDMS-based mold. As the polyester textile can obstruct the light to pass through, we prepared the transparent mold to provide a transparent side for UV exposure. This design of the transparent mold can further enhance the flexibility of fabrications of pressure sensors on the textiles. Our approach, therefore, offers a facile approach to achieve reliably capacitive pressure sensors, which paves an important way for the development of health-monitoring smart textiles.

2. Results and Discussion

The used polymer, PVDF-HFP, is a fluorinated elastomer, commonly cross-linked using benzoyl peroxide at relatively high thermal-curing temperatures (>150°C). However, this is not practical for the fabrication of polyester textiles. Therefore, we chose to use a photo-curing process to avoid the high-temperature processes. The chemical structure of PVDF-HFP, TBAP, triallyl isocyanurate (TAIC) as a cross-linker, Irgacure 184 as a photoinitiator, and the device structure of capacitive pressure sensors are shown in Figure 1a. The cross-linking reaction was achieved using UV light to initiate Irgacure 184, leading to the highly efficient combination with the trivinyl TAIC and the cross-link site moiety (CSM) of PVDF-HFP. The CSM group of PVDF-HFP is a vinyl bromide structure (CH\(_{4} = \text{CFBr})\), which is capable of acting as a reactive site. After UV exposure for a few minutes, the PVDF-HFP/TAIC can be quickly solidified and not be dissolved in common solvents, indicating successful cross-linking. Note that after cross-linking, PVDF-HFP is still elastic and suitable for capacitive pressure sensors.

PVDF-HFP is also a high-dielectric-constant material. The relatively high dielectric should be able to improve the performance of the pressure sensors. To further enhance the capacitance...
response, we doped the ionic dopant, TBAP, into the polymer. TBAP is an organic salt, which easily dissolved in common organic solvents, e.g., methyl ethyl ketone (MEK), acetone, and chloroform. It can be fully dissolved in PVDF-HFP solution using MEK as a process solvent. The well-mixing capability of TBAP facilitates to enhance the capacitance response in the PVDF-HFP film. The capacitance–frequency relationship of the ion-doped PVDF-HFP is shown in Figure 1b. The addition of the TBAP dopant indeed increases the capacitance of the PVDF-HFP film. After doping only 2 wt% TBAP, the dielectric constant of the PVDF-HFP increases to 37.5, which is sixfold higher than the value of the pristine polymer without any dopant. Intriguingly, the capacitance of the ion-doped PVDF-HFP significantly increases in the low-frequency regime (20 Hz to 1 kHz). At the low frequency of 20 Hz, the capacitance of 2 wt% TBAP-doped film can achieve 3 μF, while the capacitance is only around 20 nF at the frequency of 10 kHz. The thickness of these doped film prepared by spin-coating is 1.5 μm, but we could observe large changes with different dopant ratios. This dramatic change may be attributed to two possible reasons: 1) electrochemical double layers or 2) polarization of PVDF-HFP. PVDF-HFP itself can act as a solid-state electrolyte. In the literature, many groups preferred to blending a large amount (>40%) of ionic liquids to enhance ionic conductivity (>0.1 mS cm$^{-1}$) and capacitance (>1 μF).\cite{2018-224} Furthermore, the capacitance of the electrochemical-double-layer capacitors is commonly independent of film thicknesses, because the electrochemical double layers tends to be only formed nearby the surface of electrodes.\cite{24} However, in our devices, we have found that the capacitance of the TBAP-doped PVDF-HFP film is dependent on the thicknesses (Figure S1, Supporting Information). The thinner TBAP-doped film gives higher capacitance. These results are against the hypothesis that electrochemical double layers dominate the capacitance response. Therefore, we believe that the large capacitance of the TBAP-doped film should be contributed by the polarization of PVDF-HFP. PVDF-HFP is a polarizable material. The C-F group of PVDF-HFP is a strong polar group and would be potentially oriented by an electrical field. When doping with ionic species, the interaction between ions and PVDF-HFP may facilitate the polarization of the C-F group under an electrical

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Figure 1. a) Chemical structures of PVDF-HFP/TAIC/Irgacure 184 and the ionic dopant, TBAP; b) Capacitance–frequency relationship of the TBAP-doped PVDF-HFP films with different dopant ratios. c) The layer structure of the capacitive pressure sensors on the textiles with printed Ag electrodes; d) picture, e) optical microscopy, and f) SEM images of PVDF-HFP pyramids on the textiles.
field, thus leading to significant increases in the dielectric constant of PVDF-HFP. This feature is actually great for capacitive pressure sensors, because the capacitance would be sensitive to changes in the dielectric distance. Furthermore, the polymer film only requires a small amount of organic salts. Then, the devices would show complete solid-state and reasonable capacitance response. Unlike the ionic-liquid-based devices, despite large capacitance, the large amount (40–60 wt%) of ionic liquids may easily form a liquid-like and sticky film, which may not be favorable for device fabrications.

To realize pressure sensors based on the ion-doped PVDF-HFP film, we fabricated a capacitive pressure sensor on the textiles using a pyramid structure. Schematic processes for the device fabrication are shown in Figure 2. As the polyester textiles cannot survive during thermal curing processes, we chose photocuring processes for device fabrication. To use UV exposure, it is necessary to fabricate a transparent mold. To obtain a transparent mold with the pyramid structure, we transferred the PDMS films twice from the structured wafer (Figure 2a). First, we thermally cured a PDMS film with a pyramid-structured wafer. The microscale pyramids were transferred onto the surface of the PDMS substrate. Then, the target transparent PDMS mold was obtained by transferring PDMS films again from the microstructured PDMS substrate. Note that the surface of the mold was modified by self-assembly monolayers of octadecyltrimethoxysilane (OTS) as a release layer to improve the yield of transfer processes.

The pressure sensing textiles were fabricated by sandwiched with the microstructured PVDF-HFP/Ag/textile and Ag/textile electrode. The schematic process steps of the pressure-sensing textile are shown in Figure 2b. A printed Ag electrode was defined on the PET-based textile using dip-pen printing with a silver paste (Dupont PE-872). The defined area of the Ag electrode is $2 \times 2 \text{ cm}^2$. Due to the rough surface of the PET textile, we deposited the two different PVDF-HFP layers on the textile. The first layer is a buffer layer of PVDF-HFP used to flatten the surface of the textile (the thickness of the flattened textile is around 100 μm). The PVDF-HFP buffer layer was introduced to improve the uneven surface and fulfill the voids inside the PET textiles. The second layer is the pressure-sensing PVDF-HFP layer with microscale pyramids. The pyramid structure of PVDF-HFP was fabricated by transferring a surface topology from a transparent PDMS mold. The PVDF-HFP pyramids were photo-crosslinked by a UV light at the wavelength of 365 nm for 1 min to solidify the pyramid shape. The micro-scale pyramid structures on the PVDF-HFP are uniform and periodic across the textiles. The pyramids can be observed by optical microscopy (Figure 1d,e) and scanning electron microscopy (SEM) (Figure 1f). The height and width of each pyramid were 12 and 20 μm, respectively. The distance between pyramids was $7–8\text{ μm}$. Note that, even though the textiles were flattened by the first PVDF-HFP buffer layer, the textures of the PET textiles were still observable (Figure 1e).

Figure 3 shows the capacitance response curves for the pressure-sensitive textiles. We compared three different pressure-sensitive textiles, including PDMS, PVDF-HFP, and ion-doped PVDF-HFP. The capacitance response curves were measured after loading and unloading various pressures from

![Figure 2. Schematic processes of a) the PDMS mold and b) capacitive pressure-sensing textiles.](image-url)
can detect a pressure as low as 0.01 kPa to 4.9 kPa for a few seconds. The devices exhibited a stable response under a static pressure. Furthermore, the ion-doped PVDF-HFP sensors can detect a pressure as low as 9.8 Pa, which is 100 mg loaded on a 1 × 1 cm² surface. Even under a high pressure of around 2 kPa (20 g loaded on a 1 × 1 cm² surface), the relative capacitance changes still display a stable signal without any significant hysteresis or degradation. This suggests that our pressure-sensitive textile is capable of detecting a wide range of pressures. The pressure sensitivity was estimated from the slope of the ratio of capacitance changes versus pressure \( S = \frac{\Delta C}{C_0} \frac{\partial p}{\partial p} \), where \( p \) is the external pressure, \( C \) is the capacitance. For comparison, a PDMS-based pressure-sensitive textile was made as a control device. As shown in Figure 3a,b, the PDMS-based device exhibited a very small capacitance response in the level of \( \approx 0.4 \) pF. The capacitance change ratio \( \Delta C/C_0 \) of the PDMS is less than 0.1. The estimated sensitivity of the PDMS-based devices was only 0.19 kPa⁻¹, which is lower than the values (0.3–0.5 kPa⁻¹) reported from the thin-film devices in the literature.[3] The lower capacitance response and sensitivity of the PDMS-based device are mainly attributed to the thick PDMS layer and non-uniform distance between the electrodes. Although the textiles were planarized by the elastomers (PDMS for the PDMS-based devices and PVDF-HFP for the PVDF-HFP-based devices), the addition of buffer layer still causes the increase in the thickness of the dielectric layer. Therefore, the increased thickness significantly reduces the capacitive signals, because the capacitance is inversely proportional to the dielectric thickness. Despite the high-k PVDF-HFP as the dielectric layer, the capacitive signals only show a slightly increased sensitivity of 0.3 kPa⁻¹, which is still too low for practical applications. To enhance the capacitance, we doped the quaternary ammonium salt, TBAP, with a small amount. Hence, we mixed PVDF-HFP with different concentrations of TBAP. Herein, we did not use the higher ratio of TBAP for pressure sensors, because the 2 wt% TBAP devices exhibited more stable performance, compared with the devices using the higher ratio of TBAP. This may be attributed to the increased leakage current at the higher ratios. As shown in Figure 3, the PVDF-HFP device doped with TBAP with 2 wt% exhibited an order of magnitude higher capacitive response with an improved pressure sensitivity of 0.73 kPa⁻¹, as compared with the pristine PVDF-HFP- and PDMS-based devices. Table 1 is the summary of the capacitive pressure-sensing textiles. The results show that TBAP plays a critical role in the performance of capacitive pressure-sensing textiles. As the amount of TBAP is increased, the sensitivity of the PVDF-HFP textile devices increased from 0.31 to 0.73 kPa⁻¹ at low pressure regime (below 0.5 kPa) (Figure S2, Supporting Information). The increased sensitivity may originate from the increased dielectric constant of the TBAP-doped PVDF-HFP. Note that, when the pressure is higher than 0.5 kPa, the pressure sensitivity showed a decrease in trend. The decrease in trend may originate from the enhanced elastic resistance of the pyramid structure under a large compression.

The response time of the pressure-sensitive textiles was also investigated. The devices exhibited fast response and relaxation times below 0.1 s at the pressure of 0.49 kPa (Figure 4a,b). The capacitive response of the TBAP-doped PVDF-HFP is much higher than the one made by PDMS. Note that, the response time of TBAP-doped PVDF-HFP reaches the limitation of the time resolution of our equipment and software, suggesting that our device can be potentially used for fast pressure measurements. Intriguingly, the time response and relaxation times of the devices with and without TBAP are similar. The fast response and relaxation time suggests that the ion-induced polarization

![Image](https://www.advancedsciencenews.com/)

**Figure 3.** a) Capacitance curves of the pressure sensors based on PDMS, PVDF-HFP, and the ion-doped 2 wt% PVDF-HFP pyramids under different pressures; b) Capacitance–pressure curves of the pressure-sensitive devices made with different ratios of dopants and PDMS, respectively.

### Table 1. Summary of pressure sensitivity of capacitive sensing textiles.

| Dielectric layer material | TBAP [%] | Dielectric constant \(1\,\text{kHz}\) | Sensitivity \(1\,\text{kPa}^{-1}\) |
|--------------------------|----------|--------------------------------|-------------------------------|
| PDMS                     | 0        | 2.7                            | 0.19                          |
| PVDF-HFP                 | 0        | 6.0                            | 0.31                          |
| PVDF-HFP                 | 0.2      | 7.7                            | 0.58                          |
| PVDF-HFP                 | 2        | 37.5                           | 0.73                          |

*The sensitivities were estimated from the pressure regime below 0.5 kPa⁻¹.*
did not cause any restriction for fast speed measurements of capacitive sensors. The fast response time allows the collection of physiological information in human body. The ion-doped PVDF-HFP sensors exhibited excellent repeatability and stability as demonstrated by cycle testing. Figure 4c shows 100 cycles of capacitance–pressure curves of the ion-doped PVDF-HFP sensors under different pressures. Figure 5 shows the real-time pulse measurements recorded by our pressure-sensitive textiles. The device was attached on the neck or wrist for pulse detection. The flexible and lightweight pressure-sensitive textiles can be attached easily on the nonflattened surface of human skin. The heart pulses in human ranged from 60 to 80 beats per minute. This suggests that our pressure sensor can be potentially used for health monitoring.

It is noteworthy that the RC time constant of the ionic-doped pressure sensors is relatively small. The RC time constant of the capacitive sensors is the product of electrode resistance and capacitance. Typically, a small time constant (τ) is desirable for practical applications, because the possible maximum operation speed of the devices is restrained by the time constant. In the pressure-sensitive textiles, the resistance of the electrode is around 1 Ω, and the capacitances are from a few tens to hundreds of pF. This implies that the time constant should be as fast as 0.1 ns. Our system is potentially applicable for high-speed electronics.
3. Conclusion

In this work, we developed fast and low-temperature photo-curing processes for pressure-sensitive textiles with a high capacitive sensitivity. Through UV exposure and transparent PDMS mold, the high-k fluorinated elastomer can be fast solidified on the PET-based textiles. Our photo-curing approach is a promising pathway for the development of smart clothes on PET-based textiles, because the poor thermal stability and low Tg of the PET-based textiles limit thermal-curing processes. Furthermore, the addition of ionic dopant, TBAP, significantly improves the sensitivity of the pressure-sensing textiles due to the dramatic increase in the dielectric constant. The interaction between the ionic dopant and the polar C-F group enhance the polarization of the fluorinated polymer, thus leading to the increase in the dielectric constant and capacitive response. Our work shown here, therefore, not only provides an approach to fabricate highly sensitive pressure sensors on textiles but also sheds light on the design strategy for high-k materials.

4. Experimental Section

Preparation of Fluorinated Elastomer Dielectric: PVDF-HFP (DAI-EL G-801) is a fluoroelastomer copolymer purchased from Daikin. It can be photo-cured by Iracure 184 and TAIC. The mixture of the fluoroelastomer, initiator, and cross-linker with the mass ratio of PVDF-HFP:Iracure 184/TAIC = 100 mg:3 mg:9 mg was dissolved in MEK (purchased from Sigma-Aldrich). The ionic dopant, TBAP (purchased from TCI), with different amounts was added in the fluoropolymer mixture, and stirred overnight until completely dissolved. The conductive textile electrode was fabricated using dip-pen printing (Figure 2b) with a silver paste (Dupont PE-872). The size of the Ag electrode was 2 x 2 cm². To fill the void of the textile electrode, we poured 300 mL of the fluoropolymer mixture and photocured the textile. After photo-crosslinking, we prepared PVDF-HFP pyramids using the transparent PDMS mold. To obtain the pyramid microstructure, 300 mL of the mixture was poured onto the octacdecytrimethoxysilane (OTS, purchased from Gelest)-modified transparent PDMS mold with the pyramid microstructure. The PDMS mold was prepared by transferring twice from a silicon template. To obtain PVDF-HFP films with different thicknesses, the fluoropolymer solution was spin-coated at 1000–4000 rpm for 10 s on an ITO/Glass (sheet resistance = 10Ω/square). Then, the cured fluoropolymer film was made for pressure-sensing measurement.

PDMS Dielectric Layer Preparation: PDMS (Dow Corning, Sylgard 184 A) and a curing agent were mixed well at a ratio of 10:1 (w/w). Then, 300 μL of the mixture was poured onto an OTS-modified mold and covered by PET textiles. The PDMS samples were cured at 90 °C for 24 h. After curing, the PDMS samples with pyramids were obtained.

Characterizations: Capacitance measurement was performed using Agilent 4395A precision LCR meter. Capacitance was measured at 1 kHz. Thicknesses of the elastic pressure-sensing film was performed by a Microfigure Measuring Instrument (Surfcoeter ET3000, Kosaka Laboratory Ltd.). Surface morphology of the fluoropolymer films was obtained using a field-emission scanning electron microscope (FE-SEM, JEOL JSM-6510).

Ethical Considerations: Written informed consent was obtained from the subject involved in the real-time pulse measurement experiments illustrated in Figure 5.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

capacitive pressure sensors, fluorinated elastomers, high dielectric constant materials, ionic dopants

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