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Effect of $BaTiO_3$ on the sensing properties of PVDF composite-based capacitive humidity sensors

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A B S T R A C T

Capacitive humidity sensors consisting of materials such as polymers, ceramics, and piezoelectrics are widely used to monitor relative humidity levels. The effect of barium titanate ($BaTiO_3$) nanoparticles on the humidity sensing properties, dielectric response, thermal stability, and hydrophilicity of the polyvinylidene fluoride (PVDF)-$BaTiO_3$ composite films is investigated. Hydrophilicity and surface morphology of the PVDF-$BaTiO_3$ composite films are modified for the development of a good humidity sensor. The nanocomposite solutions are prepared by mixing an optimized concentration (2.5 wt%) of PVDF with different concentrations (0.5, 1, and 2 wt%) of $BaTiO_3$ nanoparticles. X-ray diffraction, thermogravimetric analysis, field emission scanning electron microscopy, and contact angle measurements are used to characterize the structure, morphology, thermal stability, and hydrophilicity of the spin-coated sensing films. The dielectric study of PVDF-$BaTiO_3$ composite film shows that as the concentration of $BaTiO_3$ particles increase, the dielectric constant of the composite films increases as well. PVDF-$BaTiO_3$ (2.5 wt%-1 wt%) based capacitive sensors show stable capacitive response and low hysteresis as compared to the other concentrations of the PVDF-$BaTiO_3$ composites. The maximum hysteresis of the capacitive PVDF-$BaTiO_3$ (2.5 wt%- 1 wt%) humidity sensor is found to be ~2.5%. The response and recovery times of the PVDF-$BaTiO_3$ (2.5 wt%-1 wt%) based capacitive sensors are determined as 40 s and 25 s, respectively, which are significantly lower than those reported for the other PVDF composite based sensors.

1. Introduction

Capacitive humidity sensors are widely used to monitor the relative humidity levels in various industrial processes [1]. The key advantages of capacitive humidity sensors include low cost, ease of fabrication, and linear response with respect to the change in relative humidity levels. The working principle of these sensors is based on the variation in dielectric permittivity of the active sensing layer which results in a change in the capacitance of humidity sensors. To increase the sensitivity of the capacitive sensors, different materials have been used including polymers, ceramics, nanocomposite, metal oxide, and piezoelectric have been extensively studied by Zhang et al. [2-6]. Polymer composite film-based sensors have attracted significant attention owing to their interesting chemical and physical sensing properties.

Among the polymers, polyvinylidene fluoride (PVDF) is a piezoelectric polymeric material which is flexible, thermally stable, and chemically resistive [7]. Because of these properties, the PVDF polymer has been extensively investigated as a sensing polymeric film. The introduction of piezoelectric nanoparticles within the polymer matrix results in enhanced dielectric properties, thermal stability, and good flexibility, compared to the properties of only the polymeric film [8,9]. Among the piezoelectric materials, $BaTiO_3$ is as an excellent candidate for capacitive sensors because of its high dielectric constant, good ferroelectric properties, high hydrophilicity, and good thermal stability. Zhou et al. [10] studied the dielectric properties of the PVDF-surface-hydroxylated $BaTiO_3$. They reported a high dielectric constant and lower dielectric loss for the PVDF-$BaTiO_3$ composite film. Chanmal et al. [11] prepared the PVDF-$BaTiO_3$ composite by melt compounding method, and observed that by increasing the concentration of $BaTiO_3$, the dielectric property of the composite film could be improved.

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Most of the prior studies on PVDF-$BaTiO_3$ composite were focused on determining the effects of $BaTiO_3$ nanoparticles in improving the dielectric properties, enhancing the thermal and mechanical stability, and investigating the hydrophilic properties of the PVDF-$BaTiO_3$ composite film. However, no significant research was performed on the electrical and humidity sensing responses of the PVDF-$BaTiO_3$ composite-based humidity sensors. This could be owing to the hydrophobic characteristics of PVDF, because of which it does not sufficiently adsorb the water molecules and has low sensitivity towards the variation in the relative humidity levels. Notably, the hydrophilic polymeric materials possess high affinity for the detection of the change in humidity levels; however, some hydrophilic polymers are soluble in water and are not very useful for humidity sensors [12,13].

Considering the unique properties of the PVDF-$BaTiO_3$ composite films, a systematic study is conducted herein to improve the hydrophilicity and surface morphology of the PVDF-$BaTiO_3$ composite film-based capacitive humidity sensors, while retaining the thermal stability and dielectric properties.

2. Experimental

$BaTiO_3$ (−325 Mesh Powder) and PVDF powder with an average molecular weight, Mw, of 534,0000 were purchased from Sigma-Aldrich (St. Louis, Missouri, United States) and used without further treatment. The PVDF-$BaTiO_3$ composite solution was prepared in two steps. First, 2.5 wt% of a concentrated solution of PVDF was prepared in N, N-dimethylformamide (DMF) and continuously stirred for 6 h. This concentrated solution of PVDF produced a uniform and good quality film as reported in our previous study [14]. Second, 0.5, 1, and 2 wt% suspensions of $BaTiO_3$ were prepared in DMF and stirred for 60 min. Each suspension of $BaTiO_3$ was mixed with 2.5 wt% of PVDF with 1:2 ratio. Each mixed PVDF-$BaTiO_3$ composite solution was stirred at 500 rpm for 2 h to obtain homogeneous blends. To prepare the humidity sensor devices, the PVDF-$BaTiO_3$ composite solutions were spin-coated (at 6000 rpm for 50 s) on the interdigitated indium tin oxide (ITO) electrodes purchased from Osilla (Sheffield, United Kingdom). The sizes of the substrates were 15 mm × 20 mm and each substrate consisted of five sets of 100-nm-thick interdigitated ITO electrodes. The thickness of the composite film was 150 ± 20 nm. The composite films were then etched for 20 s with acetone using the procedure described by Mallick et al. [14]. PVDF has low solubility in acetone [15]; acetone modified the surface of the sensing film by developing a rough and uneven surface of the sensing films. The capacitance value of the ITO electrodes without the sensing film was 10.8 pF at 40% RH. However, after the deposition of the PVDF-$BaTiO_3$ composite, the value increased to 13.6 pF at 40% RH.

To measure hydrophilicity, the contact angle images of the water droplets on the modified surface of the composite films were captured using a camera with high magnification (six-fold zoom lenses) and the ability to record 2450 frames/s. The sessile drop method (using SCA software) was used to measure the contact angle in the captured images. To examine the surface morphology and roughness of the composite films, field emission scanning electron microscopy (FESEM) was used. Thermal stability of the composite was determined using the Perkin-Elmer DSC 8500 analyzer (Waltham, Massachusetts, United States). Empyrean advanced X-ray diffractometer (Malvern Panalytical, Malvern, United Kingdom) was used to measure the crystallinity of the composite films. X-ray diffraction (XRD) measurements were recorded using a 2θ step size of 0.013° at a scan rate of 1 s per step in the 2θ range of 15–90°. The dielectric measurements were recorded using a broadband dielectric spectroscopic system (BDS; Novocontrol Technologies, Montabaur, Germany). These measurements were performed at room temperature over a broad frequency range (10^1–10^6 Hz). Fig. 1a shows a schematic of the preparation of the PVDF-$BaTiO_3$ composite solution, its deposition via spin coating, and the characterisation of the sensing film.

The electrical characteristics of the PVDF-$BaTiO_3$ humidity sensors were determined using a humidity-controlled sealed chamber. The humidifier and drierite were attached to the humidity-controlled chamber to humidify and dehumidify the sealed chamber, respectively. A reference humidity meter was placed inside the sealed chamber to monitor the real-time relative humidity level in the chamber. The capacitance of the sensor was measured using the MS5308 LCR meter (clipped to the electrode of the sensors). Fig. 1b shows the set up used for the electrical response measurement with components including the humidity sensor, LCR meter, humidifier, and drierite.

3. Results and discussion

FESEM analysis of the composite sensing films was performed to investigate the sensing film morphology, which is an important characteristic of the humidity sensor. The water vapor absorption is directly related to the surface roughness, porosity, and hydrophilicity of the sensing film. FESEM images of the PVDF-$BaTiO_3$ composite films with three different concentrations (0.5, 1, and 2 wt%) of the PVDF-$BaTiO_3$ composite are shown in Fig. 2. In the composite film, the PVDF concentration is maintained constant at 2.5 wt%, but the $BaTiO_3$ concentration is varied (0.5, 1, and 2 wt%). Fig. 2a shows that the $BaTiO_3$ particles are almost completely buried in the PVDF matrix, while in Fig. 2b, the $BaTiO_3$ particles are well-spread and a regular rough surface is observed. However, the $BaTiO_3$ particles agglomerate at the surface of the composite film (Fig. 2c). Therefore, the PVDF-$BaTiO_3$ composite with 1 wt% $BaTiO_3$ (Fig. 2b) concentration is selected for further investigation. The uniform distribution of the $BaTiO_3$ particles in the sensing film can facilitate the entrapment of water molecules over the surface of the sensing film and enhance the sensitivity of the humidity sensors.

To enhance the wettability and sensitivity of a humidity sensor, the hydrophilicity of the sensing films must be improved. The hydrophilicity is closely related to the film surface morphology/roughness and represents the adsorption behaviour of a sensing film. Here it is important to mention that the PVDF is working as a base material and act as a binder to hold the $BaTiO_3$ particles. $BaTiO_3$ particles have been used as an additive to develop the adsorption sites on the surface of the composite film to increase the sensitivity of the humidity sensing film. PVDF itself does not show the good sensitivity towards the change in the relative humidity levels as shown in figure S-1 (in the supplementary file). To compare the surface morphology, the AFM images of PVDF and PVDF-$BaTiO_3$ composite have been given in Figure S-2 in the supplementary data file.

Table 1 includes the contact angle measurements (hydrophilicity) of the PVDF, PVDF-$BaTiO_3$ (2.5 wt%-1 wt%) composite, and PVDF-$BaTiO_3$ composite films after acetone treatment. The contact angle of a pure PVDF film is 92.25°, which indicates its hydrophobic nature. The contact angle of the composite film (PVDF-$BaTiO_3$) is 80.95°, which reveals that the $BaTiO_3$ nanoparticles increase the surface hydrophilicity of the composite film. After treatment with acetone, the contact angle further decreases to 50.6°. Acetone etching at the surface of the sensing film results in a rough and uneven structure, which enhances the wettability of the composite film.

Fig. 3 shows the XRD data of the composite films in the $15° < 2θ < 90°$ range. XRD analysis of the nanocomposite films reveals that the reflection peaks are observed at 22.30°, 31.51°, 39.38°, 45.86°, 51.16°, 66.19°, 70.47°, 75.24°, 79.51°, and 83.62°, corresponding to the typical (100), (101), (111), (200), (201), (211), (202), (103), (230), (311), and (222) peaks of the barium titanate, respectively [16]. The peak at 2θ = 20.3 corresponds to PVDF [17]. To determine the effect of $BaTiO_3$ on the thermal stability of the PVDF-$BaTiO_3$ composite, thermogravimetric analysis (TGA) was performed. The inset in Fig. 3 shows the weight losses in the PVDF powder and PVDF-$BaTiO_3$ composite. Both thermograms exhibit almost similar behaviors; the weight loss of the PVDF-$BaTiO_3$ composite is less than 30% (~ 500 °C),

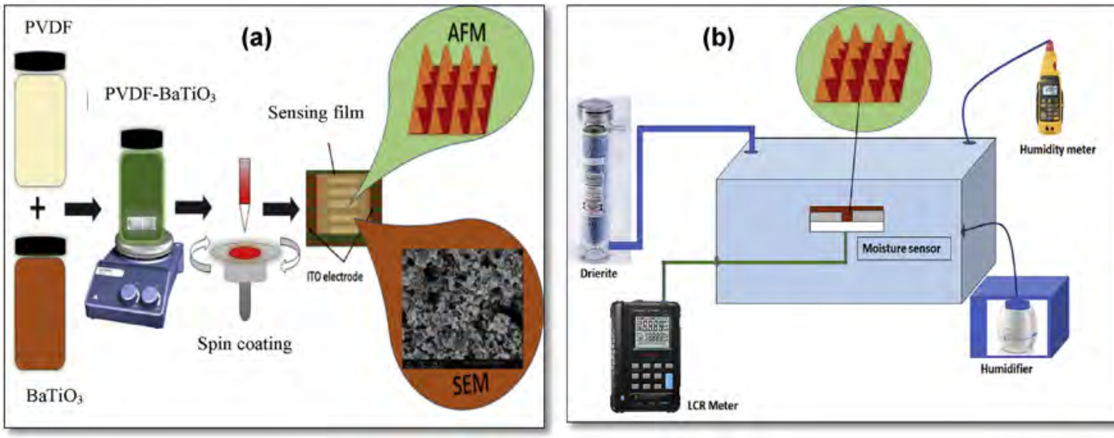


Fig. 1. (a) Schematic of the spin-coated humidity sensors and **(b)** electrical characterisation setup for capacitive humidity sensors.

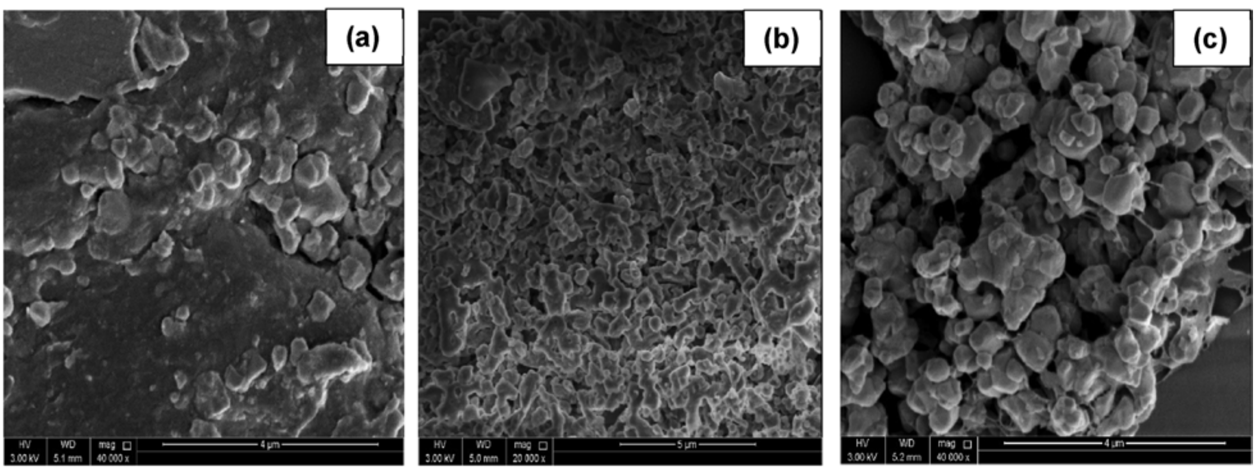
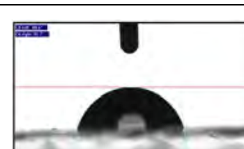


Fig. 2. FESEM images of the PVDF-$BaTiO_3$ composite films with different concentrations of $BaTiO_3$: (a) 0.5 wt%, (b) 1 wt%, and (c) 2 wt%.

whereas, for the PVDF powder, the weight loss is almost 60% at the same temperature. The final weight loss in the composite remains lower than the weight loss observed in the case of only PVDF. At 800 °C, the total thermal weight loss of PVDF is 78% in comparison to 34% for PVDF-$BaTiO_3$ composite. This is because of the remaining $BaTiO_3$ particles, which have high thermal stability compared to PVDF. The 1st step of weight loss in the PVDF-$BaTiO_3$ composite film which starts at ~100 °C can be due to the evaporation of moisture or solvent that remains even after drying the samples at room temperature.

Fig. 4 shows the variation in dielectric properties of the PVDF polymer and PVDF-$BaTiO_3$ composite film as a function of frequency. Fig. 4a shows the dielectric constants (Ɛ′) of PVDF and PVDF-$BaTiO_3$ composite. As expected, the introduction of $BaTiO_3$ particles increases the dielectric constant of the PVDF-$BaTiO_3$ composite film. The dielectric constant is slightly high in the low-frequency region and decreases with an increase in frequency from 10^1 to 10^6 Hz; this may be due to the polarisation relaxation effects. At low frequencies, as the electric field is applied, the accumulation of charges causes an increase in the interfacial polarisation. However, in the high-frequency region, the interfacial polarisation decreases as the charge carrier do not align with the applied electric field, and the dielectric constant decreases. At 10 KHz, the dielectric constant of the PVDF film is 2.07, however after the addition of $BaTiO_3$ particles within the PVDF matrix this value is found to be 2.46. This increase in dielectric constant improves the capacitance of the film. The dielectric loss (Ɛ″) describe as the energy dissipation in a dielectric material through conduction loss, dipolar loss and interfacial polarisation [18]. Fig. 4b shows the dielectric loss (Ɛ″) of PVDF and PVDF-$BaTiO_3$ composite as a function of frequency. At lower frequency region, dielectric loss is due to the interfacial polarisation and conductivity, however, the dielectric loss at higher frequency is

Table 1
Contact angle measurements of PVDF, PVDF-$BaTiO_3$ (2.5 wt%-1 wt%) composite, and PVDF-$BaTiO_3$ (2.5 wt%-1 wt%) composite films after acetone etching.

Sample Type	PVDF Film	PVDF-$BaTiO_3$ composite film	Acetone-etched PVDF-$BaTiO_3$
Contact angle image			
Contact angle	92.25°	80.95°	50.6°

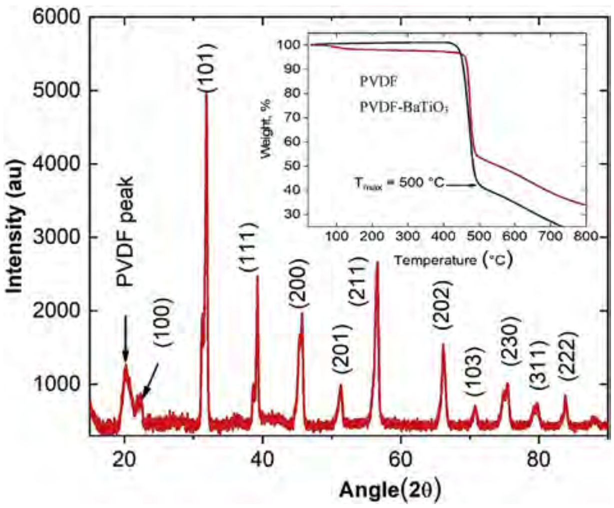


Fig. 3. XRD pattern of the PVDF-$BaTiO_3$ composite films at room temperature (25 ± 1 °C). Inset shows the TGA thermograms of PVDF and PVDF-$BaTiO_3$ composite.

associated with dipolar relaxation [19]. The dielectric loss of PVDF film measure to be 0.01 (@10 KHz), however, after the introduction of $BaTiO_3$ particles the reduction in the dielectric loss has been observed.

The capacitive response of PVDF-$BaTiO_3$ composite-based humidity sensor under varied relative humidity levels ranging from 40–90% RH is shown in Fig. 5. The capacitance responses of the humidity sensors show that the PVDF-$BaTiO_3$ composite-based sensors are more linear and sensitive compared to $BaTiO_3$, PVDF, acetone-etched PVDF, and PVDF-$BaTiO_3$ (with other ratios) composite-based sensors. $BaTiO_3$ is used to enhance the porosity and sensitivity. Acetone etching of the PVDF-$BaTiO_3$ composite film develops the surface of the sensing film, enhances the wettability, increases the adsorption sites which trap more water molecules, as well as decreases the hysteresis by facilitating the desorption process. The sensitivity ($pF/\%RH$) of the sensor is measured by the formula $S = (C_{90} - C_{40})/(90 - 40)$. Where C_{90} and C_{40} represent the capacitance value at the highest humidity level (90 %RH) and lowest humidity level (40 %RH). The sensitivity of PVDF-$BaTiO_3$ composite (acetone etched) sensor is found to 0.2416 pF/%RH.

Fig. 5 also shows the adsorption and desorption characteristics of the sensor. The adsorption and desorption curves represent the hysteresis response of the sensor. When the water contents are adsorbed on the surface of a sensing layer, the clusters of water molecules are formed, and the desorption of moisture becomes slow, leading to

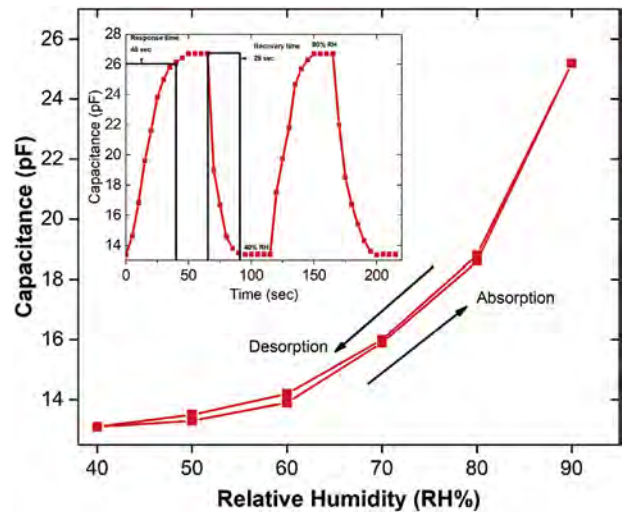


Fig. 5. Adsorption and desorption curves of the capacitive PVDF-$BaTiO_3$ (2.5 wt%-1 wt%)-based humidity sensor. Inset shows the response and recovery cycles (40–90% RH) of PVDF-$BaTiO_3$ (2.5 wt%-1 wt%)-based sensor. The measurements are performed at 25 ± 1 °C.

hysteresis. The maximum hysteresis of the capacitive PVDF-$BaTiO_3$ (2.5 wt%- 1 wt%) humidity sensor is ~2.5% at 60% RH. The hysteresis at any relative humidity level can be defined as the difference in capacitance when the sensor is exposed to a change in relative humidity from low RH to high RH (absorption process) and high RH to low RH (desorption process). The response and recovery times of the capacitive humidity sensors are also important characteristics in determining the performance and implementation of sensors. The time taken by a sensor to achieve 90% RH is defined as the response time of the sensor (in the present study: 40–90% RH). In contrast, the recovery time is the time that a sensor takes to reach 40% RH from 90% RH (initial state, in this case). The inset in Fig. 5 shows the response and recovery times of the fabricated PVDF-$BaTiO_3$ capacitive humidity sensor, which are calculated as 40 s and 25 s, respectively. The presence of the nanoparticles within the polymer matrix, develop the rough and uneven structure on the composite film. The rough and uneven structure on composite film increases its surface area and provides more adsorption sites on the surface of the sensing film [20]. These adsorption sites improve the water molecules entrapment mechanism of the sensing film. Fig. S3 (supplementary data) shows the schematic illustration of the entrapment of water molecules on the surface of the sensing film. Table 2 summarises the response and recovery times of different PVDF

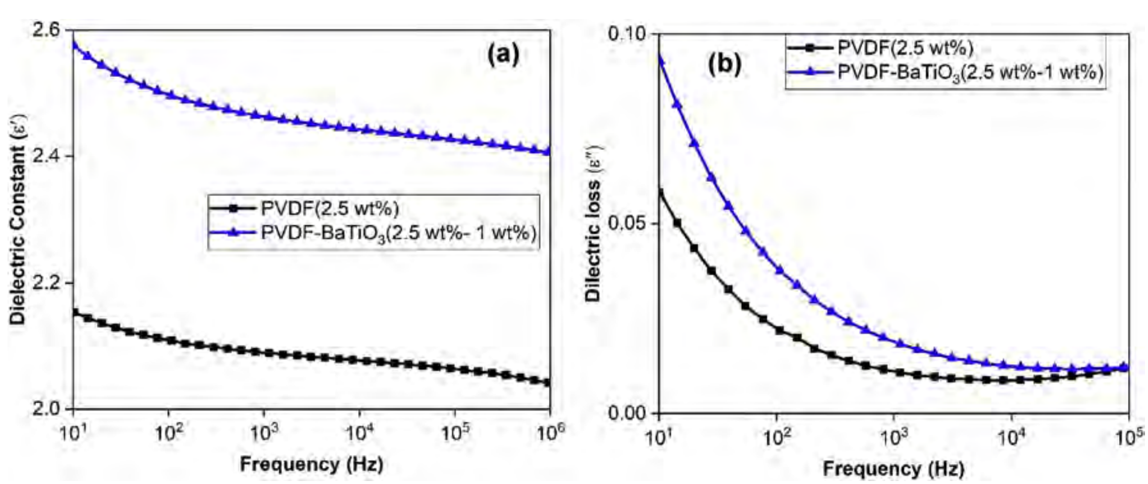


Fig. 4. Variation in (a) dielectric constants (Ɛ′) and (b) dielectric losses (Ɛ″) of PVDF and PVDF-$BaTiO_3$ composite films with frequency. Both measurements are recorded at 25 °C.

Table 2
Response and recovery times of PVDF-$BaTiO_3$ composite humidity sensors.

Material	Fabrication Method	Response time (s)	Recovery time (s)	Measure. range	Sensitivity
Polymeric RMX/$BaTiO_3$ composite [21]	Screen printing	15	120	33–98% RH	N/A
$BaTiO_3$-PMMA composite [22]	Sintering method	120	60	30–98% RH	1.9pF/%RH
$BaTiO_3$/polystyrene sulfonic sodium composite [23]	Spin coating	50	120	33–98% RH	N/A
Piezoelectric pMUT array with GO sensing layer [24]	Drop casting	78	54	10–90% RH	0.719kHZ/%RH
ZnO nanosheets [25]	HTG method	600	3	12–96% RH	220
PVDF-$BaTiO_3$ (Present work)	Spin coating	40	25	40–90% RH	0.2416 pF/%RH

composites in comparison to the data obtained in this work. These times are significantly shorter for the proposed sensor compared to other reported sensors.

4. Conclusion

The effect of $BaTiO_3$ particles on the thermal stability, morphology, hydrophilicity, and humidity sensing response of the PVDF-$BaTiO_3$-based humidity sensor is investigated at 40–90% relative humidity level. PVDF-$BaTiO_3$ (2.5 wt%-1 wt%) composite films exhibit regular and even distribution of $BaTiO_3$ in the composite films in comparison to the composite films with other ratios of PVDF and $BaTiO_3$. The wettability analyses of the composite films show that acetone treatment reduces the contact angle and enhances the hydrophilicity of the films. Dielectric study of the composite film demonstrates that the addition of $BaTiO_3$ increases the dielectric constant and decreases the dielectric loss. PVDF-$BaTiO_3$ (2.5 wt%-1 wt%) composite-based humidity sensors exhibit a stable and more linear capacitive response with reduced hysteresis over the 40–90% RH range in comparison to the sensors with 0.5 and 2 wt% of $BaTiO_3$. The optimized capacitive sensor shows 2.5% maximum hysteresis.

Declaration of competing interest

The authors declare no conflict of interest.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.ceramint.2019.09.291.

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