Flexible Pressure Sensor Based on Carbon Black/PVDF Nanocomposite

Abstract:

A piezoresistive flexible pressure sensor was fabricated using carbon black and Poly (vinylidene fluoride) (CB/PVDF) composite. The conductive CB/PVDF composite was prepared by a wet-cast method and deposited into a flexible polyethylene (PE) substrate. The surface morphology, crystal structure, and material properties were studied using SEM and X-ray diffraction methods. This flexible pressure sensor was tested in a wide pressure range of about 0 -76 kPa and its response time was less than 0.43 s. The sensitivity, response time, and recovery time were studied for different pressures and vibration modes. The repeatability and reproducibility characteristics of the sensor were studied and found that the sensor exhibits excellent characteristics. The sensor was subjected to different loading and unloading pressures and the resistance of the sensor remained stable indicating that the sensor had a high degree of reproducibility. Owing to its flexible properties and the material used, the proposed device can be applied in the next generation of smart sensors for biomedical, robotic, and automotive sensing applications.

Keywords: flexible pressure sensor, carbon black, PVDF, composite, solvent, piezoresistive.

1. Introduction

Sensors with distinct characteristics such as lightweight, high flexibility, and high sensitivity are attracting a wide range of applications. In particular, flexible pressure sensors are increasingly applied in different fields such as electronic skin, health monitoring, robotics, advanced prosthetics, virtual reality entertainment technology, and sports [1-7]. Novel materials such as conductive polymers, carbon nanomaterial, and carbon black are mainly used in these flexible pressure sensors because they possess excellent electrical conductivity, chemical stability, and low toxicity and are widely available [8-12]. These novel materials are deposited on polymeric substrates such as polyimide (PI), polydimethylsiloxane (PDMS), polyethylene terephthalate, and PE for flexible pressure sensor applications [3, 13-15]. These novel materials replace the metal foils and semiconductors that were traditionally used as the sensing platform. The rigidity and high cost of the old materials make them inefficient for current practical applications.

The sensing mechanism of the pressure sensors can be divided into different categories. The four main categories are piezoresistive, piezoelectric, capacitive, and transistor [16-18]. Piezoelectric pressure sensors have high sensitivity and stability but have limited applications in pressure measurements because they cannot measure static pressure. The surface charge produced by the applied force in the piezoelectric materials can be neutralized easily by current leakage, charges from the environment, or the resistance from the connected electronics making them require high pass filters. [14, 19-22]. Another disadvantage of these sensors is that they are sensitive to temperature, most piezoelectric materials are pyroelectric, therefore they are temperature sensitive, and the temperature may induce crystal deformation, which may in turn produce unwanted forces [22]. The piezoelectric pressure sensors are the most utilized sensors due to their remarkable advantages including simple fabrication, stability, and reliability, and they have high pixel density to measure large pressure ranges [23]. The conductive sensing material is an important parameter in evaluating the variation rate of the resistance in these resistive sensors. To improve their sensitivity, porous materials such as porous elastomers and electrospun nanofiber membranes have been used to increase the deformation of the piezoresistive sensors. Polymer materials are suitable polymer nanomaterials, including carbon nanotubes, graphene, nanowires, and other metal nanomaterials.

Various publications on piezoresistive materials, using different binders, and nanoparticles on different substrates, have been reported in the past on their sensitivity, hysteresis, reproducibility, and resistance performance. For instance, Zhang et al. experimented on carbon black/silver nanoparticles (CB/AgNPs) based strain sensors on polyuarethane substrate. They found that the sensitivity improved by 18 times for CB/AgNPs composite compared with bare carbon-based strain sensors [24]. Chang et al. developed a flexible resistive-type pressure sensor using carbonized cotton fabric (CCF). The sensors showed a large measurement range (~0.16 kPa), a low detection limit (~0.70 Pa), and excellent durability (>4000 cycles) [25]. For fluid pressure sensing on a curved microtube, Yao et al. developed a piezoresistive flexible sensor using micropatterned conductive carbon nanotube as the sensing element. A high sensitivity (0.047 kPa^{-1} in gas sensing and 5.6×10^{-3} kPa^{-1} in liquid sensing) and low consumption (<180 μW) were reported [26]. Kweon et al. fabricated PVDF-HFP/ PEDOT composited-based pressure sensors using 3D electrospinning. These samples indicated higher pressure sensitivity compared with conventional electrospinning (13.5 kPa^{-1} for 3D electrospinning whereas 5.1 kPa^{-1} for conventional). The sensors also showed a minimum detection of 1 Pa and were resilient up to 10,000 compressive cycles [27].

This paper reports the experimental results of a simple, low-cost, and highly sensitive piezoresistive flexible pressure sensor using CB/PVDF composite as the sensing element. The composite was prepared by the wet casting process and deposited on a flexible polyethylene substrate. The pressure vs resistance response was recorded, and it resulted in good stability, sensitivity, and repeatability. The details of the experimental process and the results obtained will be covered in the subsequent sections.

2. Materials and Methods

The materials used for the preparation of the sensing element are CB nanoparticles, PVDF powder, and DMF. The material properties of these materials were taken from the manufacturer's literature. Table 1 gives a summary of the material properties of CB and PVDF. CB was used as the conductive phase, PVDF was used as a binder and DMF was used as the solvent to mix up the composition.

Table 1 Properties of carbon black and PVDF used for the preparation of the sensing element.

	Specific surface area (m^2/g)	**Resistivity (Ω.cm)**	**Young's Modulus (MPa)**	**Dielectric constant**
Carbon Black	780	0.2	1.5 X10^5	
PVDF	3.76 **-** 6.61	$> 10^{14}$	2450	7.5-13.2

The process of fabricating the film and preparing the CB/PVDF nanocomposite is illustrated in Fig.1. First, CB nanoparticles were blended in DMF solvent, followed by mechanical stirring and sonication to attain a thoroughly dispersed CB suspension. A PVDF solution was prepared by dissolving 50 mg PVDF powder in the DMF solvent. The solution was thoroughly stirred using the magnetic stir bar until the powder was completely dissolved in DMF solvent. The dispersed CB solution was then combined with PVDF solution in a mass ratio of 1:1 as shown in the schematic in Fig. 1 (a-c). This ratio was chosen in order to have a high carbon: binder ratio and to provide clarity on the analysis. The binder also helps in forming a uniform film that is mechanically stable when loading and unloading pressure is applied. The solution was vigorously stirred to ensure homogenous dispersion of the conductive nanocomposite. This composition was also ultrasonicated for 1 hour with a constant interruption of 20 minutes to avoid the agglomeration of nanocomposites. The PE substrate was then prepared according to standard practice for the preparation of surfaces of plastics to enable adhesive bonding of the CB/PVDF composition. The physical surface preparation of the substrate involved wiping with methanol, sanding, wiping with a clean dry wipe, and then wiping with the methanol wipes again [28].

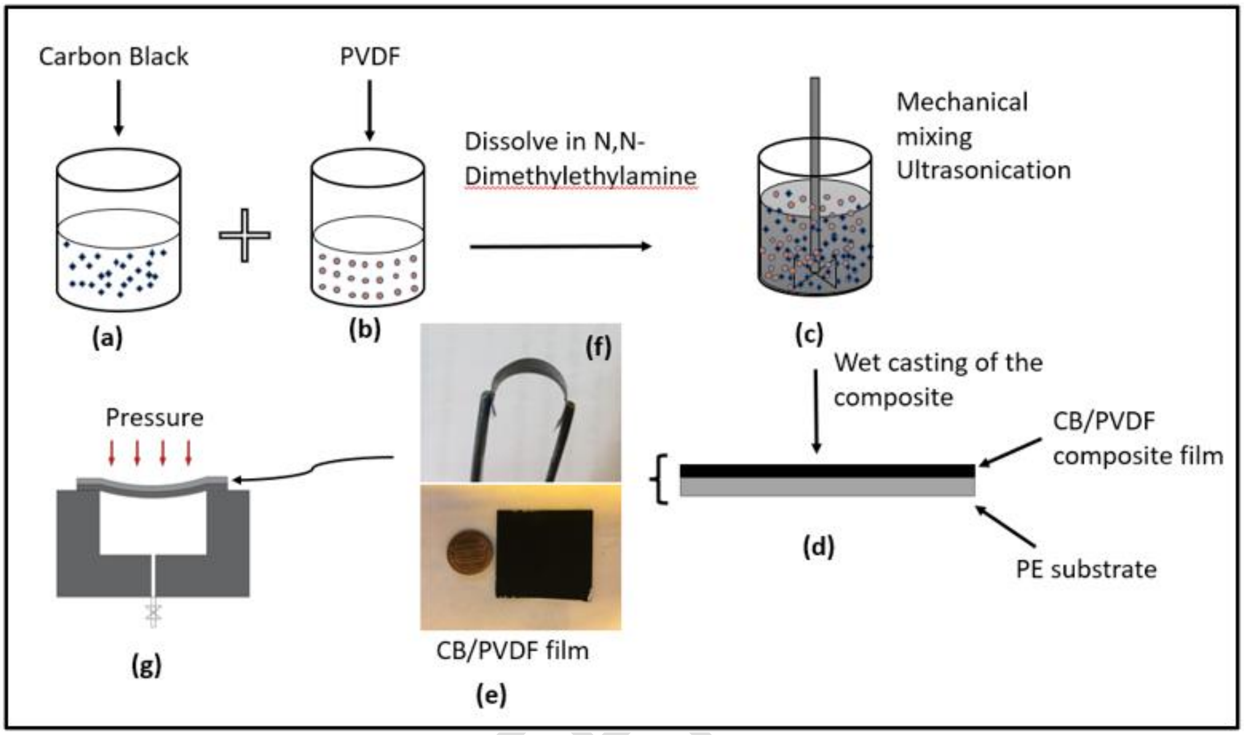

Fig. 1. The schematic illustration for the preparation of CB/PVDF composite film for fabrication of pressure sensor.

The CB/PVDF nanocomposite was wet-casted onto a flexible PE substrate to form a thin CB/PVDF film of about 0.1 mm (Fig. 1. d). The film was then placed on a warm plate at 30 ^{0}C for 20 minutes to evaporate the extra DMF solvent, furthermore, the film was set to air dry in the room for 12 hours. A sensitive thin film of CB/PVDF was formed and it had a thickness of about 30 microns, the film was etched to form a thin strip of about 2 mm thick. The dried nanocomposite film was integrated into a (20 × 20 × 10) mm acrylic PMMA cubic with an opening at the center to create a sensing device. The center diameter was measured to be 5 mm with a depth of 5 mm (Fig.1 g). The CB/PVDF conductive film was then mounted to the surface containing the 5 mm hole in the acrylic PMMA cube (Fig. 1. g). A high-pressure compressor was then connected to the acrylic PMMA fixture through the valve attached to the 5 mm hole. The pressure gauges were mounted to regulate the applied pressure on the CB/PVDF film. When the gauge pressure was applied through a valve, the thin CB/PVDF membrane was deformed into a dome-shaped structure. The radius of the dome was a function of pressure and hence the strain experienced by the film gave rise to resistance in the material, which changed as a function of applied pressure. The resistance of the film was recorded using a Keithley multimeter connected to a computer through a custom-made LabVIEW interface. The response graphs obtained are a function of resistance and time.

3. Results and Discussion

a. Morphology of Materials/Characterization

The surface morphology of the fabricated film was investigated using scanning electron microscopy (SEM). Here, small magnification (Fig.2(a)) and large magnification (Fig.2(b)) are given to understand the microstructure of CB/PVDF films. The SEM images in Fig.2 indicate a good dispersion of CB/PVDF particles. The uniform distribution of the conductive and binder matrix plays a key role in determining the piezoresistive property of the composite film. The uniform dispersion is influenced by the particle size, shape, and orientation of the particles. The XRD pattern has been carried out on CB/PVDF composite in the ratio by mass percentage is shown in Fig. 3. It is clear from the figure that there were peaks of PVDF corresponding to a 2 θ angle of 20.16 and 39.84. The presence of a dominant peak at 20.160 confirms the presence of beta-phase in PVDF material. The XRD pattern confirms the semi-crystalline nature of PVDF polymer film [29]. The crystalline size for the PVDF-110 peak was found to be approximately 0.89 nm using the Scherrer equation. The diffused peak of carbon black was observed at an angle of 23.8^0 (002) plane, this diffused peak may be caused by the lack of crystallinity of the carbon black and polymer mixture at this angle. The PVDF restricts the diffract of the film at this angle due to its polymeric structure, the peaks of PVDF dominated the CB as indicated by the XRD spectra [30-32].

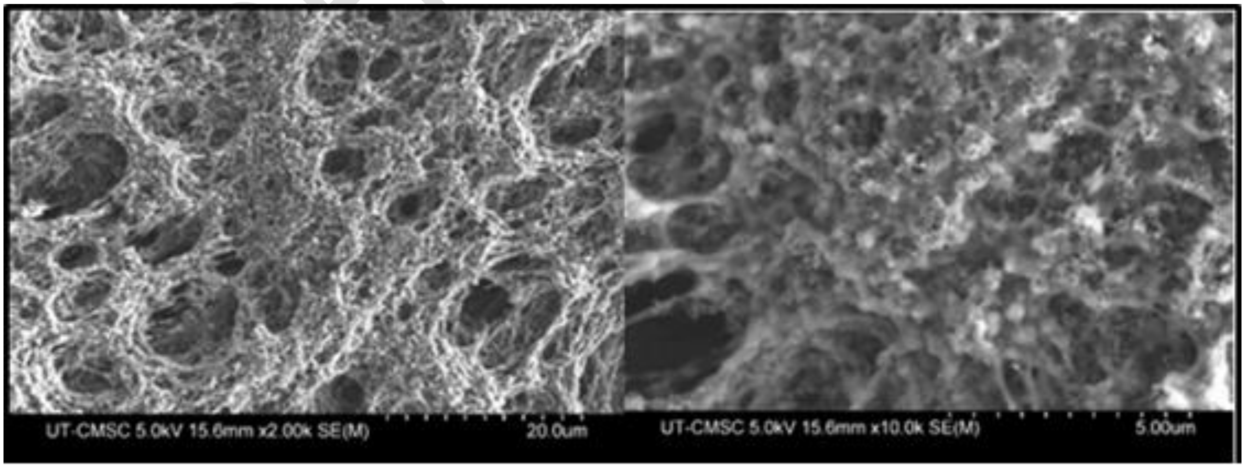

Fig. 2. The surface morphology of the CB/PVDF composite was investigated using a Scanning Electron Microscope (SEM).

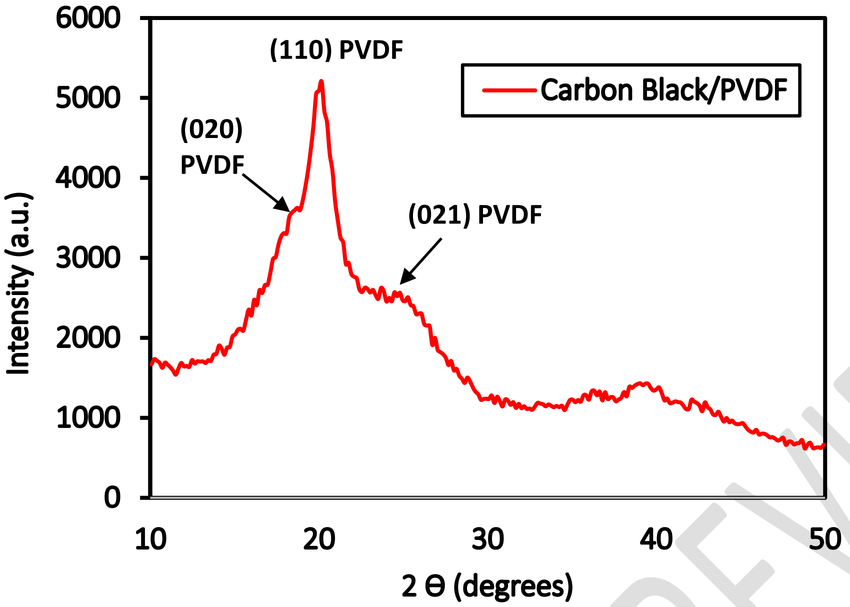

Fig. 3. X-ray diffraction patterns of CB/PVDF.

b. Pressure Sensing Characterization Sensing Mechanism

Fig. 4 shows the time response for CB/PVDF film when vacuum pressure was applied. It was noted that when pressure was applied the film resistance decreased. This decrease in resistance was because the film diaphragm deformed causing compressive stress on the film, the uniaxial pressure causes the gaps between two adjacent conductive particles to be smaller which leads to a decrease in the film's electrical resistance [33]. The PVDF binder helps the active material to mitigate the stresses of contraction and to maintain the adhesion of the CB to the conductive network. The compression destroys the conductive paths of CB by causing the transverse slippage of carbon black particles which contributes to an increase in composite resistance [33]. The electrical resistance of a single conductive path can be described by the equation below [34]:

$$R = \frac{2h^2 sL}{3a^2 e^2 \sqrt{2m\varphi}} exp\left(4\pi\sqrt{2m\varphi}\frac{s}{h}\right) \qquad (1)$$

Where, *R, L, m, e, h, s,* φ*, a*^{2} are the resistance of the single conductive path, the number of particles forming the conductive path, the electron mass, the electron charge, the Plank's constant, the thickness of insulating film, the height of potential barrier between adjacent particles, and the effective cross-sectional area respectively.

When the pressure was applied on the thin CB/PVDF film, the resistance (response) decreased with time, and when the gauge valve was turned off the response increased abruptly. Eq. 2 was used to calculate the recovery time on the exponential curve of this pressure response [35]:

$$R = R_0 + A_1\, exp\left(-\frac{t}{t_1}\right) \tag{2}$$

Where, R_O and A_1 are constants, while t and t_1 are the time response and time constants, respectively. The calculated average time constants (t_1) are 0.43 s when the valve was turned off (recovery) and 0.138 s when pressure was initially applied to the film. The time response values were estimated from the linear fit of the exponential curves. The higher recovery time t_1 can be attributed to a slow air intake on the valve but the rapid increase of the air in the chamber resulted in a small t value.

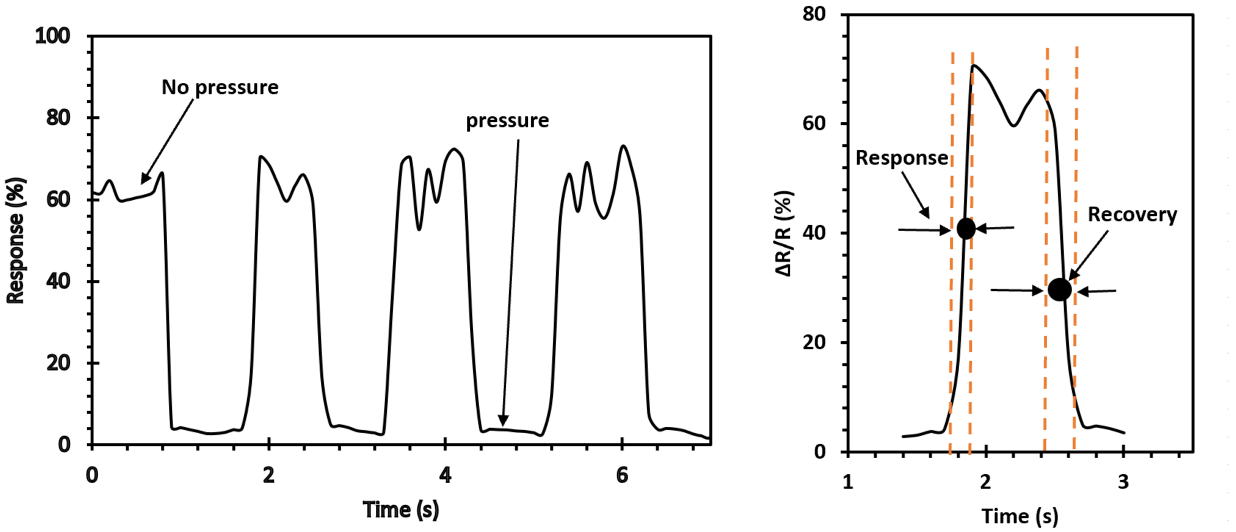

Fig. 4. Sensing performances of the pressure sensor with a thin CB/PVDF membrane under a pressure of 76 kPa. The right enlarged view is one of the responses for clarity showing the response and recovery of the sensor.

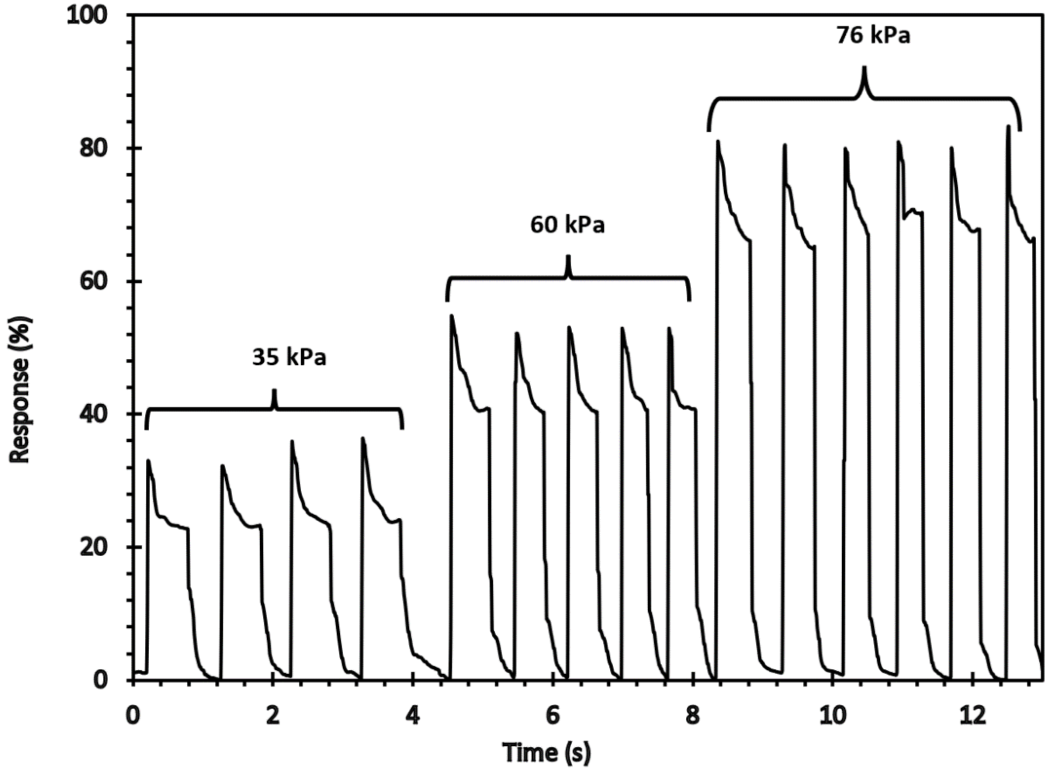

Fig. 5. Time response and repeatability test for CB/PVDF composite with different vacuum pressures applied.

The sensor was then subjected to different pressures as shown in Fig. 5, this test was used to study the repeatability and reproducibility of the sensor. The sensing element was subjected to loading and unloading for several cycles and the response at different pressures was recorded. The mean response at different pressures was recorded and marked as shown in Fig. 6. The sensor exhibited a linear response to applied pressure, as indicated by a high coefficient of determination ($R^2 = 0.9668$), which signifies the consistent and predictable relationship between the applied pressure and the sensor's output. This linearity is important for the calibration of the pressure sensor. The trendline closely aligning with the error bars enhances the credibility of the sensor's measurements, indicating minimal deviation from the expected values. The linear curve of the CB/PVDF nanocomposite pressure sensor is a critical characteristic that highlights the sensor's precision and reliability. This characteristic is important in applications where accurate and proportionate pressure sensing is needed.

The sensitivity of the CB/PVDF pressure sensor to applied pressure was defined within the range of 0 – 76 kPa. The sensor exhibited a high sensitivity of 0.9611% kPa^{-1}, indicating its capacity to detect small variations in pressure and convert them into proportional changes in electrical output. The ability to detect small changes in pressure is important, especially in applications where accuracy and responsiveness are critical, such as in the development of piezoresistive pressure sensors. The reported sensitivity indicates the potential of the CB/PVDF nanocomposite as a key component in advanced sensing technologies, promising reliable and accurate pressure measurements across a broad pressure spectrum.

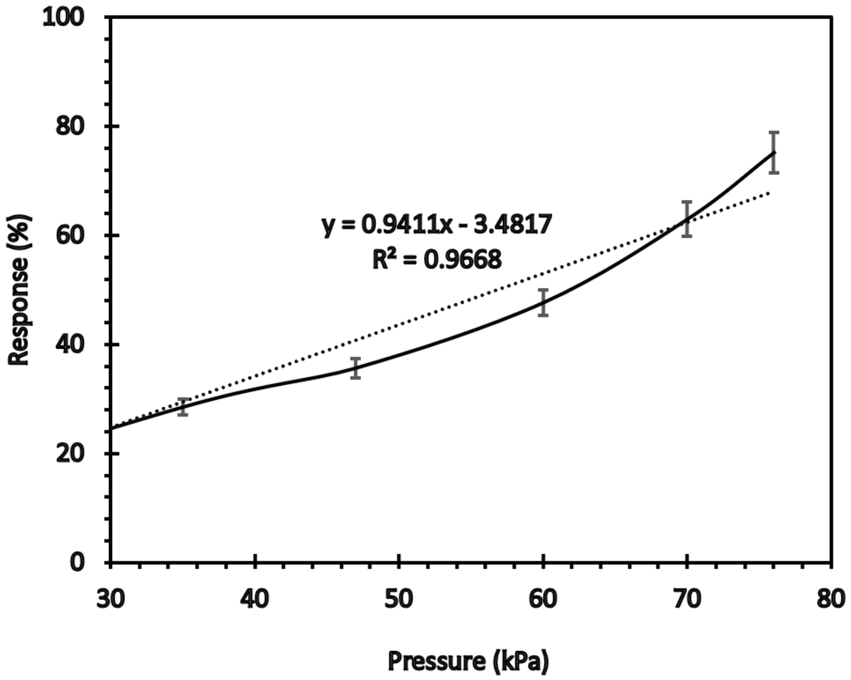

Fig. 6. Dependence of piezoresistive response on the pressure with error bars for the experimental data.

The response remained stable after a series of loading-unloading tests at the same pressure, this indicated that CB/PVDF had a high degree of repeatability and reproducibility which could be attributed to the firmness of the CB conductive layer. The sensing element was also evaluated by subjecting it to a vibrating device that had different vibrating modes suitable for the test. The vibration response was given in Fig. 7. showing that the sensor had a high degree of reproducibility and repeatability. The calculated frequency with that specific vibration mode was found to be about 16 Hz. Subjecting the sensing element of carbon black/PVDF nanocomposites to a vibrating device with different modes provides a comprehensive assessment of the sensor's performance, reliability, and suitability for applications in dynamic and vibrating environments. It aids in optimizing the sensor's design and materials to meet the specific demands of diverse real-world scenarios [36].

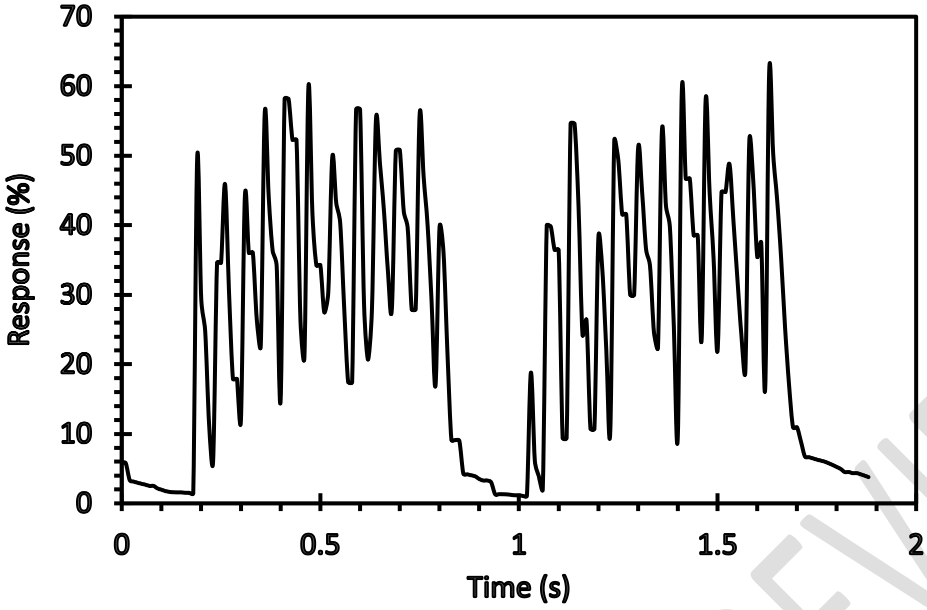

Fig. 7. Vibration response of the sensor when the thin CB/PVDF membrane was mounted to a vibrating device, the frequency was estimated to be about 16 Hz.

Combining CB and PVDF nanocomposites can leverage the strengths of both materials, creating sensors with enhanced properties such as improved sensitivity, selectivity, and response times. CB/PVDF composites exhibit high sensitivity due to the excellent electrically conductive nature of CB and the high surface area, providing a large active area for interactions with analytes. The piezoelectric properties of PVDF also enhance the sensitivity thus resulting in accurate pressure sensing. Kai et al prepared PVDF nanocomposites filled with CNTs and CB by melting them using a small-scale compounder. They were able to demonstrate that the sensor prepared hard improved conductivity and piezoresistive sensitivity. Their composite combines both a relatively wide strain sensing range as well as a high sensitivity [37].

The flexibility and lightweight nature of the nanocomposite contribute to faster response times compared to some traditional sensors. These properties are important for applications where sensors need to be utilized on complex and curved surfaces such as in biomedical and robotics. The fabrication process and the scale of production are crucial factors in achieving a cost-effective outcome for this sensor. Additionally, the materials used are relatively cost-effective, positioning this sensor as competitive and advantageous when compared to existing sensor technologies [38, 39].

The unique property combination of CB and PVDF nanomaterial opens avenues for advanced sensing technologies. To fully optimize this sensor performance, researchers should explore techniques that enhance sensitivity, optimize mechanical properties, cost-effective fabrication techniques, and promote multifunctionality. Notably, the annealing process has been shown to improve the electrical conductivity of the nanocomposite, further enhancing its overall performance [40]. The nanocomposite can also be integrated with smart materials and investigate energy efficiency designs of the sensor. Further work can be done on exploring wearable health monitoring devices through improved human-machine interfaces. Additionally, lightweight and fast response time flexible pressure sensors can be integrated into automotive safety systems, such as smart airbags or seat occupancy sensors. Finally, interdisciplinary collaboration and innovative approaches will be instrumental in unlocking the full potential of carbon black/PVDF nanocomposites.

4. Conclusion

A flexible pressure sensor was designed based on CB/PVDF nanocomposite and fabricated by the wet-cast technique. The nanocomposite was deposited onto a flexible PE substrate with remarkable flexibility. The addition of PVDF into the conductive CB not only enhanced the adhesion of the active material but also provided a crosslink between CB particles. This composite material exhibited a substantial change in resistance during loading, corresponding to film stretching. PVDF also provides an excellent mechanical property essential to generate a sensor signal under loading and unloading cycles. In-depth investigations into the dynamic characteristics revealed that the sensor has a high sensitivity, rapid response time, and the ability to measure a wide pressure range. The linear response to the applied pressure ($R^2 = 0.9665$), indicates the sensor's linearity, a crucial feature for calibration. The pressure was applied within a range of 0 – 76 kPa and showed a sensitivity of 0.9611% kPa^{-1}. The average response and recovery times were determined to be 0.43s and 0.138s, respectively. The reported pressure sensor demonstrates its high potential for developing piezoresistive pressure sensors and applications in flexible electronics such as wearable devices to monitor the movements of the human body.

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