Soft Capacitive Pressure Sensor With Enhanced Sensitivity Assisted by ZnO NW Interlayers and Airgap

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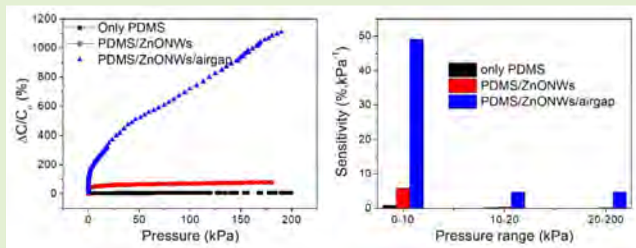
***Abstract*—Highly sensitive capacitive pressure sensors with wide detection range are needed for applications such as human-machine interfaces, electronic skin in robotics, and health monitoring. However, it is challenging to achieve high sensitivity and wide detection range at the same time. Herein, we present an innovative approach to obtain a highly sensitive capacitive pressure sensor by introducing a zinc oxide nanowire (ZnO NW) interlayer at the polydimethylsiloxane (PDMS)/electrodes interface in the conventional metal-insulator-metal architecture. The ZnO NW interlayer significantly enhanced the performance with ∼7 times higher sensitivity (from 0.81%kPa^{-1} to 5.6452%kPa^{-1} at a low-pressure range (0-10 kPa)) with respect to conventional capacitive sensors having PDMS only as the dielectric. The improvement in sensitivity is attributed to the enhanced charge separation and electric dipole generation due to the displacement of Zn^{+} and O^{-} under applied pressure. Further, the orientation of ZnO NWs and their placement between the electrodes were investigated which includes either vertical or horizontal NWs near the electrodes, placing a third ZnO NW interlayer in the middle of dielectric PDMS and introducing an air gap between the ZnO NWs/electrode. Among various combinations, the introduction of air gap between the electrode and ZnO NW interlayer revealed a significant improvement in the device performance with ∼50 times enhancement at a low-pressure range (0-10 kPa) and more than 200 times increase at a high-pressure range (10-200 kPa), in comparison with the conventional PDMS-based pressure sensor.**



***Index Terms*—Flexible pressure sensors, capacitive sensors, ZnO NW, electronic skin, PDMS, soft touch sensor.**

I. INTRODUCTION

HUMAN skin can detect external stimuli such as pressure, strain, chemicals and temperature etc. because of the presence of various receptors at different depths and positions within the skin [1]–[3]. For an electronic skins (e-skin) with the capability to mimic the functionalities of the human skin, it is essential to have an array of flexible and stretchable sensors. The research on e-skin has led to rapid development of sensing technologies, particularly related to flexible pressure sensors, which are needed in a wide range of applications such as health monitoring, tactile displays, wearable systems etc. [4]–[9]. There are different types of pressure sensors based on the sensing mechanism used, such as, capacitive, piezoelectric, piezoresistive and triboelectric etc. [10]–[14]. Among these the capacitive pressure sensors are widely used because of their simple fabrication, low cost, high sensitivity, good stability and simple read out electronics [15], [16].

The structure of a typical capacitive sensor includes a dielectric material sandwiched between the two parallel metal conducting plates [17]. The output of these sensors is highly dependent on the properties of the dielectric material. Typically, capacitive sensors have rigid dielectric material, but in the case of e-Skin elastomeric materials such as Ecoflex, polydimethylsiloxane (PDMS), and polyurethane (PU) are commonly used [18]–[20] as they enable soft touch. Further, they have good thermal and chemical stability, biocompatibility, and mechanical properties. Several methods are adopted for the fabrication of high-performance capacitive pressure sensors using these elastomeric dielectric materials. These methods have either used microstructured pyramids, pillars or wrinkles in the dielectric material or the fibrous dielectric materials with porous structures and an air gap. In some cases,

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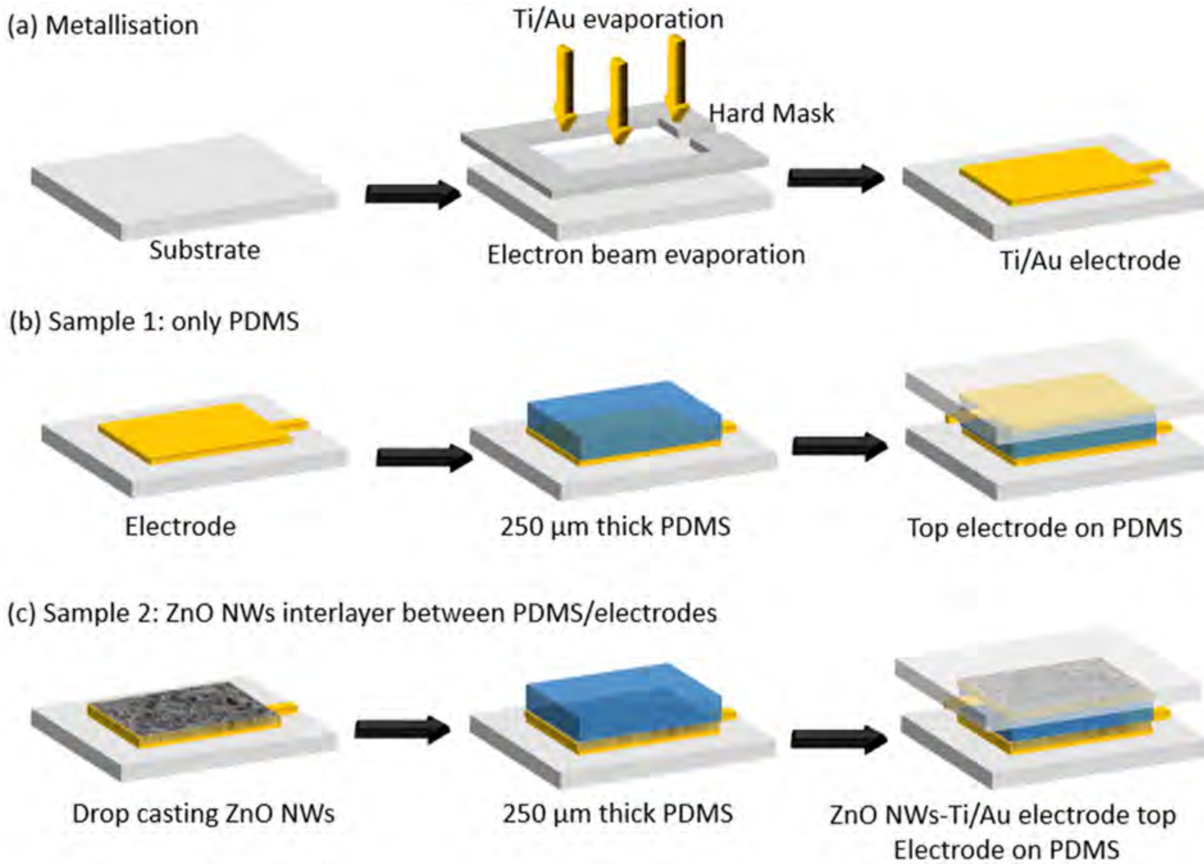

Fig. 1. Schematic diagram showing the fabrication of capacitive pressure sensor. (a) Electrode realization with electron beam evaporation. Fabrication of capacitive pressure sensor (b) without and (c) with ZnO NWs interlayer.

a composite dielectric incorporating filler material such as carbon nanotubes, silver nanowires (NW) etc. in the elastomeric matrix have also been explored [12], [21]–[25]. For example, micro-arrayed PDMS structure based capacitive pressure sensors show better sensitivity (0.6-2 kPa^{-1}) compared to the flat PDMS (0.01-0.016 kPa^{-1}) in the low-pressure region (0-2 kPa) [26], [27]. Likewise, porous dielectric materials using three dimensional (3D) sacrificial templates have been achieved by mixing a sacrificial salt inside PDMS [28]. The capacitive sensor based on porous microstructures exhibited excellent sensitivity of 44.5kPa^{-1} for pressures less than 100Pa [29]. It has been reported that the porosity and microstructures are responsible for an abrupt capacitance change under small pressure [30]. Similarly, the combination of porous dielectric material with air gaps have been explored to enhance the sensitivity [31]. However, the fabrication of air gap with microporous structures requires complicated fabrication process such as lithography techniques and silicon-based molds/templates. Further, a reduction in the working pressure range was also observed for porous and microstructure-based sensors [32], [33]. To enhance the sensitivity as well as the working pressure range, filler materials such as carbon nanotubes, graphene, graphite, silver nanowires, high dielectric nano particles etc. are inserted within the dielectric elastomeric material. Similarly, piezoelectric materials like ZnO nanostructures are embedded in the composite material to enhance their performance [18]. However, to achieve a repeatable sensing performance, a homogeneous and uniform dispersion of the filler materials in the elastomeric dielectric matrix is required which is highly challenging.

In this work, to address the existing challenges, we present a novel design of capacitive pressure sensor by introducing ZnO NW interlayers and an air gap between PDMS/electrodes interfaces to enhance the sensitivity and working pressure range. The sensing performance of capacitive pressure sensors without ZnO NW interlayers (named as sample 1) and with the ZnO NW interlayers (named as sample 2) were investigated by measuring the relative change in capacitance under applied pressure. This work is the extension of our recent paper presented at IEEE FLEPS 2021 [34]. Here we have carried out detailed study by investigating the influence of ZnO NW orientation and their location, which includes either placing third ZnO NW interlayer in the middle of PDMS or introducing a 150 μm air gap between ZnO NWs and the Ti/Au electrode (named as sample 3). An increase in sensitivity by 7 times was observed in sensors with the introduction of ZnO NW interlayers compared to only PDMS based sensors in the low-pressure range (0-10 kPa). This is a significant rise in the sensitivity when compared with the recent reports [35], [36]. Interestingly, the sensitivity of the device was significantly improved at both pressure ranges (low pressure range and high-pressure range) after introducing the air gap with an unprecedented linear sensitivity of ~4.7%kPa^{-1} between 10-200 kPa pressure.

This paper has been organized as follows: Section II presents the fabrication methodology for the capacitive pressure sensors. The results related to the device characterization and performance have been discussed in Section III and the key outcomes of this work have been summarized in Section IV.

II. Materials and Methods

Fig.1 shows the fabrication scheme for the various sensors realized in this work. Two sensors with different metal-dielectric-metal architecture were fabricated using a standard metallization (Fig.1a) and spin-coating process; one without ZnO NW interlayers (Fig.1b) and other with ZnO NW interlayers (Fig.1c). The two types of samples were developed to study the impact of ZnO NW on their capacitance. Initially, 10/80 nm thick Ti/Au contact electrodes were deposited on to ~175μm-thick polyvinyl chloride (PVC) sheet, using an electron-beam (E-beam) evaporator. The dimensions of top and bottom electrodes were defined using the hard mask containing a small opening of 1 cm x 1 cm to facilitate the deposition of Ti/Au in the defined region. Finally, a dielectric layer, namely PDMS, prepared by mixing the elastomer and curing agent in 10:1 ratio was sandwiched between the electrodes as shown in Fig. 1b and 1c.

In the case of sample 1, a 250 μm-thick PDMS layer was directly spin-coated over one of the electrodes (bottom) and annealed at 80°C for 15 mins. Subsequently, the top electrode was firmly placed over the semi-cured PDMS layer with an intention to achieve a proper adhesion as shown in Fig 1b. Likewise, sample 2 was prepared by drop-casting ZnO NWs over both electrodes prior to the spin-coating of PDMS layer (Fig. 1c) and then the entire device was assembled. The drop casted ZnO NWs were annealed at 80°C for 10 mins (Fig. 1c). Then, 250 μm-thick PDMS layer was spin-coated on the top of previously made ZnO NW/bottom electrode. Subsequently, another ZnO NW coated top electrode was placed on the top of semi-cured PDMS/ZnO NW-bottom electrode. In doing so, we can take advantage of piezoelectric behavior of ZnO NWs to enhance the amount of electric current transferred from the electrodes.

Device characterization for the prepared sensors was carried out by placing the devices under a single point 1004 aluminium load cell, and the force was applied using a square-shaped plastic probe attached to a linear stage driven by a motor having a resolution of ~0.1mm. To read the output of the sensors, its electrodes were attached to an E4980AL precision LCR meter (Keysight Technologies, Santa Clara, CA, USA), and a PC running a LabVIEW 2018 Robotics v18.0f2 (National Instruments, Texas, USA) was used to record the changes in thee capacitances of devices under different loading conditions.

III. Results and Discussion

Fig. 2 shows the scanning electron microscope (SEM) image of sample 2. The densely distributed drop casted ZnO NWs over the metal electrode are clearly captured in the cross-sectional SEM image shown in the inset of the Fig. 2a. The PDMS layer spin coated on the top of ZnO NWs-based Ti/Au electrode forms a smooth top surface (Fig. 2b). Over this, the top electrode was placed in a way that the pressure sensitive PDMS layer gets sandwiched between the two ZnO NWs-based Ti/Au electrodes (Fig. 2c).

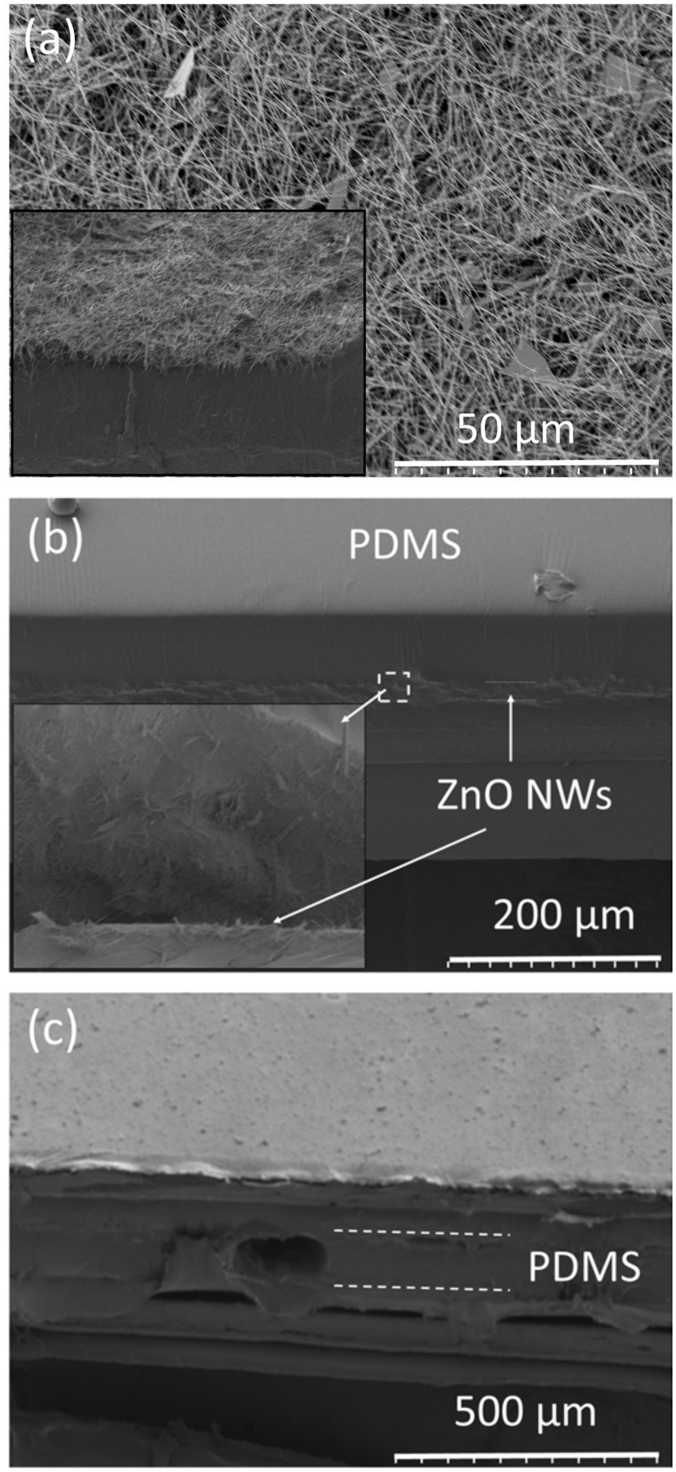

Fig. 2. (a) The cross-sectional SEM image of capacitive pressure sensor with the PDMS layer sandwiched between ZnO NWS/Ti/Au electrodes (sample 2). (b) SEM top view of drop casted ZnO NWS on Ti/Au electrode and its cross-sectional image in the inset. (c) The cross-sectional SEM image of PDMS layer spin coated over the ZnO NWS/Ti/Au electrodes and its magnified image in the inset.

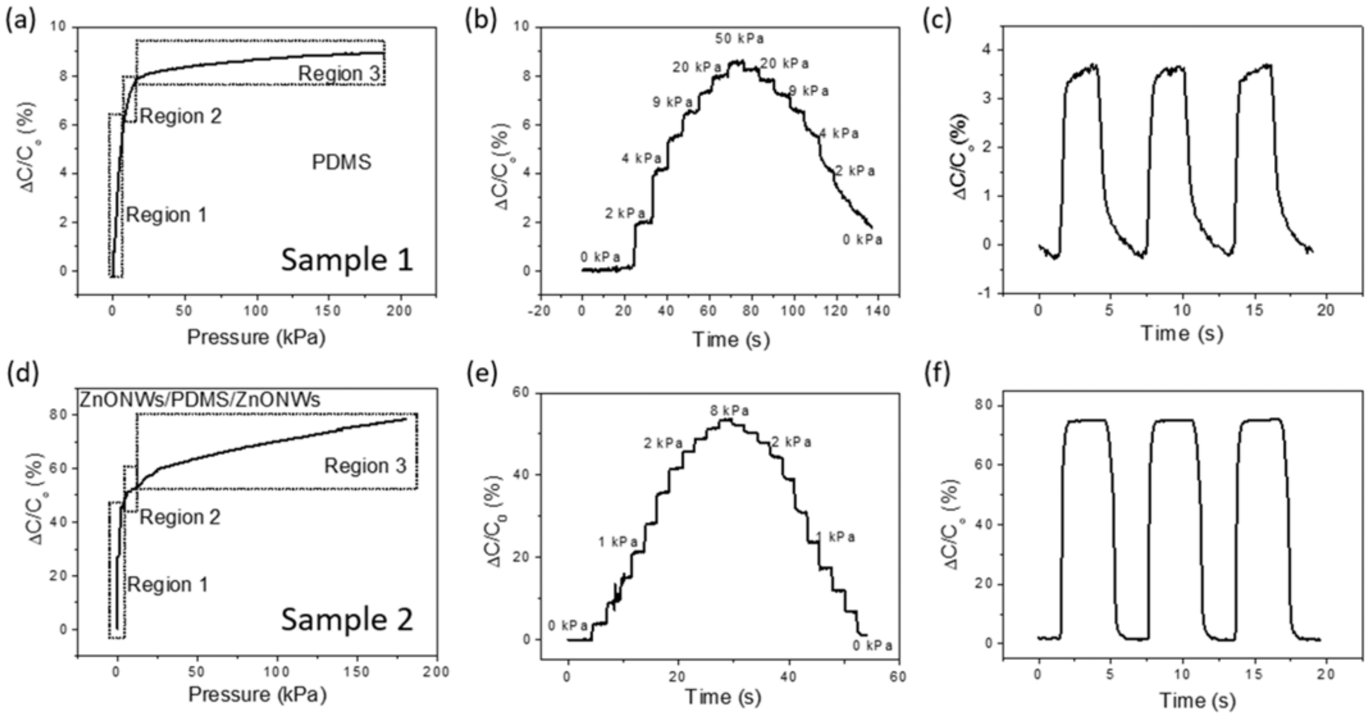

Fig. 3. Characterisation of capacitive pressure sensor with and without ZnO NWs interlayers. (a-c) Sensor characteristics of PDMS-based pressure sensor without ZnO NWs interlayers: (a) relative change in capacitance with respect to the applied pressure, (b) step response under various pressure varied from 0 to 50 kPa, and (c) the cyclic response at 4 kPa. (d-f) Sensor characteristics of PDMS-based pressure sensor with ZnO NWs interlayers: (i) relative change in capacitance with respect to the applied pressure, (ii) step response under various pressure varied from 0 to 8 kPa, and (iii) the cyclic response at 150 kPa.

To evaluate the sensor performance in terms of the influence of ZnO NWs interlayers between the electrode and PDMS, both samples were subjected to different pressure ranges varied from 0 to 200 kPa and their relative change in capacitance were measured as shown in Fig. 3a and 3d. Further, the step response was observed under systematic loading/unloading (Fig. 3b and 3e) and the cyclic response at constant applied pressure (Fig. 3c and 3f). In both the samples, the response curve (Fig. 3a and 3d) shows three linear regions in the pressure range varied between 0-10 kPa, 10-20 kPa and 20-200 kPa, in which the sensors demonstrated high response at low pressure region. This is common due to the elastic nature of PDMS. At low pressure region, the elasticity of PDMS is good and results in large capacitance variation. On the contrary, the elasticity of PDMS reaches its limit at the high-pressure range between 20 to 200 kPa which resulted in small capacitance variation. The standard sample (sample 1 without interlayers) revealed a relative change in capacitance of less than 10% between the applied pressure range (0-200 kPa). On the other hand, the sample with ZnO NW interlayers (sample 2) revealed a relative change in capacitance of ∼80% for the same pressure range.

Based on these observations, it can be said that the performance of the capacitive pressure sensor has been significantly improved by the addition of ZnO NWs interlayer. This performance enhancement is attributed to the piezoelectric effects introduced by ZnO NW interlayer placed next to the electrodes to reinforce the electrical connection from the electrodes to the dielectric material (Fig. 4a). The response of the capacitive pressure is significantly affected by two parameters - the change in distance between the electrodes of MIM structure and the dielectric constant. The base capacitance of the PDMS-based sensor is 8pF/cm^2, while the ZnO-PDMS-based sensor is 6.9pF/cm^2. Similar decrease in the base capacitance with the incorporation of ZnO nanoparticles inside the PDMS has been reported earlier [18]. This change in base capacitance is attributed to the chance in dielectric constant between the pure-PDMS and ZnO NW-PDMS. In standard PDMS-based pressure sensor, the change in capacitance is directly related to the change in distance between the two electrodes in the metal-insulator-metal (MIM) architecture under the applied pressure. This phenomenon is due to the physical deformation of the elastomeric dielectric layer. Accordingly, the change in distance is very small for the low-pressure range. Therefore, the relative change in capacitance is less than 7% at pressure range below 10 kPa. However, in sample 2, ZnO is known to become polarized under an external electric field due to its non-central symmetrical structure [37], [38]. In this case, under the influence of vertical pressure, ZnO NWs become more compact. Meanwhile, the induced charge carriers are generated because of the polarization effect which sequentially interact with PDMS and strengthens its dielectric property due to the Maxwell-Wagner-Sillars interfacial polarization [39]. As a result, at low pressure, the charge separation and electric diploe generation [40] takes place and the relative change in capacitance is enhanced to 60% at 10 kPa which is more

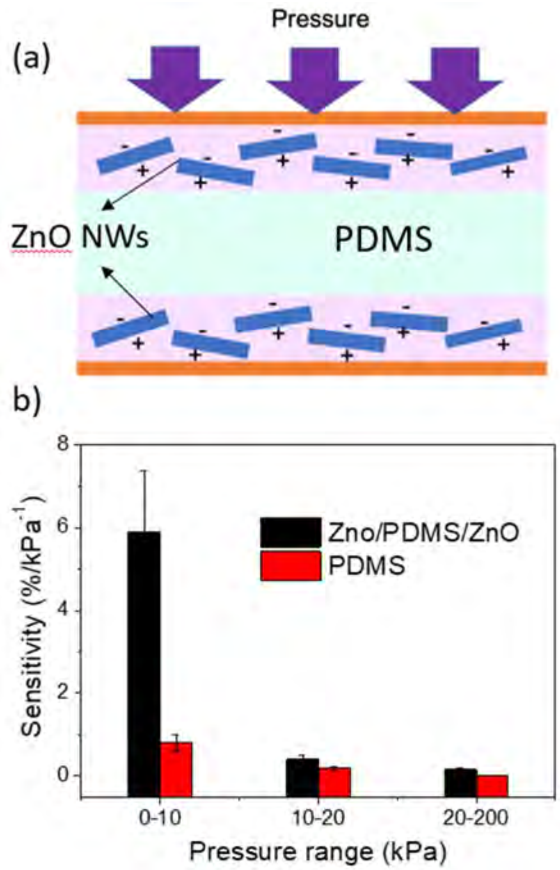

Fig. 4. (a) Schematic representing the working mechanism of ZnO NW interlayer-based capacitive pressure sensor. (b) Comparison of sensitivity under various pressure range for sample 1 and sample 2 extracted from Fig 3a and 3d.

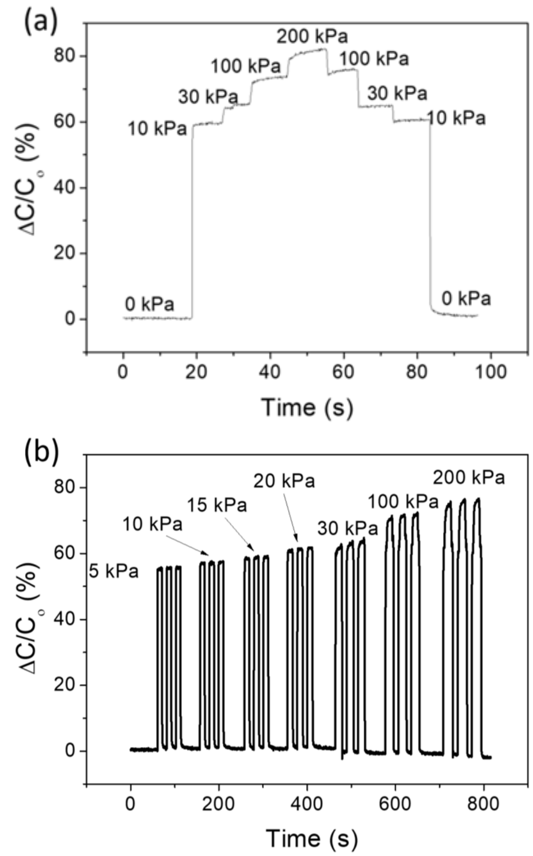

Fig. 5. Sensor characterisation of Sample 2. (a) Relative change in capacitance with stepwise increase/decrease in applied pressure varied from 0 kPa to 200 kPa. (b) Relative change in capacitance with various loading and unloading cycles under pressure range varied from 0, 5, 10, 15, 20, 30, 100 and 200 kPa.

than 7 times higher than the standard PDMS based pressure sensor. In other words, the change or increase in dielectric constant under applied pressure is due to the polarization effect of ZnO NWs and the change in distance between the electrodes of MIM structure results in the performance enhancement of PDMS-ZnO based sensor. Additionally, the device sensitivity was extracted from Fig. 3a and Fig 3d by using the equation (1) [41]:

$$S = \delta[(C - C_0)/C_0]/\delta P \tag{1}$$

where, C and C_0 are the capacitances with and without applied pressure, and P is the amount of pressure applied. The sensitivity for different pressure ranges (1-10 kPa, 10-20 kPa, 20-50 kPa) was then derived from Fig. 3a and 3d. As shown in Fig. 4b, the PDMS-based capacitive pressure sensor with ZnO NW interlayers shows a higher sensitivity for all the pressure ranges varied from 0 to 200 kPa, indicating a dramatically improved sensing performance. Particularly, at the lower pressure range, from 1-10 kPa, sample 2 demonstrates the much highest sensitivity of 5.6452%kPa^{-1}, which is significantly higher when compared with the 0.81%kPa^{-1} from sample 1. In addition, it is noticeable from Fig. 3b-c and 3e-f that sample 2 shows a higher stability and reliability by presenting a full recovery response, less noise and a larger range of capacitance change.

Fig. 5a shows the step response of the sample over a long pressure range of loading/unloading varied from 0, 10, 30, 100 and 200 kPa. The sensor demonstrated negligible hysteresis with similar change in capacitance value during loading and unloading steps. Moreover, the reliability of sample 2 over various pressure ranges was assessed by carrying out a cyclic loading test by loading and unloading the samples using 5, 10, 15, 20, 30, 100 and 100 kPa at a frequency of 0.33Hz (Fig. 5b). The sample 2 demonstrated a stable performance over various pressure values. Finally, the long-term stability of the sensor was evaluated by subjecting the sensor to loading and unloading cycle under 30 kPa at 0.33 Hz frequency for 1100 sec (Fig. 6 and supporting video V1). Based on the observation, the addition of ZnO NWs interlayer significantly enhanced the sensor performance in comparison with the standard PDMS-based pressure sensor with reliable performance over long cyclic test.

The influence of ZnO NW orientation and their location between the electrodes were also investigated by fabricating three different combinations: 1) the capacitive pressure sensor with vertically grown ZnO NW at both the electrodes; 2) the placement of third ZnO NW layer in the middle of PDMS in addition to the drop casted NW layers on both the electrodes

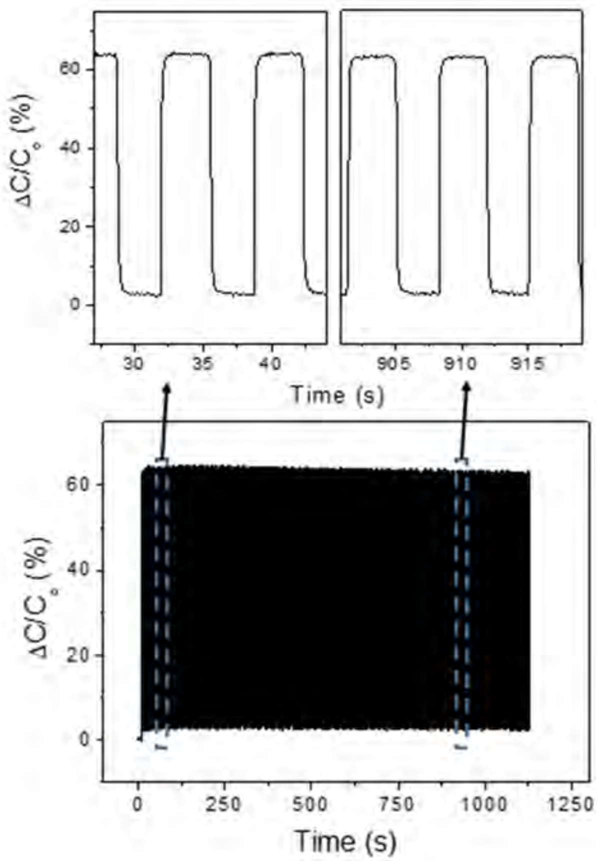

Fig. 6. Relative change in capacitance under cyclic loading and unloading with 30 kPa pressure at 0.33Hz frequency for 1100 sec and its magnified graph between 30-40 sec and 905-915 sec at the top.

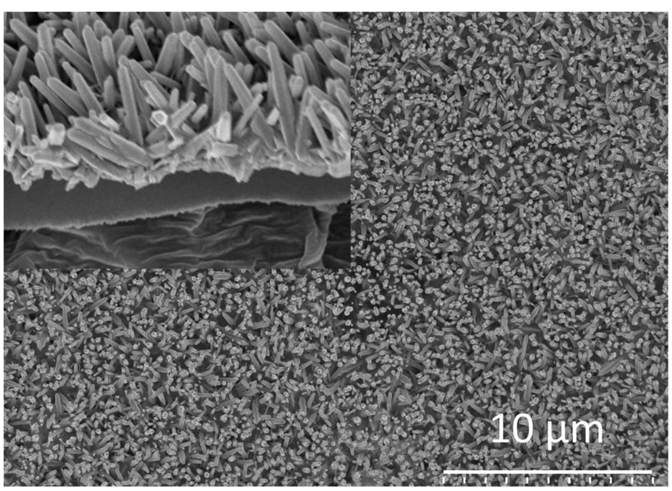

Fig. 7. SEM image of vertically grown ZnO NWs on Ti/Au electrode and its cross-sectional image in the inset.

(named as three-layered ZnO NWs); and 3) the introduction of 150 μm thick airgap between the ZnO NW and the top-electrode (Sample 3). In case of vertically grown ZnO NW based pressure sensor, instead of drop casting, the ZnO NWs were directly grown on the Ti/Au electrode as shown in Fig. 7. In all the combinations, 3 batch of devices were fabricated to understand the device-to-device variation (Fig. 8 and 9). The sensor response of vertically grown ZnO NW based pressure sensor was measured by subjecting it to pressure range from 0 to 200 kPa range. Fig. 8a shows the relative change in the capacitance with respect to applied pressure. The sensor with vertically grown ZnO NW interlayer demonstrated similar trend as sample 2 with negligible variation in the relative change in capacitance. Among three devices, the device-to-device variation is less than 10%. Further, the sensor demonstrated stable step response (Fig. 8b) and reliable cyclic response under various pressure range including 2.5, 5, 10, and 30 kPa (Fig. 8c). Likewise, the sensor performance of three-layered ZnO NW-based device was characterised by measuring the change in capacitance when subjected to the external pressure of 0- 200 kPa as shown in Fig. 8d. The step response and the cyclic response at different step pressure were investigated as shown in Fig. 8e and 8f. We noticed that neither the orientation of ZnO NWs nor the inclusion of third layer in the middle of PDMS have any significant influence on the sensor performance with respect to sample 2. Overall, the addition of ZnO NW interlayer enhanced the sensor performance in comparison with the pristine PDMS based pressure sensor.

In case of sample 3, a 150 μm airgap between the ZnO NWs and top-metal electrode was introduced by placing spacer on the top electrode as shown in Fig. 9a. The performance of the sensor was evaluated by measuring the change in capacitance under the pressure varied from 0-200 kPa (Fig. 9b). Interestingly, the capacitance of the device drastically increased from ~760 fF to >9 pF with a relative change in capacitance of ~1100% at 200 kPa pressure, which is ~14 times higher than the ZnO NW-PDMS hybrid pressure sensor without the airgap. The reason for such a large change in capacitance is the mechanical deformation and the change in dielectric property of the dielectric layer [31], [42]–[44]. In previous reports, the increase in air gap/porosity has been found to decrease the dielectric constant of the material,which reflects their base capacitances [30], [42], [45]. Accordingly, the presence of air gap increased the distance between both the electrodes which eventually decreased the base capacitance from 6.9 pF (without airgap) to 760 fF (with airgap). While subjected to the external pressure, the airgap reduced and the electrode come in contact with the ZnO NWs embedded on the PDMS surface. Sequentially, the piezopolarisation of the ZnO NWs takes place to significantly increase the capacitance value after certain pressure range. Due to these reasons, the sensor with the air gap have large mechanical deformation and the dielectric change in comparison with its non-air gap counterpart, when subjected to same pressure range. Accordingly, the device demonstrated significant increase in capacitance from 700 fF to 2pF at 10 kPa pressure as shown in the supporting video V2 and the Fig. 9c. The cyclic performance of the device was measured by subjecting the sensor to different cyclic pressure varied from 1.5, 3, 5, 20, 25 and 50 kPa (Fig. 9d and 9e). The device demonstrated stable performance under cyclic pressures with negligible hysteresis (Fig. 9f). Similar performance enhancement using the airgap-based structures have been reported but they are limited to the low-pressure range [30], [45]. It is worth mentioning that with the influence

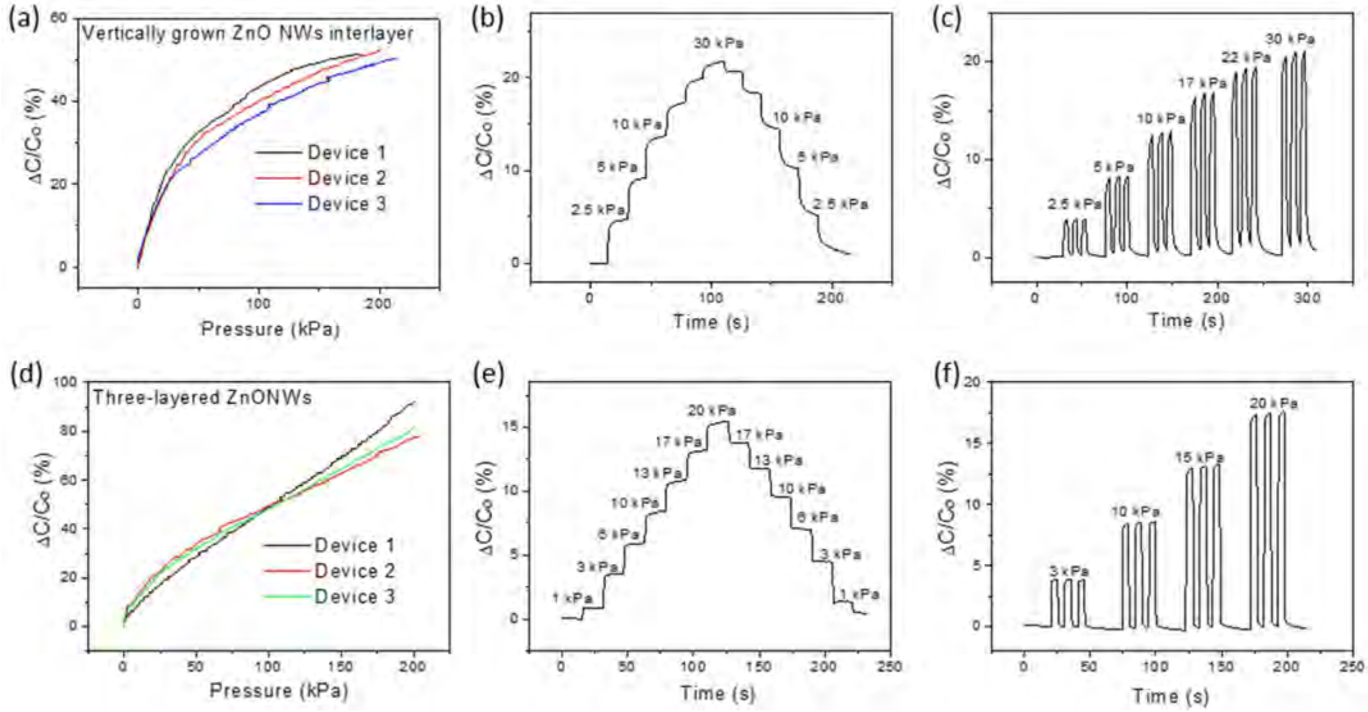

Fig. 8. (a-c) Sensor characterisation of vertically grown ZnO NW interlayers: (a) Relative change in capacitance at different applied pressure varied from 0 kPa to 200 kPa, (b) the step response and (c) the cyclic response under different loading/unloading cycles. (d-f) Sensor characterisation of three-layered ZnO NWs: (d) Relative change in capacitance at different applied pressure varied from 0 kPa to 200 kPa, (e) the step response and (f) the cyclic response under different loading/unloading cycles.

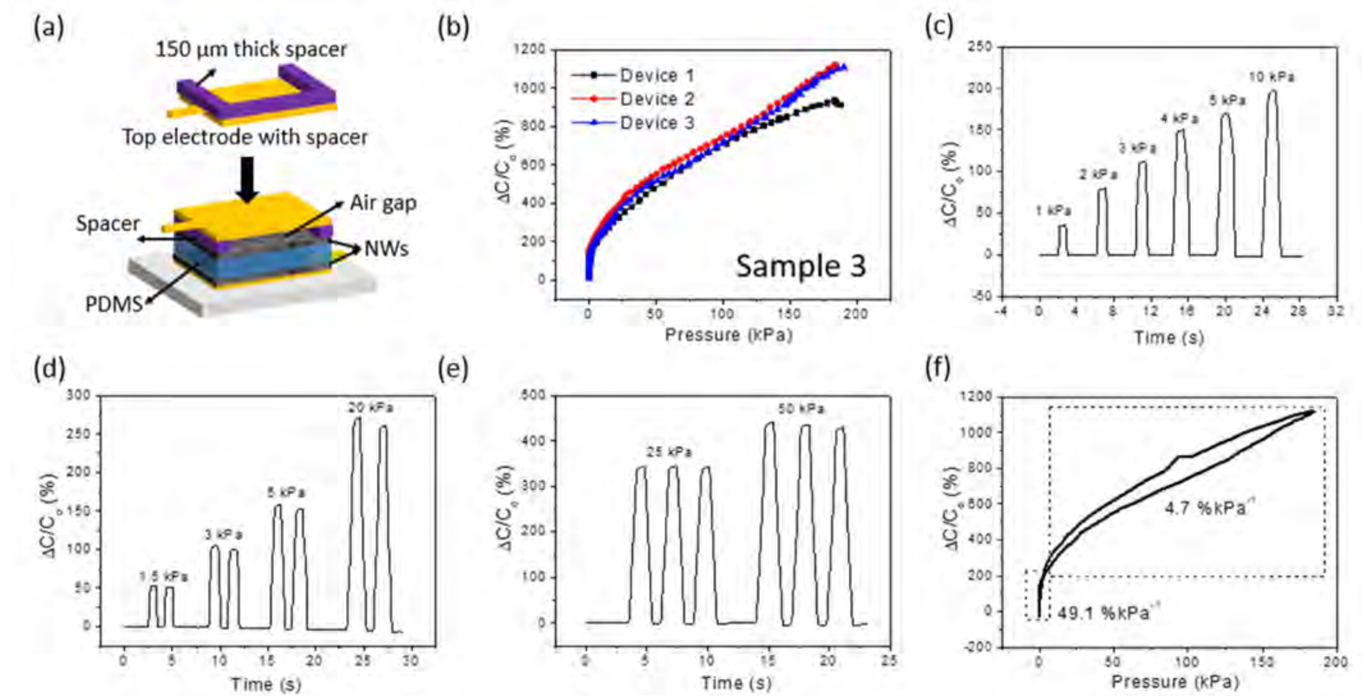

Fig. 9. Sensor characterisation of ZnO NW interlayers based pressure sensor with 150 μm thick airgap: (a) Schematic representation of pressure sensor with the ZnO NW interlayer and airgap, (b) relative change in capacitance at different applied pressure varied from 0 kPa to 200 kPa, (c-e) step response under loading/unloading cycles at different pressure range varied from (c) 1, 2, 3, 4, 5, and 10 kPa, (d) 1.5, 3, 5, and 20 kPa, and (e) 25 and 50 kPa and (f) hysteresis curve while loading and unloading from 0-200 kPa pressure range.

of ZnO NWs and air gap, our sample 3 demonstrated superior performance with an unprecedented sensitivity of 49%kPa^{-1} at low pressure range (0-10 kPa) and 4.7%kPa^{-1} at high pressure range (10-200 kPa).

IV. Conclusion

In this work, we presented two PDMS-based capacitive pressure sensors with and without ZnO NW interlayers and investigated the effect of ZnO NW on the output. The device performance was evaluated by comparing the experimental results and it is found that the pressure sensor with ZnO NW interlayer exhibited an enhanced capacitance, sensitivity, and reliability due to its piezoelectric effects. The ZnO NW orientation and their placement between the electrodes were investigated with three batches containing vertical grown ZnO NW interlayer, by placing third layer in the middle of PDMS dielectric and introducing air gap between the ZnO NW/electrode. It is observed that the introduction of airgap decreased the base capacitance and increased the sensor performance. A high-sensitivity ($\sim$49%kPa^{-1}) of the capacitive pressure sensor was demonstrated in the pressure range of 1-10kPa. Further investigation in terms of the density, NW orientation and the number of layers can be carried out to tune the performance of presented capacitive pressure sensor to improve their usefulness for mimicking the functionality of human skin.

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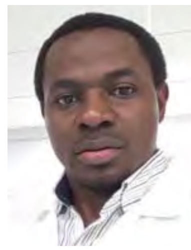

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