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A highly sensitive biodegradable pressure sensor based on nanofibrous dielectric

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ABSTRACT

High performance biodegradable electronic devices are becoming popular due to their enormous applications as biomedical implants or short term communication devices. They are biocompatible and eco-friendly. For example wearable or implantable biodegradable pressure sensors are very useful for cardiovascular or blood flow monitoring. But unfortunately very few previous researches have focused on developing biodegradable pressure sensors. Here we propose a composite of Polylactic-co-glycolic acid (PLGA) and Polycaprolactone (PCL) for biodegradable piezo-capacitive pressure sensor fabrication for low pressure measurement in tactile ranges of ($0 < P < 5$ kPa). An electrospun PLGA-PCL composite membrane has been used as elastomeric dielectric sandwiched between two biodegradable iron-zinc (Fe-Zn) bilayer electrodes, deposited on degradable Polyvinyl alcohol (PVA) substrate using electron beam deposition, and encapsulated in PLGA thin films for device fabrication. PLGA-PCL nanofibrous dielectric membrane is highly compressible and porous having tunable mechanical as well as dielectric properties and has been reported first ever for biodegradable pressure sensor application. The sensitivity of the sensor was found to be 0.863 ± 0.025 kPa^{-1} in the low pressure region ($0 < P \leq 1.86$ kPa) which is quite high as compared to previous literature on biodegradable sensors so far while it adapted to a value of 0.062 ± 0.005 kPa^{-1} for high pressure region (1.86 kPa $< P \leq 4.6$ kPa). The lowest detected value was 1.24 Pa (at 10mgf). Sensor showed good average response and recovery times of 251 ms and 170 ms respectively. In vitro degradation studies of the sensor were performed in PBS solution and sensor lost 60% of its initial weight during first two weeks of degradation and continued to degrade even after that. After one week of incubation sensor showed a 19.5% decrease in low pressure measurement range while there was no significant difference in upper detection range. It has been used to measure the arterial pulse wave on wrist and a 4×4 pressure sensor array was made to demonstrate its use in mapping different pressure points.

1. Introduction

Biodegradable electronics have played an important role in developing a new class of wearable, small sized, light weight and ecofriendly devices which is a big shift in terms of biotechnological advancements [1–3,8,10,11]. These devices are made up of completely biodegradable materials and they function for a certain period of time then degrade leaving no harmful leftovers [4,5]. They are biocompatible with human body as compared to current devices which being much advanced and high performing are laced with toxic materials like precious metals harmful to the human health and the environment around. This makes conventional devices incompatible for medical purposes where they may possibly be in direct contact with human body or being used for monitoring the food items. Sensing devices are quite often used outside the body in medicinal treatments, but the ones that are surgically implanted into the tissues or organs have the drawback of painful surgical removal which can lead to damaging or infecting the particular area and long recovery time. On the other hand biodegradable sensing devices help improve usefulness of implantable devices and reduce waste associated with wearable and point of care diagnostics [6,13]. There are quite a few examples of biodegradable sensing devices which were used for medical purposes [14,15] and monitoring the food items [7].

Pressure sensors are one of the most important vital monitoring devices. Several types of pressure sensors including piezocapacitive

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[1,16–26], piezoelectric [6,27–31], and piezoresistive [2,3,32–44] type have been developed for their potential uses in various applications including health monitoring devices, electronic skins, human machine interfaces, and soft robotics. A lot of them are currently under evaluation for market ready applications. Capacitive pressure sensors are mostly popular because of simple and easy to fabricate structure, their stability and cost effectiveness. Many non-biodegradable of piezo-capacitive pressure sensors have been reported recently having polymeric composites as dielectric material in different structures to change the dielectric properties upon application of pressure. Only a few attempts have been made to make completely biodegradable capacitive pressure sensors. For example, Luo et al. reported a radio frequency (RF) based pressure sensor using costly fabrication techniques of electroplating and lithography in a complex parallel plate type structure suffering from poor performance in low pressure region [12]. Boutry et al. reported a highly sensitive biodegradable pressure sensor using micro-structured biodegradable elastomer Poly Glycerol Sebacate (PGS) as dielectric layer for cardiovascular monitoring [9]. Curry et al. have reported a biodegradable piezoelectric force sensor using Poly L-Lactic Acid (PLLA) film as piezoelectric sensing layer [6]. But similarly Kwon et al. have reported a flexible wearable capacitive pressure sensor (not biodegradable) based on a microporous elastomeric dielectric for this purpose [28]. Micro-structured dielectric approach or dielectric casting approach in these biodegradable devices account for changing the dielectric response of the sensing materials. But micro-structuring the dielectric is a complicated and improvident process. In some of the recent researches nanofiber membranes (NMs) are being used as dielectric layers [1,20,25] for pressure sensor applications as well as active layer for strain sensor applications [45] due to their excellent flexibility, inherent high porosity, lightweight and exceptional compatibility with printed electronics [1,46]. Polymeric nanofiber membranes are mostly prepared using electrospinning which is an efficient, and cost effective method. Pressure sensors using NMs as dielectric membranes showed good sensitivities and reliable performance for the specific applications.

This work focuses on employing the NM of a biodegradable composite PLGA-PCL, prepared using state of the art electrospinning technique as dielectric layer for completely biodegradable pressure sensor fabrication. It has tunable mechanical, dielectric, as well as degradation properties because the composition can be changed to any desired ratio promising enormous potential for biodegradable electronics. Pure Iron and Zinc have been chosen as electrode materials for bilayer (Fe-Zn) electrodes with iron just to promote adhesion of main electrode to the substrate. Electrodes have been deposited onto the commercially available PVA substrate using e-beam evaporation process. PLGA having a dielectric constant in the range of 3.3–4.4 for different molecular weights [47] has been chosen as the main constituent of the composite to impart desired dielectric properties while PCl as secondary constituent to impart mechanical properties in small optimized ratio. The proposed composite nanofibrous dielectric has required mechanical (around less than 10 MPa modulus of elasticity) and degradation properties [48,49] guiding to use low concentrations for PCL for the sensor fabrication and performance in low pressure measurements.

The device was prepared in a layered structure comprising of the biodegradable dielectric nanofibers sandwiched between the biodegradable electrodes on the biodegradable substrate. Finally it was encapsulated in biodegradable PLGA film using-spin coating to avoid rapid degradation of the actual substrate. The device (n = 3) has a sensitivity of 0.863 ± 0.025 kPa^{-1} in low pressure region and 0.062 ± 0.005 kPa^{-1} for high pressure region over a tactile pressure range of 0–4.6 kPa. It showed good average response time of around 251 ms and average recovery time around 170 ms. In the end, the sensing device has been utilized for measuring arterial pulse on wrist artery. Also it has been used for spatial pressure distribution by making a 4×4 array and custom built instrumentation. Mostly the characteristic experiments and analyses were performed using the device before degradation studies. In vitro degradation studies of the sensor were performed in PBS medium kept in incubator and sensor lost 60% of its initial weight during first two weeks of degradation due to relatively fast degradation of PVA substrate after degrading PLGA encapsulation and it further continued to degrade as observed until 18 days. Sensor's performance gradually decreased with degradation at first and then became quicker when the main substrate PVA started degrading which showed the device degradation time can be controlled by encapsulation layer of PLGA without compromising much of the device's performance. And after one week of incubation sensor showed a 19.5% decrease in low pressure measurement rang while there was negligible effect on sensitivity for upper detection range. All the materials used are FDA approved and given the controlled degradation concept makes the device suitable for in-vivo applications as well. It can also find many other applications as well like biodegradable capacitive touch modules, touch sensitive displays and pressure sensitive electronic skins etc.

2. Methods

2.1. Materials

Poly-lactic acid-co-glycolic acid (PLGA(75:25), Mw = 66000–107000) and Polycaprolactone (PCL, M_w = 10,000) were bought from Sigma Aldrich to be used as the dielectric material for sensors fabrication. 2,2,2-Trifluoroethanol to be used as solvent for ink preparation was also bought from Sigma Aldrich. A commercially available 30 μm thick PVA sheet has been used as the biodegradable substrate for biodegradable pressure sensor. Pure Fe and Zn both in granular form (for bilayer electrodes fabrication) (for e-beam deposition process) were bought from Kurt J. Leskar Company.

2.2. Fabrication of sensor

The commercially available 30 μm thick PVA sheet, after plasma treatment for 5 min to modify surface properties for electrodes deposition, was masked at the non-deposition area with inkjet-printed sticker paper readily available. After that a 10 nm thin layer of Fe followed by 100 nm layer of Zn was deposited using the e-beam evaporation system to make bilayer electrodes onto the substrate. Thin layer of Fe was used to promote the adhesion between Zn electrodes and the main substrate. Two as prepared electrode-substrate samples were used to make a sandwich like structure for the sensor later. For solution electrospinning of the dielectric material first a high viscosity ink of PLGA:PCL in 8:2 was prepared in 2,2,2-Trifluroethanol. 0.8 g of PLGA and 0.2 g of PCL were dissolved in the 7.3 g of solvent and stirred magnetically for 6 h at 40 °C and 1500 rpm to form homogeneous solution. This ink formulation was chosen after comparing with 5:5 PLGA:PCL composition (which resulted in non-uniform fiber diameters, no performance improvement and could have possibly resulted in long degradation time of the composite membrane because of more PCL content). Then this high viscosity ink was used in electrospinning process (Applied Potential: 8.7 kV, Collector distance: 14 cm, Flow rate: 0.4 ml/hr) to form ~8 μm thick membrane of nanofibers and cut into 10mm*10 mm pieces as the composite biodegradable dielectric layer. After making electrical wiring connections, for measurement, using silver epoxy to the electrodes the device was glued in a sandwich type structure after placing dielectric membrane using PVA solution as adhesive. At the end it was encap-

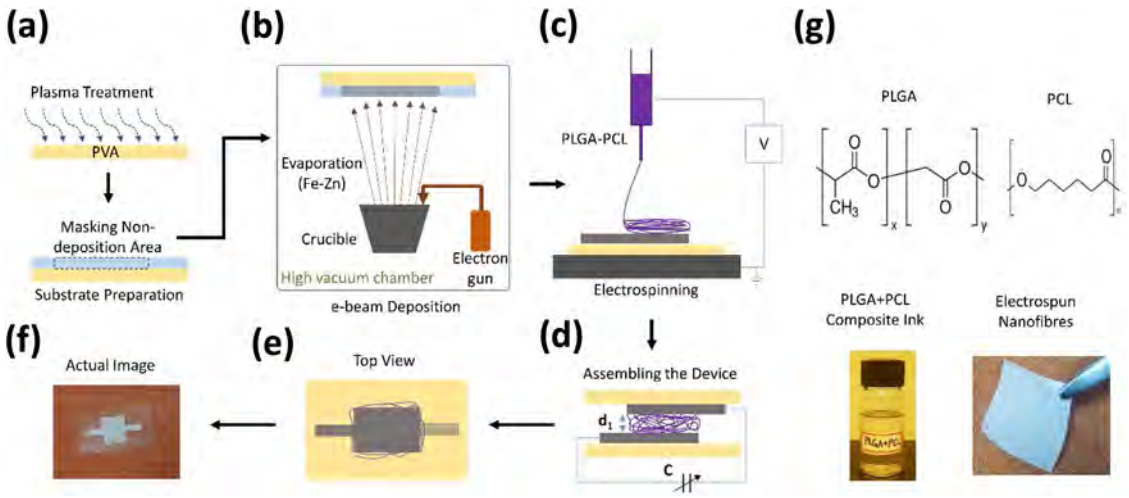

Fig. 1. Fabrication process of biodegradable Pressure Sensor with structures of the components of dielectric membrane shown on right – **(a)** Substrate preparation (plasma treatment for 5 min and then masking the non-deposition area) for electrodes deposition **(b)** e-beam deposition 10 nm of Fe and 100 nm of Zn bilayer electrodes on to the masked substrate **(c)** Electrospinning the PLGA-PCL solution to form nanofibers membrane **(d–f)** Assembled device using PVA as adhesive and encapsulated using PLGA solution (schematic and actual images) **(g)** Structure of individual constituents, composite ink and prepared nanofibers of PLGA-PCL.

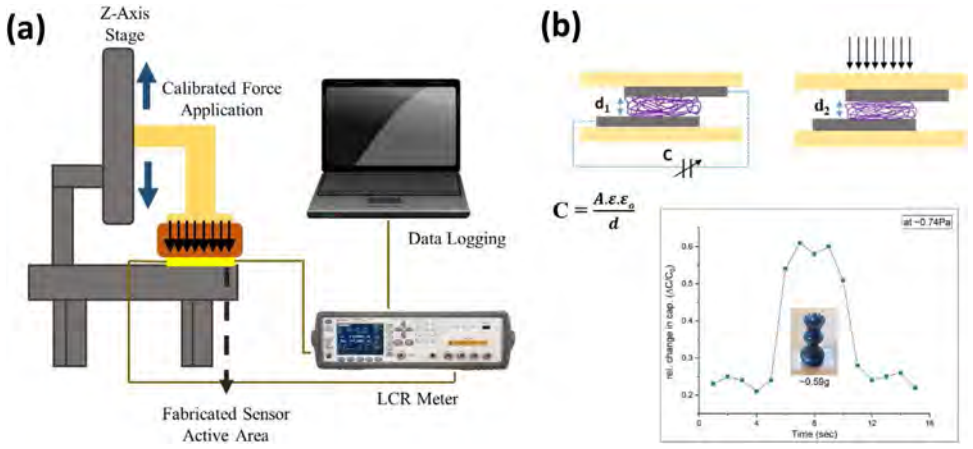

Fig. 2. **(a)** Mechanical test setup for sensor response: A highly precise pre-calibrated force application platform and an LCR meter attached with the laptop to log capacitance data **(b)** Working Principle with a test experiment using a chess board wooden queen placing for 5 s and removed afterwards.

sulated using spin coating the around 5 μm thin layer of PLGA (which counted towards the degradation time of device). The whole fabrication process has been demonstrated in Fig. 1.

2.3. Characterizations

SEM and FTIR characterization of as prepared nanofibrous dielectric were performed using MIRA3 TESCAN Field Emission Scanning Electron Microscopy (FE-SEM) and Nicolet 6700 FT-IR spectrophotometer to know the morphology as well as chemical composition of the electrospun nanofibers respectively.

Sensitivity analysis and test result experiments were performed using a Keysight E4980 LCR meter and a highly precise pre-calibrated motorized vertical force (1gf to 500gf) application stage as shown in Fig. 2. A glass sheet and a shape memory foam cut of same size as sensing area were glued together and gram force compensated in pre-calibration of the force application stage before start of the testing and analysis to apply equal force on the active area. First the device was calibrated in a tactile range of $0 < P < 4.6$ kPa by logging the relative change in capacitance data against the pressure changes. The response of the device ($n = 3$) was recorded and presented for sensitivity analysis. Then the device ($n = 3$) was subjected to response and recovery times' analysis. Device ($n = 3$) was subjected to 1 kPa pressure using the same setup to get the transient response data for response and recovery times' computation. The experiment on each sample was repeated for at least three times to confirm the repeatability of the sensor performance characteristics. Secondly small test weights were examined and time response of different pressure values was investigated. Cyclic response was determined using the same vertical stage to show high repeatability of sensor operation at different pressure applications with time. For cyclic response the sensor was subjected to a particular pressure point for 5 s interval and then released for another 5 s in each cycle. The sensor was subjected to the degradation in PBS media for 18 days in incubator to recapitulate the in-vitro degradation. All the experiments were performed at 10 kHz test frequency of the LCR meter.

After that a prepared sample of device was utilized to monitor the arterial pulse on wrist artery using capacitance to digital converter (CDC). For this purpose the commercially available CDC being AD7746 was used with an Arduino microcontroller to record the pulse signal from pulse artery on computer screen. The device pasted on wrist using an adhesive plastic bandage. In the end a 4×4 array sample of sensors was prepared using the same fabrication approach as described above involving first masking the substrate, then e-beam deposition of the patterned array electrodes and then

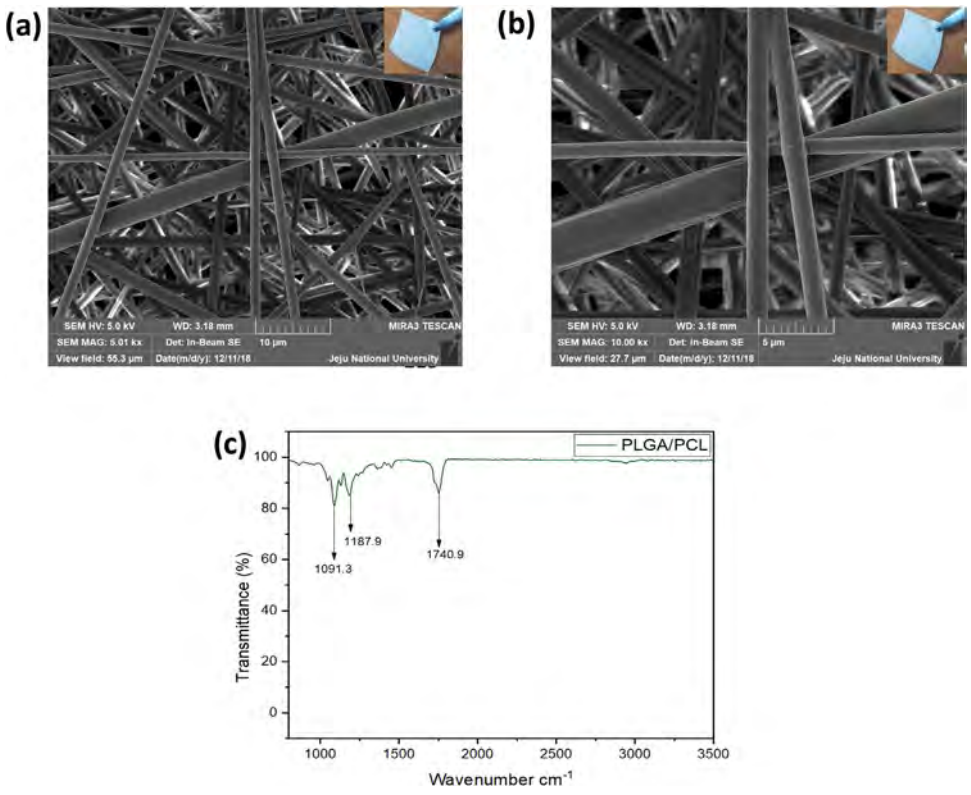

Fig. 3. FE-SEM micrographs of PLGA-PCL composite nanofibers: **(a)** scale bar 10um **(b)** scale bar 5um, and **(c)** FTIR spectra showing the chemical properties of the nanofibrous dielectric membrane.

in the end making sandwich structure by placing the dielectric in between. Connections were made using copper electrodes and a PLGA encapsulation layer was also spin coated on the prepared array samples. A commercially available pressure mapping platform from Kitronyx (which also employs the same capacitance to digital conversion approach) was used to characterize the pressure distribution on sensor array by placing various objects on it one by one.

3. Results and discussion

The SEM images of the composite dielectric nanofibers are shown in Fig. 3(a,b). The SEM power of more than 5.0 kV was burning the organic sample because it was first sputter coated with Pt for preparing samples for characterization. As it can be seen in the Fig. 3(c) that there are three characteristics absorption bands at 1091.3 cm^{-1}, 1187.9 cm^{-1} and 1740.9 cm^{-1} respectively which are attributed to the stretching of esters group (simple and conjugated) present in the PCL and PLGA respectively. Sharp peaks at 1091.3 cm^{-1} and 1187.9 cm^{-1} showed the stretching of R—O—R group in the structure, while the multiple absorption peaks between 1200 cm^{-1} and 1500 cm^{-1} have shown the presence of C—O—C groups. The other sharp peak in the spectrum at 1740.9 cm^{-1} has depicted the stretching of C=O group present in the blend of PCL and PLGA. This suggested that the blend of two polymers has been successfully synthesized into the nanofibrous mat fabricated using electrospinning.

As fabricated device when subjected to mechanical characterization was analyzed using an LCR meter. The response of the device (n = 3) has been plotted as relative change in capacitance against applied pressure as shown in Fig. 4(a), and it shows that sensors' response to pressure changes is quite linear in both operating regions of ($0 < P \leq 1.86$ kPa & 1.86 kPa $< P < 4.6$) and highly repeatable. It has a sensitivity of 0.863 ± 0.025 kPa^{-1} in ≤ 1.86 kPa region while the sensitivity further decreased to 0.062 ± 0.005 kPa^{-1} in upper region 1.86 kPa $< P < 4.6$. The higher sensitivity obtained can be subjected to the increased deformation in dielectric layer because of porosity of the nanofibrous structure rendering the device with decreased stiffness of the structure. [20] The device is quite flexible and adaptive to the contour of human body. The sensor samples were vacuum sealed in a polythene bag for later experiments and studies of degradation to protect from the environment. Small deviations from the trend response correspond to uneven membrane thickness in electrospinning process at different area in the active region, device assembly process, instability, minimal viscoelasticity, and loss of iron conductivity with the passage of time.

Then the sensors time response to small pressure values was investigated as shown in Fig. 4(c–d). Fig. 4(b) shows the typical time response curve for the fabricated sensor (n = 3) at 1 kPa pressure application and removal. As can be seen the average response time was computed as the time required to reach from 10% of the baseline value to 90% and similarly average recovery time as the time required to reach from 90% of the baseline value to 10%. The average response and recovery times were computed to be 251 ms and 170 ms. The deviation from the response shown was within ±5 ms range to that of the average response presented already. As can be seen that small weights at 1.24 Pa and 6.2 Pa showed around 0.01 pF and 0.05 pF changes. The lowest detected pressure value was 1.24 Pa, while the maximum value that was detected was 4.58 kPa which corresponds to a capacitance value of around 2.07 μF. After that sensor was subjected to cyclic response characterization at two different pressure values of 62 Pa and 1 kPa as shown in Fig. 4(e–f). The experiment was performed using the same calibrated vertical stage. Each of the cyclic response was carried out for a 5 s interval of loading and unloading. As can be seen from

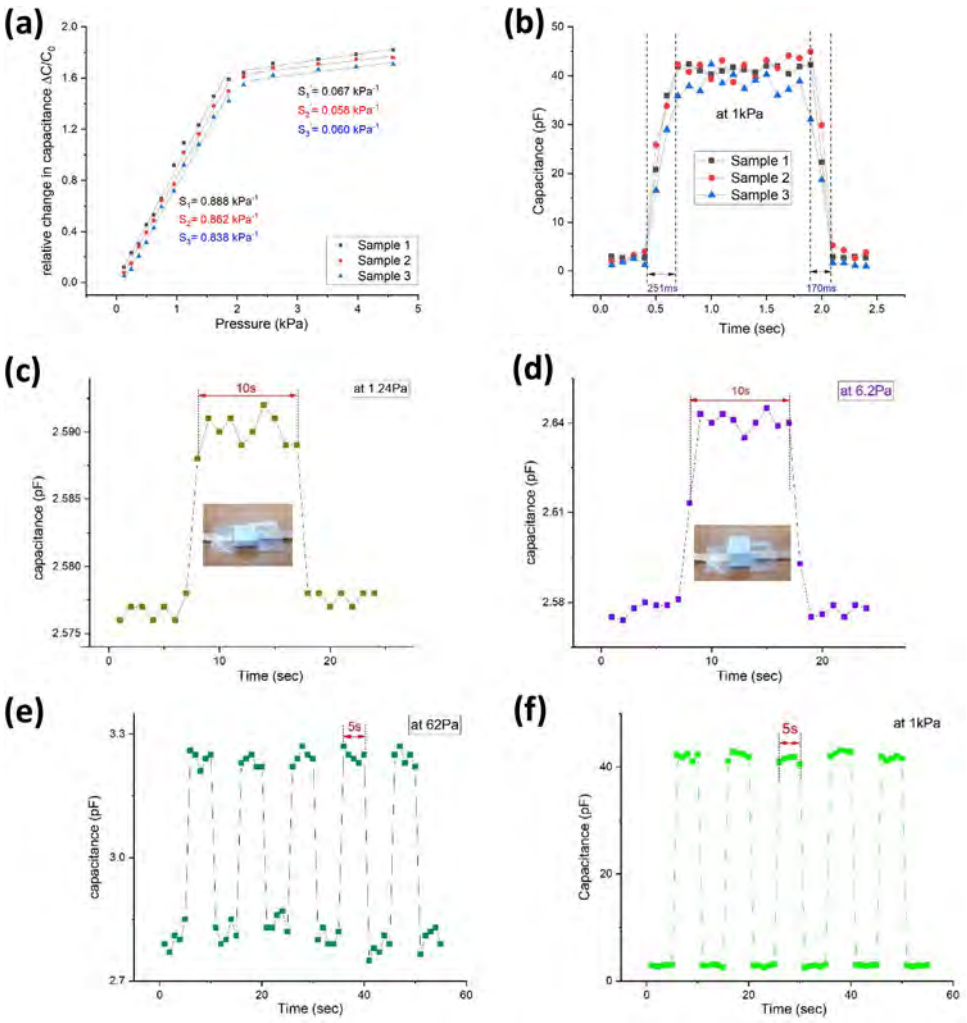

Fig. 4. **(a)** Relative change in capacitance vs. change in pressure (n = 3) **(b)** Response and recovery time computation using graphical analysis (n = 3) **(c–d)** Change in capacitance when polystyrene cubes of ~10mgf (1.24 Pa) and ~50mgf (6.2 Pa) are placed to affect all the effective area respectively **(e–f)** Time response of sensor to different objects exerting 62 Pa and 1 kPa pressure respectively with 5 s interval of loading and unloading.

the figure that the sensor response is quite reproducible which is mainly due to the increased adaptability to deformation because of low Young's modulus, negligible viscoelastic behavior at small pressure values for the specific range of operation.

As fabricated device was used to monitor the wrist pulse. For this purpose a custom built setup using a 24bit capacitance to digital converter AD7746 was used along with an arduino microcontroller to record the capacitance data. The data was recorded at 40 Hz sampling frequency keeping in view the response and recovery times to produce minimum lag. The data for a normal pulse and pulse after 5 min of exercise were recorded to demonstrate the sensor's usefulness in vital monitoring. Sensor response for a 5 s interval has been shown in Fig. 5. As can be seen in the figure that sensor is showing higher relative change in capacitance and higher frequency for pulse signal after 5 min of exercise because of the rise in blood pressure in wrist artery and in the heart beat.

An array of 4 × 4 pressure sensors was fabricated and utilized for demonstrating its use in spatial pressure distribution as shown in Fig. 6. A commercially available pressure mapping platform along with their software was used to characterize the fabricated sensor array for this purpose. Different objects were placed on it to prove its usefulness in mapping pressure points.

3.1. Degradation behavior

Sensor was characterized for its degradation in PBS media at neutral pH for 18 days of observation. After first week the media was replaced with new one and device was characterized again to analyze performance of the device. The first layer to degrade is the PLGA encapsulation which degrades by hydrolysis or biodegradation through cleavage of ester bonds into oligomers and monomers, once degraded the monomeric components are expelled from natural path ways [50]. PVA degrades by the microbial oxidation of 1,3-diols and enzymatic hydrolysis [5]. After the substrate is degraded enough to allow the degradation of Fe-Zn bilayer electrodes, they will degrade completely as has been studied by Luo et al. in RF pressure sensor fabrication [12]. PCL also goes to hydrolytic de-esterification but takes years to degrade depending on the quantity. For this study, very small ratio of PCL has been used to impart mechanical properties to the dielectric membrane and hence can degrade quickly due to nanofibrous nature of the membrane.

PVA substrate has been used, upon its oxidation and enzymatic hydrolysis, to quickly start degradation of the metallic bilayer electrodes and then the dielectric membrane itself. Since rapid degradation of PVA might affect the device performance significantly the device has been encapsulated in PLGA using spin coating.

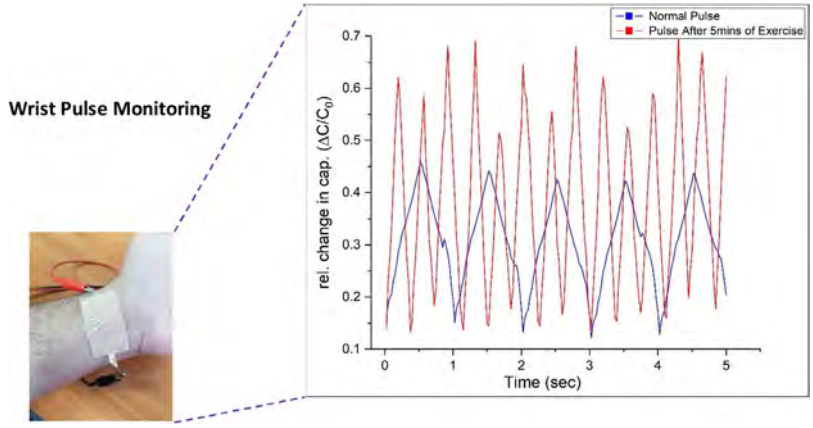

Fig. 5. ADWrist pulse recording at a sampling frequency of 40 Hz using custom built CDC CE 7746 with Arduino microcontroller based setup: blue indicating the normal pulse and red indicating the pulse after 5 min. of exercise (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).

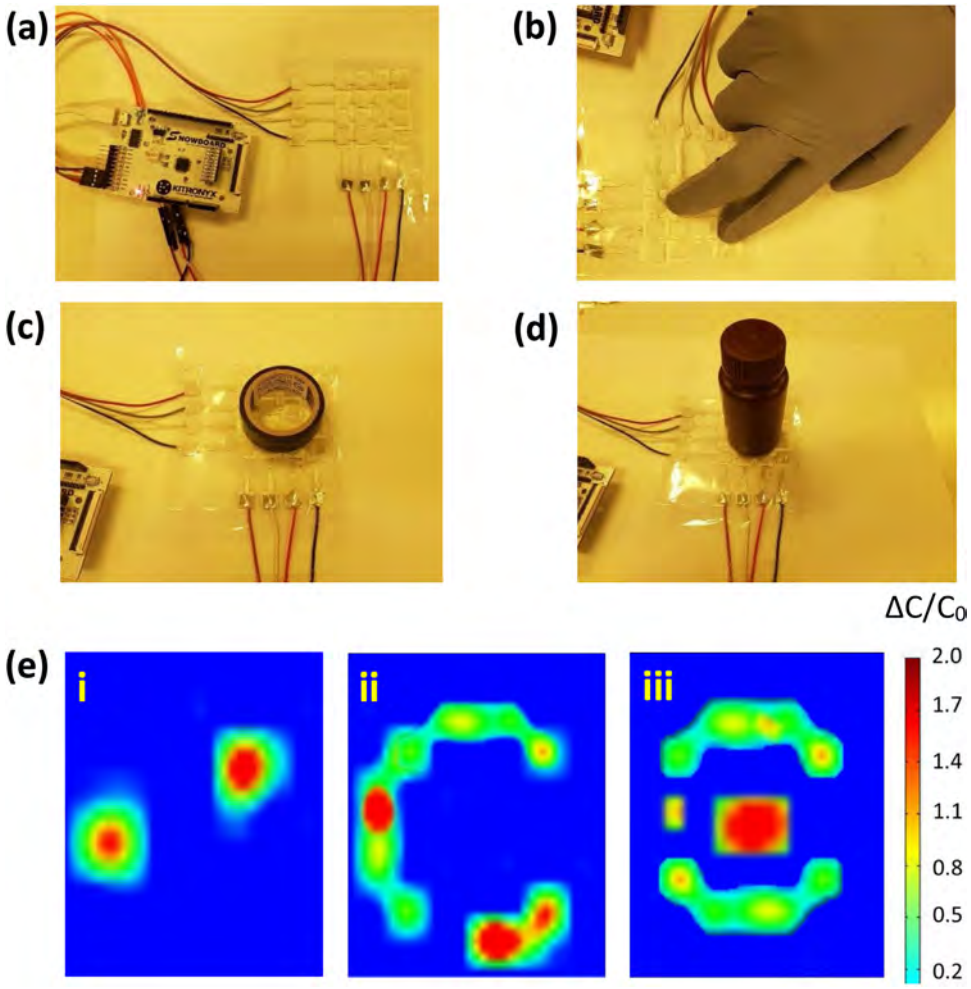

Fig. 6. **(a)** 4 × 4 array prepared using dielectric membrane connected to commercially available pressure mapping platform, and Application of pressure **(b)** By using fingers **(c)** By adhesive tape **(d)** By a (powder) filled bottle, and **(e)** Snapshots of pressure profiles indicated in i, ii, iii, for (b), (c), and (d) respectively from the software with scale bar showing relative change in capacitance with respect to color.

Around 5 μm thick PLGA encapsulation was applied to control the degradation without affecting the device performance significantly as compared to without PLGA encapsulation. The degradation images of the device for 18 days and degradation profiles (n = 3) have been shown in Fig. 7(a and b). It can be seen that sensor continues degrading as shown in degradation images and profiles up to 18 days and around less than 20% weight of the whole device (n = 3) remains. Further weight measurements were not possible to be conducted due to scale limitation and ultralight weight of what was left of the device (mainly the small footprint of the bottom electrode and the dielectric membrane which would probably take more time to degrade because of PCL content but degrade eventually). The

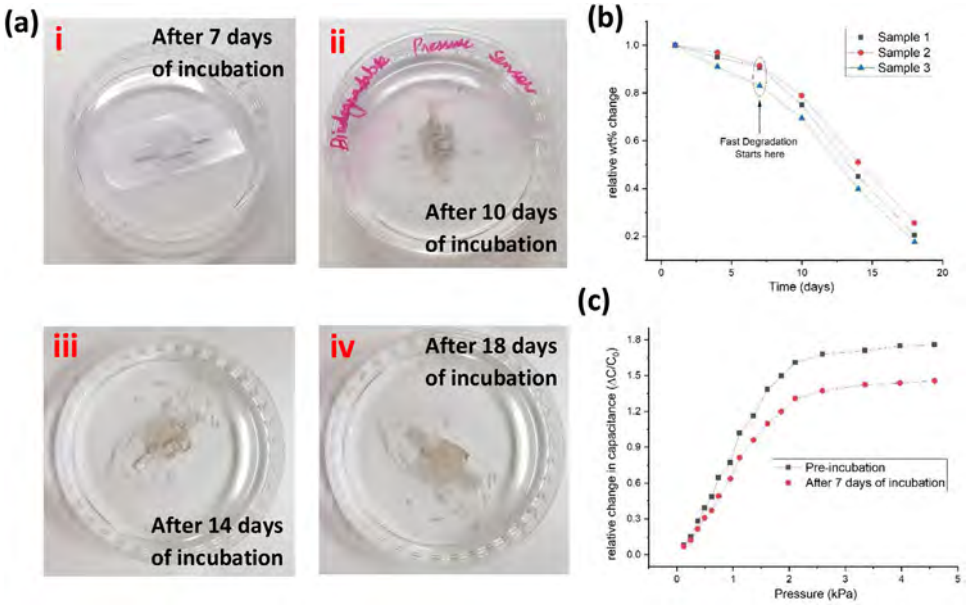

Fig. 7. **(a)** Images of device degradation in PBS media at different times **i.** after 7days of incubation in PBS media, **ii.** After 10 days of incubation, **iii.** After 14 days of incubation, iv. After 18 days of incubation **(b)** Degradation profiles of the device (n = 3) up to 18 days **(c)** Effect on device performance after 1 week of incubation – representing a sensitivity decrease of 19.5% in low pressure region with negligible change in sensitivity in comparatively high pressure region.

18th day data value was an average of 5 readings for one sample and included in the degradation curve as the reading becomes uncertain at such low weights because of the degrading chunks going away while picking up the sample for weighing analysis. The degradation profiles are almost showing similar degradation behavior with small changes that can be attributed to the fabrication margins. Since the sensor is a biodegradable one, it is going to lose its performance with the passage of time and become unstable with time. But the concept of adding PLGA encapsulation was to avoid compromising the sensor performance and stability in the first week of degradation. For performance evaluation of the sensor it was immersed in PBS media for 1 weak of incubation and its characteristic curve has been obtained and compared with initial results as shown in Fig. 7(c), and it is quite evident from the results that sensor showed very small instability in overall performance in terms of sensitivity decrease only. It showed a sensitivity decrease of 19.5% in low pressure measurement region with negligible change in comparatively high pressure region. Since the device degrades in less time as compared to the literature reported before it can be used as ecofriendly and disposable electronic device for in-vitro applications.

4. Conclusion

In this work we have developed a highly sensitive biodegradable piezo-capacitive pressure sensor for low pressure measurements. PLGA-PCL composite nanofiber membrane prepared using electrospinning technique has been used as elastic dielectric membrane to fabricate the biodegradable piezo-capacitive pressure sensor by sandwiching it into pure iron-zinc bilayer electrodes (top and bottom) deposited on biodegradable substrate using e-beam deposition. Nanofibrous dielectric membrane was subjected to morphological and optical characterizations using FE-SEM and FTIR analyzer. Sensor was characterized using an LCR meter and a highly precise and motorized pre-calibrated vertical stage. The sensitivity of the sensor was computed to be 0.863 ± 0.025 kPa^{-1} in the low pressure region ($0 < P \leq 1.86$ kPa) while it decreased to a value of 0.062 ± 0.005 kPa^{-1} for high pressure region (1.86 kPa $< P \leq 4.6$ kPa). The lowest detected value was 1.24 Pa (at 10mgf). Sensor's average response and recovery times were recorded to be 251 ms and 170 ms respectively. Its degradation study was investigated in PBS solution and it lost around 60% of its initial weight in 2 weeks with minimal loss in device performance after one week of incubation decreasing sensitivity by 19.5% in low detection range. While there was no significant change was observed in upper detection range. The device continued to degrade further observed until 18 days. All FDA approved biodegradable materials were used for the fabrication of device which is why it is a potential candidate for biodegradable, and disposable wearable devices. It was used to measure the arterial pulse monitoring on wrist artery using custom built instrumentation. Also a 4×4 array of sensors was fabricated to demonstrate its application in pressure mapping using a commercially available pressure mapping controller and software.

Competing interests

Authors declare no competing interests.

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