A bioprotein-based flexible and self-powered pressure sensor towards a biomimic of an artificial Pacinian corpuscle

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***Abstract*—The demand for low-cost, flexible, biocompatible, biodegradable, and self-powered pressure sensors is increasing, particularly for applications in soft robotics, prosthetics, and implantable electronic applications. However, most of the pressure sensors lack in terms of biodegradability and biocompatibility resulting in increased electronic waste. Further, most of the state-of-the-art pressure sensor need an external power supply for its operation and are bulky further making it not suitable for the wearable and implantable electronic applications. To address these limitations, there is a need to develop a novel self-powered pressure sensor that utilizes bio-proteins i.e., Chitosan offering both flexibility, biodegradability, and high sensitivity. This sensor will enable accurate and efficient detection and response to varying levels of pressures, with high mechanical flexibility, and repeatability. The strategy outlined here introduce an innovative biomaterial, offering significant utility for various applications such as next-generation wearables, implantable devices, soft robotics, and biomedical.**

Keywords— flexible, soft robotics, implantable, pressure sensor, self-powered, chitosan, biomaterial.

I. INTRODUCTION

Pacinian corpuscles are mechanoreceptors located subcutaneously and are responsible for detecting high-frequency vibrations and pressure [1]-[3]. With growing interest in biomimetic technologies, mimicking the properties of Pacinian corpuscles is of great interest in the development of advanced pressure sensors with potential applications in areas such as robotics, prosthetics, and wearable devices [4],[5]. There are several reports on the development of different artificial mechanoreceptors and electronic skin (e-skin), using both static and dynamic pressure sensors as slow/static adapting (SA) and rapid/fast adapting (RA) mechanoreceptors [6]-[10]. Different sensor types, such as resistive, capacitive, piezoelectric, optical, and triboelectric pressure sensors, using different organic, inorganic and polymer materials, have been explored for wearable, implantable, soft robotics, biomedical, implantable, and e-skin applications [11],[12]. However, the former (static) sensors typically rely on relatively high levels of power consumption since they generally require an external power supply or battery for operation. The latter (dynamic) sensors are typically less stable in their response, and non-degradable, thus exacerbating the problem of electronic waste (e-waste) [13]. However, there is progress in this, where for example Han et.al recently demonstrated a self-powered artificial mechanoreceptor using a sustainable triboelectric nanogenerator (TENG) integrated with a biristor, thus creating a neuromorphic tactile system [14-15].

Biodegradable electronics are electronic devices that can decompose and be safely absorbed by the environment over time. These devices are an important step towards reducing e-waste and creating more sustainable technology [16],[17]. Some of the well-explored biodegradable materials such as cellulose, silk, chitosan, and egg albumin offer excellent physical, mechanical, and chemical properties. Chitosan is a natural polymer derived from chitin, a substance found in the shells of crustaceans such as shrimp and crabs and has been explored as a candidate for flexible and transparent substrates and dielectric layers for various applications including transistors, sensors, memristors, and photodetectors [18]. Chitosan offers numerous advantages in terms of natural abundance, low cost, solution processability, mechanical flexibility, biocompatibility, and biodegradability. Furthermore, chitosan has exhibited desirable piezoelectric properties for piezoelectric nanogenerators or wearable dynamic pressure sensors [19]. Recently, Ensieh et al. demonstrated a piezoelectric nanogenerator based self-powered dynamic pressure sensor using chitosan-glycine based biodegradable nanofibers which exhibit good sensitivity, cyclic stability, and biodegradability.

Despite these advances, there remain performance concerns including long-term stability, miniaturisation, and compactness regarding current chitosan-based devices. To address these challenges, in this research we present the fabrication of a flexible, biocompatible, and self-powered pressure sensor utilizing the protein-based polymer chitosan. The response characteristics of the sensor are investigated under various external pressures and frequencies. The sensor exhibits excellent sensitivities for

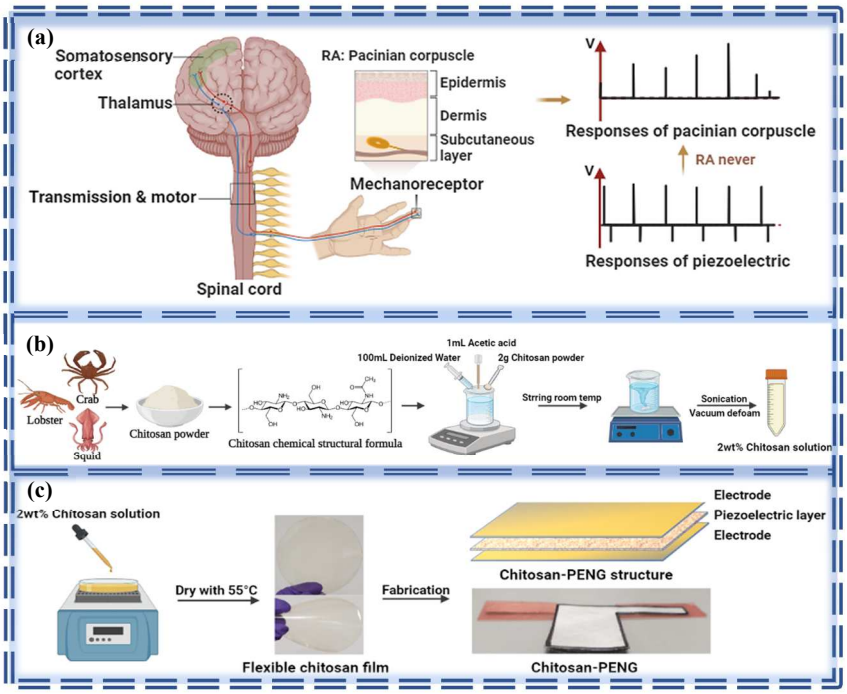

Fig. 1. (a) The artificial Pacinian corpuscle system emulating the human biological equivalent; (b) The chitosan solution preparation process; (c) The fabrication process of the PENG.

low and high frequencies, along with a wide pressure range. Finally, repeatability and mechanical flexibility studies are conducted. It is shown that the innovative chitosan-based films exhibit potential as both substrates and active layers for diverse applications in next-generation flexible electronics and wearable devices, such as e-skin, photodetectors, and nanogenerators.

II. Fabrication

A. Preparation of Chitosan Film

To prepare chitosan film, a low molecular weight chitosan powder (Sigma Aldrich 9012-76-4, $C_{18}H_{35}N_3O_{13}$) was selected which was made from 75-85% deacetylated chitin [20]. The schematic representation of the biological tactile perception system with the mechanoreceptors inside the human skin (slow adapting and rapid adapting) shown in Fig. 1(a). The schematic representation of the preparation of the large-area flexible chitosan films is illustrated in Fig. 1(b). First, the 2 g chitosan powder was mixed with 1 mL acetic acid solution and 100 mL to obtain a 2 wt.% chitosan solution, after which a magnetic bead was added to the solution. The solution was placed on a magnetic stirrer and heated to 60-70°C. The chitosan solution was then sonicated at 37 kHz for 30 min, and after thorough mixing the prepared solution was placed in a vacuum desiccator and degassed for 1-2 hours until all the air bubbles in the solution had disappeared. After this, the chitosan solution was drop-casted onto a polystyrene petri dish and subsequently spin coated for 30 s at a speed of 1000 rpm to for a uniform thin film of chitosan of thickness of 100 microns. This was followed by a drying process in a heater at 55°C. When the film completely dried as shown in Fig. 1(c), the biodegradable chitosan film is mechanically flexible and optically transparent, exhibiting relatively high flexibility and mechanical robustness under repetitive mechanical bending and twisting.

B. Fabrication of the Chitosan-based PENG

The PENG device was created using a spin coating process, which is a simple and cost-effective technique. The device has the sandwich structure as depicted in Fig. 1(c). The electrodes consist of aluminium, serving as both the top and bottom layers, while the active piezoelectric layer is made of chitosan. To fabricate the device, the chitosan film is first prepared and then peeled off from the petri dish and then cut into a size of 1 x 2 cm. The top and bottom electrodes, made of aluminium foil, are cut into shapes that match the dimensions of the active layer. Next, the chitosan film is sandwiched between the two electrodes, creating the PENG sensor. To ensure proper insulation and avoid short circuits, the device was encapsulated in polyamide.

C. Characterization and Testing

To evaluate the dielectric properties of the chitosan films, their capacitance and impedance were measured by impedance spectroscopy (Agilent 4294A, Keysight Technologies). To evaluate the performance of the sensor in terms of output voltage and sensitivity, in addition to the stability of the PENG at various frequencies and magnitudes of pressure were applied to the PENG (TV 50018 Vibration Test System, TIRA GmbH), with the output signal collected and recorded via a digital storage oscilloscope (DSOX3014T).

III. Results And Discussion

To understand the dielectric properties of the chitosan film, the variation of the capacitance (C) with frequency (f) was obtained, and these results are shown in Fig. 2(a). The results show that there is a decrease in C from 0.91 nF to 0.82 nF, as f increases from 10 Hz to 10 kHz. Through the impedance data shown in Fig. 2(b), the resonance is observed at 35 MHz and the anti-resonance at 52 MHz.

The dielectric constant of the device can be calculated using (1).

$$\varepsilon_r = \frac{\varepsilon_c}{\varepsilon_0} = \frac{Cd}{\varepsilon_0 A} \quad (1)$$

ε_c is the permittivity of the chitosan film, ε_0 is vacuum permittivity, which is equal to 8.85×10^{-12} F/m, and ε_c can be calculated from C and the dimensions of the chitosan film, where the thickness of the chitosan film d is 10 µm and the area A is 2 cm^2. It is evident that the dielectric constant of the material decreases as f increases, as shown in Fig. 2(c). At a relatively high frequency of 10 kHz, the dielectric constant of the material is 4.61, whereas the dielectric constant at the lower frequency of 100 Hz is 5.16. The combination of different frequencies with high and low values of dielectric constant may be due to the ion and electron polarisation occurring in the material.

The piezoelectric characteristics of the PENG device were evaluated by measuring the output voltage under different pressures (low and high). The sensor's output response to pressures ranging from 10 kPa to 80 kPa at a constant frequency of 6 Hz, with a pressure sensitivity S_P of 16.8 ± 0.2 mV/kPa, are shown in Fig. 3(a). Additionally, the frequency sensitivity S_f of the pressure sensor at frequencies ranging from 1 Hz to 8 Hz at a constant pressure of 60 kPa, with an S_f of 200 ± 0.2 mV/Hz are shown in Fig. 3(b.), calculated using equations (2) and (3).

$$S_P = \frac{\Delta Output\ voltage}{\Delta Pressure} \quad (2)$$

$$S_f = \frac{\Delta Output\ voltage}{\Delta Frequency} \quad (3)$$

The output response characteristics of the nanogenerator when subjected to cyclic increments of pressure ranging from 40 kPa to 80 kPa at a constant frequency of 6 Hz are illustrated in Fig. 3(c). The results show a linear increase in the output voltage with the applied pressure. At 40 kPa, the output voltage is 540 mV, which reaches 1.41 V at 80 kPa. The deformation of the piezoelectric film under increasing pressure aligns the dipoles inside the chitosan, generating charges on the film's surface and transporting them through the electrodes. This process creates a potential difference, resulting in an output voltage. The cyclic incremental change in the output voltage of the pressure sensor is

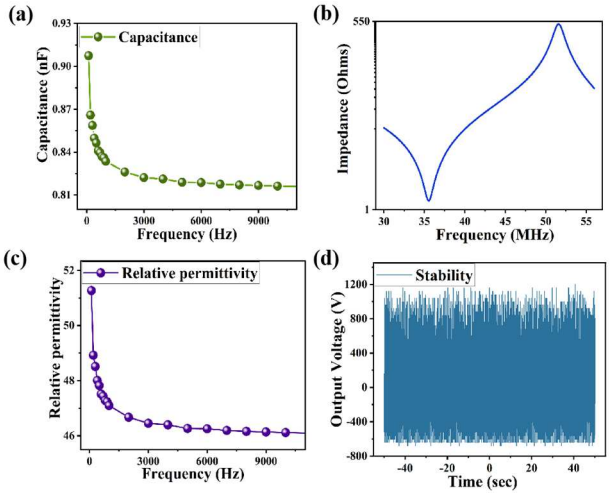

Fig. 2. Impedance spectroscopic analysis of the chitosan film with a potential of 0.5 V, showing (a) capacitance with frequency; (b) impedance variation with frequency; and (c) relative permittivity with frequency. (d) cyclic stability of the PENG device.

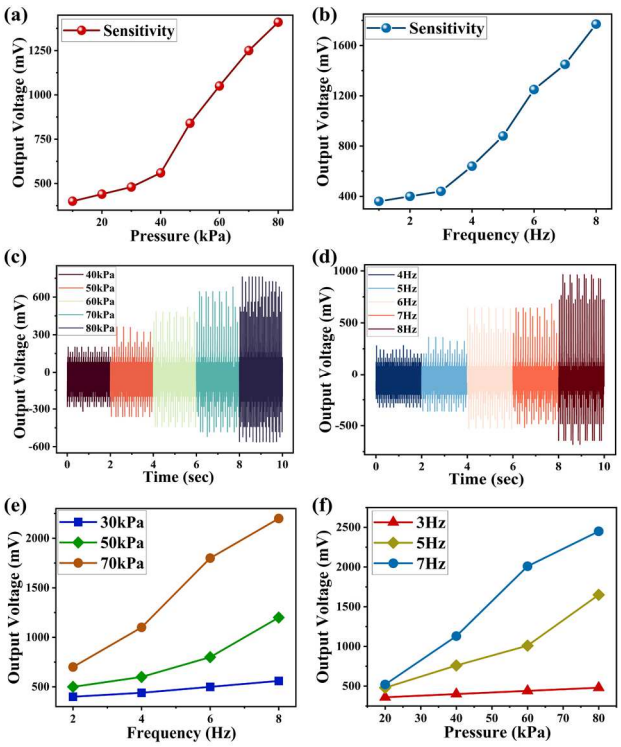

Fig. 3. (a), (b) Pressure and frequency sensitivities of the PENG under different pressures at a constant frequency of 6 Hz, and different frequencies at a constant pressure of 60 kPa, respectively; (c), (d) output voltage as a function of pressure and frequency, respectively; (e), (f) change in the output voltage of the PENG as a function of different frequencies and pressures.

depicted in Fig. 3(d) for different frequencies (4-8 Hz) at a constant pressure of 60 kPa, ranging from a minimum of 640 mV_{P-P} at 4 Hz to a maximum of 1.77 V_{P-P} at 8 Hz.

The piezoelectric response of the PENG is influenced by the variation in applied pressure and frequency. The device demonstrates linear output response characteristics under both low and high-frequency regimes. The piezoelectric response of the PENG improves at higher frequencies, enhancing its sensitivity to external vibrations. The results in Fig. 3(e) and Fig. 3(f) show the voltage increases generated by the PENG at different frequencies (3, 5, and 7 Hz) and pressures (30, 50, and 70 kPa). Higher frequencies result in higher output voltages, while an increase in pressure leads to more significant deformation within the piezoelectric layer and consequently higher piezoelectric potential. The high linear piezoelectric sensitivity of the device enables the detection of subtle physiological signals and minor external forces, simulating the perception of externally applied pressure like Pacinian corpuscles. The mechanical stability and repeatability of the chitosan-based pressure sensor when subjected to an externally applied pressure of 60 kPa for extended durations of up to 100 seconds is shown in Fig. 2(d).

The PENG exhibits exceptional stability and repeatability in generating output voltage during multiple cycles of operation at a cyclic pressure of 60 kPa and 6 Hz. This signifies the device's outstanding stability in maintaining its output signal over time. These findings can be compared to the current state-of-the-art in the field. Table I provides an overview of important properties of other piezoelectric pressure sensors developed using biodegradable materials as reported in the literature.

Table I. Comparison of piezoelectric pressure sensors fabricated using biodegradable materials.

Material	Pressure (Max)	Frequency (Max)	Output Voltage (V)	Ref.
Silk	82.2 kPa	3.1 Hz	3.1	[21]
Onion skin	34 kPa	3 Hz	18	[22]
Glycine–Chitosan	30 kPa	30 Hz	0.2	[23]
Ginkgo tree leaves	40 kPa	3 Hz	6.3	[24]
Chitosan	**80 kPa**	**8 Hz**	**2.73**	**This work**

Among the self-powered pressure sensors based on chitosan, specified in Table I, it has been observed that the chitosan-based PENG sensor exhibit good performance in detecting moderately high pressure and frequencies as compared to literature. This demonstrates their wide pressure sensing range and suitability for high-frequency applications. Additionally, the fabrication methodology for these sensors is relatively low in cost and simple to implement compared to other methods.

IV. Conclusion

In summary, a flexible and self-powered pressure sensor has been developed using simple, low cost and solution-processed techniques. The sensor exhibits excellent pressure and frequency sensitivities of 16.8 mV/kPa and 200 mV/kPa respectively, for operation across a wide range of pressure and frequency limits. The fabricated sensor exhibits the potential for use in various applications such as e-skin, prosthetics, and soft robotics. Furthermore, the chitosan film can be used as a flexible, transparent, biocompatible, and biodegradable substrate for implantable and biomedical applications.

V. Acknowledgment

This research was supported by EPSRC Grant No. EP/V049658/1 and the European Commission through grant reference H2020-MSCA-ITN2019-861166.

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