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Self-healing pseudo–piezoelectric pressure sensors from sustainable and recyclable materials

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Advances in pressure sensing technologies are driven by the growing demand for flexible, lightweight, and highly sensitive devices for wearable electronics, robotics, and human–machine interfacing. While conventional piezoresistive, capacitive, and piezoelectric sensors have been successfully used in applications, they often struggle with mechanical rigidity, signal instability, and limited compatibility with soft systems. In this study, we present a pseudo–piezoelectric pressure sensor that combines both self-healing and sustainable materials while being designed for touch-level detection. Conductive and flexible films were prepared by blending sustainable and recyclable materials in water: a biopolymer (chitosan) and a natural deep eutectic solvent (NADES) as a plasticizer. The advantage of the elastomer is its mechanical degradation (scars, cracks, and tears) can be self-healed. A proof-of-concept sensor was fabricated using a simple bilayer architecture by integrating a conducting polymer (**pPDS**) into one of the elastomer layers to enable asymmetric charge dynamics. The sensitivity of the resulting device was 128.4 mV kPa^{-1}. The sensitivity was also reliable and the sensor's physical damage caused by over stretching could be self-healed. These results underscore the promise of iontronic sensors that exploit interfacial ionic mobility and electrostatic coupling, offering a sustainable pathway toward next-generation tactile sensors for soft, low-force applications.

1. Introduction

Human–machine interfacing for soft robotics is spurring demands for flexible, sustainable, lightweight, and highly sensitive pressure sensors.[1–4] This is further compounded by the ever-increasing use of wearable smart technologies, such as health monitoring devices that require sensors and components that are stretchable for their operation.[5,6] New materials are required to meet this insatiable appetite for mechanically compliant soft materials with high sensitivity of touch and movement. Transducers based on electrostatic interactions are emerging as alternative technologies to meet these stringent sensitivity demands for low-force and touch-responsive applications compared to traditional transduction.[7,8] Their emergence overcomes the limitations of current transduction. Indeed, pressure sensing has come to rely on piezoresistive, capacitive, and piezoelectric transduction.[9] While each transduction has met a given performance demand, they each suffer from their respective limitation. For example, piezoresistive sensors are limited by drift and temperature sensitivity.[10] Capacitive sensors are highly responsive, but their vulnerability to electromagnetic interference curbs their reproducibility. They further require complex signal conditioning.[11,12] While piezoelectric sensors excel for dynamic pressure detection, they typically depend on crystalline materials that are brittle, making them incompatible with flexible substrates.[13] These challenges have prompted alternative sensing paradigms that offer mechanical compliance, high sensitivity, and compatibility with both soft and stretchable systems.[2,14–16]

Electrostatic pressure sensors have emerged as promising candidates to address the shortcomings of conventional pressure transducers.[17–20] They offer charge redistribution, interfacial polarization, and electrostatic coupling between layers. This contrasts with piezoelectric sensors that primarily generate charge through lattice deformation.[18] Iontronic sensors further build upon electrostatic pressure interactions by taking advantage of ion migration and interfacial dielectric modulation to transduce mechanical stimuli.[17,21,22] These collective mechanisms enable iontronics to detect subtle pressure changes with high fidelity beyond traditional transducers, with the added advantage of being both flexible and lightweight. The electrostatic and ion transport of iontronic sensors makes them highly sensitive toward applied force. These attributes can be preserved when integrating into flexible substrates and biocompatible systems. This was demonstrated by Cheng *et al.*, who developed an iontronic pressure sensor with a

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microstructured ionogel dielectric layer. Pressure sensing was based on ion migration and electrostatic coupling. Their design was tailored for wearable electronics with high resolution.[21] Similarly, a rose-structured ionogel, paired with porous electrodes, was developed to improve the pressure sensitivity and signal stability.[23] The role of soft ionic materials and electrostatic mechanisms for ultrasensitive flexible pressure sensors, required for human–machine interfacing has further been higlighted.[24] These highlight the importance of ionic layer patterning, structural optimization and hybrid sensing mechanisms, combining ionic mobility with electrostatic charge dynamics for ultrasensitive electrostatic response.

The sensitivity of iontronic sensors is underpinned by the material used for the conductive layer. Graphene, carbon nanotubes (CNTs), and reduced graphene oxide (rGO) have been choice materials because they meet the collective electrical and mechanical requisites of flexible electronics.[25,26] They are also excellent conductors that can be processed into either thin films or composites. Metallic silver nanowires are also widely used because of their high conductivity and macroscopic transparency. These properties are ideally suited for touch-sensitive displays and wearable devices.[27–29] Conductive polymers such as PEDOT:PSS offer a balance between flexibility and conductivity. They can also be easily integrated into stretchable electronics.[30–32] Its self-doped water-soluble counterpart (**pPDS**) offers the advantage of homogeneous processing in water with a range of sustainable and biodegradable biopolymers that can be leveraged to increase recyclability by composting at the sensor end-of-life.[33,34]

The device architecture further plays an important role in the sensor performance of iontronics. Both the signal intensity and the mechanical resilience can be enhanced by layering multiple ionic and conductive materials.[14] However, consistent contact between the layers is quintessential for the synergistic interfacial dielectric and electrostatic coupling to affect the iontronics.[35–37] Therefore, a minimalist iontronic sensor consisting uniquely of two layers with a high surface area would be ideal for evaluating new materials for sensing by interfacial mobility. A bilayer sandwich type architecture would ensure maximum interfacial polarization and charge redistribution for identifying ideal materials for iontronics. Given repetitive movements of wearable sensors eventually cause irreversible damage, undesired loss of sensitivity, and ultimately device failure, it is important to determine whether self-healing materials could replace conventional elastomers that are otherwise irreversibly damaged without compromising iontronic sensing. Bearing these in mind, a bilayer sensor consisting exclusively of a self-healable biopolymer elastomer was prepared and evaluated. Further ensuring the environmental circularity of the iontronic sensor, a natural deep eutectic solvent (NADES) was chosen to plasticize the chitosan film and make it flexible, stretchable, and self-healable after breakage. Herein, a sustainable iontronic pressure sensor that combines high-sensitivity and consistent performance after healing from mechanical damage is presented toward addressing the need for elastic and responsive sensors in soft robotics. The collective attributes position electrostatic and iontronic pressure sensors to become integral transducers for enabling tactile and wearable technologies along with facilitating real-time monitoring of smart systems.

2. Results and discussion

Chitosan was used to prepare the sensors because of its natural abundance and its biodegradability.[38–40] The choline chloride/citric acid NADES was chosen as a plasticizer because of its similar sustainability and degradability attributes as well as its ionic conductivity, required to enable the iontronic sensing.[41] The self-doped **pPDS** conducting polymer (Fig. S1) was preferred to PEDOT:PSS because of its water solubility and because it could be homogeneously blended with chitosan and NADES (Fig. S1), a requisite for iontronic sensing. The **pPDS** polymer was blended into the chitosan matrix to confer electrical conductivity while further preserving the sought after elastic properties. These were expected to confer asymmetric charge dynamics to the corresponding iontronic sensor. The conducting polymer was incorporated into the chitosan/NADES blend in varying amounts (0.025, 0.05, 0.1, 0.15 and 0.20 wt%) to give a homogenous solution. Similarly, an elastomer prepared from chitosan and NADES without the conducting polymer was also prepared to benchmark the effect of the polymer and the iontronic effect when two different films were sandwiched together. In all, 1.5 wt% of chitosan was used for the formulated elastomers. This was according to known formulations that give films that are highly transparent and ductile (Fig. S2) along with chitosoan solubility for film casting.[42] The effect of the conducting polymer on the optical transparency of the elastomer film was determined by measuring the transmittance of the films (Fig. 1A). Although optical transparency is not a requisite for pressure sensing, absorption measurements provide qualitative information about the homogeneous dispersion of the conductive polymer in the elastomer. More importantly, the conductive polymer had a characteristic absorption that can be tracked to confirm its consistent doping (*ca.* 700–1200 nm), and hence its conductivity. This is of importance to ensure blending both the NADES and chitosan do not dedope the conductive polymer.[43] The film without the **pPDS** had the highest transmittance at *ca.* 85%. The transmittance in the visible region decreased contingent on the **pPDS** loading amount. This is a result of the intrinsic blue/green color of the doped polymer, resulting in a decrease in transmittance upwards of *ca.* 75% with 0.20 wt% polymer loading. The films made were between 200–300 μm thick, with a transverse sheet resistance of 0.8–0.07 MΩ (Table S2).

The effect of the conducting polymer loading on the mechanical properties of the films was also assessed from the stress–strain measurements. According to the stretching data in Fig. 1B, adding the conducting polymer did not alter the mechanical properties of the films when compared to the blank without the polymer. For example, a maximum tensile strain of *ca.* 80% was obtained for 0.20 wt% of **pPDS** loading in the

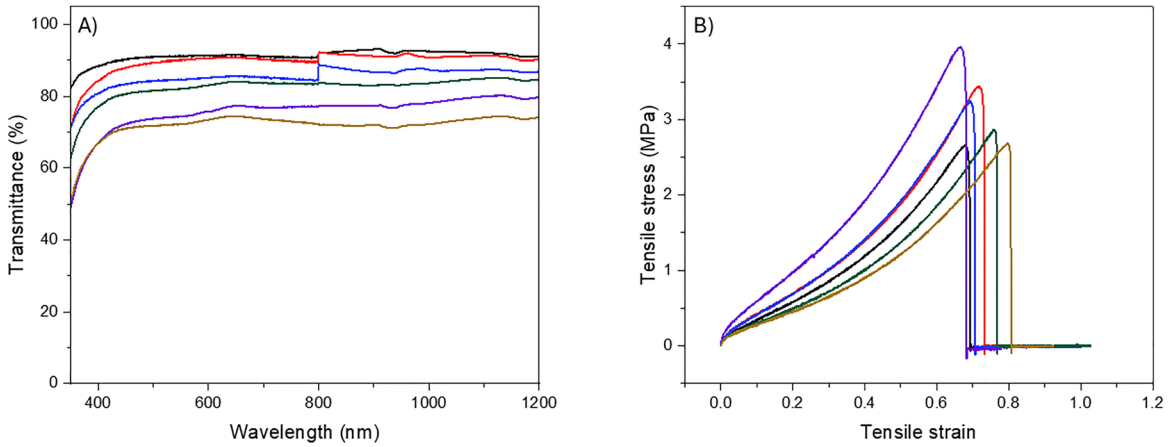

Fig. 1 (A) Absorption spectra of pseudo–piezoelectric films. (B) Stress–strain curves of the pseudo–piezoelectric films: 0.0 (black), 0.025 (red), 0.05 (blue), 0.1 (green), 0.15 (purple), and 0.20 wt% (gold) **pPDS** per volume of water in the chitosan-NADES films.

elastomer matrix compared to 70% from the blank film (Fig. 1B and Fig. S3). This provides sound evidence that the conducting polymer indeed does not affect the tensile strength or the Young's modulus of the polymer. Rather, the polymer improves the robustness and the elasticity of the elastomer, both requisite qualities for wearable sensors. Also, the tensile strength of the films was unchanged within the 2–4 MPa range, with a Young's modulus of 2.2–4.4 MPa (Table S1).

The pseudo–piezoelectric properties of the films were examined according to their electrostatic charge generation potential (Fig. 2A). The mechanism of the sensor was mostly due to the generation of charge with applied pressure, resulting in a decrease in the electrical double layer and a difference in voltage output. For this, a simple bilayer architecture was used, consisting of an ionic and an electrically conductive layer that were sandwiched together. The conductive layer consisted of the maximum loading of the conducting polymer (0.20 wt%) while the ionic layer consisted uniquely of the elastomer without **pPDS**. Measurements were made by connecting the two layers to an oscilloscope *via* copper wire leads (Fig. 2B). Both layers were shaped into circular disks with a diameter of approximately 2 cm.

The pressure measured as the voltage output from the electrostatic charge generation with constant mechanical tapping are summarized in Fig. 2C. The maximum output from the sensor was *ca.* 2.0 V and it was consistent over the 10 s

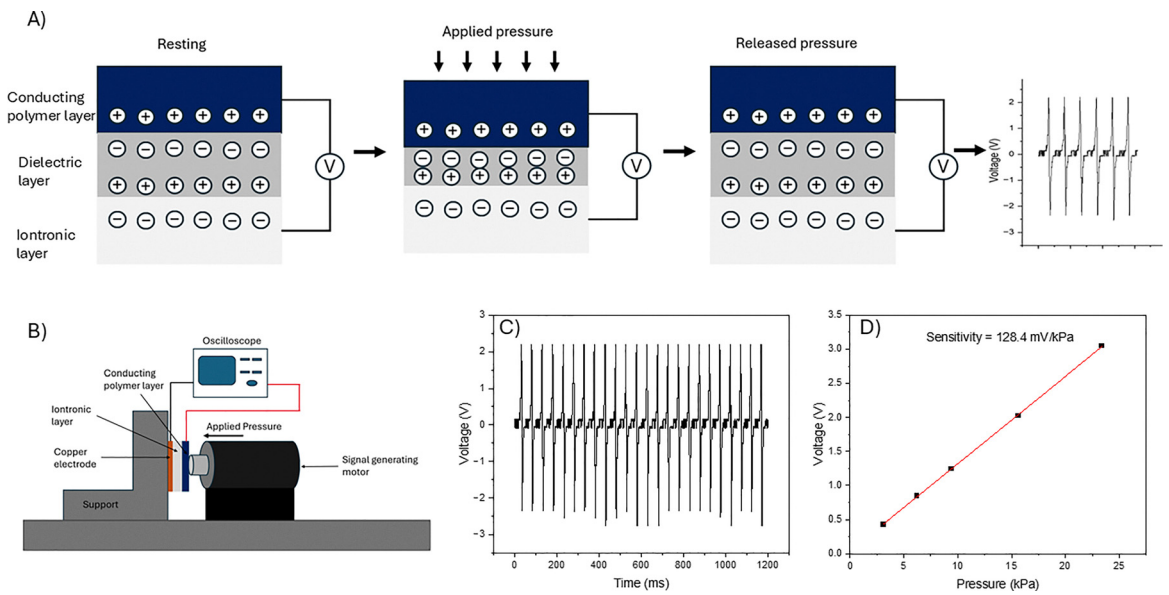

Fig. 2 (A) Schematic representation of the mechanism of operation of pseudo–piezoelectric sandwich pressure sensor *via* electrostatic induction: conductive polymer layer (blue) with electrical double layer at the interface sandwiched between the iontronic layer (grey). From left to right: sensor at rest, sensor engaged with applied pressure, pressure released, and expected output with applied pressure and release. (B) Schematic representation of the setup for pseudo–piezoelectric measurement. (C) Voltage outputs over time of the pseudo–piezoelectric sandwich device measured at 15 kPa. (D) Sensitivity measurements of the sensor contingent with applied forces.

measurement window at a 10 Hz tapping frequency with a signal-to-noise ratio of *ca.* 18.5 dB. The sensitivity of the sensor was also determined. This was assessed by measuring the voltage with different forces of applied loads of 3, 6, 9, 15 and 23 kPa (Fig. 2D). The transduction arises from a change in internal resistivity with film compression. This decreases both the interlayer ion migration and electrostatic interactions, improving conductivity and increasing the voltage output. The change in voltage between 0.5 and 3.0 V correlated linearly with the pressure (Fig. 2D). The direct voltage/pressure correlation is ideal for accurately detecting pressure according to a calibrated pressure response curve. The sensitivity of the pressure sensor can further be determined from the voltage/pressure linear relationship. The sensitivity of the sensor was 128 $\pm$ 1.0 mV kPa^{-1}, confirming moderate sensitivity. To frame the sensitivity of the simplistic sustainable device, the sensitivity of other iontronic sensors is summarized in Table 1. The sensitivity of the chitosan/DES sensors falls within the lower sensitivity range of piezoelectric sensors involving PDMS, polyurethane, and polyacrylamide materials. This aside, it has the benefits of being both derived principally from sustainable/degradable materials and fabricated using a simple bilayer sandwich architecture. Collectively, the simple design and straightforward film preparation by mixing bulk materials and solution casting are significant advantages compared to complex device architectures that are reliant and materials that are available in limited amounts. Readily available and sustainable bulk materials can nonetheless be used to prepare sensitive pressure sensors with the potential of upscaling to large area devices. The collective inertness and biocompatibility of the sensor make them ideally suited for safe usage across a range of consumer applications, including medical implants, wearable sensors, and elementary electronic devices.

The advantage of NADES as the elastomer plasticizer is it confers self-healing. Chitosan plasticized with NADES is known to heal after their rupture, with their mechanical properties being restored to their pre-ruptured state.[50] To evaluate whether this was indeed the case with the pressure sensor, with pressure sensing being restored to its original capacity after healing a rupture, controlled damage was introduced to the elastomer *via* incisions both before and after stretching. Initial tensile/strain measurements of the blank films showed a 70% elongation prior to the onset of damage (black line Fig. 3A). Elastomer healing after rupture is moisture dependent. The humidity accelerates hydrogen bond formation of the NADES at the interfaces, subsequently restoring the mechanical integrity of the elastomer. This influences the elongation of the relative to the original film before rupture. Hydrogen bond reformation at the interface along with the dynamic nature of the bonds promote interfacial interdiffusion of the NADES and the biopolymer for distributing the load. These contribute to increasing the elongation at break and improving the ductility. Therefore, the Young's modulus is reduced due to the stronger reinforced bond at the healed junction. This aside, the strength of the original/unruptured elastomer is only partially recovered upon healing with the elastomer becoming softer and more extensible when healed (Table S3).

The elongation of the healed film before rupturing was reduced to *ca.* 30% after five minutes of healing. This elasticity is sufficient for most flexible sensor applications that do not require high degrees of flexibility/stretchability. However, the film's mechanical tenacity improved significantly with increased healing time. The elongation at break increased to *ca.* 80% with 30 min. of healing, surpassing its original tensile strain. The elongation at break further doubled the original elongation, *ca.* 140% (green line Fig. 3A and Fig. S5), after healing for 60 min. This is a result of progressive strengthening from increased hydrogen bonding over time. Additionally, the tensile strength and Young's modulus decreased post-healing, suggesting reduced stiffness and improved flexibility at the healed junction (Table S3). These results confirm that the film composed of environmentally friendly materials is capable of autonomous healing at ambient conditions. Longer healing times toughen the films while also making them more stretchable. The self-healing is not limited to a unique break/healing cycle. Rather, the elastomers can potentially be perpetually healed, providing the elastomer maintains its self-standing mechanical integrity. Therefore, the lifespan of the sensor can potentially be prolonged indefinitely by self-healing of any micro- and macroscopic tears that form during repeated stress/release cycles. The sensor film exhibited moderate hysteresis with applied pressures of *ca.* 0.4–0.6 MPa (Fig. S6). This means it can withstand moderately applied loads without undergoing irreversible deformation. This recovery after

Table 1 Sensitivity of flexible pressure sensor compared with other sensors

Sensor type	Sensitivity range (V kPa^{-1})	Component	Ref.
Self-healing pseudo–piezoelectric sensor	0.13	Chitosan–choline-chloride-citric acid–**pPDS** films	This work
Flexible piezoelectric sensor with microstructure	0.5–1.5	PDMS substrate with microdome architecture, multi-walled carbon nanotubes (MWCNTs), ionic additives	44
Pseudo piezoelectric hydrogel-based sensor	0.1–0.8	Polyacrylamide hydrogel, graphene oxide nanosheets, borax crosslinker	45
MXene-based self-healing sensor	0.3–1.0	MXene ($Ti_3C_2T_x$) nanosheets, polyurethane matrix	46
PVDF/ZnO nanofiber sensor	$\sim$1.9	Electrospun PVDF–HFP/ZnO composite nanofibers	47
Ionic elastomer sensor	0.5–1.3	Silicone elastomer, ionic liquid-grafted silicone oil, microstructured dielectric layer	48
Polyurethane–CNT sensor	$\sim$0.25	Polyurethane sponge matrix coated with carbon nanotube (CNT)	49

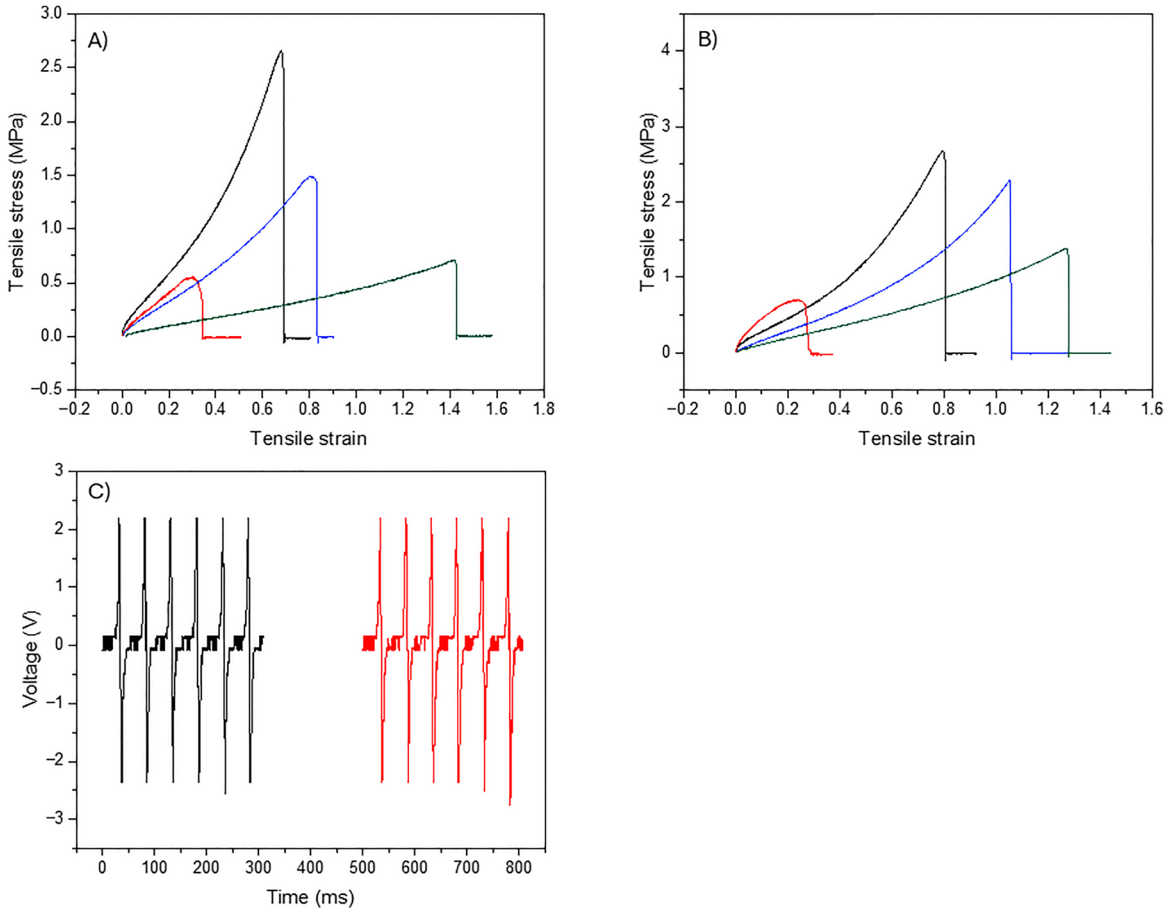

Fig. 3 Stress–strain measurements of elastomers without (A) and with 0.20 wt% **pPDS** (B) measured after given healing times: 5 (red); 30 (blue), and 60 mins (green) along with the corresponding film before damage and healing (black). (C) Output performance dependent on self-healing: before rupture (black) and healed films after rupturing (red).

applied load is ideally suited for detecting and transducing weakly applied pressures.

The effect of integrating the conductive polymer into the sensor on the self-healing was also assessed (Fig. 3B). The strain value across all the time points (5, 30, and 60 min.) was reduced for the films with the conductive polymer (0.20 wt%). The overall healing trend remained unchanged compared with the films without **pPDS**. The elongation at break was *ca.* 28% after 5 min. healing and increased with increasing healing times. Indeed, the elongation was *ca.* 100% at 30 min. and *ca.* 130% at 60 min. The conductive polymer did not inhibit the film from healing and it even enhanced its mechanical robustness. Regardless of the film composition, moisture was a critical factor in healing any cracks with only a minimal amount required to fuse the joints.

The pseudo–piezoelectric (piezoresistive) response of the sensor before and after healing is illustrated in Fig. 3C. The voltage output was consistent after healing, confirming the film's surface properties, and hence its ionic conductivity is not comprised by the break junction and subsequent healing. No significant ion loss and structural disruption were macroscopically observed and the induced cut was fully restored without visible scarring. In some cases, the reverse voltage post-healing was slightly increased compared to the pre-healing measurements, suggesting enhanced interfacial bonding.

To validate the sensor's performance under true usage, it was tested with human stimuli. For this, the sensor was assembled and subjected to single, double, and triple-tapping patterns with a finger. According to Fig. 4, the signal-to-noise ratio with tapping is *ca.* 16 dB. Single taps produced distinct voltage spikes, while double taps generated closely spaced signals, reflecting the rapid succession of the contacts. Notably, the second tap in a double sequence was often weaker than the first. This trend was also observed with triple tapping. Each tap produced a unique signal, with the first typically being the strongest due to natural human variability in applied pressure.

3. Materials and methods

3.1 Chemicals and reagents

All chemicals, reagents, and solvents were used as received. Chitosan (200–600 mPa s^{-1}), citric acid, and choline chloride were obtained from commercially available sources. The conducting polymer, **pPDS**, and choline chloride deep eutectic solvent (DES) were prepared according to known procedures.[33,34,50]

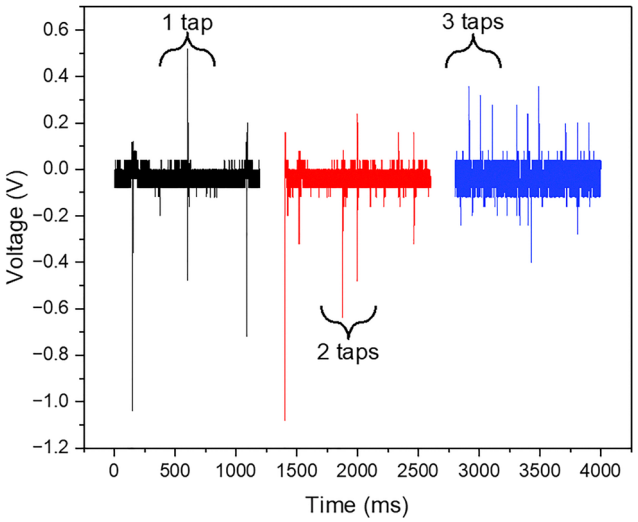

Fig. 4 Finger pressure tapping measurements. Output voltages *vs.* frequency of tapping: single (black), double (red), and triple (blue) taps.

3.2 Fabrication of films

The NADES solution was prepared first by mixing citric acid and choline chloride in a 0.7 : 0.3 molar ratio in DI water (50 mL). Then, high molecular weight chitosan was dissolved in this solution at 1.5 wt% relative to the total solvent weight. The mixture was stirred overnight until to fully dissolve the chitosan. The conductive polymer (**pPDS**) was added to the solution to afford a homogenous blend. The polymer was added in varying amounts (0.025, 0.05, 0.10, 0.15, and 0.20 wt% to the volume weight) by stirring for 30 minutes. The given solution was cast into a mold and cured at room temperature for about 3 days by the slow evaporation of water. Once completely dry, the films were cut into dog bone shapes using a mechanical punch for stretching, tensile strength, and elongation at break tests. The anhydrous/dry films were cut in half for the self-healing tests. Both pieces were then rejoined by slightly humidifying the contact area with a moist cotton swab and then drying at ambient conditions for 5, 30, and 60 minutes to promote polymer chain mobility and bond reformation. The healing efficiency was determined by mechanical testing.

3.3 Pseudo piezoelectric output measurement

The pseudo–piezoelectricity by electrostatic charge generation was measured *via* tapping between two electrodes in close proximity under mechanical pressure (see Fig. 2B). The chitosan/NADES films were cut into a circular shape with a diameter *ca.* 2.0 cm and used as one layer, while the conductive layer had the same dimensions. A linear pressure actuator was employed to provide periodic and consistent pressure of 15 kPa at a distance of *ca.* 0.8 cm on sandwiched layers. The output voltage was measured using a digital oscilloscope connected with various resistances, while the actuator was left to run for 10 s before the output data were recorded. This was to allow the system to reach mechanical stability. Similar measurements were conducted under the same conditions for the self-healed bilayer films. Sensitivity measurements were conducted by attaching the sensor on a rigid base, while a known force (weight/area) was applied perpendicular to the sensing surface using a small flat plate to distribute weight evenly. The sensor was connected to a source meter unit for measuring the output voltages.

4. Conclusion

A sustainable and self-healing pressure sensor was developed to have consistent performance when healed after mechanical damage. The sensor was enabled by electrostatic interactions at the interfaces of two elastomers having different electron affinity, producing a pseudo–piezoelectric effect when compressed. This mechanism enabled consistent and sensitive pressure detection without relying on conventional piezoelectric materials. This proof-of-concept underscores the viability of combined sustainable and biodegradable materials for advanced sensing applications, aligning with the growing emphasis on environmental circularity and resilient tactile electronics without compromising sensitivity and mechanical compliance. The development of pressure sensors that are not reliant on piezoelectric materials is an opportunity to fill the void of flexible and stretchable materials combined with touch sensitivity for tactile sensors to meet the needs for devices that offer both high precision and mechanical adaptability, especially for integration into wearable and soft electronic systems. Emerging pseudo–piezoelectric technologies using electrostatic and iontronic materials are ideal alternatives for replacing otherwise rigid piezoelectric materials for enhanced sensitivity by leveraging interfacial charge redistribution and ionic mobility with the benefits of elasticity and sustainability.

Author contributions

M. J. R. H. and W. S. prepared and characterized elastomers. W. S. and C. A. measured piezoelectric and mechanical properties of films. C. A. and W. G. S conceptualized the project, supervised the research, and drafted the manuscript. All authors reviewed the manuscript.

Conflicts of interest

The authors have no competing interests to declare.

Data availability

The data supporting the findings and the conclusions are included in both the text and the supplementary information. Supplementary information containing the chemical structures, absorption spectra, mechancial and electrical data is available. See DOI: https://doi.org/10.1039/d5ma01489a.

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