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Sensor for Human Motion Monitoring Utilizing Tough, Self-Healing, Polyurethane Elastomer

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Flexible strain sensors have made significant progress, but challenges remain in developing sensors that combine robust self-healing capability, adhesive properties, and recyclability for enhanced durability, ease of use, and reduced electronic waste. This study presents a novel self-healing polyurea-based elastomer synthesized from bis(aminopropyl) terminated polydimethylsiloxane (PDMS) and a mixture of aromatic and aliphatic diisocyanates, methylene bis (phenyl isocyanate) and isophorone diisocyanate. The polymer exhibits excellent mechanical strength, high elasticity, and autonomous self-healing properties by leveraging a balance of weak and strong hydrogen bonding interactions. The self-healing polyurea elastomer was applied in stretchable strain sensors integrated with carbon nanotubes (CNTs) via $\pi-\pi$ stacking interactions. Initial results of the sensor to test the pressure, angle changes, and tensile measurements are shown in the report, and applications will be detecting human motion and facial expressions, human-machine interaction, and electronic skin technologies.

I. Nomenclature

PDMS = *Polydimethylsiloxane*
SW-CNT = *single walled carbon nanotubes*
THF = *Tetrahydrofuran*
Mn = *Number-Average Molecular Weight*
MPa = *Megapascals (a unit of pressure or stress)*
FTIR = *Fourier-Transform Infrared Spectroscopy*
SHE = *Self-Healing Efficiency*
MPU = *Methylenebis(phenyl isocyanate)*
IU = *Isophorone Diisocyanate Urea*

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II. Introduction

Flexible and stretchable strain sensors have gained considerable attention due to their lightweight design, high sensing performance, and ability to endure large strains [1-4]. These features make them suitable for diverse applications such as healthcare monitoring, electronic skin, soft robotics, and human motion detection. These sensors rely on changes in piezoelectric capacitive sensors [5-11] or electrical resistance caused by mechanical deformation in conductive elastomers, which are often composites of elastomers with conductive fillers like carbon nanotubes (CNTs) [12], silver nanowires [13], MXene [14], or graphene [15].

Self-healing materials are an innovative class of substances designed to autonomously repair damage, extending their lifespan and functionality. Self-healing occurs through use dynamic covalent bonds, such as disulfide reactions, ester exchange, and Diels–Alder reactions, which can reversibly break and reform under specific conditions like heat or light [16-20]. Additionally, noncovalent interactions, including hydrogen bonding [21], host–guest interactions [22], and metal coordination, contribute to healing by enhancing elasticity and energy dissipation [23]. These combined strategies enable materials to recover from physical or chemical damage while maintaining mechanical performance, offering sustainable solutions to reduce waste and improve durability in advanced technologies. Conductive fillers as mentioned above such as carbon nanotubes (CNTs), silver nanowires, MXene, and graphene significantly enhance the functionality of self-healing materials, particularly in flexible electronics and sensors. These conductive fillers provide superior electrical conductivity, mechanical strength, and durability. CNTs are valued for their exceptional tensile strength, lightweight nature, and ability to maintain robust conductivity through π-π stacking interactions with the polymer matrix, even after repeated damage and repair cycles [24]. Silver nanowires offer excellent electrical conductivity and flexibility, forming efficient conductive networks that ensure stable performance in stretchable and self-healing devices [13]. MXene, a class of 2D materials with high conductivity and customizable surface properties, contributes to both the electrical and mechanical performance of self-healing polymers [14]. Similarly, graphene, with its outstanding electrical, thermal, and mechanical properties, provides a large surface area for strong interaction with the polymer matrix [15], maintaining conductivity and flexibility after self-repair. Together, these fillers enable self-healing materials to combine conductivity, strength, and adaptability, supporting advanced applications in sensors, wearable electronics, and other high-performance technologies. This paper explores the synthesis, characterization, and application of self-healing PDMS-based polymers with conductive fillers, focusing on their potential for use in soft electronics and jaw motion sensors.

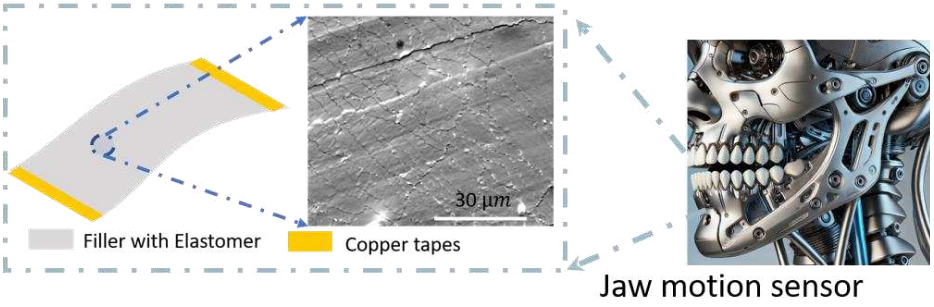

Fig 1. Schematic Representation of the Self-Healing Sensor Construction for Jaw Motion Sensing with a Scanning Electron Microscope Image of the Sensor Surface.

III. Methodology

The materials are as follows: Bis(3-aminpropyl) terminated poly(dimethylsiloxane), H2N-PDMS-NH2, Mn = 2500; Mn 850-900, Mn = ~5000, Triethylamine, anhydrous tetrahydrofuran (THF), methylene bis (phenyl isocyanate), isophorone diisocyanate, methanol was purchased from Sigma Aldrich and used as received. The single walled carbon nanotubes were purchased from American Elements as used as is.

A. The Self-Healing Polymer Synthesis and Characterization

In a nitrogen atmosphere, 15.0 grams of bis(3-aminopropyl)-terminated poly(dimethylsiloxane) (Mn = 2500, 1 equivalent) were added to a flame-dried 250 mL Schlenk flask, followed by 30 mL of anhydrous tetrahydrofuran (THF). The solution was cooled to 0 °C, and triethylamine (0.80 mL, 1 equivalent) was introduced. Separately, a 100 mL flame-dried round-bottom flask was charged with Methylenebis(phenyl isocyanate) (0.60 grams, 0.4 equivalents) and Isophorone diisocyanate (0.80 grams, 0.6 equivalents) dissolved in 15 mL of anhydrous THF. The diisocyanate solution was added dropwise to the reaction mixture over 15 minutes. The reaction mixture was allowed to warm gradually to room temperature and stir overnight. After 12–15 hours, the reaction was quenched with 0.5 mL of anhydrous methanol (10 equivalents). Stirring continued overnight, and the solvent was removed via rotary evaporation, yielding a solution containing approximately 40% polymer in THF. The polymer solution was poured into a silicone mold and cured under vacuum at 100 °C for 2 hours [25]. The self-healing PDMS and Sylgard 184 was characterized using Agilent Cary 630 Fourier-transform infrared (FTIR) spectroscopy with attenuated total reflectance (ATR). Spectra were averaged over 128 scans at a resolution of 4.0 cm^{-1}. As shown in Fig 2, the disappearance of the MPU and IU peaks containing the N=C=O end-groups around 2300 cm^{-1} confirms that the material synthesis has been completed, and all isocyanate groups have been consumed and reside within the new polymer chain [26]. Additionally, the successful preparation of PDMS-based poly(urea)s was proven by the appearance of typical peaks of the introduced urea moieties between 3250 to 3400 cm^{-1} related to the antisymmetric and symmetric stretching vibration νN-H and between 1500 to 1700 cm^{-1} typical for the urea amide I and amide II region. The urea amide I mode is mainly composed of the stretching vibration νC=O of the carbonyl group coupled to the stretching vibration νC-N and the deformation vibration δC-C-N of the urea moiety. Such bonds are not seen in the commercial Sylgard 184 PDMS.

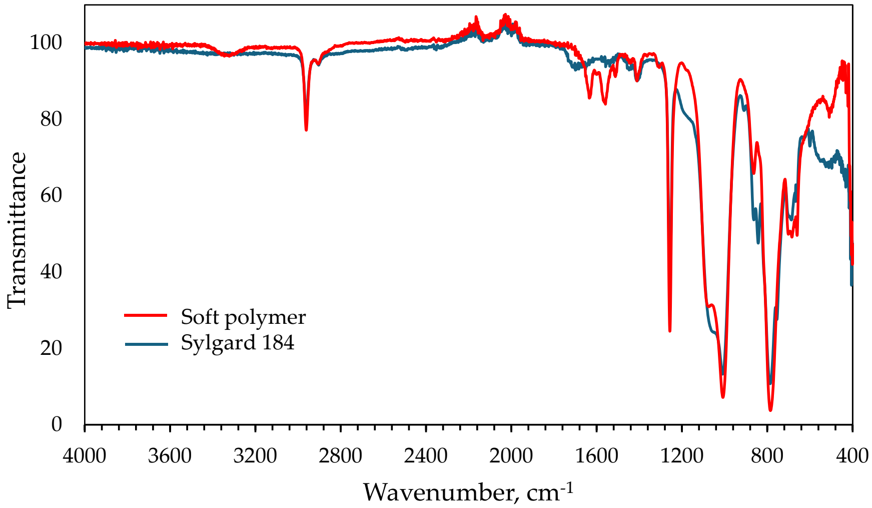

Fig 2. FTIR spectra of the developed Self-healing polymer compared to Sylgard 184.

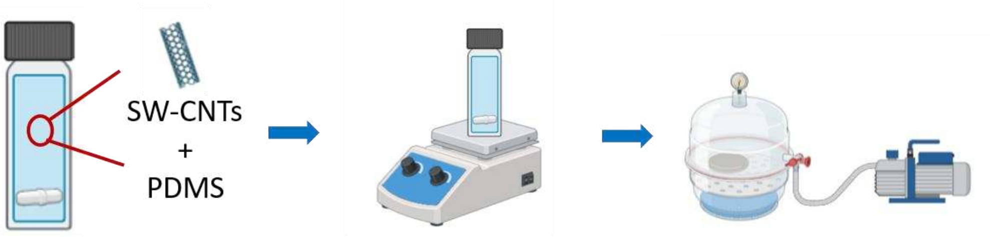

Fig. 3 Preparation Process of the Sensor: Development Stages.

B. Fabrication of Sensor

The first stage involved the fabrication of the sensor is the mixing of the specific amounts of CNTs with self-healing PDMS. The concentrations of 1.0 wt. %, 0.5 wt. %, 0.25 wt.%, and 0.125 wt. % were made by measuring the weight equivalent of single-walled carbon nanotube CNT (SW-CNT) and self-healing polymer. If the concentration was hard to mix, the THF was added to make a malleable mixture. The mixture was kept at slow mixing 150 rpm on a magnetic stirrer for 24 hours. The solution was filled into silicon mold which is degassed and cured in the vacuum for the next 24- 48 hours. This process ensures the bubble-free layer and removes the THF curing the self-healing PDMS, as shown in Fig 3. As a control Sylgard 184 was used to make similar molds with the same wt. % of the SW-CNT.

IV. Results

A. Mechanism Underlying the Concept

In this study, the self-healing mechanism and functionality of a sensor system composed of carbon nanotubes (CNTs) embedded in a polymer matrix. Initially, the CNTs form a conductive network within the matrix, enabling electrical conductivity. When damage occurs, the conductive pathways are disrupted, leading to a loss of functionality. During the healing process, polymer segments mobilize to bridge the gaps, reconnecting the CNT network. Once healed, the sensor regains its mechanical and electrical properties as shown in Fig 4A. Fig 4B and Fig 4C are the pictures of the diode that regains its electrical functionality after the self-healing process, demonstrating the successful restoration of conductivity and the reusability of the sensor system.

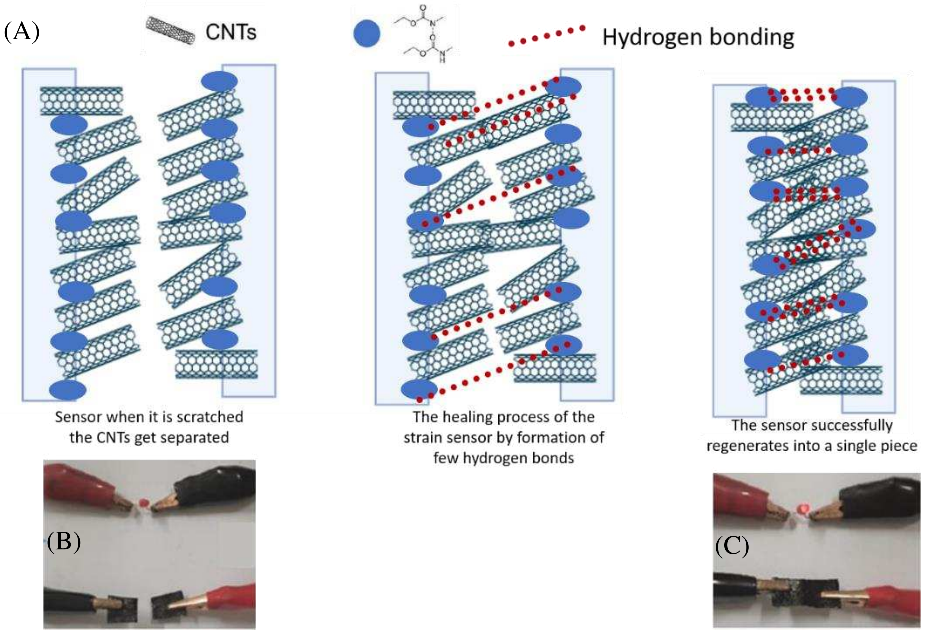

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Fig. 4 (A) Schematic diagram illustrating the self-healing mechanism of the strain sensor (B) Diode is not lit as the circuit is broken (C) diode is lit as there is the complete circuit.

A. Validation of Sensing Performance

The plots Fig 5A and 5B highlight the relationship in Sylgard 184 PDMS (control) between percentage change in resistance (%ΔR/R) and two variables: pressure and angle, for materials with 0.1 wt.% and 1 wt. filler concentrations. Under increasing pressure, as shown in the left plot, the material with 0.1 wt.% filler exhibits a pronounced response, with %ΔR/R rising steeply and reaching approximately 70% at 5 MPa, indicating high sensitivity to applied pressure. In contrast, the 1 wt.% filler demonstrates a more gradual change, with %ΔR/R peaking at 22% under the same conditions, reflecting reduced pressure sensitivity. The Fig 5B plot depicts %ΔR/R behavior with varying angles. For the 0.1 wt.% filler, %ΔR/R shows a steady increase considering 180° to be the control, while the 1.0 wt.% filler experiences a sharper increase in resistance as the angle changed from 180° to 60°. These findings suggest that the 0.1 wt. % filler provides greater sensitivity to pressure and angle response. This behavior underscores the influence of filler concentration on the material's mechanical and electrical responsiveness.

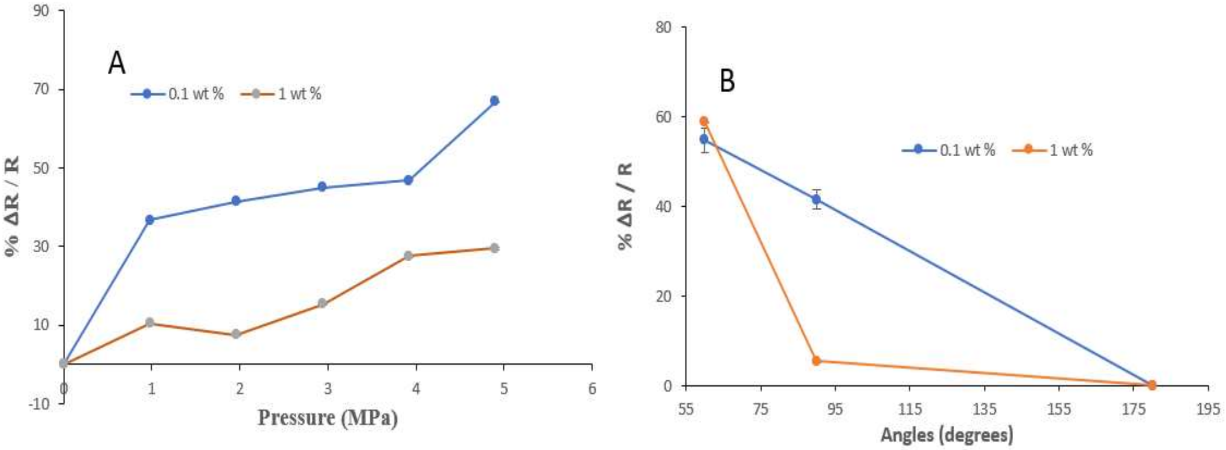

Fig. 5 Effect of Filler Concentration on Resistance Change for Sylgard 184 PDMS (control) under (A) Pressure and (B) Angular Deformation.

Given the information for the control PDMS, self-healing PDMS concentrations of the filler were limited to 0.125 wt % and 0.25wt %. Fig 6 illustrates the percentage change in resistance (%ΔR/R) under varying mechanical conditions for materials with 0.125 wt.% and 0.25 wt.% filler concentrations. In the Fig 6A plot, %ΔR/R increases with pressure, with the 0.125 wt.% filler showing a pronounced response, reaching approximately 60% at 5 MPa, indicating high-pressure sensitivity. In contrast, the 0.25 wt.% filler exhibits minimal changes, remaining nearly constant across the pressure range, reflecting significantly lower sensitivity. This is similar to the control sample as well. The Fig 6B, plot depicts %ΔR/R as a function of angle for 0.125 wt. % sample, showing a sharp decline as the angle increases from 80° to 180° (sample control). At 80°, the material demonstrates extremely high angular sensitivity, with %ΔR/R exceeding 2000%. These results highlight the substantial sensitivity of the 0.125 wt.% filler to pressure changes and the material's strong responsiveness to angular deformation, particularly at lower angles.

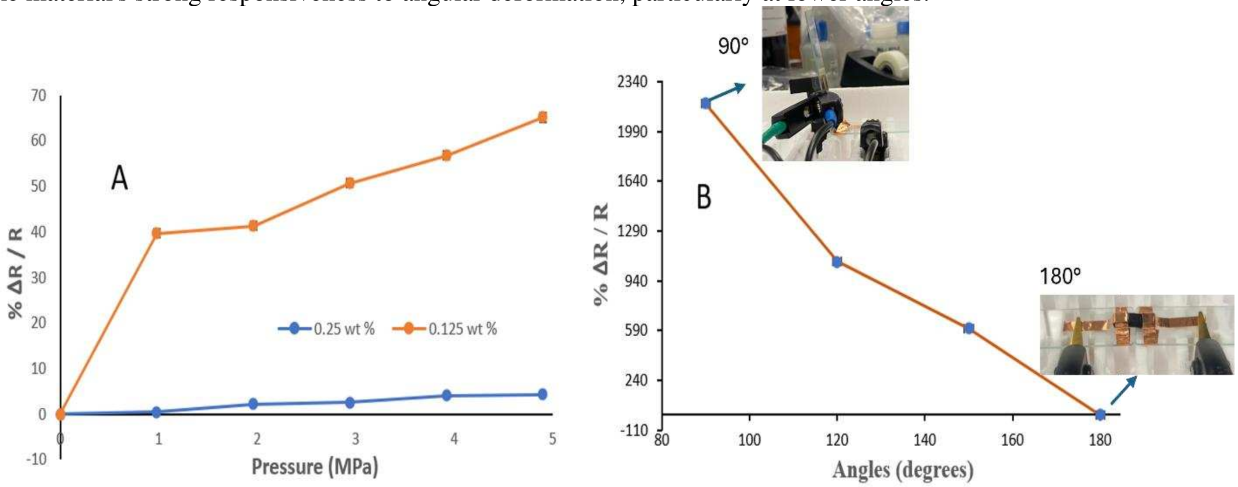

Fig. 6 Effect of Filler Concentration on Resistance Change for Self-healing PDMS (control) under (A) Pressure and (B) Angular Deformation (0.125 wt. %).

B. Validation of Self-Healing

Fig 6 illustrates the self-healing capability of a conductive polymer with integrated filler, evaluated through changes in resistance and microscopic analysis. In Fig 6A, the resistance remains stable in the "No scratch" state, indicating intact conductivity. However, upon scratching, a sharp increase in resistance is observed, reflecting a disruption in the conductive network. After 20 minutes, the resistance nearly returns to its original value, demonstrating effective self-healing and restoration of electrical conductivity. The accompanying microscopic images provide visual evidence of this process. The top image Fig 6B, taken immediately after scratching, shows a visible mark (2.5 mm) on the polymer surface caused by mechanical damage. In contrast, the bottom image, captured after 20 minutes, reveals substantial recovery of the scratched area, with the surface appearing almost intact. Together, these results highlight the polymer's ability to autonomously repair damage, making it suitable for applications requiring durability and electrical reliability.

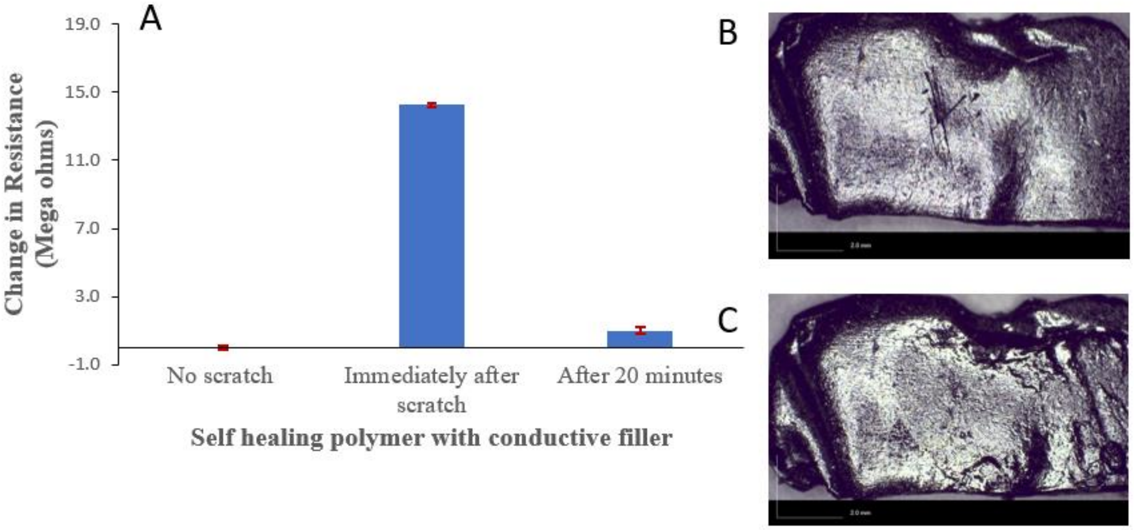

Fig. 7 Self-Healing Performance of Conductive Polymer: Electrical and Microscopic Analysis. (B) Scratch of the 2.5 mm on the surface (C) After 20 min.

V. Conclusion

This study successfully demonstrates the synthesis and characterization of self-healing PDMS-based polymers with integrated conductive fillers, showing their potential for applications in soft electronics and jaw motion sensors. The material exhibits exceptional self-healing properties, restoring electrical conductivity and structural integrity within 20 minutes of damage, making it ideal for flexible and durable devices. Furthermore, the polymer demonstrates high sensitivity to pressure and angular deformation, particularly at lower filler concentrations (0.125 wt%), which is crucial for detecting subtle motions such as jaw movements. These findings underscore the suitability of the self-healing PDMS for use in soft, wearable electronics and responsive sensing technologies for precise biomechanical monitoring.

Acknowledgments

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