

**Flexible Pressure Sensors Based on Electrospun PAN Fiber
Films Incorporating Graphene/Polypyrrole Composites and
Engineered PDMS Microstructures**

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Supplementary Information

Supplementary Notes

Supplementary Note S1

1 g of graphite powder was mixed with 10 mL of honey and magnetically stirred at 800 rpm for 30 min. Subsequently, the mixture was transferred to a three-roll mill. While the three-roll mill rotates, the high-viscosity medium, honey, was ensured to uniformly distributed across the rollers, and the natural graphite was mechanically peeled off for 5 h. After that, the mechanically stripped black material was transferred to a beaker, and the resulted viscous medium was rinsed with distilled water at 80 °C to obtain single-layer and multi-layer graphene. Lastly, single-layer graphene powder was obtained after the above-prepared material was centrifuged at 4000 rpm for 10 min and dried..

Supplementary Note S2

A homogeneous mixed solution was prepared by blending 33.468 g of N-methylpyrrolidone (NMP) with 4.005 g of 4,4'-diaminodiphenyl ether (ODA). 4.362 g of pyromellitic dianhydride (PMDA) was then added to the mixed solution and stirred thoroughly, yielding a polyimide (PI) precursor solution. This precursor solution was spin-coated onto a silicon wafer, and subsequently two-step curing processes were conducted at 50 °C for 5 min and 250 °C for 1 h, respectively. As a result, a PI film was fabricated. Through a photolithography process followed by thermal evaporation, interdigitated electrodes were deposited and patterned on the PI film surface, featuring an electrode width of 200 μm , a spacing of 100 μm , and an effective pressure-sensitive area of 3 mm $\times$ 3 mm. The resulted samples were then immersed in acetone, isopropanol, and deionized water to remove the photoresist and realize clean gold interdigitated electrodes.

Supplementary Figures

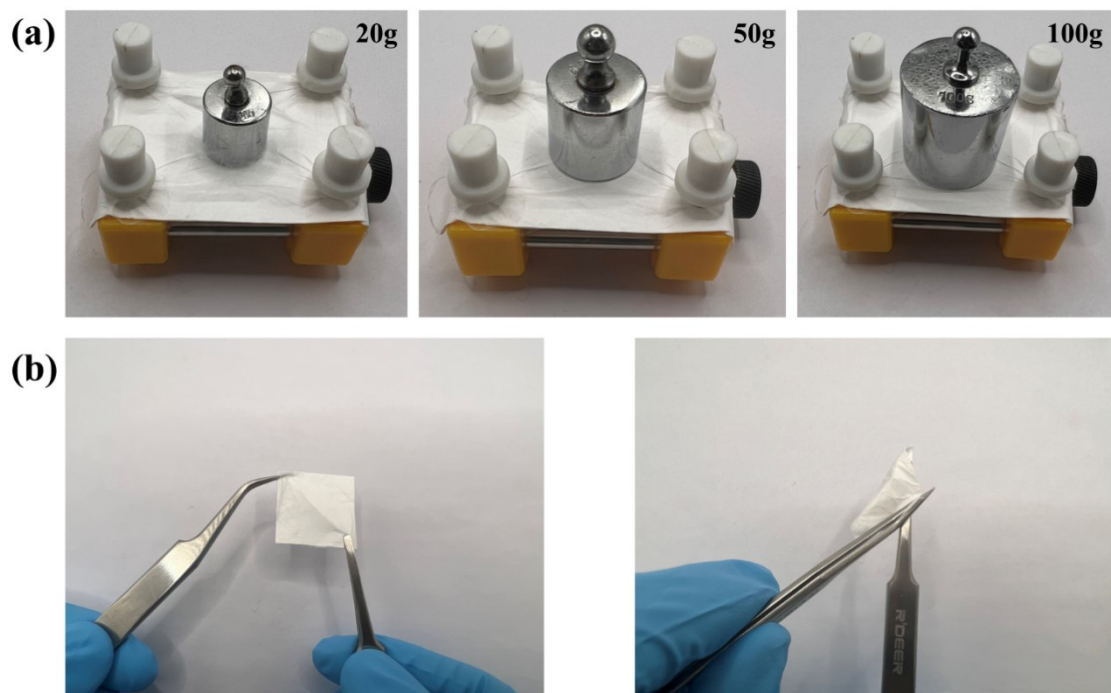


Fig. S1. (a) Demonstration of mechanical strength of the graphene (GR)/polypyrrole (PPy)@PAN nanofiber film. (b) Photos of the GR/PPy@PAN nanofiber film and demonstration of its mechanical flexibility.

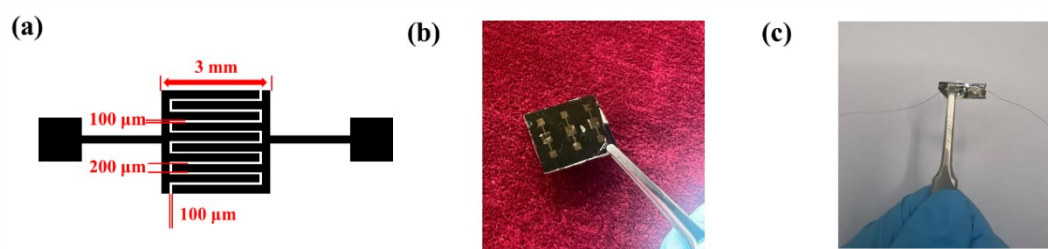


Fig. S2. (a) Schematic and dimensions of an interdigitated electrode. (b) The interdigitated electrodes printed on the PI film. (c) Piezoresistive material integrated on the interdigitated electrode and encapsulated with a polyethylene (PEN) film.

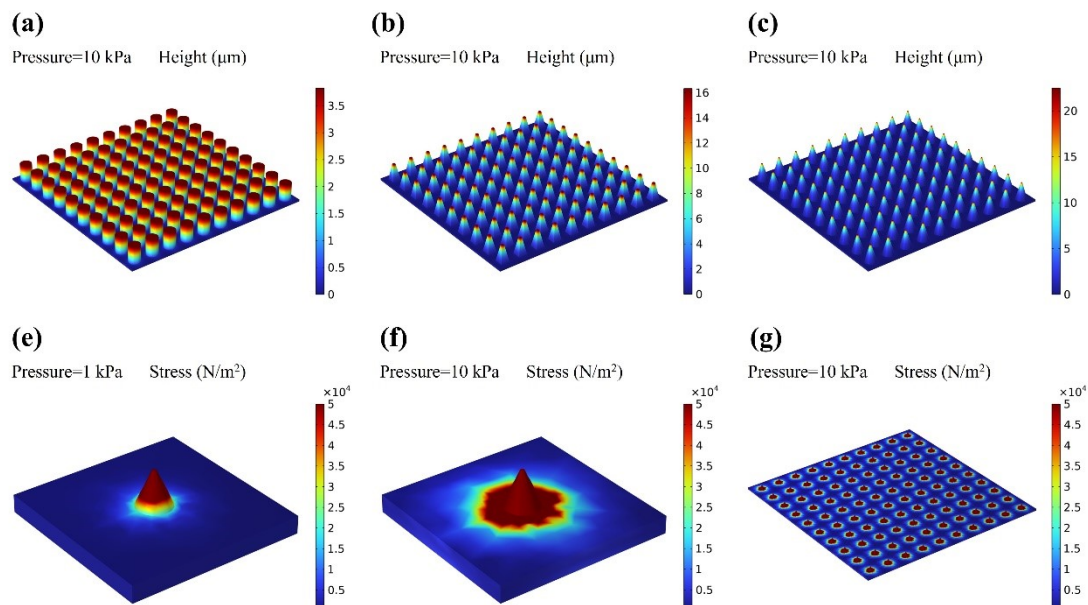


Fig. S3. (a-c) Simulated displacement distribution of samples with cylindrical, pyramidal, and conical microstructures under 10 kPa. (e-f) Simulated displacement distribution of a conical microstructure under 1 kPa and 10 kPa, respectively. (g) Simulated displacement distribution for an array of conical microstructures at 10 kPa.

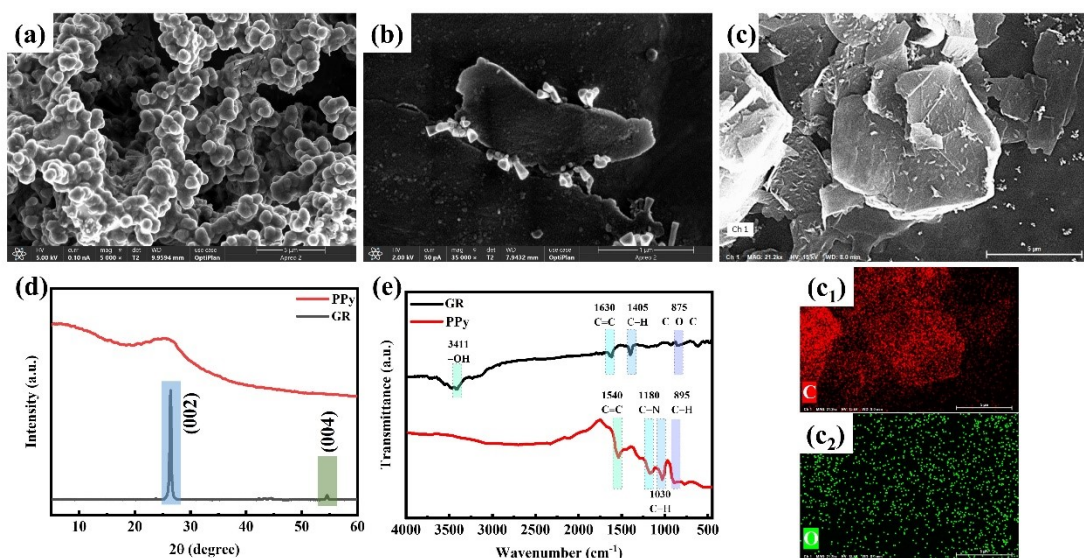


Fig. S4. (a) SEM image of self-polymerized PPy under the oxidant $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$. (b-c) SEM images of monolayer graphene. (c₁-c₂) EDS images of monolayer graphene showing the distribution of C and O elements. (d) XRD spectra of PPy and GR. (e) FT-IR spectra of PPy and GR.

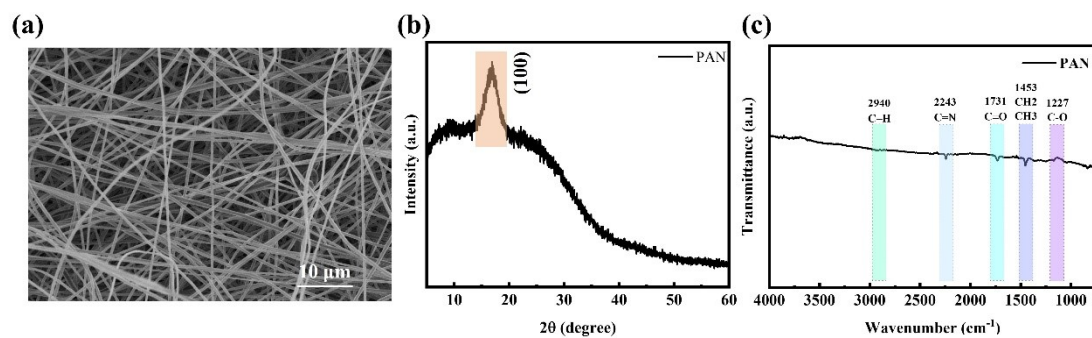


Fig. S5. Structural characterization results of electrospun PAN fibers. (a) SEM image. (b) XRD image. (c) FT-IR spectrum.

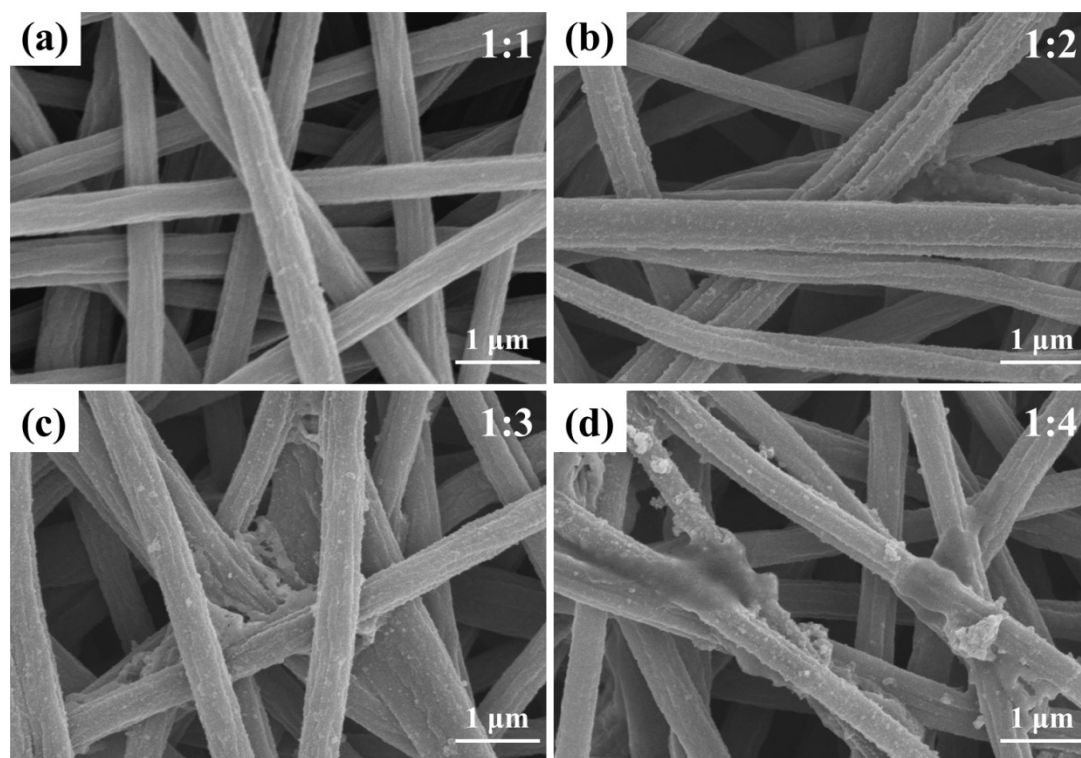


Fig. S6. SEM images of electrospun PAN fibers loaded with PPy nanoparticles prepared at different molar ratios of Py:FeCl₃·6H₂O. (a) 1:1. (b) 1:2. (c) 1:3. (d) 1:4.

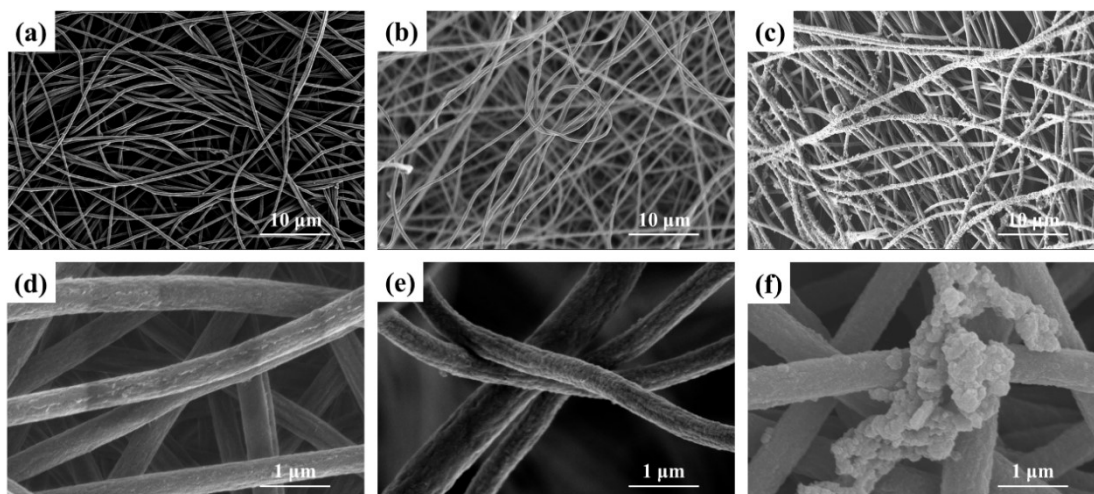


Fig. S7. SEM images of electrospun PAN fibers loaded with PPy nanoparticles prepared at different molar concentrations and Py at a molar ratio of $\text{Py}:\text{FeCl}_3 \cdot 6\text{H}_2\text{O} = 1:2$. (a-c) Images of electrospun PAN fibers loaded with 0.01 M, 0.03 M, and 0.05 M Py, respectively. (d-f) High-resolution SEM images of electrospun PAN fibers loaded with 0.01 M, 0.03 M, and 0.05 M Py, respectively.

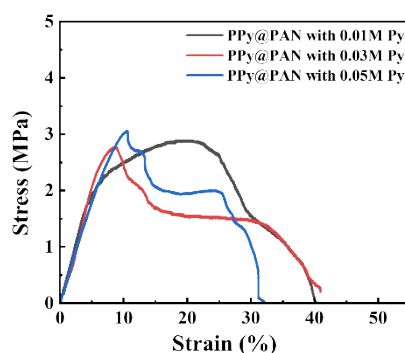


Fig. S8. Stress-strain curves of electrospun PAN fiber films prepared with different Py molar concentrations.

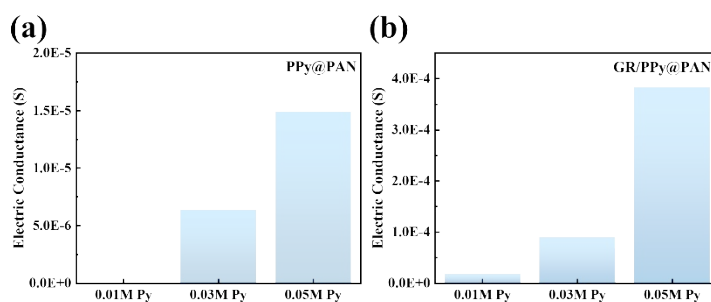


Fig. S9. (a) Electric conductance of electrospun PAN fibers included with PPy at different molar

concentrations. (b) Electric Conductance of electrospun PAN fibers included with PPy at different molar concentrations under GR loading.

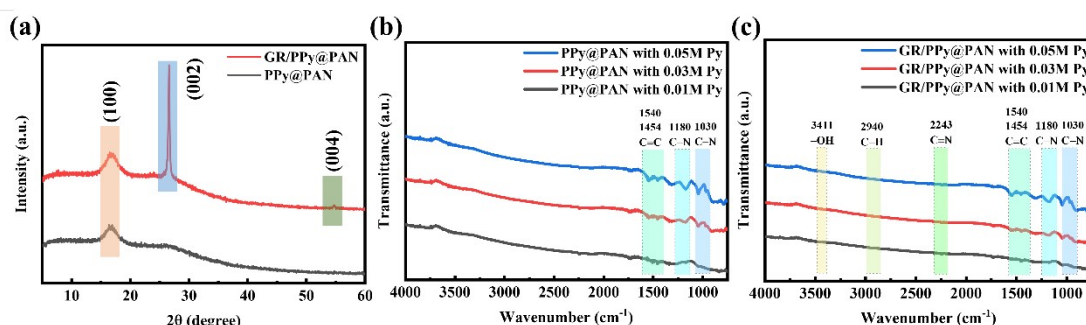


Fig. S10. Structural characterization of PAN fibers included with various filling materials. (a) XRD spectra of PPy@PAN and GR/PPy@PAN fiber films. (b) FT-IR spectra of PPy@PAN fiber films. (c) FT-IR spectra of GR/PPy@PAN fiber films.

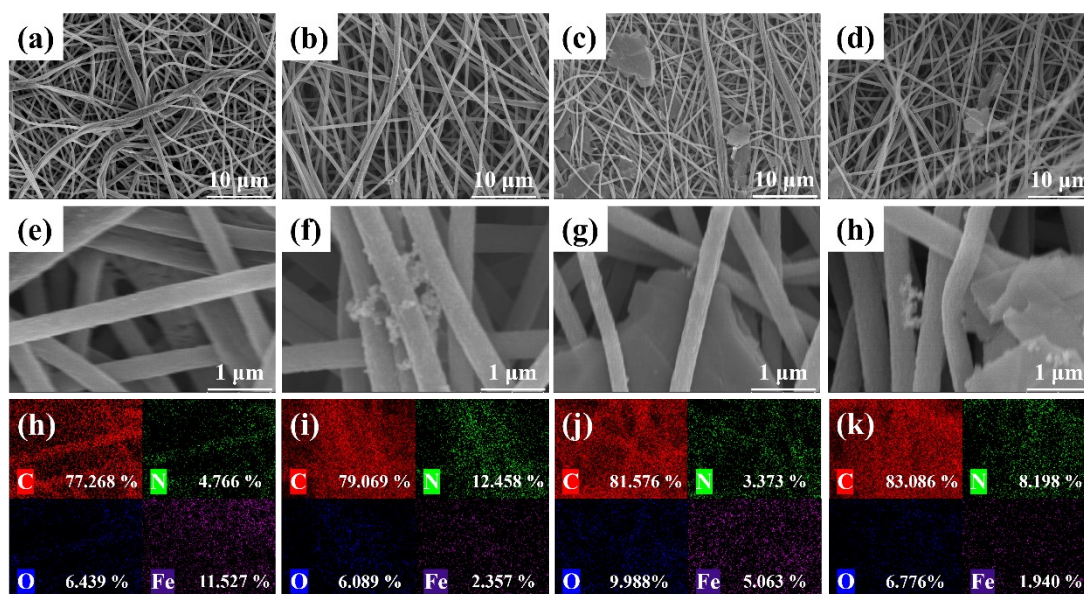


Fig. S11. SEM, high-resolution SEM, and EDS images of electrospun PAN fiber films included with PPy and GR. (a, e, i) 0.01 M Py, (b, f, j) 0.05 M Py, (c, g, k) 0.01 M Py and GR, (d, h, l) 0.05 M Py and GR.

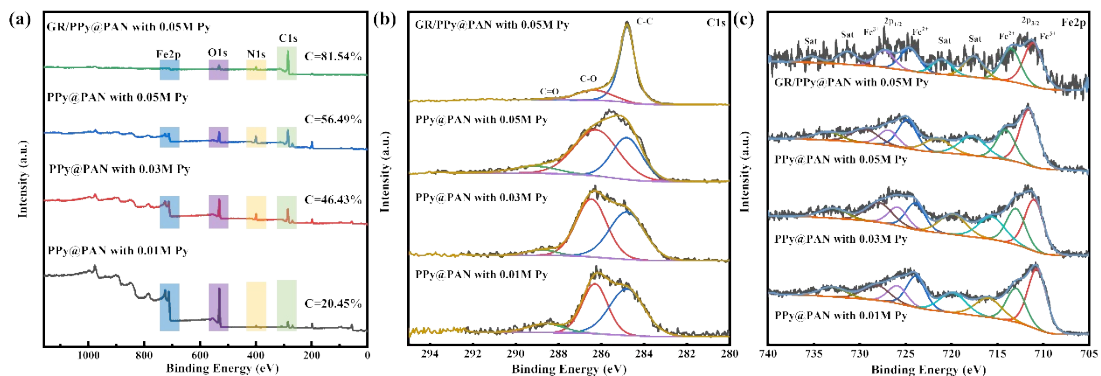


Fig. S12. XPS characterization for PAN fiber films included with PPy and GR. (a) XPS spectra of samples prepared with Py at molar concentrations of 0.01 M, 0.03 M, and 0.05 M. (b) C 1s XPS spectra and deconvoluted peaks. (c) Fe 2p XPS spectra and deconvoluted peaks.

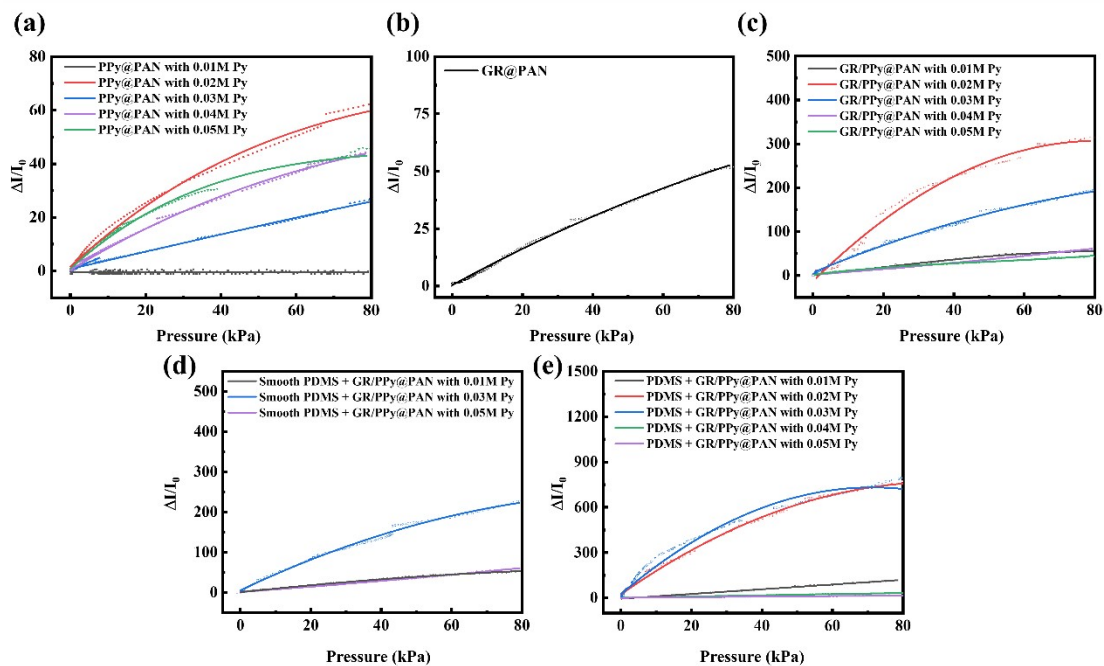


Fig. S13. Detection sensitivity of the piezoresistive pressure sensors based on various sensing layers. (a) PPy@PAN fiber films. (b) GR@PAN fiber film. (c) GR/PPy@PAN fiber films. (d) GR/PPy@PAN fiber films added with smooth PDMS layers. (e) GR/PPy@PAN nanofibers added with microstructured PDMS layers.

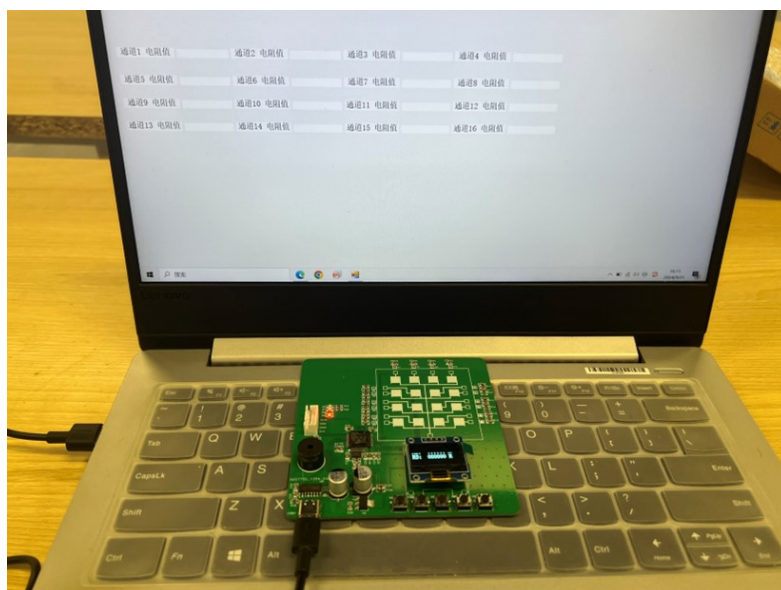


Fig. S16. Circuit board for the resistance detection array system implemented through the STM32 microcontroller and its connection to the host computer via serial port.

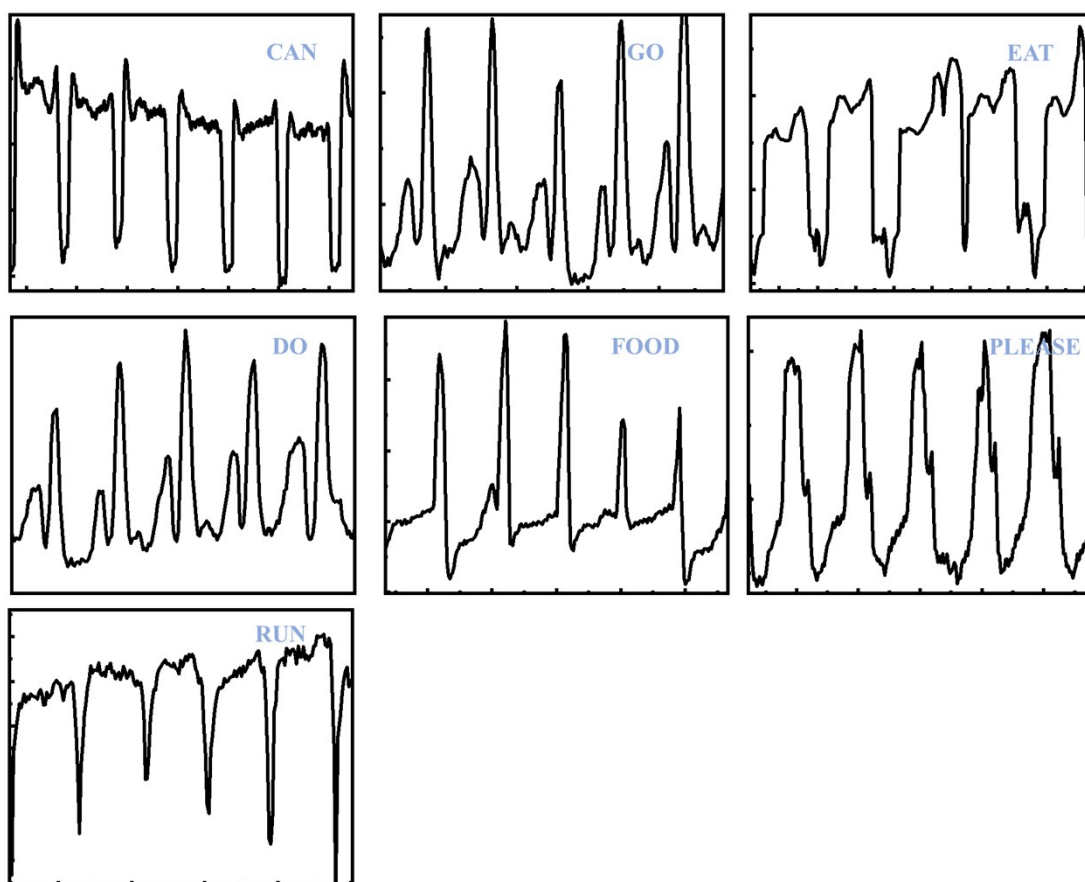


Fig. S17. The featured waveforms collected by the sensor when seven words: "CAN", "DO", "EAT", "FOOD", "GO", "PLEASE", and "RUN" are read and repeated.

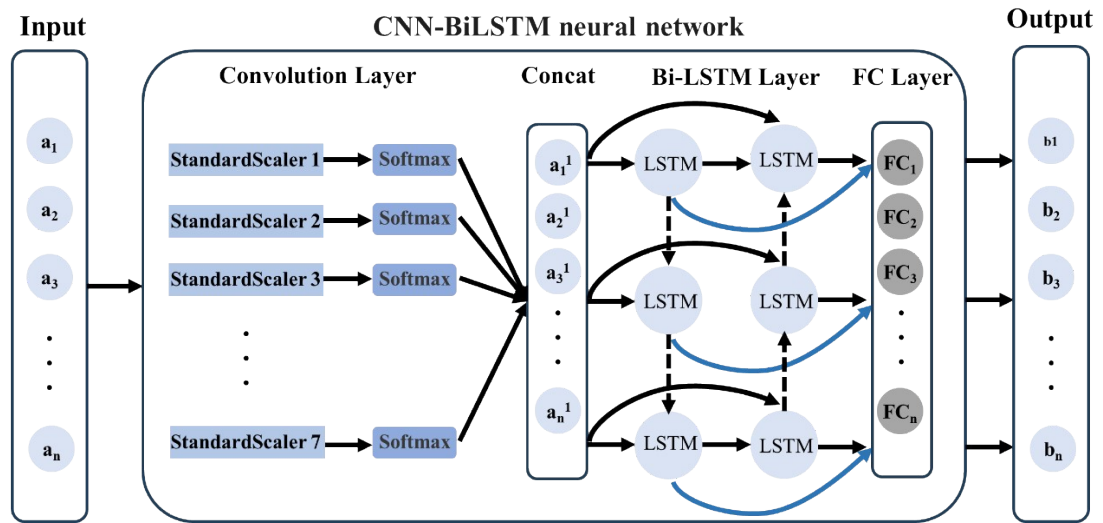


Fig. S18. Flowchart for speech recognition process using the long short-term memory (LSTM) algorithm.