Supplementary Materials for

**Heterogeneous integration of rigid, soft, and liquid materials for self-healable, recyclable, and reconfigurable wearable electronics**

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(available at advances.sciencemag.org/cgi/content/full/6/45/eabd0202/DC1)

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Supplementary Text

Chemical characterization of polyimine

To demonstrate the imine bond exchange reactions, compounds **aa** and **bb** were mixed in 1:1 mole ratio, and the model reaction was run at room temperature or 80 °C, as shown in Fig. S2B. ¹H-NMR characterization was conducted in CDCl₃ after mixing under room temperature for 5 minutes, 12 hours and at 80°C for 12 hours. The only distinguishable signal is the H on N-Me, 2.25ppm vs. 2.27ppm, due to the similarity of the structures. A new signal corresponding to the proton nuclei in the N-Me moiety of the target product **ab** showed up after mixing for only 5 minutes. The 12-hour reactions, at room temperature or 80 °C, showed similar ratios of the three species, indicating an equilibrium reached. These results clearly showed the imine bond exchange reaction is efficient at both room temperature and 80 °C.

The FTIR spectra of terephthaldehyde, original polyimine and recycled polyimine are shown in Fig. S2C. The disappearance of C=O stretch at 1681 cm⁻¹ and appearance of C=N stretch at 1640 cm⁻¹ in both original and recycled polyimine suggest a full conversion of aldehyde into imine linkage. Also, the high similarity between the FTIR spectra of the original polyimine and recycled polyimine support the notion that these two films are chemically identical.

Self-healing test and mechanical characterization

We investigated the influence of applied weight on the self-healing time of the polyimine encapsulated LM wire at room temperature, as shown in Fig. S6, C~G. It shows that the required self-healing time for polyimine encapsulated LM wire decreases with the increase of applied weight. For example, the required self-healing time with a 90 g weight was 144 minutes, which drastically decreased to 13 minutes with a 1000 g weight. To further demonstrate the self-healing performance, the cutting and self-healing process at the same location of a polyimine encapsulated LM conductor was repeated for 5 times. As shown in Fig. S6B, the resistance recovers to the original value after 5 times cutting and self-healing. As demonstrated in Fig. S6H, the LM conductor can still be stretched by 100% after 5 times self-healing. The stress-strain curve of the self-healed device was cut to rectangular with scissors (10 × 50 mm²) then measured using an Instron mechanical testing system (Fig. S6A).

Isolation of wires at intersection

To avoid short circuit, the red dotted line in Fig. S13A shows the isolation of wires on intersection. As shown in the right image, polyimine was inserted between the crossing wires for insulation.

Sensing performance test
The upper insets shown in Figs. 2, B ~ D, are completed devices laminated onto the skin of the forearm, forehead and neck, respectively, using 50μm thickness VHB double side tape. The device could be connected to the signal processing system through LM cable in the device and an etched copper foil cable out of the device (~18 × 82 mm²) (Fig. S13B). A thin lithium polymer battery (3.7 V, 45 mAh, GMB, China) was used to supply power and the total power consumption of our device is ~2.06 mW.

For ECG, three electrodes (AUVON TENS Unit Pads) with Electrode Gel (Spectra PAR12-02 Parker Laboratories 360 Electrode Gel) were connected to Chest1, Chest2 and Leg inputs. The Out1 and Out2 (ECG outputs) were connected to a PC microphone jack, and low-pass filter and high-pass filter were realized by digital filter using a python program.

For temperature sensing, Arduino as well as a 16 bit analog to digital converter (ADS1115) were used for measuring the voltage from Out3 (thermometer output) every 0.1 second.

For motion sensing, Arduino was used for measuring the voltage from Xout, Yout and Zout (accelerometer outputs) every 0.1 second.

For acoustic sensing, the ground and Z outputs from the accelerometer were plugged into a PC microphone jack, and the signal was recorded and analyzed by a python program.

**Design of the circuit**

Figure S13C illustrates the circuit of the multifunctional wearable electronics. For ECG sensing, chip AD8505 (Operational amplifier), chip R4 (100 kΩ resistor) and chip R5 (2 kΩ resistor) were used to amplify the voltage difference between chest1 and chest2 electrodes by 50 times. Chip R1 (2 kΩ resistor) and chip R2 (2 kΩ resistor) were used to form dual supply for the amplifier. Chip R3 (10 MΩ resistor) was used for the protection of electrical circuits.

Chip ADXL335 (accelerometer) was used for acceleration sensing. Chip C1 (0.1 μF capacitor) and an internal resistor (32 kΩ) of Xout formed low-pass filter for antialiasing and noise reduction. Chip C2 (0.1 μF capacitor) and chip C3 (0.1 μF capacitor) had the same functions as Chip C1 for Yout and Zout, respectively. The equation for the frequency is

\[ F = \frac{1}{2\pi \times R_i \times C_i}, \tag{S1} \]

where \( F \) is the low-pass frequency, \( R_i \) and \( C_i \) are the resistor and capacitor, respectively, and \( i \) is the output of X, Y or Z. Chip C4 (0.1 μF capacitor) placed close to the ADXL335 supply pins adequately decouples the accelerometer from noise on the power supply.
For thermometer sensing, chip MCP9700 was used to measure the body temperature.
Supplementary Figure Captions

(A) Multilayer construction of the multifunctional wearable electronics. (B) Optical images of the multifunctional wearable electronics from top view (top), tilted view (bottom left) and back view (bottom right). (C) Optical microscope images of the LM connection with the pins of chips at different configurations. Photo Credit: Chuanqian Shi, University of Colorado Boulder.
Fig. S2. Polymerization and characterization of polyimine. (A) Polymerization of polyimine. (B) Model reaction design. Model reaction scheme includes the two reactants aa and bb, and the product ab (top). $^1$H-NMR of aa, bb, mixture after 5 minutes at room temperature, mixture after 12 hours at room temperature, and mixture after 12 hours at 80°C. A new peak corresponding to the N-CH$_3$ around 2.26 indicates the formation of ab.
(C) FTIR of terephthalaldehyde (black), original polyimine (red) and recycled polyimine (blue).

Fig. S3. Sensing performance comparison between the wearable electronics and commercial devices. (A) Body temperature measurements from the wearable electronics compared with those from a commercial thermometer. (B) Acoustic data measured by the wearable electronics (top) and by a PC microphone (bottom). Photo Credit: Chuanqian Shi, University of Colorado Boulder.
**Fig. S4.** Optical images of skin before (top left) and after wearing a polyimine film (top right) for 12 hours (bottom left) and 24 hours (bottom right). Photo Credit: Pengcheng Zhu, University of Colorado Boulder.
Fig. S5. Mechanical properties of polyimine film. (A) The uniaxial tension stress-strain curve of a polyimine film (left) and the linear fitting at small strain (right). (B) The loading and unloading stress-strain curves of the polyimine film. (C) Optical microscope image of the cross section of an 100 μm-thick polyimine film (left), and the optical image of this film being stretched by 100% (right). Photo Credit: Pengcheng Zhu, University of Colorado Boulder.
Fig. S6. Self-healing performance of polyimine encapsulated LM conductors. (A) Stress-strain curves of the original and self-healed films. (B) Resistance data of a polyimine encapsulated LM conductor during self-healing. Optical images of the LM conductor before (C) and after (D) cutting. The required self-healing time of polyimine encapsulated LM conductor with three different applied weights, 90 g (E), 400 g (F) and 1000 g (G). (H)
The polyimine encapsulated LM conductor after 5 times self-healing. The self-healed films can be stretched by 100%. Photo Credit: Chuanqian Shi, University of Colorado Boulder; Pengcheng Zhu, University of Colorado Boulder.

Fig. S7. Resistance data of a polyimine encapsulated LM conductor subjected to rubbing (A) and pressing (B). (C) Resistance data of a polyimine encapsulated LM conductor
subjected to cyclic 60% uniaxial strain. Photo Credit: Pengcheng Zhu, University of Colorado Boulder.

**Fig. S8.** Max principal strain contours in polyimine when the multifunctional electronics is subjected to 30% biaxial strain (top), 60% uniaxial strain along vertical (middle) and horizontal (bottom) directions.
Fig. S9. Sensing performance of the devices under different loading conditions. (A) Motion data when the multifunctional electronics is subjected to 30% biaxial strain (top left), 60% uniaxial strain along horizontal (top right) and vertical (bottom left) directions. (B) Temperature data when the multifunctional electronics is subjected to 30% biaxial strain (top left) and 60% uniaxial strain along horizontal (top right) and vertical (bottom left) directions. The measurements using commercial thermometer are also shown for comparison. Photo Credit: Chuanqian Shi, University of Colorado Boulder.
Fig. S10. Sensing performance of the self-healed and recycled device. (A) Sensing performance of the self-healed device: Motion data (top left), Acoustic sensor data (top right), and Temperature measurements (compared with commercial thermometer results, bottom left). (B) Sensing performance of the recycled device: ECG signal (top left), Acoustic sensor data (top right), and Temperature data (compared with commercial thermometer results, bottom left). Photo Credit: Chuanqian Shi, University of Colorado Boulder.
Fig. S11. The time and temperature dependent mechanical properties of polyimine films. (A) Storage modulus, loss modulus and tan δ of polyimine versus temperature. (B) Relaxation test of polyimine at different temperatures.
Fig. S12. Sensing performance of multifunctional wearable electronics in different configurations. (A) ECG signals of the multifunctional wearable electronics in its second (left) and third (right) configurations. (B) Motion data of the multifunctional wearable electronics in its first (left) and third (right) configurations. (C) Acoustic sensor data of the multifunctional wearable electronics in its first (left) and second (right) configurations.
Fig. S13. Optical images of the LM circuitry and cable, and circuit design. (A) Optical image the LM circuitry (left), and optical microscope image of two crossing wires insulated using a polyimine interlayer (right). (B) Optical image of the cable for connecting the multifunctional wearable electronics with data acquisition equipment. (C) Circuit design of the multifunctional wearable electronics, the red lines mark the crossing wires. Photo Credit: Chuanqian Shi, University of Colorado Boulder.
| Type       | Number | Values | Manufacture       | Unit Price |
|------------|--------|--------|-------------------|------------|
| Resistor   | 3      | 2KΩ    | Bourns Inc.       | $0.04      |
| Resistor   | 1      | 100KΩ  | Bourns Inc.       | $0.1       |
| Resistor   | 1      | 10MΩ   | Bourns Inc.       | $0.074     |
| Capacitor  | 4      | 0.1μF  | Yageo             | $0.19      |
| ADXL335    | 1      | N/A    | Analog Devices Inc.| $6.08     |
| MCP9700    | 1      | N/A    | Microchip Technology| $0.32     |
| AD8505     | 1      | N/A    | Analog Devices Inc.| $1.61     |

Table S1. A list of the chip components used for the multifunctional wearable electronics.
Supplementary Movie Captions

**Supplementary Movie S1.**
Real-time recording of the ECG data during exercising.

**Supplementary Movie S2.**
ECG data during cutting and self-healing.

**Supplementary Movie S3.**
Self-healed device subjected to stretching and bending.

**Supplementary Movie S4.**
Motion data measured from a recycled device.

**Supplementary Movie S5.**
Motion data of the device in its second configuration and wrapped around the ankle.

**Supplementary Movie S6.**
Acoustic sensor data of the device in its third configuration and wrapped around the neck.