Conducting Polymer Films and Bioelectrodes Combining High Adhesion and Electro-Mechanical Self-Healing

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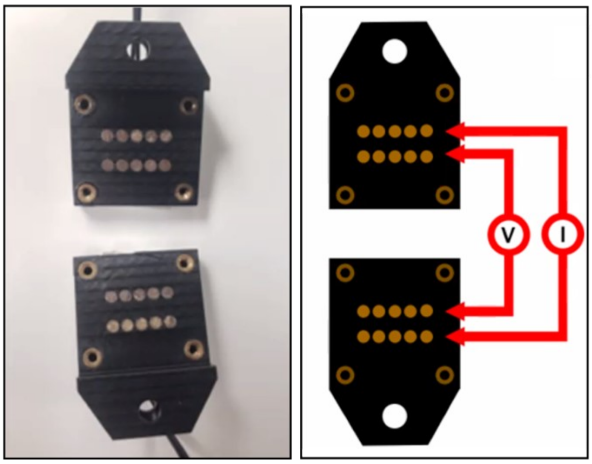

Fig. S1 The 4-points probe electrical tensile grips used in this study.

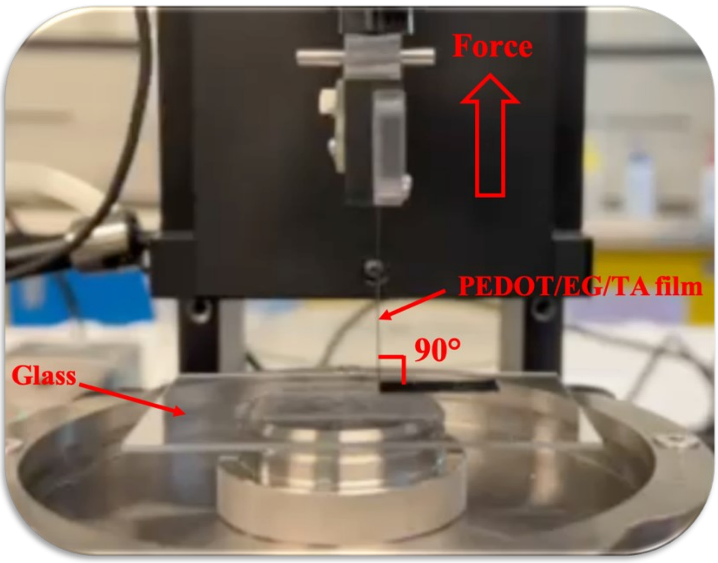

Fig. S2 90° peel-off testing configuration. The test involves peeling PEDOT/EG/TA films away from substrates (glass and porcine skin) at a 90-degree angle, forming a T-shape.

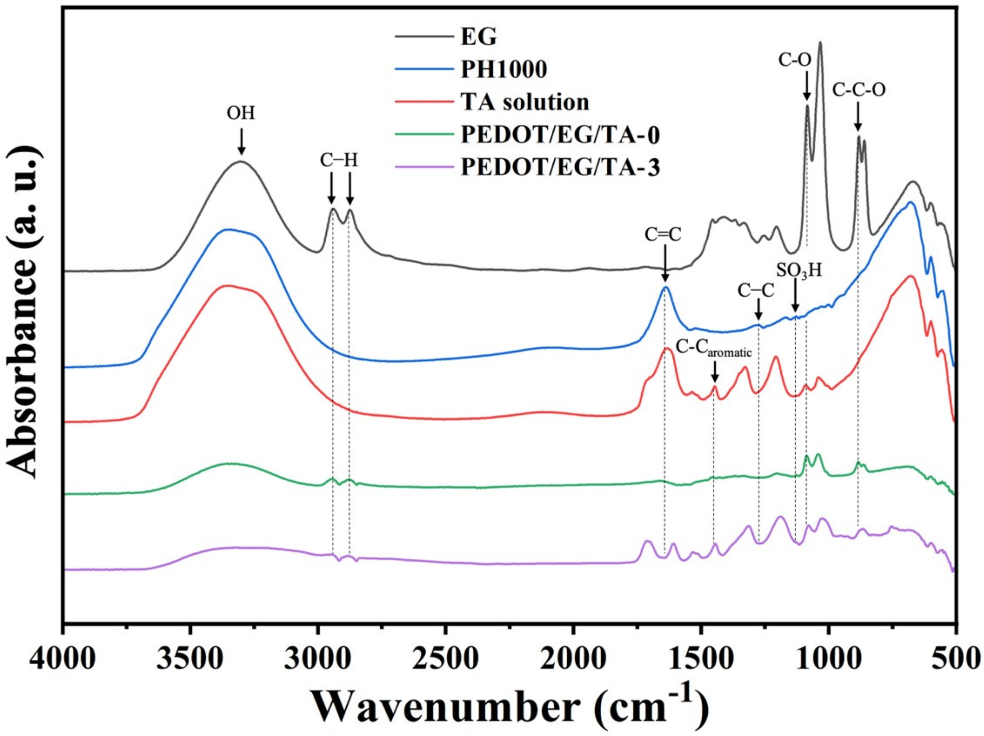

Fig. S3 FTIR spectrum of PEDOT/EG/TA films, EG, TA aqueous solution (0.12 g/mL), and PEDOT:PSS aqueous solution (PH1000).

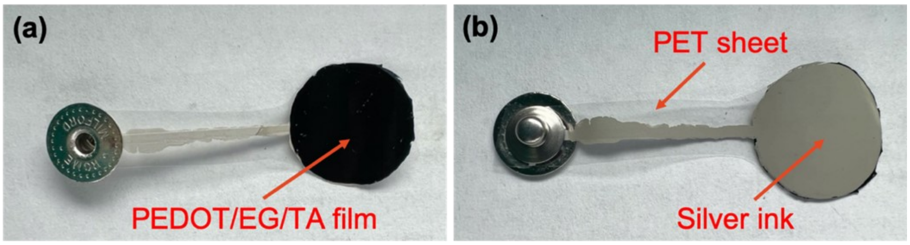

Fig. S4 PEDOT/EG/TA film electrodes. Front (a) and back view (b).

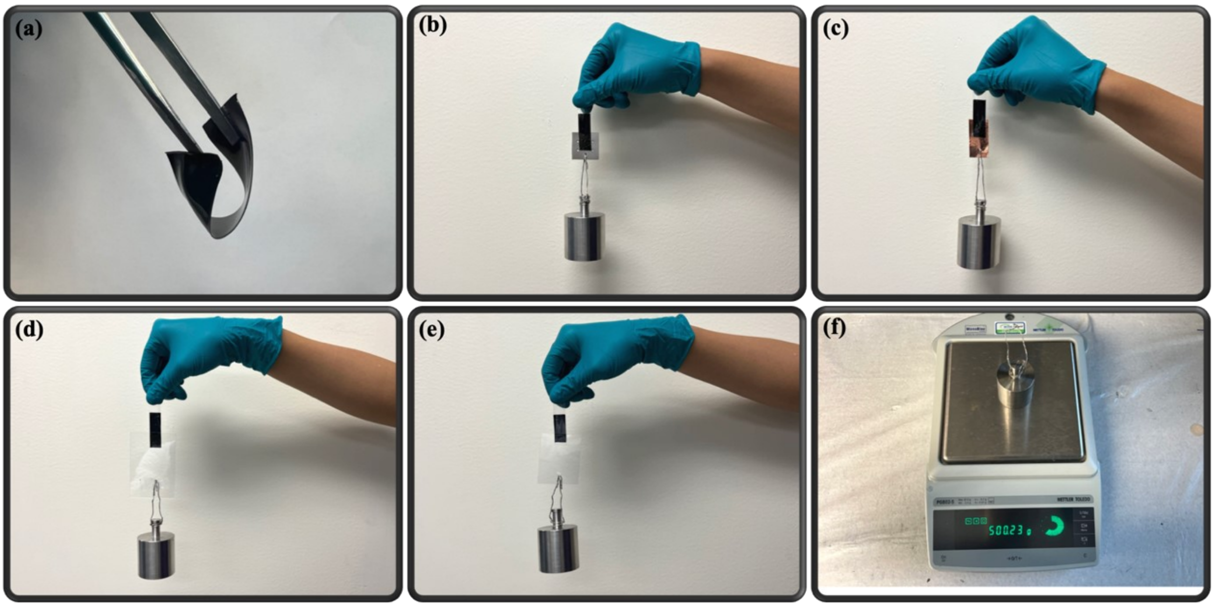

Fig. S5 Digital images. (a) Freestanding PEDOT/EG/TA-3 film adhering to tweezers. PEDOT/EG/TA-3 film adhering to various materials: (b) stainless steel, (c) copper, (d) glass, and (e) plastic PET sheet. (f) A weight of 500 g.

Table S1 Electrical tensile resistance change (*R/R_0*) of PEDOT/EG/TA films at 30% strain and break. Data for *R/R_0* (n=3) are reported as the mean ± standard deviation. *R* and *R_0* are the resistance at a strain and initial strain (0%), respectively.

Samples	R/R_0	
	at 30% strain	at break
PEDOT/EG/TA-0	1.022 ± 0.004	1.060 ± 0.013
PEDOT/EG/TA-1	1.027 ± 0.004	1.123 ± 0.007
PEDOT/EG/TA-2	1.012 ± 0.002	1.161 ± 0.016
PEDOT/EG/TA-3	1.016 ± 0.003	1.235 ± 0.018

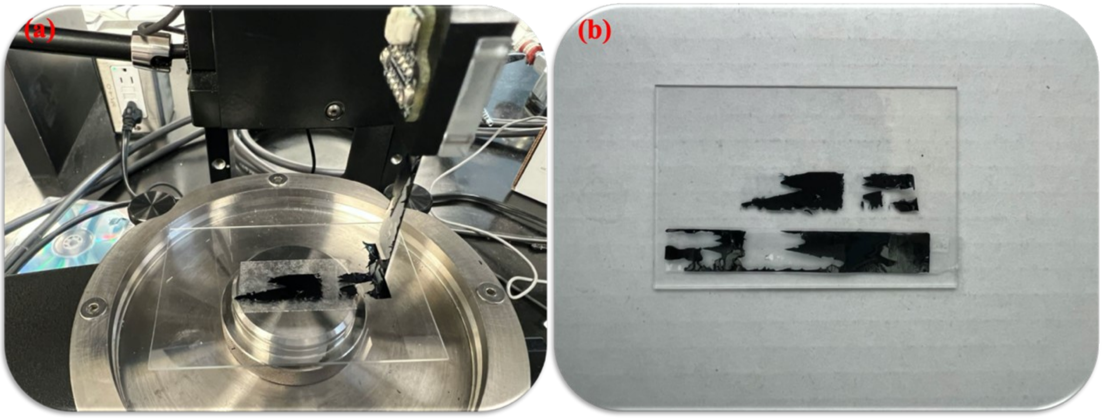

Fig. S6 PEDOT/EG/TA-3 film broke during 90° peeling off test. (a) 90° peeling off testing process and (b) the broken PEDOT/EG/TA-3 film on the glass.

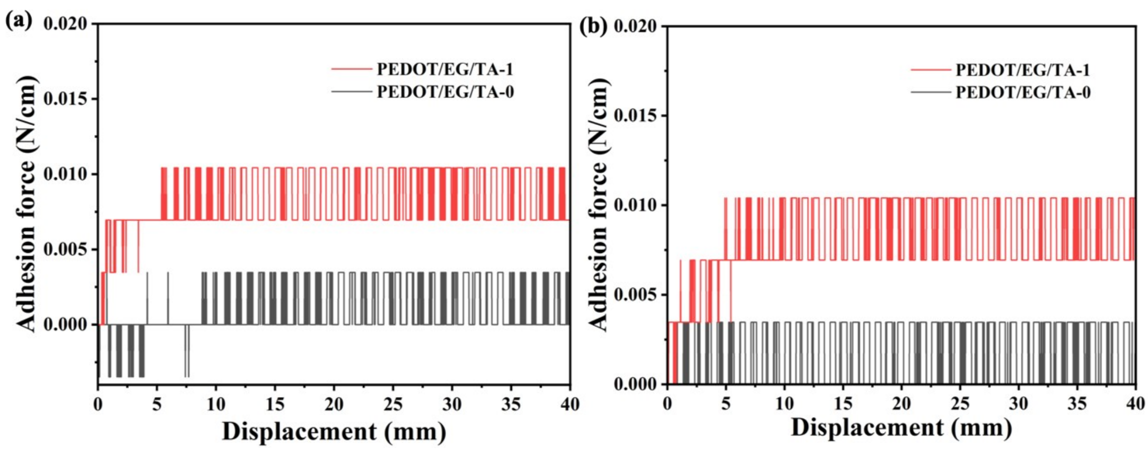

Fig. S7 90° peeling-off measurements of PEDOT/EG/TA films. Force versus displacement of PEDOT/EG/TA-1 and PEDOT/EG/TA-0 films on the glass (a) and on the pigskin (b).

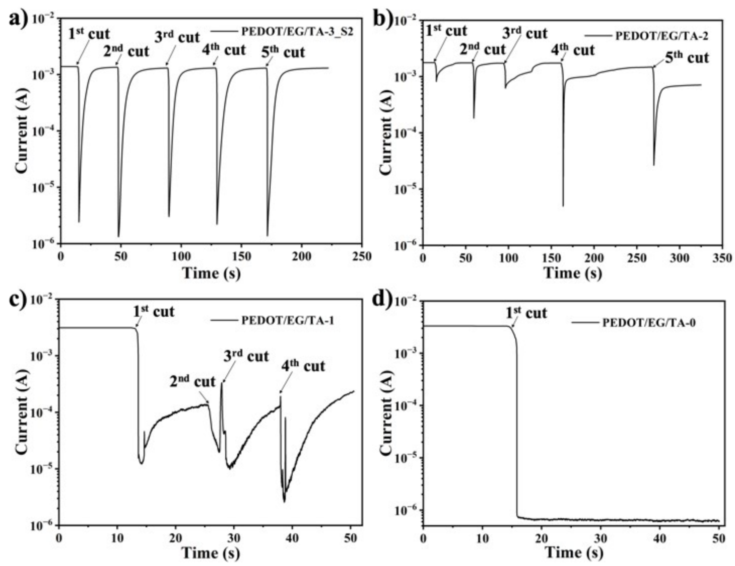

Fig. S8 Electrical autonomous healing properties of PEDOT/EG/TA films. Current versus time measurements of a) PEDOT/EG/TA-3 (sample 2), b) PEDOT/EG/TA-2, c) PEDOT/EG/TA-1, and d) PEDOT/EG/TA-0 films under a constant voltage of 0.02 V upon various cutting cycles. For PEDOT/EG/TA-1 sample, since the film had relatively low adhesion and self-healing properties and the current was very low after the 1st complete cut, there were more noises observed for the consecutive cuts.

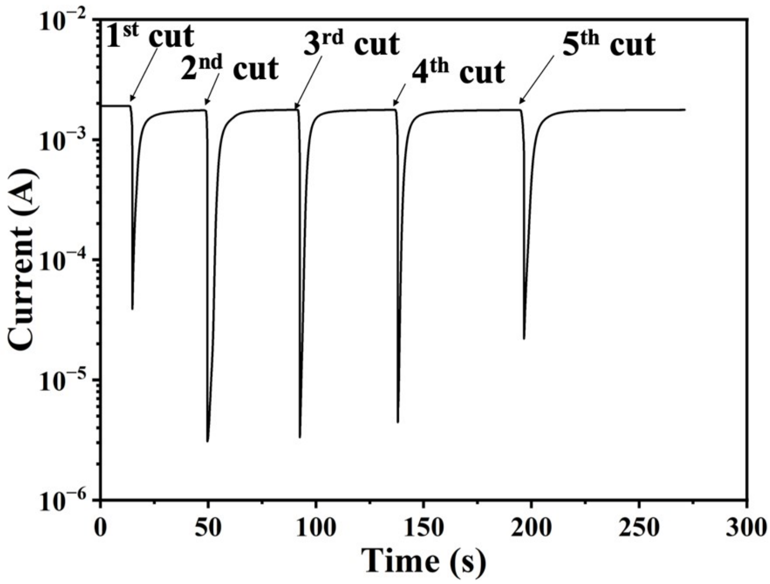

Fig. S9 a) Electrical autonomous healing process of PEDOT/EG/TA-3 films stored in a fridge for 21 days: current versus time measurements were performed under a constant voltage of 0.02 V upon various cutting-healing cycles.

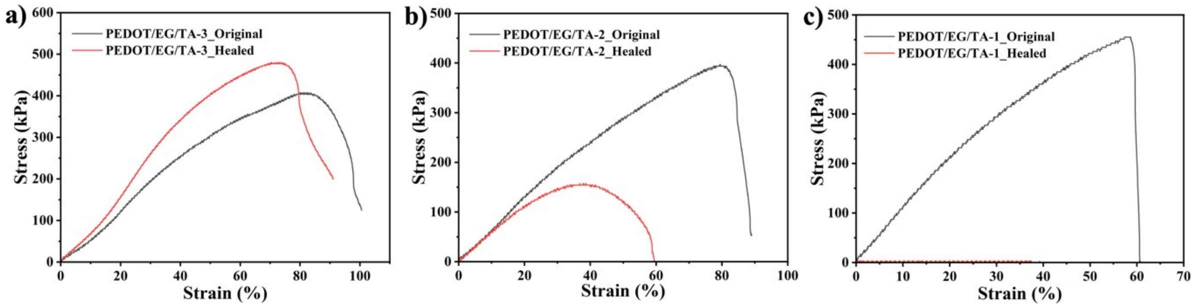

Fig. S10 Mechanical cut-stick healing process: tensile stress-strain plots of original and cut-stick healing of a) PEDOT/EG/TA-3 (sample 2), b) PEDOT/EG/TA-2, and c) PEDOT/EG/TA-1 films. The stress was calculated based on the thickness of nonoverlap area.

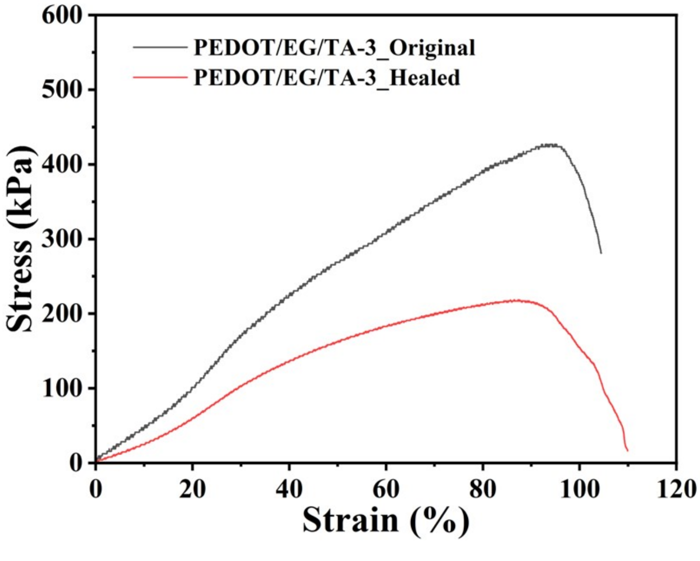

Fig. S11 Mechanical cut-stick healing process: tensile stress-strain plots of original and cut-stick healing of PEDOT/EG/TA-3film. The stress was calculated based on the thickness of overlap area.

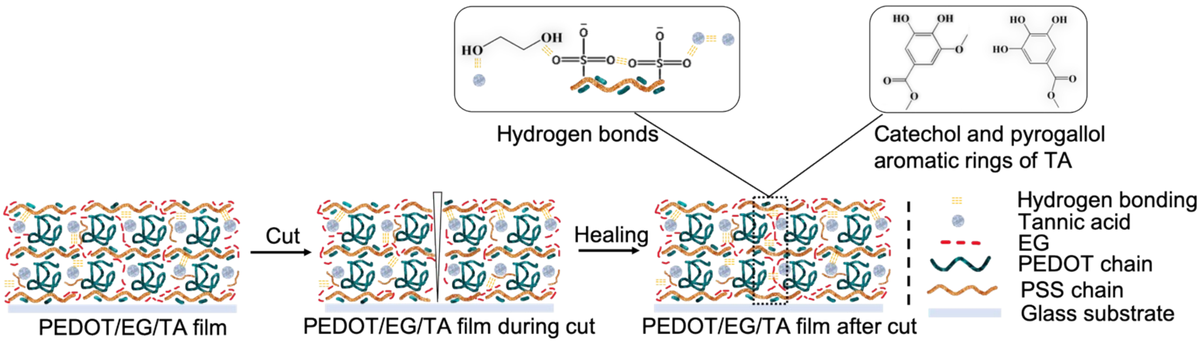

Fig. S12 Scheme of the proposed mechanism for the autonomous and cut-stick healing of PEDOT/EG/TA films.

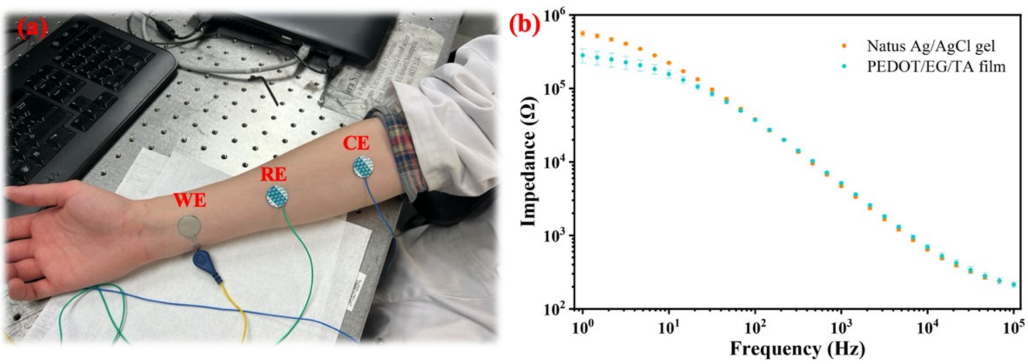

Fig. S13 Characterization of skin-electrode impedance. (a) Configuration of the skin-electrode impedance measurements of a volunteer. The electrode under test acted as the working electrode (WE), and commercial Ag/AgCl gel disk electrodes (Natus®) were used as the reference (RE) and counter (CE) electrodes. (b) Skin-electrode impedance versus frequency for PEDOT/EG/TA-3 film and commercial Natus® Ag/AgCl gel electrodes. Each electrode has a diameter of 18 mm. Measurements were performed on the same people, locations, and on the same day. Data for skin-electrode impedance are reported as mean ± standard deviation (n = 3).

Table S2 Comparison of skin-electrode impedance of Natus and PEDOT/EG/TA-3 film electrodes. Data for the impedance (n = 3) are reported as the mean ± standard deviation.

Frequency (Hz)	Impedance (KΩ)	
	Natus	PEDOT/EG/TA-3 films
1	558 ± 51	282 ± 61
10	223 ± 5	157 ± 20
100	38 ± 2	37 ± 1

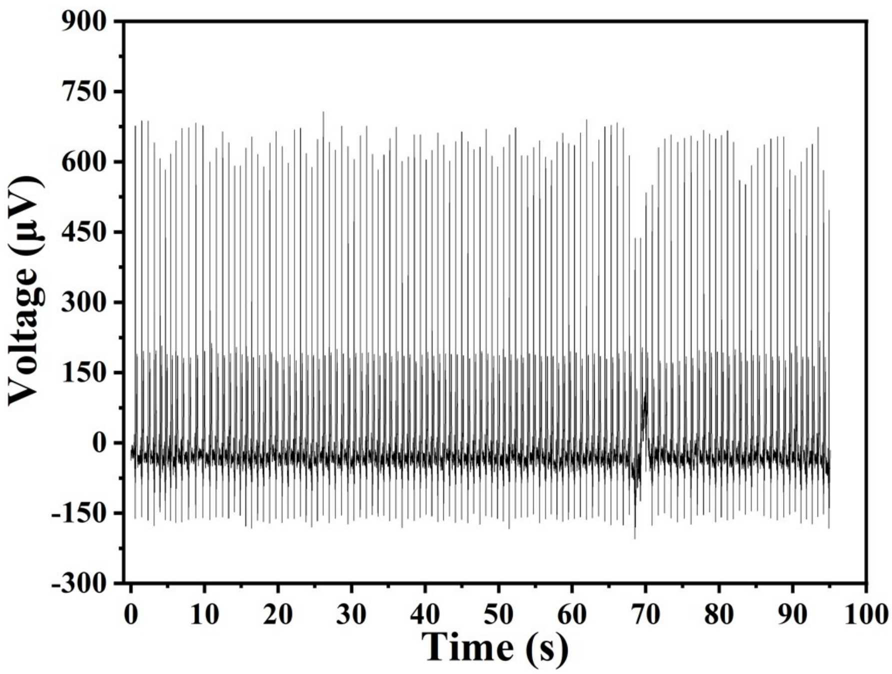

Fig. S14 ECG biopotentials (voltage versus time) recording with using PEDOT/EG/TA-3 film electrodes for over 90 seconds.

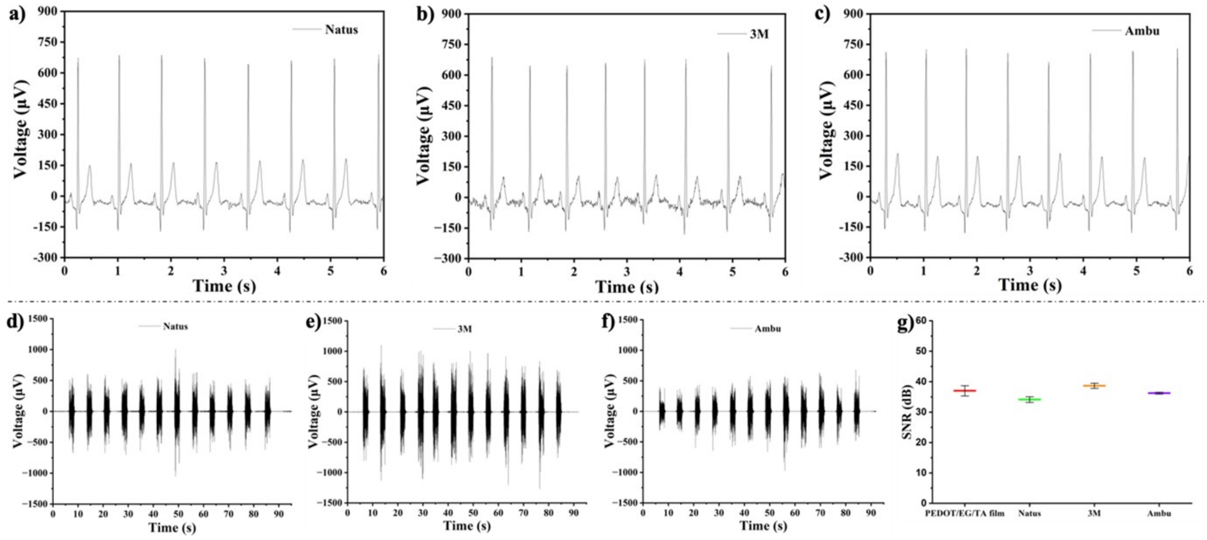

Fig. S15 ECG and EMG signal recordings for different commercial Ag/AgCl gel electrodes. ECG biopotentials (voltage versus time) were recorded using commercial Natus® (a), 3M® (b), and Ambu® (c) Ag/AgCl gel electrodes. EMG monitoring using commercial Natus® (d), 3M® (e), and Ambu® (f) Ag/AgCl gel electrodes during the relaxation and clenching phases. (g) Computed SNR values over the 90 s recording period for PEDOT/EG/TA film electrodes and three different commercial Ag/AgCl gel electrodes. SNR data are reported as mean ± standard deviation (n = 3).