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Electronic Supplementary Material

Autonomous Self-Healing, Self-Adhesive, Highly Conductive Composites Based on Silver-filled Polyborosiloxanes/Polydimethylsiloxane Double-Network Elastomer

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Materials

Hydroxy-terminated poly(dimethylsiloxane) (PDMS-OH) precursors with kinematic viscosity ranges of 65 cSt, 750 cSt and 18000-22000 cSt were purchased from the Sigma-Aldrich Company. Boric acid (BA) was purchased from Sinopharm Chemical Reagent Co., Ltd., and grinded, sieved *via* a 100-mesh sieve aluminium screen and dried for 24 h before used. Silver nanowires (diameters around 30 nm, length around 30 μm) were purchased from Zhejiang Kechuang Advanced Materials Technology. Silver micro-flakes (SF-01C, 2~7μm in diameter and 100~150 nm in thickness) were obtained from Sichuan Zhongchai Great Wall Precious Metal Co. Ltd.. Siloxane prepolymers (dimethylhydrogen siloxane, dimethylvinyl-terminated dimethylsiloxane) was obtained from Dow corning. All other chemicals were analytical grade reagents and used directly without further purification.

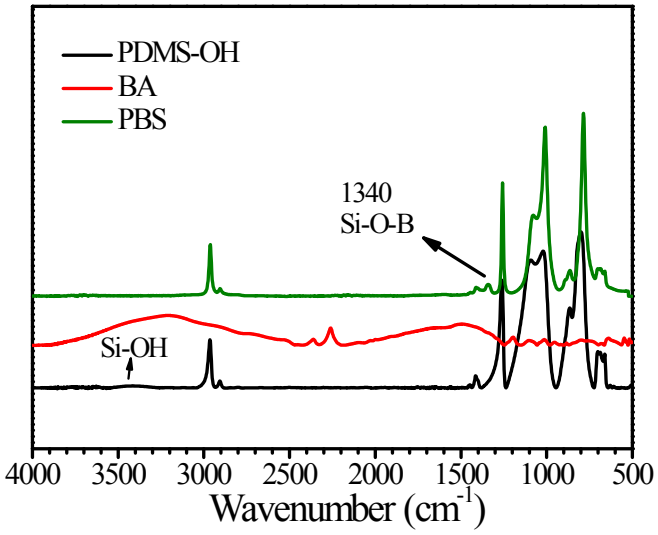

Figure S1. FTIR spectra of pure PDMS-OH, BA and obtained PBS.

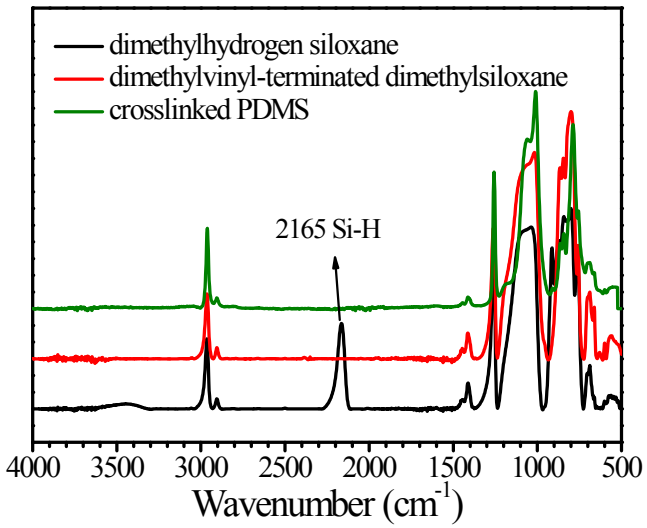

Figure S2. FTIR spectra of dimethylhydrogen siloxane, dimethylvinyl-terminated dimethylsiloxane and crosslinked PDMS. After cross-linking through hydrosilylation reaction, the absorption band at 2165 cm^{-1} for Si-H groups disappeared, indicating the hydrosilylation reaction between the Si–H groups and vinyl groups in siloxane prepolymers.[1]

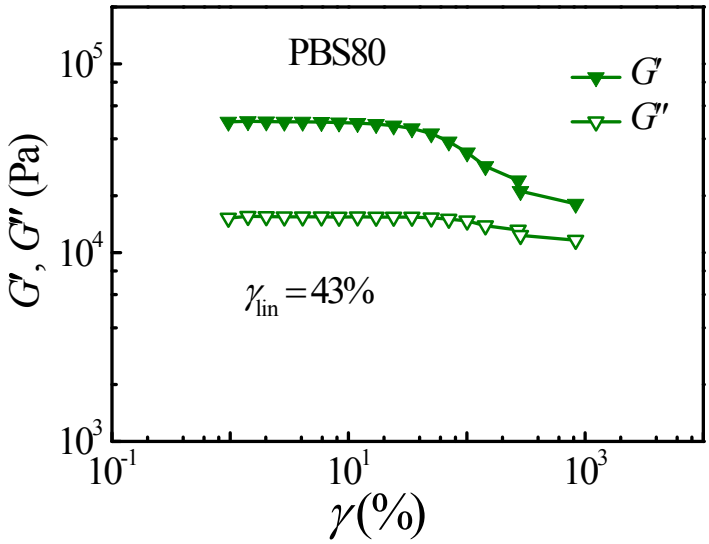

Figure S3. Strain sweep measurements of PBS80 at 25 °C with a frequency of 1.0 Hz. The storage (*G*′) and loss (*G*″) moduli were recorded as a function of strain (*γ*).

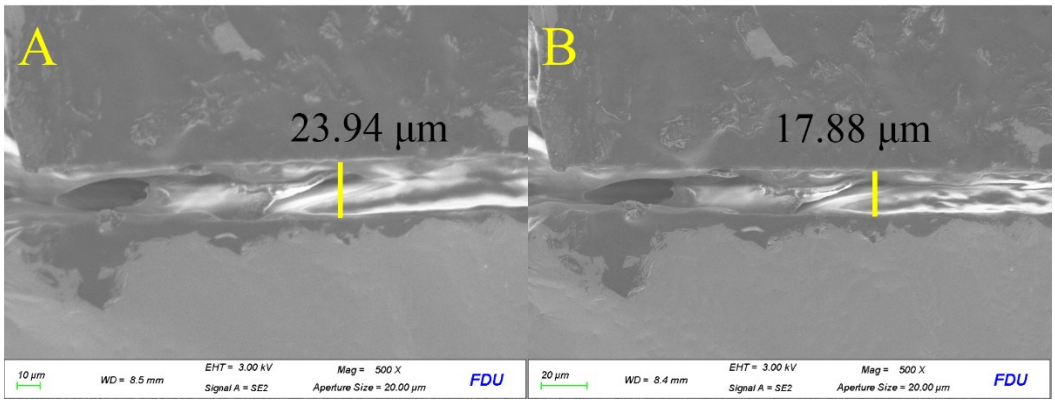

Figure S4. SEM images for PBS85 during self-healing process. The healing speed can be calculated according to the time interval (16 min) between A and B. (0.38 μm/min).

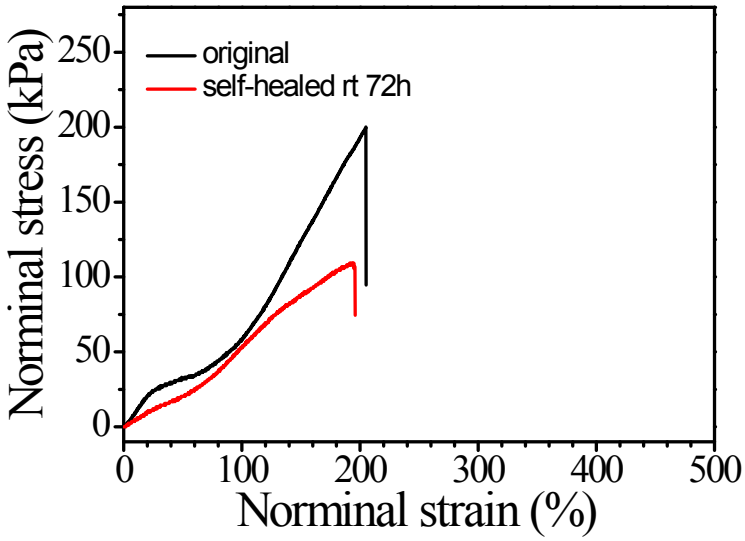

Figure S5. Stress-strain curves of original PBS85-m and its self-healed sample after 72h at room temperature

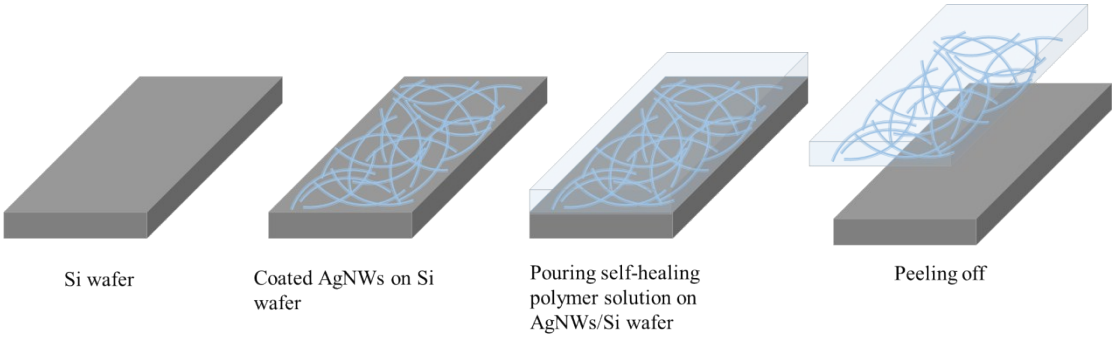

Figure S6. Schematic illustration of the fabrication process for the self-healable stretchable electrode by embedding AgNWs conductive network into self-healing polymer matrix.

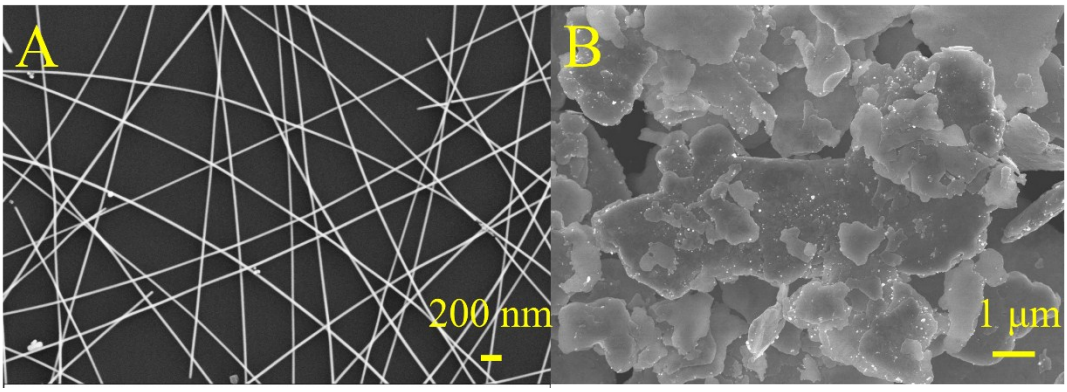

Figure S7. SEM images of (A) AgNWs and (B) KI treated silver flakes.

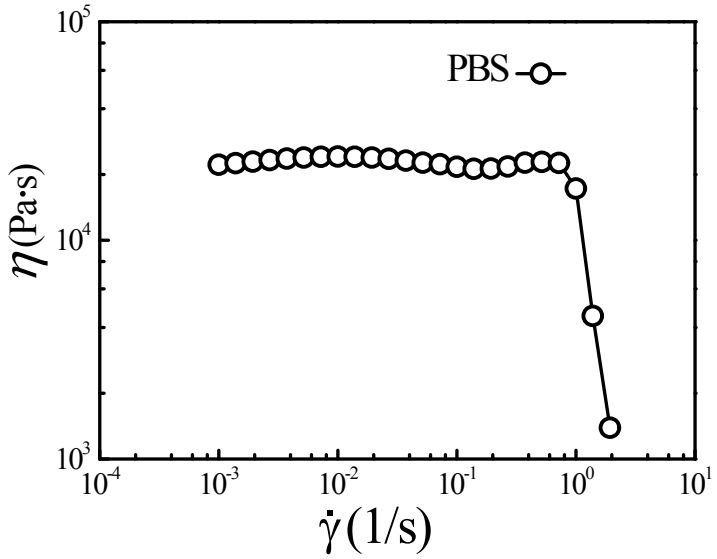

Figure S8. Steady shear viscosity profiles for PBS.

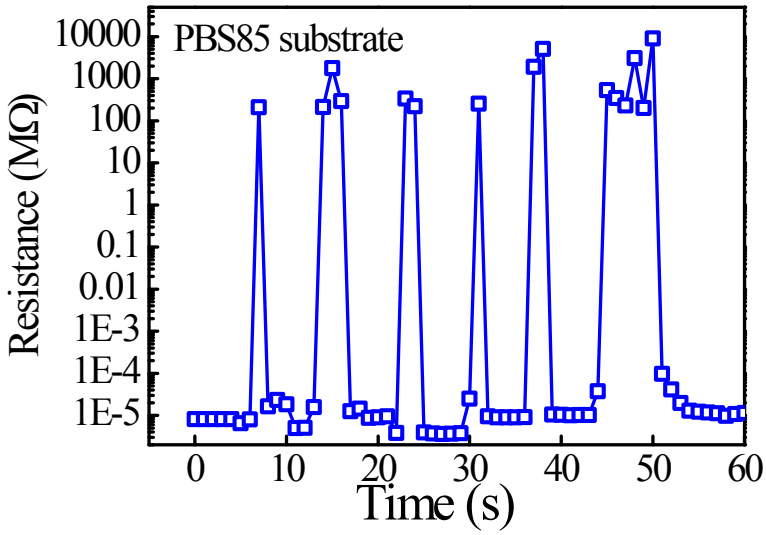

Figure S9. Plots of resistance of damage-resistant electrode prepared by silver flakes/PBS85 with PBS85 substrate as a function of time while making six consecutive cuts.

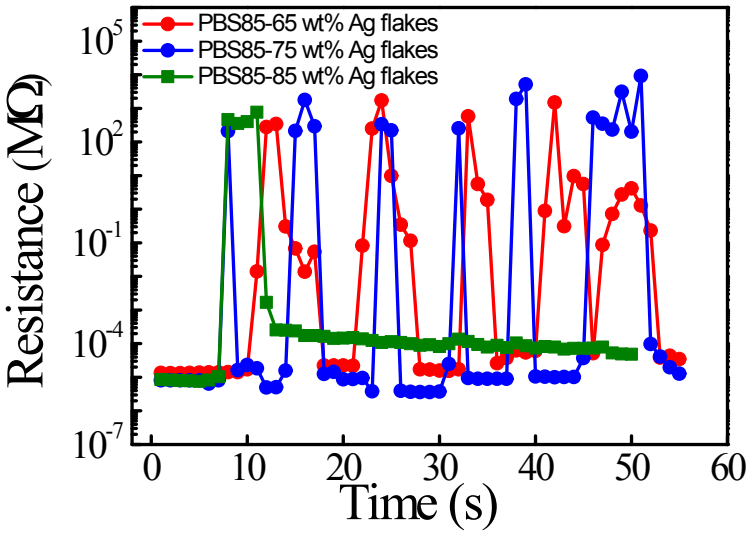

Figure S10. Plots of resistance of damage-resistant electrodes prepared by silver flakes/PBS85 with different filler amounts as a function of time while making several consecutive cuts.

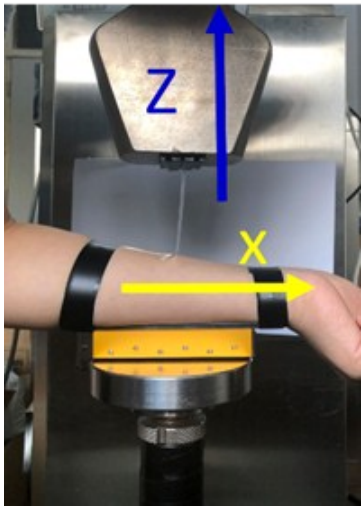

Figure S11. Photograph of adhesion measurement for PBS/PDMS DN elastomer.

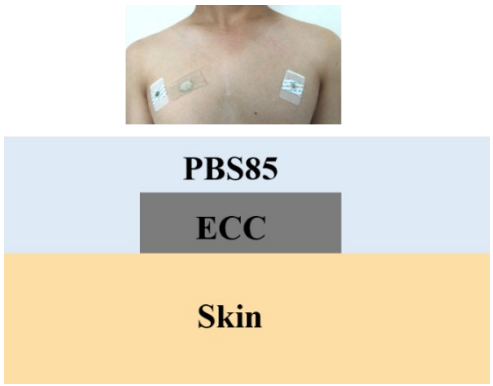

Figure S12. Digital image and corresponding schematic of self-healable ECG sensor.

Equation S1: The capacitance of parallel-plate capacitors with Ag flakes-PBS85 electrodes should have a linear variation with strain. ε_0 is the dielectric constant of vacuum, ε_r is the relative dielectric constant, λ is the strain, and A_0 is the initial overlapping electrode area and d_0 is the initial thickness of PBS85, the dielectric part of capacitors. $-0.5\ \lambda$ is the crosswise strain, so d under strain is $(1-0.5\ \lambda)\ d_0$, A is $(1+\lambda)\ (1-0.5\ \lambda)\ A_0$. Consequently, the capacitance should have linear variation with strain if the electrode keeps stable electrical performance under strain.

$$C = \varepsilon_0\varepsilon_r\frac{A}{d} = \varepsilon_0\varepsilon_r\frac{A_0(1+\lambda)(1-0.5\lambda)}{d_0(1-0.5\lambda)} = C_0(1+\lambda)$$

Table S1. Mechanical properties of self-healed PBS85 under room temperature and 60 °C. (Tensile speed:10 mm/min)

PBS85	Properties	Original	6 h	24 h	48 h	72 h	Recovery efficiency (%)
RT	Strength (kPa)	175±3	37±4.5	65±3	118±7	150±10.5	86
	Elongation at break (%)	314±29	243±13.5	251±31	269±1	305±29.5	
60 °C	Strength (kPa)	175±3	123±4	125±2.5	129±7	154±9	88
	Elongation at break (%)	314±29	286±9	285±49	293±39	305±8	

Table S2. Comparison of the performance of self-healing PBS/PDMS DN-silver flakes with reported self-healing conductive composites.

Year	Materials	Healing mechanism	Healing condition	Original performance (Given in conductivity or resisitance)	Performance after healing
2016	Sh-μAg-PU[2]	Hydrogen-bonding & reversible S-S bonds	25 °C, 40 min	20 Ω	400 Ω
2017	CNTs/PEI/CNC[3]	Ionic hydrogen bonding	160 °C, 10 min	$2.6x10^{-3}$ $S·m^{-1}$ 1	Not specified
2018	Polyaniline, polyacrylic acid, and phytic acid composite[4]	Hydrogen bonding and electrostatic interactions	Room temperature for 24 h	12 $S·m^{-1}$	≈99% efficiency
2018	PU, Ag flakes[5]	Dynamic metal-coordinated bonds and hydrogen bonds	25 °C for 48 h	4600 $S·m^{-1}$	98% efficiency
2018	PU-EDM/rmGO[6]	Diels-Alder bonds	NIR irradiation(5min, 1 W cm^{-2}), 2 min	2.23 Ω	3.24 Ω
2019	rGO and acrylate monomers[7]	Hydrogen bonding and ionic interactions	25 °C for 40 s	27.2 $S·m^{-1}$	Initial value
2019	PPy-coated hydrogels[8]	Host–guest interactions	Not specified	$(1.0 \pm 0.3) \times 10^{-1}$ $S·m^{-1}$	$(0.8 \pm 0.1) \times 10^{-1}$ $S·m^{-1}$
2019	PPy/PEG–5UPy composites[9]	Hydrogen bonding	Ambient condition, 5 min	0.88 $S·m^{-1}$	Not specified
2019	PAAMPSA/PANI/PA [10]	Hydrogen bonding and electrostatic interactions	Ambient conditions, 3 h	2 $S·m^{-1}$	98.6 ± 2.2%
2019	rGO/ P(NIPAM-co-MEA) [11]	Hydrogen bonding	Room temperature for 2 h	$1.26×10^{4}$ $S·m^{-1}$	100%
This work	**PBS/PDMS DN-silver flakes**	**Supramolecular interactions**	**Room temperature for 25 min**	**5×10^{4}** $S·m^{-1}$	**100%**

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