Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supplementary Information

Highly Efficient Self-Healable and Dual Responsive Hydrogel-Based Deformable Triboelectric Nanogenerators for Wearable Electronics

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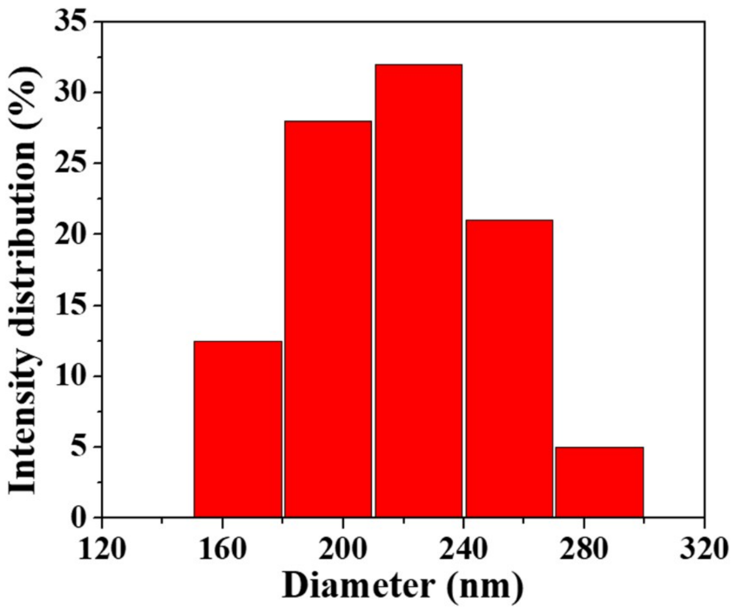

Figure S1. Diameter distribution of PDAP obtained from the oxidative polymerization of dopamine.

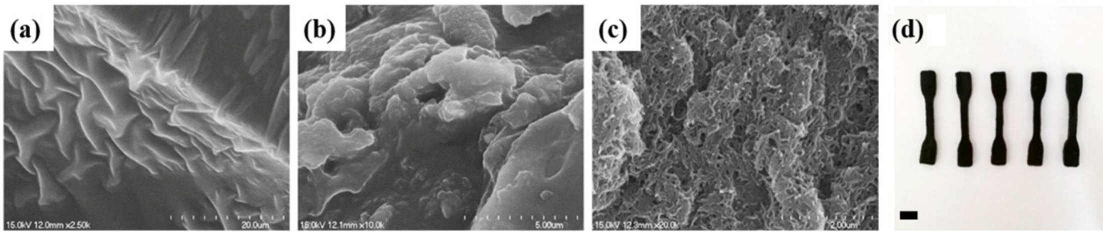

Figure S2. SEM images of cryogenically fractured hydrogel samples. (a) PVA, (b) PVA/PDAP, and (c) PVA/PDAP/MWCNT. (d) Digital image of PVA/PDAP/MWCNT hydrogel tensile test samples, (scale bar, 1 mm).

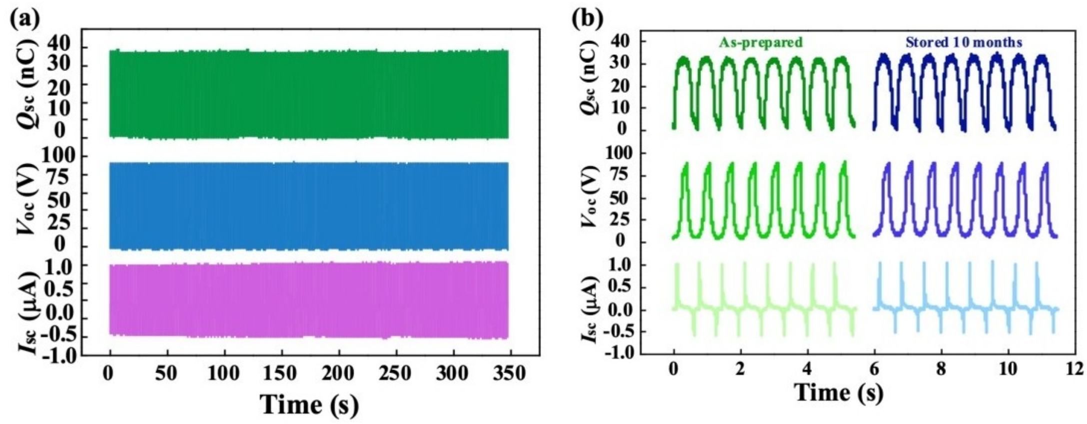

Figure S3. (a) The durability of the HS-TENG was tested for 500 cycles. (b) The stability of HS-TENG stored for 10 months was also evaluated.

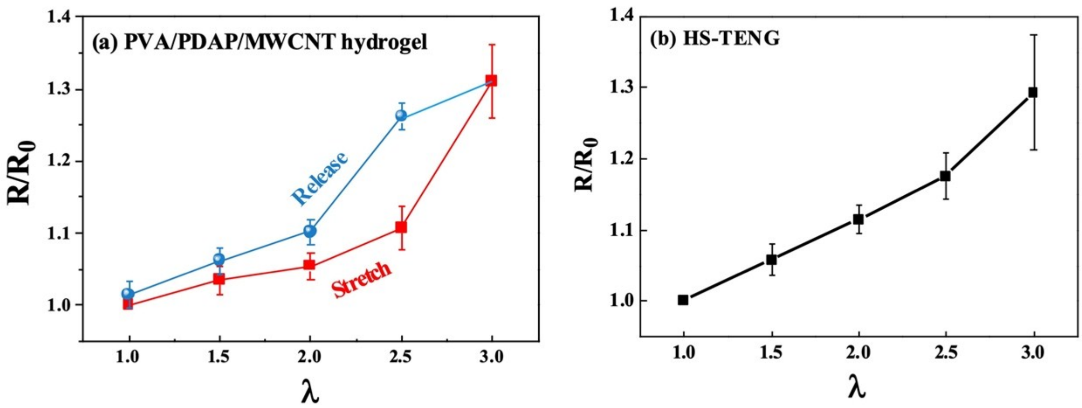

Figure S4. Resistance variation of PVA/PDAP/MWCNT hydrogel during stretch-release cycle (a) and HS-TENG (b) as a function of stretch ratio.

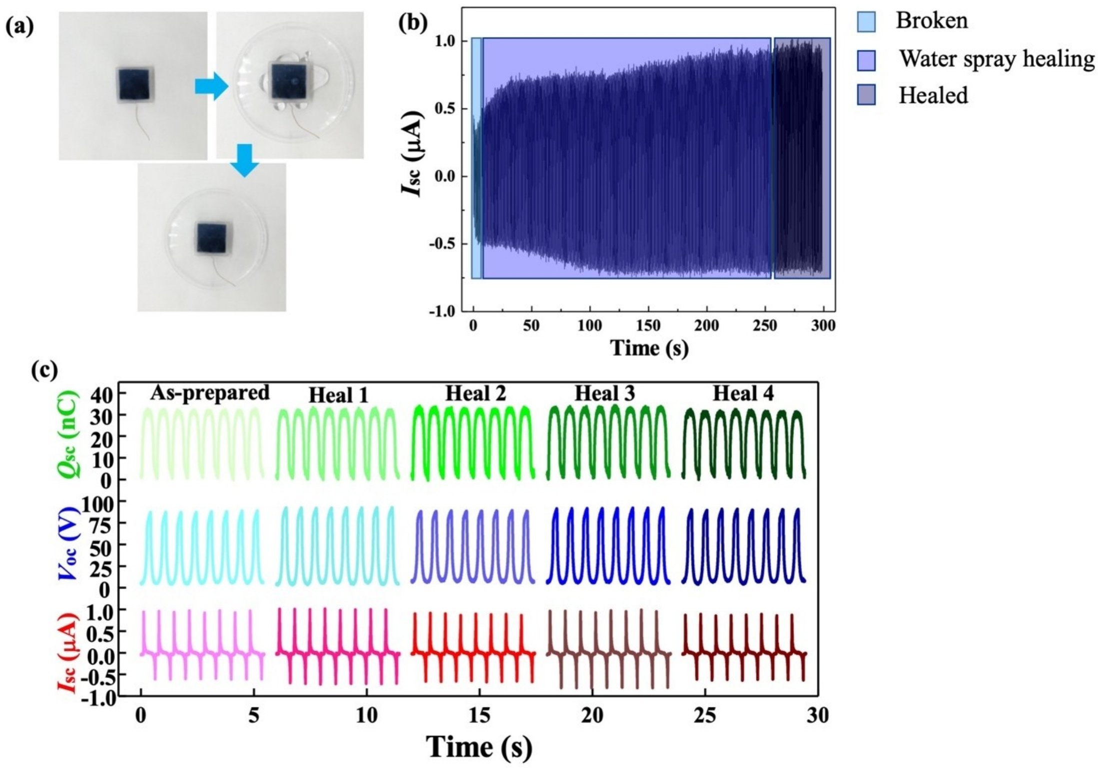

Figure S5. (a) Digital images of self-healing process induced by water for the PVA/PDAP/MWCNT HS-TENG after cutting by a razor blade. (b) The water triggered self-healing process of the output current of the PVA/PDAP/MWCNT HS-TENG was taken as a representative example. (c) The electrical output performance of the PVA/PDAP/MWCNT HS-TENG with four consecutive water triggered self-healing cycles.

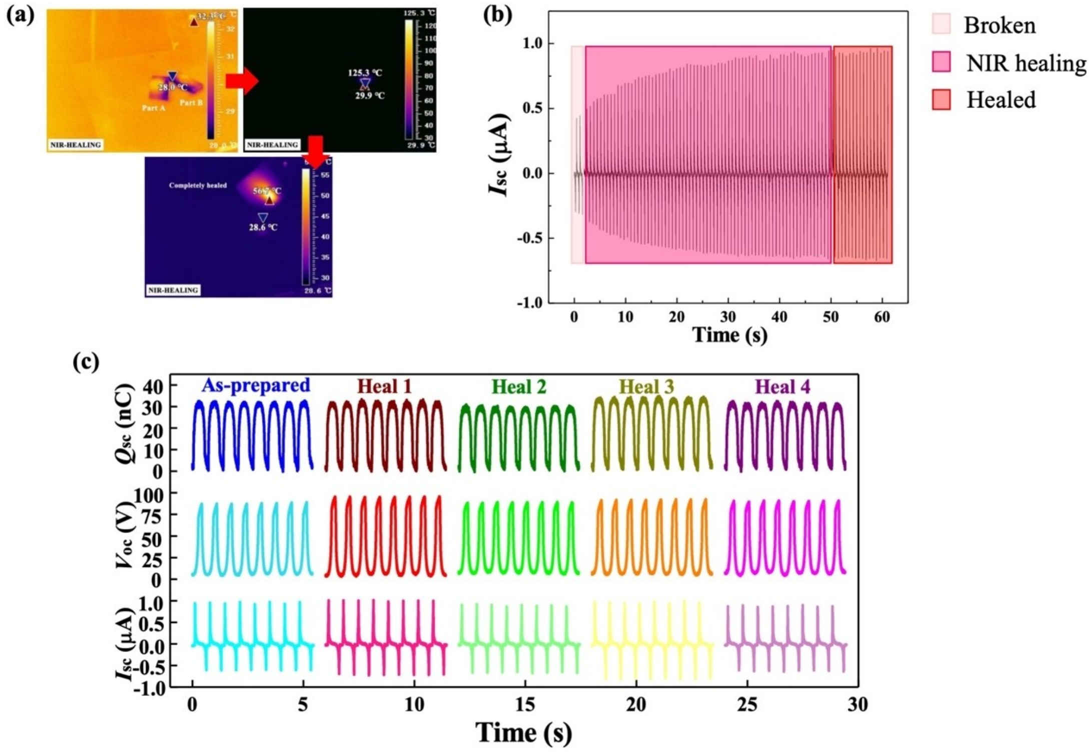

Figure S6. (a) Digital images of self-healing process induced by NIR for the PVA/PDAP/MWCNT HS-TENG after cutting by a razor blade. (b) The NIR triggered self-healing process of the output current of the PVA/PDAP/MWCNT HS-TENG was taken as a representative example. (c) The electrical output performance of the PVA/PDAP/MWCNT HS-TENG with four consecutive NIR triggered self-healing cycles.

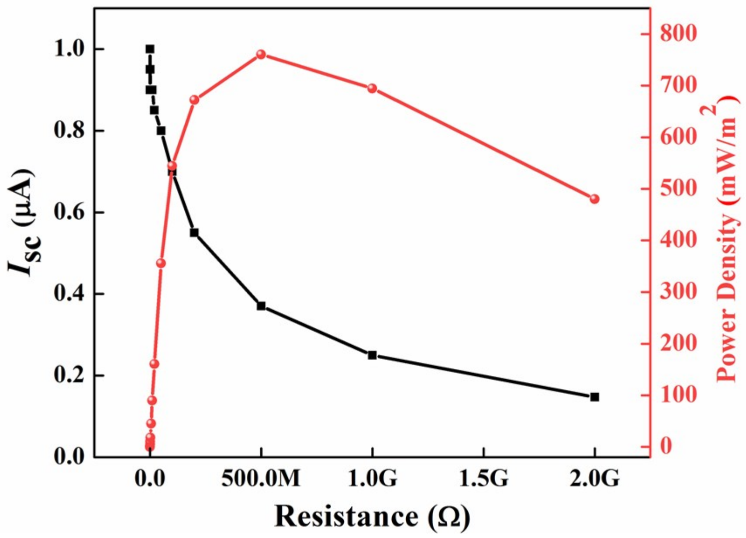

Figure S7. Output current density and output power density of HS-TENG with various external loads.

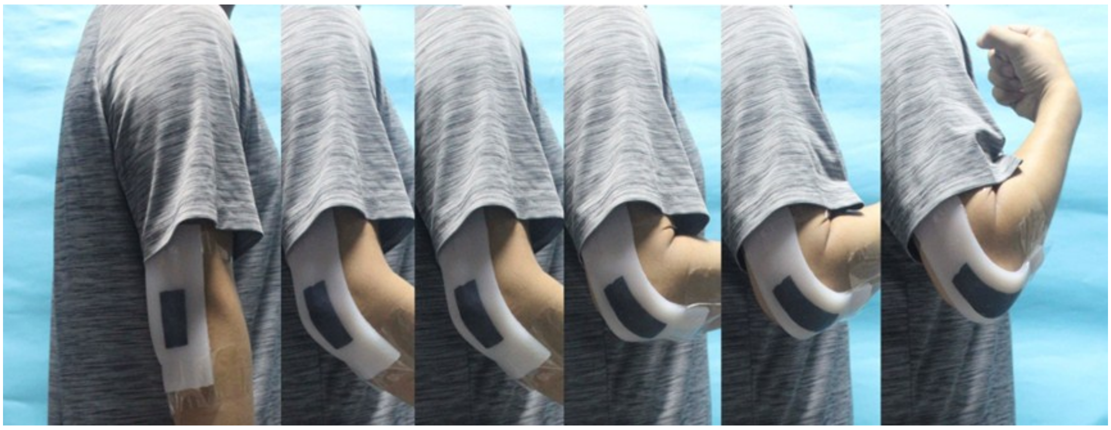

Figure S8. Photographs of stretching HS-TENG as a wearable device.

Table S1 Typical self-healing properties and electrical performance of self-healable triboelectric nanogenerators (TENGs)

Sample name	Self-healing component	Self-healing condition	Self-healing efficiency (%)[a]	Tensile strength (KPa)	Stretchability (%)	Electrical performance of TENG[b]			Ref
						Q_{sc}, Q'_{sc} (nC)	V_{oc}, V'_{oc} (V)	I_{sc}, I'_{sc} (μA)	
PDMS-PU TENG	Polydimethylsiloxane-polyurethane with disulfide bond	Thermal treatment at 65 °C for 2 h.	90	870	2745	-[c], -[c]	21.5, 21	2, 1.95	[1]
IS-TENG	Positive (Na^+) and negative ions ($B(OH)_4^-$)	Pressed with hand at 25 °C for 25 min.	81	3	700	17, -[c]	50, 50	6.5, -[c]	[2]
VTENG	Epoxy based vitrimer elastomer with dynamic disulfide bond	Thermal treatment at 65 °C for 4 h. or 95 °C for 0.5 h.	100	700	110	15, 14	55, 55	0.6, 0.5	[3]
H-PDMS/ Ag-PEDOT TENG	Dynamic covalent imine bonds	Curing at 21 °C for 12 h.	94	116	50	34, -[c]	100, 100	1, -[c]	[4]

Table S1 Continued

Sample name	Self-healing component	Self-healing condition	Self-healing efficiency (%)	Tensile strength (KPa)	Stretchability (%)	Electrical performance of TENG			Ref
						Q_{sc}, Q'_{sc} (nC)	V_{oc}, V'_{oc} (V)	I_{sc}, I'_{sc} (μA)	
SH/CNT TENG	Epoxy resin-based polysulfide elastomer	Upon exposure of NIR light for 1 min.	90	2600	3	16, 16	50, 50	0.6, 0.6	[5]
HS-TENG	1st route: photothermic - active polydopamine particle and multiwalled carbon nanotube 2nd route: water-active dynamic borate bond	**Upon exposure of NIR light for 1 min. or water spraying at 25 °C for 5 min.**	93	80	430	32, 32	95, 95	1, 1	**This work**

[a] Self-healing efficiency is characterized based on the tensile strength.

[b]Q_{sc}: short-circuit transferred charge, V_{oc}: open circuit voltage, I_{sc}: short-circuit current. Q'_{sc}, V'_{oc}, and I'_{sc} are the electrical performance of TENGs obtained after self-healing. Note: For the comparison of electrical outputs of different TENG devices, it is extremely important to have the same magnitude and frequency of force application. Moreover, the device dimensions and the mode of force application should also be same. For effective comparison, the relative value of TENGs before and after healing should be compared instead of their absolute value.

[c]N/A

Reference

[1] Xu W, Huang L-B, Hao J. Fully self-healing and shape-tailorable triboelectric nanogenerators based on healable polymer and magnetic-assisted electrode. *Nano Energy*. **2017**; *40*: 399-407.

[2] Parida K, Kumar V, Jiangxin W, Bhavanasi V, Bendi R, Lee PS. Highly transparent, stretchable, and self-healing ionic-skin triboelectric nanogenerators for energy harvesting and touch applications. *Advanced Materials*. **2017**; *29*, 1702181.

[3] Deng J, Kuang X, Liu R, Ding W, Wang AC, Lai YC, et al. Vitrimer elastomer-based jigsaw puzzle-like healable triboelectric nanogenerator for self-powered wearable electronics. *Advanced Materials*. **2018**; *30*: 1705918.

[4] Sun J, Pu X, Liu M, Yu A, Du C, Zhai J, et al. Self-healable, stretchable, transparent triboelectric nanogenerators as soft power sources. *ACS Nano*. **2018**; *12*: 6147-6155.

[5] Guan Q, Dai Y, Yang Y, Bi X, Wen Z, Pan Y. Near-infrared irradiation induced remote and efficient self-healable triboelectric nanogenerator for potential implantable electronics. *Nano Energy*. **2018**; *51*: 333-339.

Movie S1: Water triggered self-healing behavior of hydrogel sample. (AVI)

Movie S2: Self-healing behavior of hydrogel sample realized by NIR irradiation. (AVI)

Movie S3: Lighting 15 LEDs driven by tapping HS-TENG under various deformation. (AVI)

Movie S4: Wearable HS-TENG. (AVI)