

## Supporting Information

### Visual Pressure Interactive Display of Alternating Current

### Electroluminescent Devices Based on Hydrogel Microcapsules

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## Experimental details

### 1. Microcapsule Preparation:

Initially, poly (2,3-dihydrothieno-1,4-dioxin)-poly (styrene sulfonate) (PEDOT:PSS) dispersion (1 mL, 40 mg/mL, Hunan Nanoup Technology Co., Ltd.), silver nanowires (AgNWs) dispersion (4 mL, 10mg/mL, Chengdu Innoshines Technology Co., Ltd.), and single-wall carbon nanotubes (SWCNTs) dispersion (4 mL, 10 mg/mL, Suzhou Tanfeng Technology Co., Ltd.) were mixed in deionized water (20 mL) to give a homogenous suspension solution. The deionized water and the above suspension solution (PEDOT:PSS, AgNWs, SWCNTs) were taken out in equal amounts (2 mg/mL) into the beaker, and 1wt% sodium alginate (0.2 g, 1.05-1.15 Pa\*s, Guanfu, Tianjin) was added to obtain four core material solutions with the same viscosity during magnetic high-speed stirring (1000 rpm/min, VRERA, China) over 1 h. A mixture of urea (0.5 g, AR, XILONG) and formaldehyde solution (1 g, containing stabilizer methanol, 37%, TCI) was diluted with a certain amount of deionized water (2 g), and the pH of the solution was adjusted to 8.5 with ethanolamine (AR, SCR). After magnetic stirring in a water bath (70 °C, 300 rpm/min) for 30 min, the urea-formaldehyde prepolymer (PUF) was taken out and cooled to obtain transparent PUF. Then, the core material solution was dropped into 4wt% anhydrous CaCl<sub>2</sub> (99.9%, Aladdin) solution through electrostatic spray (10 kV, 50 mm/h) at a height of 20 mm. During the electrostatic spray, the electrostatic field was formed by the high-voltage DC power supply (Dongwen High-voltage Power Supply Tianjin Co., Ltd., 10 kV), and the injection pump (Changsha Nai Instrument Technology Co., Ltd., 50mm/h) pushed the syringe (Suzhou Jiaqicheng, 30G, 38 mm, 0.16). At this time, the ratio of calcium salt solution as a continuous phase to dispersed phase core material solution was 6:1wt, and cured (15 min) to obtain core material calcium alginate hydrogel. Glacial acetic acid (0.5 g, AR, Aladdin) was diluted with

deionized water (49.5 g) to obtain 1wt% glacial acetic acid solution, and add chitosan (CS) (1 g, < 200 mP\*s, Innochem) to obtain CS shell material solution. Then, in the newly solidified hydrogel microsphere system, 1.5 g CS shell material solution and 3.5 g PUF shell material solution were slowly dropped into the continuous phase in turn. The resulting solution was added a small amount of resorcinol (AR, Innochem) as a curing agent, adjusted pH to 4.5 with glacial acetic acid, magnetically stir in a water bath (60 °C, 200 rpm/min, 3 h), filter, wash and dry to obtain hydrogel microcapsules (HMC). It is worth mentioning that constant temperature heating magnetic stirring (DF-101S Shanghai Lichen Bangxi Instrument Technology Co., Ltd.) was used to synthesize prepolymer and microcapsule. The shell material is chitosan-urea-formaldehyde resin (CSUF) and the core material was hydrogel. HMCs of different core material types were also prepared according to this method. (In this paper, PEDOT-HMC is used as the abbreviation of HMC prepared with PEDOT:PSS as filler)

In this paper, PEDOT:PSS is compared with AgNWs and SWCNTs for three main reasons: (1) they meet the requirements of conductivity and light transmittance of devices at the same time. As shown in the table (Table S3), they have both bending and stretching mechanical properties, providing conditions for pressure detection; (2) PEDOT:PSS, AgNWs and SWCNTs are excellent representatives of polymer conductive materials, metal conductive materials and non-metal conductive materials respectively, and can judge the impact of three types of conductive materials on visual detection; (3) Another reason why PEDOT:PSS is selected for comparison with AgNWs and SWCNTs is that PEDOT:PSS adopts hole transmission conduction, while the latter two adopt electron transmission conduction, so the difference between them can be judged in detail.

## 2. VPI-ACEL Device Fabrication:

FTO-glass (25\*100 mm, 7 ohms/m<sup>2</sup>, Wuhan Jinge Solar Energy Technology Co., Ltd.) was ultrasonically cleaned with acetone (AR, BEIJING SHIJI), ethanol (95%, Chemical) and deionized water for 15 min, and treated with an ultraviolet lamp for 15 min. Then, the coplanar electrode was laser engraved (current 19.9 mA, speed 350 mm/s, OS3230, OUSAI LASER) on the cleaned FTO glass according to the pattern. PB glue (5 g, Shanghai Keyan Photoelectric Technology Co., Ltd.) was used as a dispersant to mix and disperse luminescent (ZnS:Cu) particles (5 g, 25 μm, D502CT) and dielectric particles (5 g, BaTiO<sub>3</sub>, 100 nm) evenly in the ratio of 1:1wt to obtain luminescent slurry (PL) and dielectric slurry (DL). With a screen plate (300 mesh, Suzhou Shifeng Screen Printing Equipment Co., Ltd.), finally, three layers of DL and two layers of PL were screen printed on the conductive glass with etched electrodes in turn by the screen printing. Each layer was dried on an electric heating plate (120 °C, 30 min) to obtain the coplanar electrode ACEL device. Subsequently, one layer of acrylic resin (AR, Jiangshan Weir) was screen printed on the coplanar electrode ACEL to protect the device. Two layers of polydimethylsiloxane (10 g) and crosslinking agent (1 g) mixture (DC84, SYLGARD) were screen printed on another glass or film. Next, place a customized screen-printing plate (as shown in Fig.S5) above the PDMS to ensure that each pore diameter is greater than most microcapsules. The microcapsules were gently transferred to the surface of PDMS through the pore diameter (1 mm), and then the PDMS was cured at sealed room temperature for 10~20 min. Finally, the two parts were bonded face-to-face, and the VPI-ACEL was obtained by packaging and curing with PDMS at the surrounding edges.

### 3. Characterization:

Freeze-dryer instrument (FD1A50- Beijing Boyikang Experimental Instrument Co., Ltd.) tested the SEM morphology of aqueous microcapsules after freeze-drying. Scanning electron

microscope (SU8020, HITACHI, Japan) and optical microscope (DM2700M, LEICA, Germany) were used to observe the morphology of microspheres and the cross-sectional morphology of light-emitting devices. The functional groups of composited microcapsules, core materials and shell materials were measured by Fourier transform infrared spectrometer (NICOLETIS10, Thermo Fisher Scientific Shier Technology Company). The particle size distribution of microcapsules was analyzed by a Laser particle size analyzer (Mastersizer 2000, malvin, UK). The corresponding thermal properties of microcapsules were analyzed by differential scanning calorimeter (DSC214, NETZSCH, Germany) and thermogravimetric analyzer (TG209F3, NETZSCH, Germany). The compressive properties of microcapsules were tested by a compressive strength machine (C5000, Xinsansi compressive strength machine). The luminous brightness, luminous wavelength and color coordinates were recorded using a scanning spectrometer (PR655, Photo Research, USA). AC programmable power supply (IT7622, ITECH electronics, co., ltd.) was used to provide voltage and frequency. The electrical impedance was tested by an electrochemical workstation (AUT86832, Mrtrohm China Ltd).

**Table S1.** The particle size distribution of microcapsules with different core and shell.

No.	Core <sup>a)</sup>	Shell <sup>b)</sup>	D10 [ $\mu\text{m}$ ] <sup>c)</sup>	D50 [ $\mu\text{m}$ ] <sup>d)</sup>	D90 [ $\mu\text{m}$ ] <sup>e)</sup>	MZ [ $\mu\text{m}$ ] <sup>f)</sup>	PDI <sup>g)</sup>	Refs.
1	PEDOT:PSS	MF	9.070	18.166	32.576	19.509	1.294	[1]
2	Graphene	MF	20.793	36.980	60.625	38.672	1.077	[1]
3	Fe <sub>3</sub> O <sub>4</sub>	MF	8.347	15.423	25.931	16.201	1.140	[1]
4	H <sub>2</sub> O	MF	4.974	9.385	15.874	9.895	1.161	[1]
5	H <sub>2</sub> O	CSUF	578.055	801.652	1139.778	807.778	0.70	This work
6	PEDOT:PSS	CSUF	586.274	770.442	1208.167	798.442	0.81	This work
7	AgNWs	CSUF	574.421	776.456	1090.881	786.164	0.66	This work
8	SWCNTs	CSUF	587.38	815.877	1149.114	825.877	0.69	This work

a) Core: core material of microcapsules (contain sodium alginate hydrogel); b) Shell: shell materials of microcapsules; c) D10: the cumulative distribution of particles is 10% of the particle size; d) D50: the cumulative distribution of particles is 50% of the particle size; e) D90: the cumulative distribution of particles is 90% of the particle size; f) MZ: the average particle size; g) PDI: polymer dispersity index.

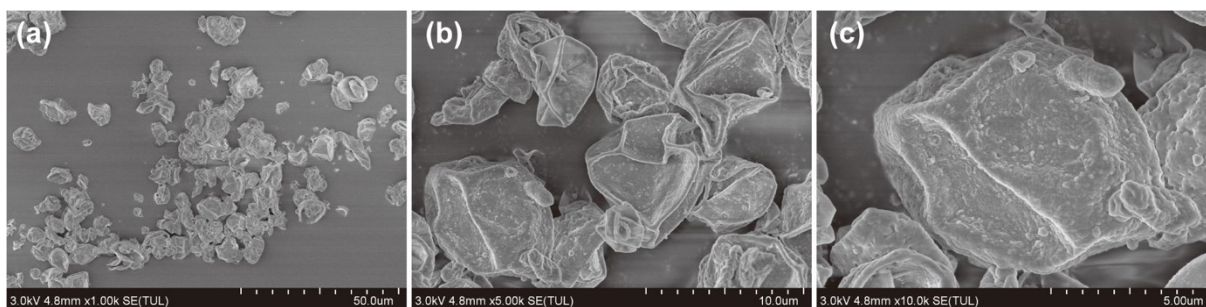
**Table S2.** Particle size of hydrogel microcapsules prepared by different types of sharp holes.

No.	Core <sup>a)</sup>	Model	ID [mm] <sup>b)</sup>	OD [mm] <sup>c)</sup>	MZ [ $\mu$ m] <sup>d)</sup>	PDI <sup>e)</sup>
1	H <sub>2</sub> O	23G	0.34	0.64	807.778	0.70
2	H <sub>2</sub> O	25G	0.26	0.52	561.433	0.89
3	H <sub>2</sub> O	27G	0.21	0.42	258.12	0.77
4	H <sub>2</sub> O	30G	0.16	0.31	N/A	N/A
5	H <sub>2</sub> O	32G	0.11	0.23	N/A	N/A
6	H <sub>2</sub> O	34G	0.06	0.23	N/A	N/A

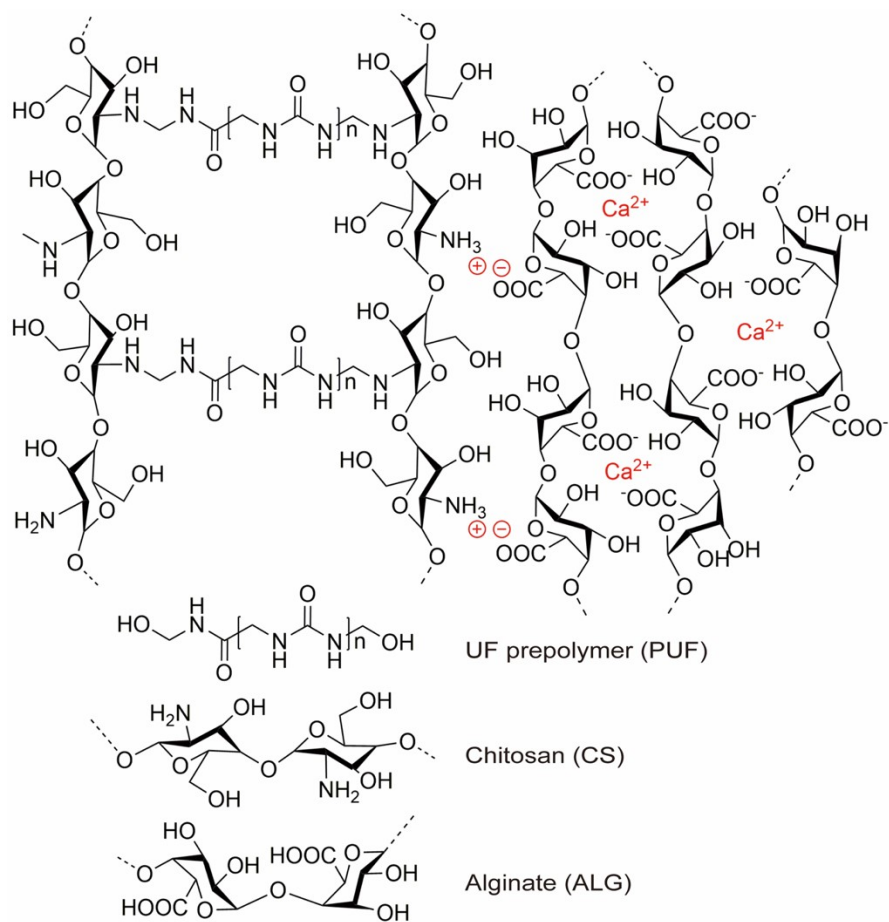
a) Core: core material of microcapsules (contain alginate hydrogel); b) ID: Inside diameter of sharp hole; c) OD: Outside diameter of sharp hole; d) MZ: the average particle size; e) PDI: polymer dispersity index.

**Table S3.** Properties of different transparent conductive materials.

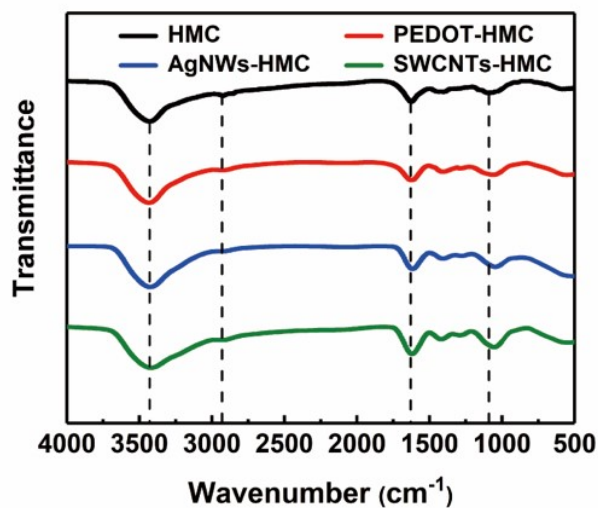
Material	Sheet resistance	Transparency	Flexibility	Cost	References
ITO/FTO	10 $\Omega$ /sq	>80% in 400-700 nm	Bending	High	[2]
PEDOT-PSS	10 $\Omega$ /sq	>80%	Bending and Stretching	Medium	[3]
Ag NWs	10 $\Omega$ /sq	60~80%	Bending and Stretching	Medium	[2]
SWCNTs	5.8 k $\Omega$ /sq	~75% at 600 nm	Bending and Stretching	Medium	[4]
Graphene	230 k $\Omega$ /sq (single layer)	>80% in visible region	Bending	Low	[5]
Ionic conductor	N/A	~100% in 350-850 nm	Bending and Stretching	N/A	[6]



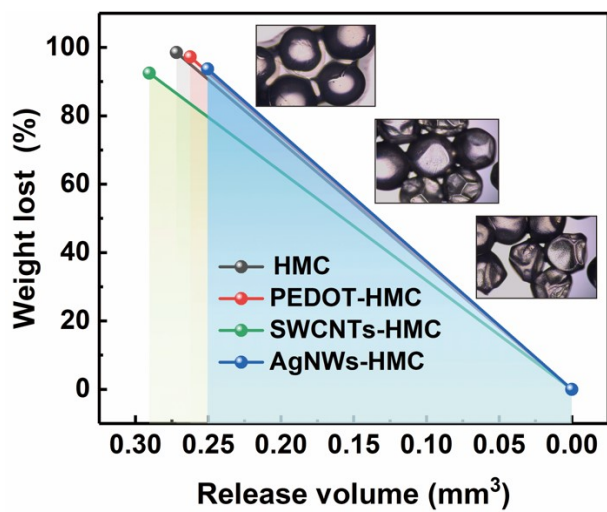
**Figure S1.** a) SEM morphology characterization of multiple water coated microcapsules; b) SEM morphology characterization of a small amount of water coated microcapsules; c) SEM morphology characterization of water coated microcapsule shell.



**Figure S2.** Chemical reaction mechanism of hydrogel microcapsules (HMC) in the process of synthesis.

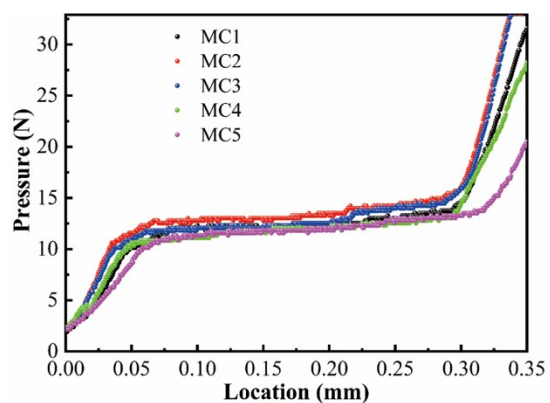


**Figure S3.** Fourier infrared analysis of the structural components of four microcapsules (HMC, PEDOT-HMC, AgNWs-HMC and SWCNTs-HMC).

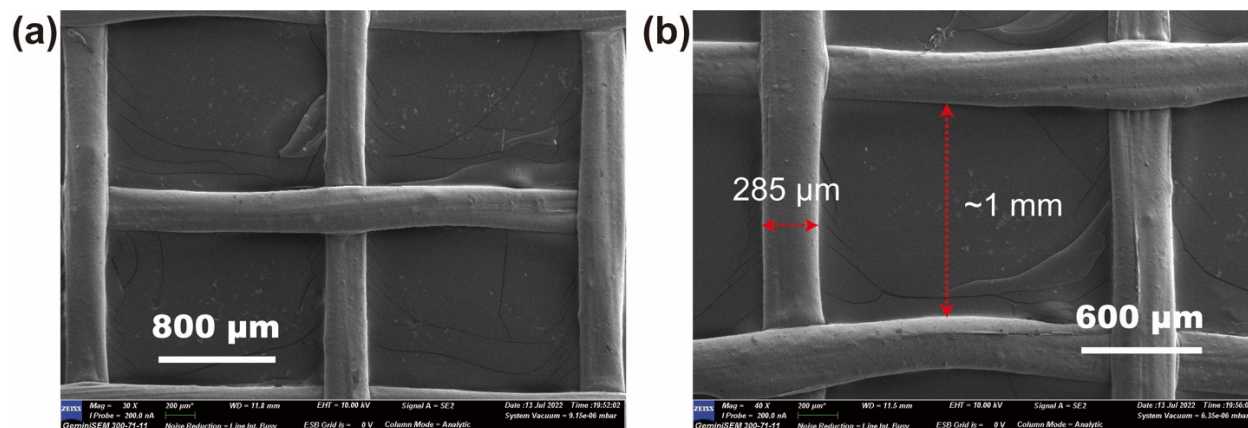


**Figure S4.** Volume and weight loss percentage curve.

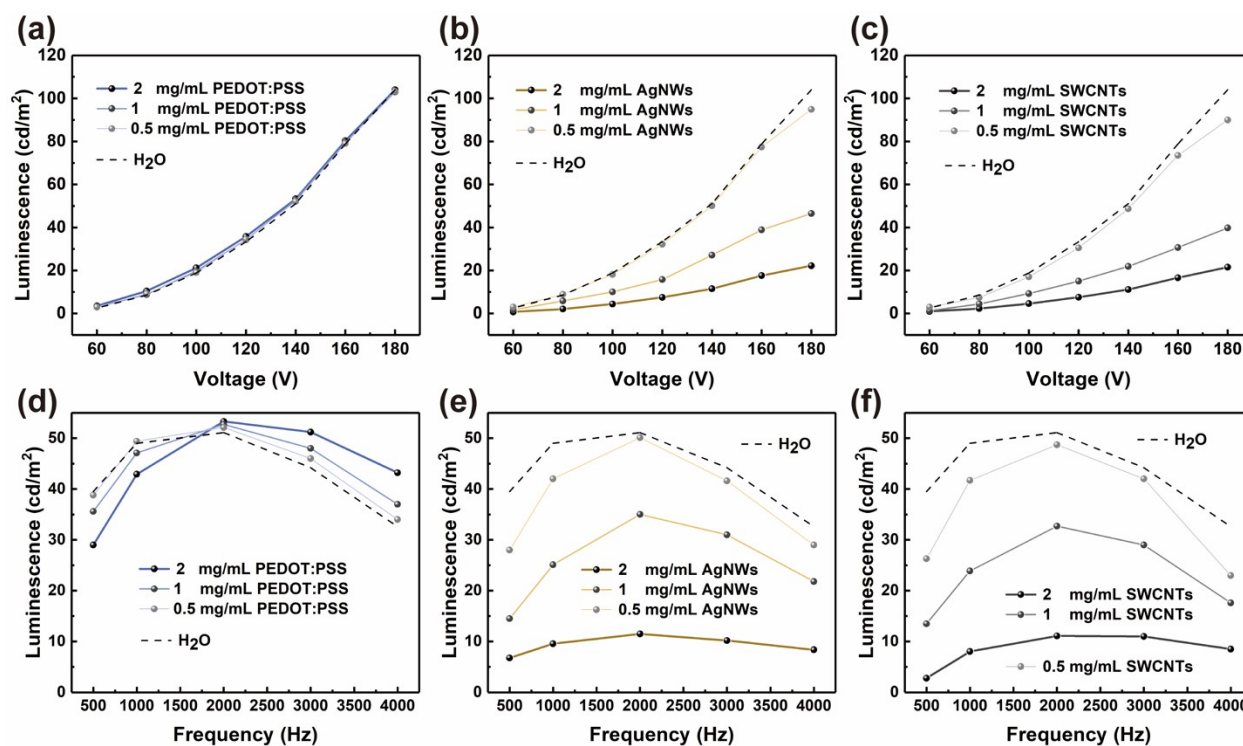




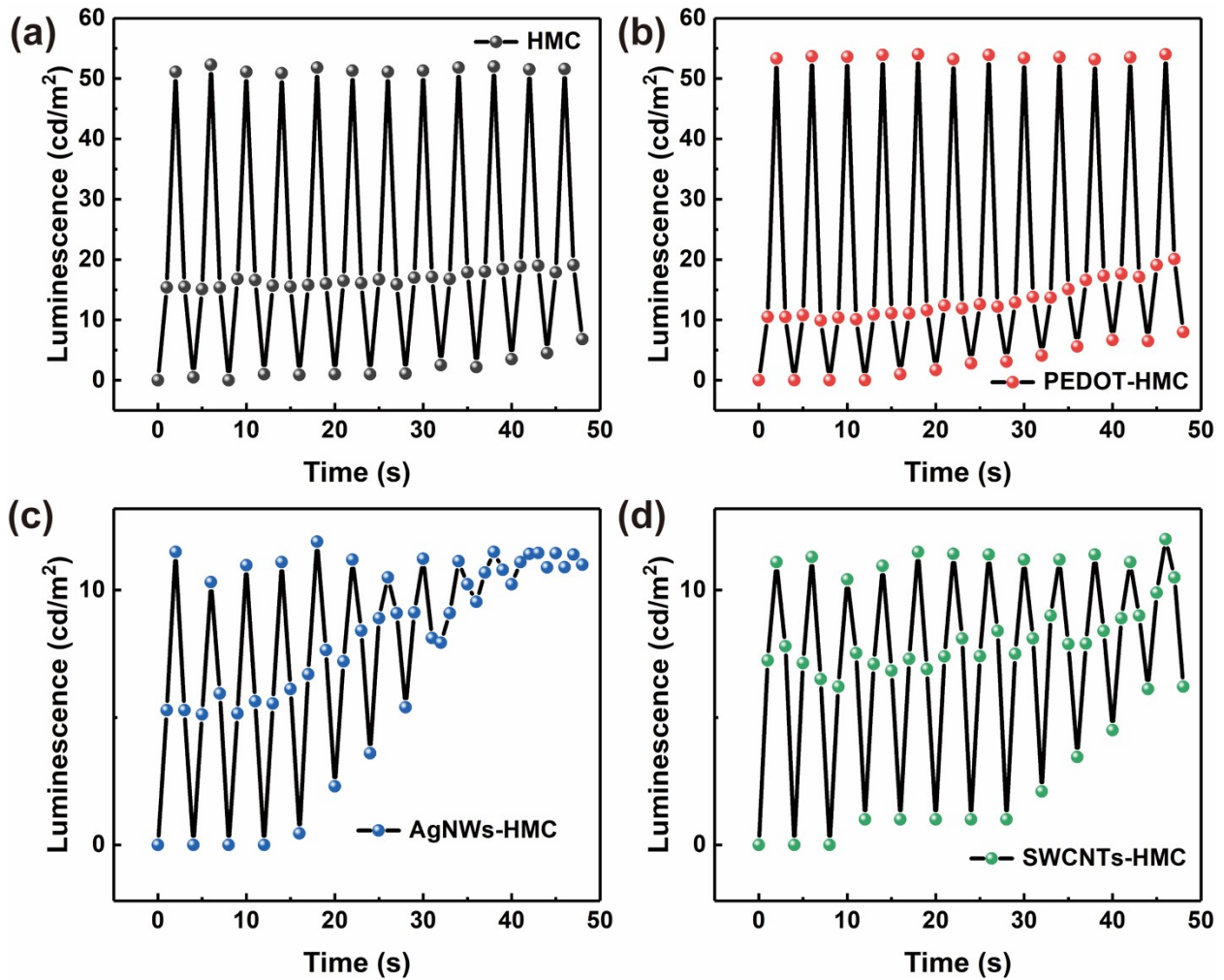
**Figure S5.** Compressive strength analysis of five kinds of self-healing microcapsules with different liquid core materials (MC1 is PEDOT:PSS; MC2 is graphene; MC3 is  $\text{Fe}_3\text{O}_4$ ; MC5 is deionized water; MC6 is tap water) and melamine resin wall materials prepared in the previous work.



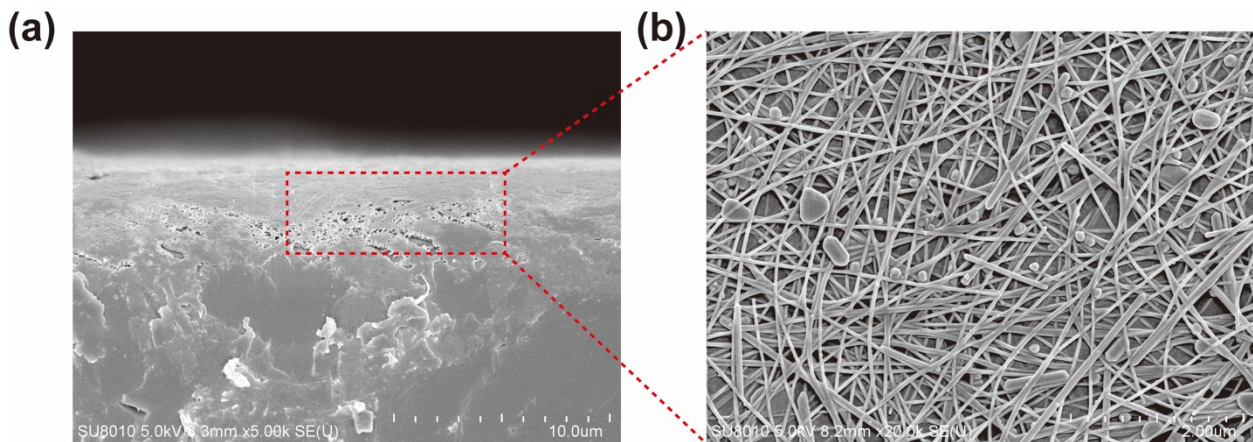
**Figure S6.** (a) and (b) is SEM of wire diameter and aperture of customized screen-printing plate.



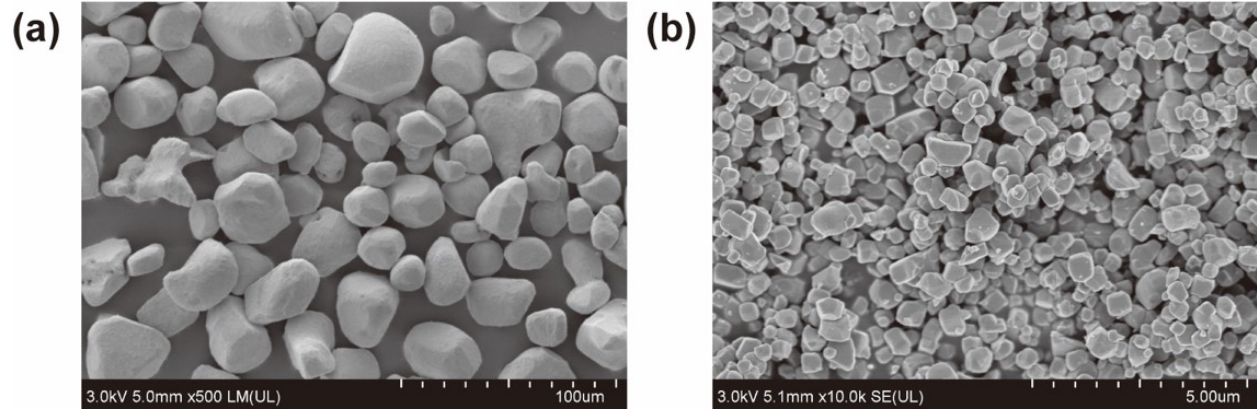
**Figure S7.** Brightness of PEDOT:PSS filling ratio of 2 mg/mL, 1 mg/mL, 0.5 mg/mL under different voltage (a) and frequency (d); Brightness of AgNWs filling ratio of 2 mg/mL, 1 mg/mL, 0.5 mg/mL under different voltage (b) and frequency (e); Brightness of SWCNTs filling ratio of 2 mg/mL, 1 mg/mL, 0.5 mg/mL under different voltage (c) and frequency (f).



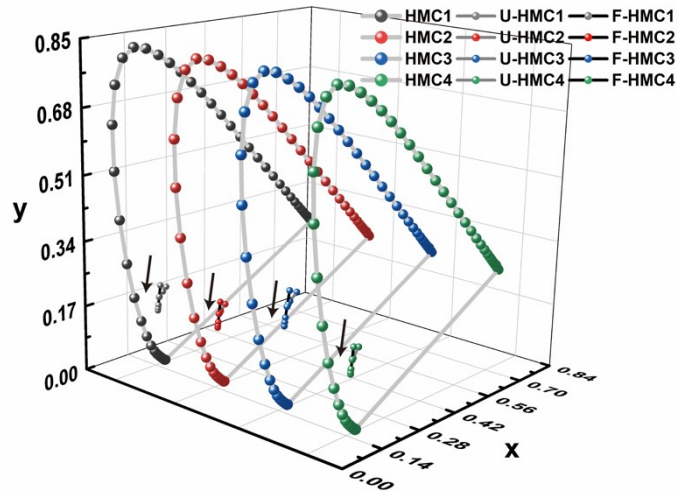
**Figure S8.** Four VPI-ACELs have 12 cycle stability test curves (4 s/time) between 0~0.35 mm



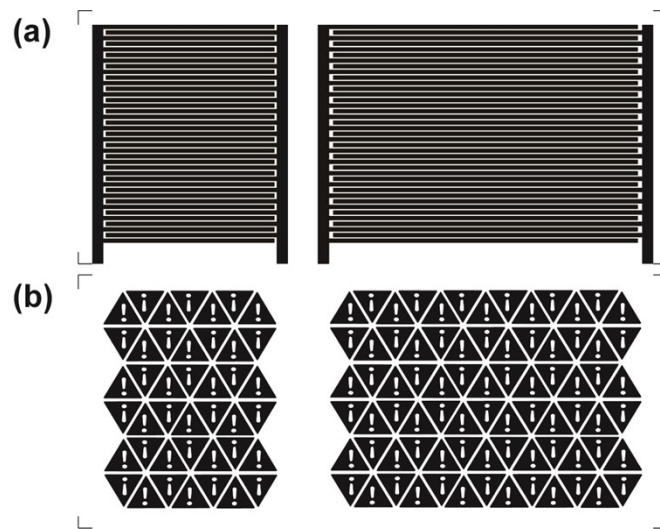
**Figure S9.** a) and b) is SEM morphology characterization of AgNWs.



**Figure S10.** a) SEM morphology characterization of luminescent powder (ZnS:Cu); b) SEM morphology characterization of dielectric powder (BaTiO<sub>3</sub>).



**Figure S11.** CIE1931 color coordinates of four different VPI-ACEL devices (HMC1 is HMC, HMC2 is PEDOT-HMC, HMC3 is AgNWs-HMC and HMC4 is SWCNTs-HMC) at different voltages (U) and different frequencies (F).



**Figure S12.** Design drawing of VPI-ACEL devices combined with packaging box: a) electrode layer; b) luminescent layer.

**Movie S1.** Practical application 1: Video of the pressing process of two icons (HMC)VPI-ACEL devices (50\*50 mm) at 140 V and 1000 Hz.

**Movie S2.** Practical application 2: (HMC)VPI-ACEL devices is applied to actual packaging boxes, and free-fall impact experiments are carried out with steel balls (175 g, 35 mm) at the heights of 5 cm and 20 cm on 80\*90 mm and 130\*90 mm packaging paper respectively at 140 V and 1000 Hz.

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