

## Supporting Information

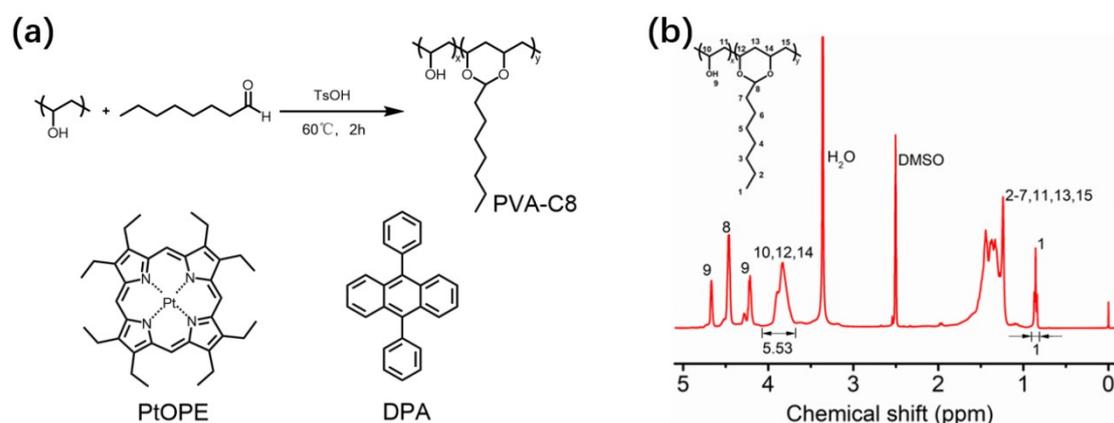
### Highly stable and air-resistant photonic upconversion organogels with self-healing and temperature responsiveness

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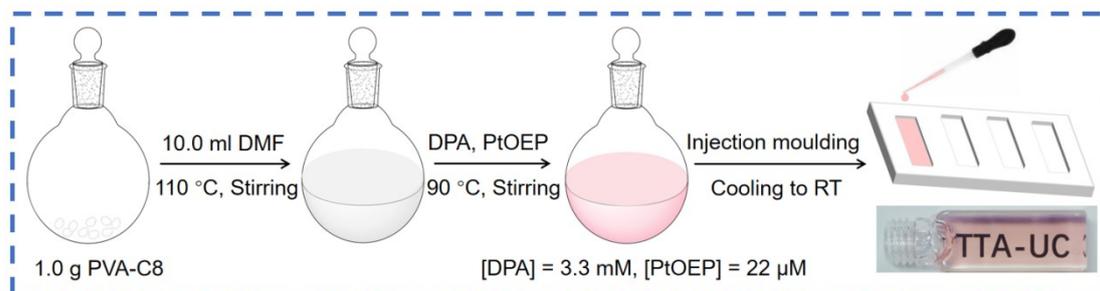
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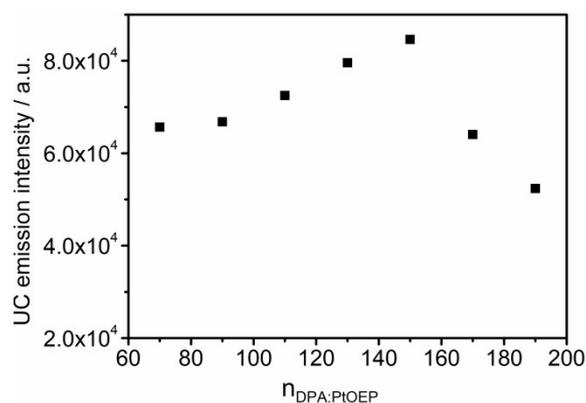
**Fig. S1** (a) The synthesis method of PVA-C8, the upconverter used in the whole study. PtOEP is a photosensitizer, and DPA is an annihilator. (B) The NMR spectrum of PVA-C8 organogel.

According to the peak area of the <sup>1</sup>H NMR spectrum, the molar ratio of the octyl group to the initial hydroxyl group was calculated to determine the degree of reaction. No. 10, 12, and 14 H can represent the amount of -OH in all the initial PVA, and its integral area is set as S<sub>2</sub>, and then according to No. 1 H being the same proton composition of the three chemical environments in

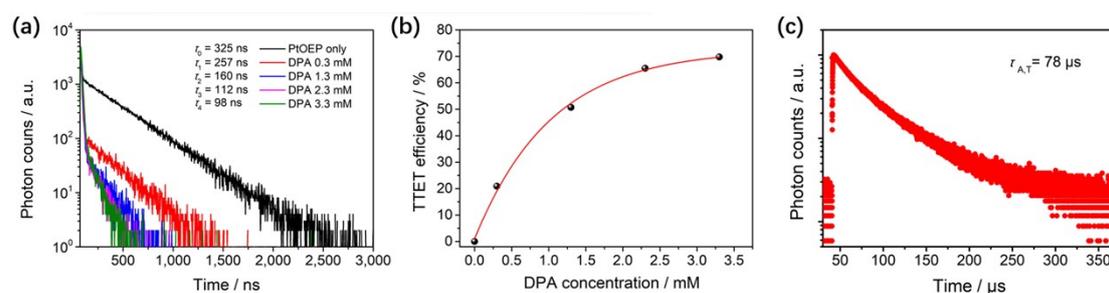
the methyl group, its integral area is set as  $S_1$ , and each side chain group should replace two -OH on the main chain. Therefore, according to the equation  $(S_1 / 3) * 2 / S_2 = \text{degree of substitution}$ . The optimal degree of substitution is about  $12 \pm 1\%$  after pre-condition optimization and screening.



**Fig. S2** Upconversion organogel preparation method and the amount of medicine in each step. The PVA-C8-PtOE-DPA organogel was injected into teflon molds and cuvettes for further characterization, and it can be seen that it has high transparency.

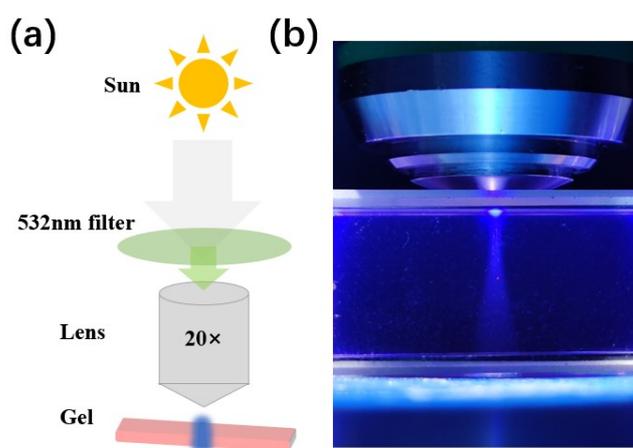


**Fig. S3** In the PVA-C8 organogel, the influence of the molar ratio of PtOEP and DPA on the upconversion intensity, the optimal ratio is 1 : 150, that is, the chromophore concentration is  $[\text{DPA}] = 3.3 \text{ mM}$  and  $[\text{PtOEP}] = 22 \text{ }\mu\text{M}$ .

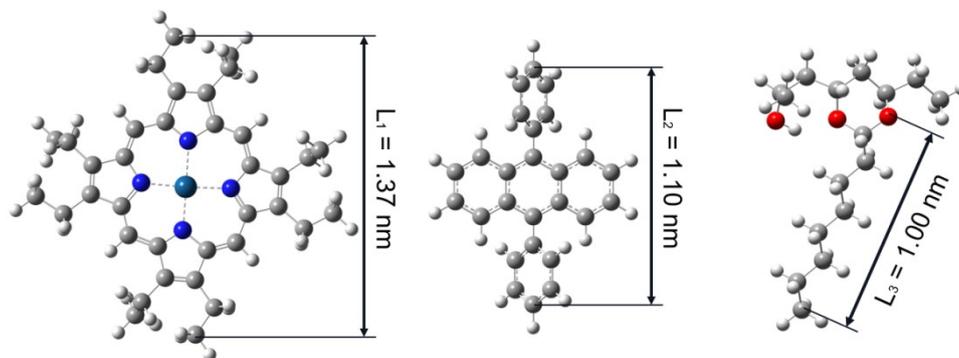


**Fig. S4** Quenching of PtOEP phosphorescence ( $\lambda_{em} = 635 \text{ nm}$ ) (a) The luminescence lifetime of the photosensitizer in the organogels with and without triplet acceptor, under excitation at 532 nm ( $[\text{PtOEP}] = 2.2 \text{ }\mu\text{M}$ ,  $[\text{DPA}] = 0\sim 3.3 \text{ mM}$ ), (b) TTET efficiency of the PtOEP /DPA pair in organogel, which was calculated according to equation 1, (c) the upconversion luminescence lifetime at 434 nm of PVA-C8-DPA-PtOEP organogel. All measurement were conducted under air conditions.

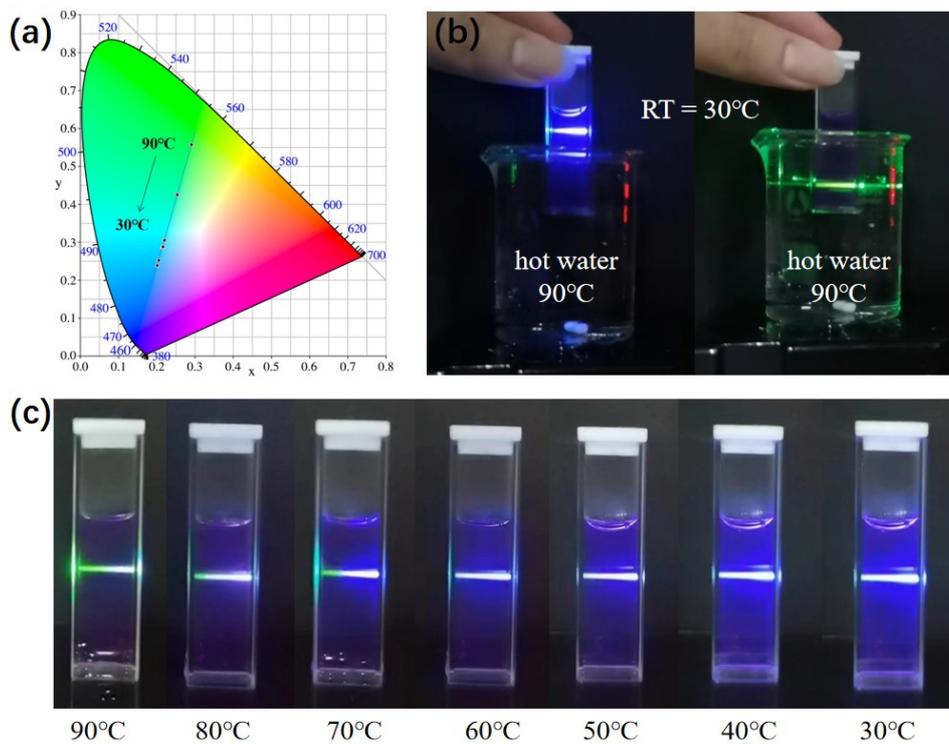
$$\Phi_{TTET} = 1 - \frac{\tau}{\tau_0} \quad \text{equation 1}$$



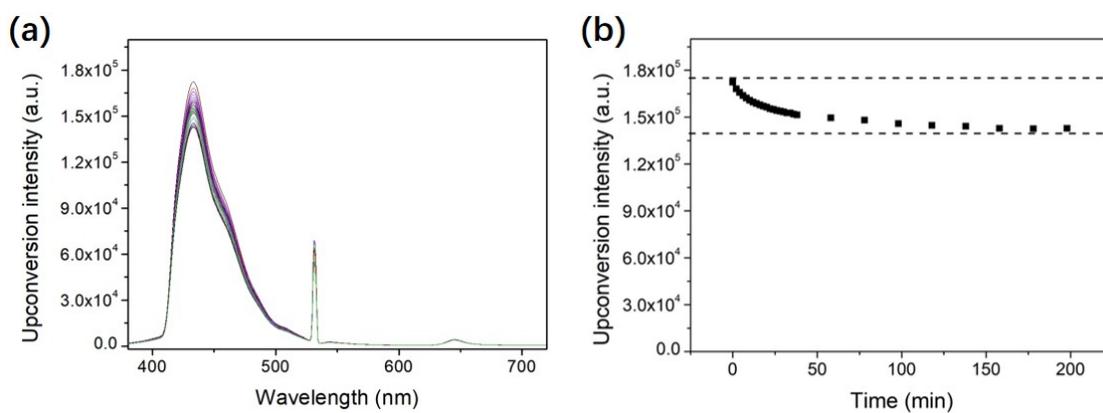
**Fig. S5** Through filtering and focusing the sunlight, the upconversion emission is realized in the air state, (a) the schematic diagram of the device and operation, (b) the PVA-C8-DPA-PtOEP organogel in the cuvette is focused by the lens, upconversion is realized under sunlight, and the sunlight intensity is  $100 \text{ mW/cm}^2$ .



**Fig. S6** the optimized structure of PtOEP, DPA and PVA-C8 model molecule (Software: G09; Solvation: DMF). According to the optimized structure, the longest distance between PtOEP molecules is  $L_1 = 1.37$  nm, the longest distance between DPA molecules is  $L_2 = 1.10$  nm, and the average length of attached octyl side chains is  $L_3 = 1.00$  nm.

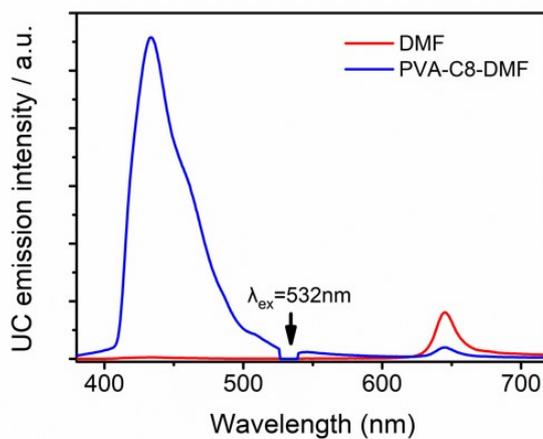


**Fig. S7** (a) The CIE chromaticity diagram of PVA-C8-DPA-PtOEP organogel, light color change from 90-30 °C. (b) At 30 °C and 90 °C, the upconversion organogels showing blue and green light, respectively. (c) Shows the change of the luminescence color of the gel from green to blue with the temperature change at 90-30 °C.

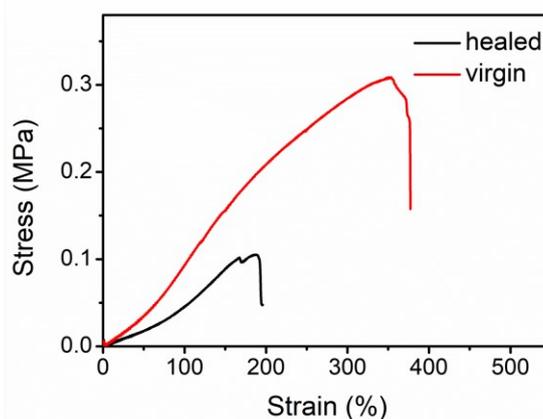


**Fig. S8** (a) Under the condition of continuous 198 min excitation intensity of  $500 \text{ mW} \cdot \text{cm}^{-2}$  with a 532nm laser in the air, the up-conversion emission spectrum of PVA-C8-PtOEP-DPA (b) at

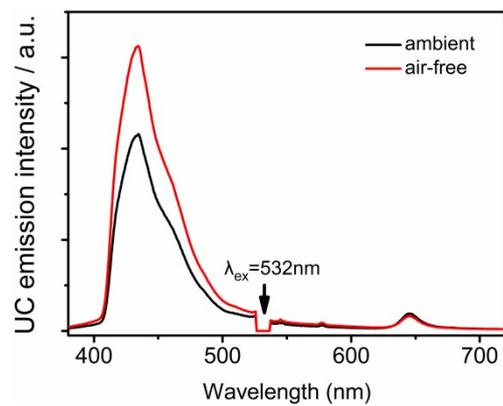
434nm, the corresponding Up-conversion emission intensity (stability) over time.



**Fig. S9** The emission spectra of PVA-C8-DPA-PtOEP organogel (blue line) and DMF-DPA-PtOEP solution (red line) under the same excitation intensity of 532 nm in air ([PtOEP] = 22  $\mu\text{M}$ , [DPA] = 3.3 mM).



**Fig. S10** The stretch curve (red line) of PVA-C8-DPA-PtOEP organogel and the stretch curve (black line) of the gel that was partially heated at 90 °C to assist healing after shearing.



**Fig. S11** The photoluminescence spectra of PVA-C8-DPA-PtOEP organogel under deoxygenation (red line) and air (black line) under 532nm laser excitation.