

Experimental section

Preparation of *m*-TiO₂/LPP photoanodes. TiO₂ colloid was synthesized according to the detailed procedures reported previously.^{S1} Colloidal TiO₂ films was fabricated by coating TiO₂ colloid onto freshly cleaned FTO glass substrates (12 Ω square⁻¹) with size of 2.5 × 2.5 cm² by a doctor-blade method. Subsequently, the FTO glass supported colloidal TiO₂ films were calcined in a muffle furnace at 450 °C for 30 min in air. The resultant *m*-TiO₂ electrodes with TiO₂ thickness of around 10 μm and active area of 0.5 × 0.5 cm² were immersed in 0.25 mM N719 ethanol solution for 24 h to obtain dye sensitized *m*-TiO₂ photoanodes. Finally, ultrafine green-emitting LPP phosphors with maximal photofluorescence decay time of 6 hours (purchased from Shenzhen HuiDuoSheng Luminous Material Co., Ltd.) were dispersed in anhydrous acetonitrile for a 0.25 g mL⁻¹ solution, which was spin-coated onto dye-sensitized *m*-TiO₂ film at a rotation speed of 2000 rpm for 20 s to obtain *m*-TiO₂/LPP photoanodes. The model number for green-emitting LPP phosphors was H-1B in the selling company, and Al, S, Y, C, Eu, S, O elements were determined for green-emitting LPPs. The average size of LPP particles was in a range of 5 ~ 10 μm.

Synthesis of PGE. The graphene tailored PAA/PEG polymer gel electrolyte (PGE) was synthesized according to the detailed procedures. Briefly, 8 g of acrylic acid, 4.4 g of PEG and 0.372 g of ultrafine graphene powders were mixed in 15 mL of deionized water. When heated to 80 °C under vigorous agitation, 8 mg of N,N'-(methylene)bisacrylamide and 0.225 g of ammonium persulfate were added for hydrous graphene tailored PAA/PEG matrix. After being dried at 60 °C, the dense matrix was immersed into deionized water for 3 days and subsequently freeze-dried under vacuum at -60 °C for 72 h. Later, the microporous graphene tailored PAA/PEG were immersed in a homogeneous mixture of graphene and redox electrolyte with graphene dosage of 1.33 g L⁻¹ to reach swelling equilibrium. The redox electrolyte consisted of 100 mM of tetraethylammonium

iodide, 100 mM of tetramethylammonium iodide, 100 mM of tetrabutylammonium iodide, 100 mM of NaI, 100 mM of KI, 100 mM of LiI, 50 mM of I₂, and 500 mM of 4-tert-butyl-pyridine in 50 ml of acetonitrile. The corresponding electrolyte included optimized compositions, which had been carefully discussed in previous literature.^{S2}

Solar cell assembly. The pyrolyzed platinum electrode on FTO glass was used as a counter electrode. Every quasi-solid-state DSSC device was assembled by sandwiching a slice of PGE with a thickness of around 500 μm between a N719 dye-sensitized TiO₂ photoanode and a Pt counter electrode.

Photovoltaic measurements. The photo photocurrent density-voltage (*J-V*) curves of solar cells with an active area of 0.25 cm² were recorded on a CHI660E electrochemical workstation under irradiation of simulated solar light from a 100 W Xenon arc lamp in ambient atmosphere. The incident light intensity was controlled to 100 mW cm⁻² (calibrated by a standard silicon solar cell). A black mask was applied on the surface of cell device to avoid stray light. Each *J-V* curve was repeated measured at least 20 times using different samples to eliminate experimental errors. Before measurement for dark *J-V* curves, the solar cells were illuminated by a simulated solar light for 1 min. Immediately, the devices were covered in a completely dark condition and the dark *J-V* curves were recorded on a CHI660E electrochemical workstation in air. The fluorescent light from *m*-TiO₂/LPP film provided solar energy to excite N719 dye, therefore the intensities of FTO glass supported *m*-TiO₂/LPP films recorded by a standard silicon solar cell were used as the light intensities for dark efficiencies. The dark cell efficiency was obtained according to $\eta_{\text{dark}} = P_{\text{max}}/P_{\text{in}}$, where P_{in} referred to fluorescent light intensity of *m*-TiO₂/LPP film.

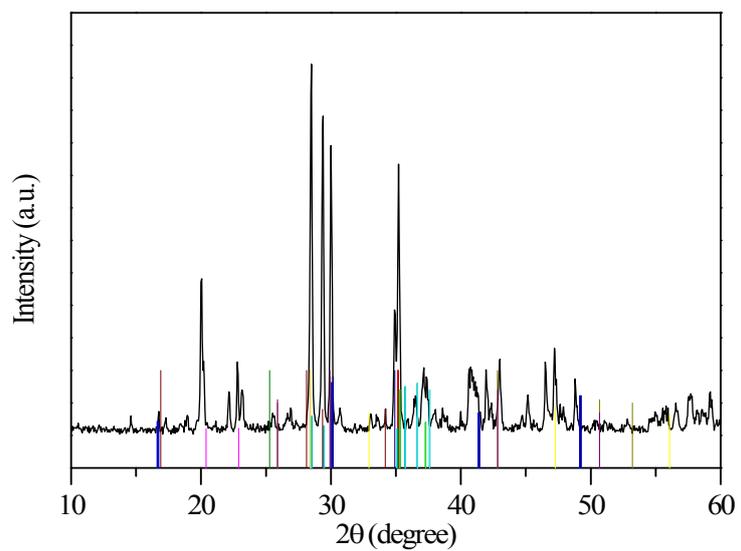


Fig. S1 The XRD pattern of TiO₂/LPP photoanode,.

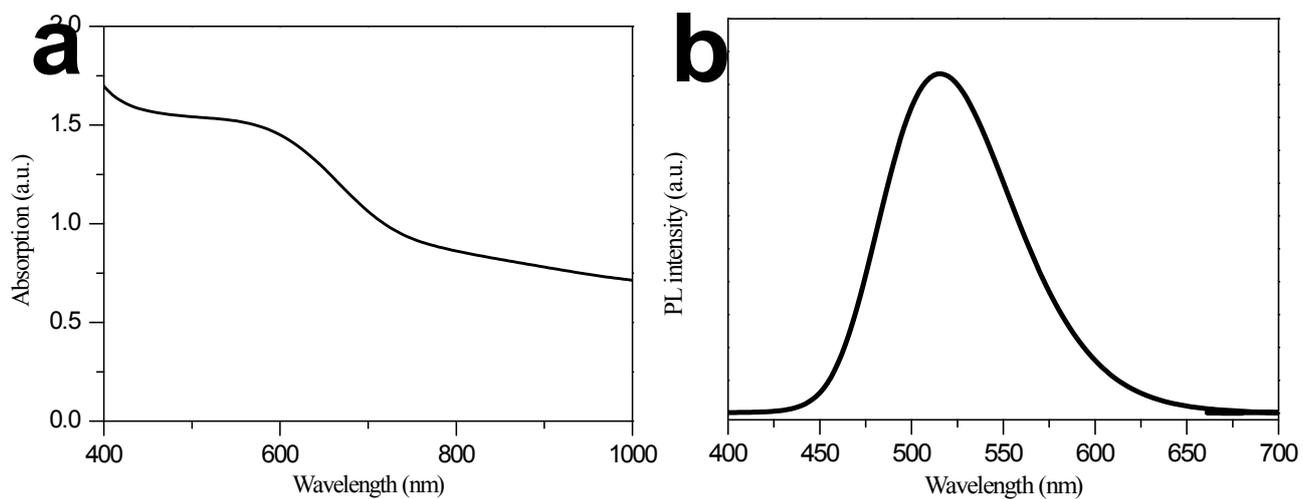


Fig. S2 (a) Optical absorption spectrum of LPP layer using dye-TiO₂ as a benchmark and (b) PL emission spectra of LPP phosphors. The PL spectrum is recorded at room temperature and excited under 330 nm light excitation.

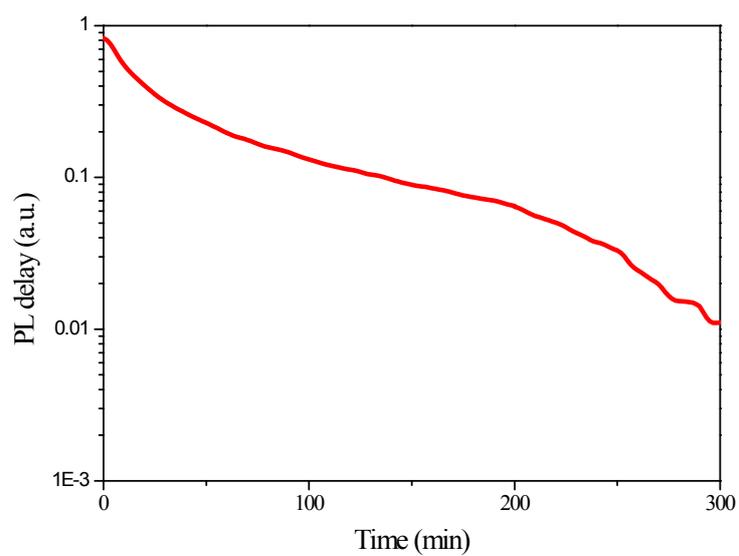


Fig. S3 The PL decay of the green-emitting LPP phosphors.

References

- S1 B. B. Hu, Q. W. Tang, B. L. He, L. Lin and H. Y. Chen. *J. Power Sources* 2014, **267**, 445.
- S2 Z. Lan, J. H. Wu, J. M. Lin, M. L. Huang, P. J. Li, Q. H. Li, *Electrochim. Acta* 2008, **53**, 2296.