

Supporting information

Perovskite Nanocrystal Luminescent Composite via In-situ Ligand Polymerization Towards Display Application

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EXPERIMENTAL SECTION

Chemicals: Cs₂CO₃ (99.9 %), PbBr₂ (99.0 %), Oleic acid (OA, 90 %), Oleylamine (OLA, 80 ~ 90 %), Octadecene (90 %, ODE), (3-aminopropyl)triethoxysilane (APTES, 99 %), Poly(methylhydrosiloxane) (15 - 40 mPa.s, PMHS), n-Hexane (98 %), and Ethylacetate (99.9 %) were purchased from Aladdin. Toluene (analytical grade, H₂O content 0.018%) was purchased from Sinopharm Chemical Reagent Co., Ltd. All the chemicals were used as purchased without further treatment.

Preparation of Cs-oleate: 0.814g Cs₂CO₃ was loaded into 100 mL three-neck flask along with 40 mL octadecene and 2.5 mL OA, dried under vacuum for 1 h at 120 °C, and alternately exposed to vacuum and argon at least six times until all Cs₂CO₃ were completely dissolved and the solution becomes clear.

Synthesis of CsPbBr₃ PeNCs with OA+OLA ligands: 0.69 g PbBr₂ and 50 mL ODE were loaded into 250 mL three-neck flask and dried under vacuum for 1 h at 120 °C. Dried 5 mL OLA and 5 mL OA

were injected at 120 °C under N₂ and maintain for 15 minutes until PbBr₂ is completely dissolved and the solution becomes transparent, and then the temperature was raised to 180 °C and 4 mL Cs-oleate solution was quickly injected. 5 s later, the mixture solution was cooled to room temperature by the ice-water bath. Finally, 8000 rpm for 4 minutes was used to isolate the aggregated PeNCs, and then ethylacetate (crude solution:ethylacetate=1:3 by volume) was added to collect the precipitate again by centrifugation, and the precipitate was dispersed in n-hexane for storage.

Synthesis of CsPbBr₃ PeNCs with OA + APTES ligands: 0.69 g PbBr₂ and 50 mL ODE were loaded into 250 mL three-neck flask and dried under vacuum for 1 h at 120 °C. Dried 5 mL APTES and 5 mL OA were injected at 120 °C under N₂ and maintain for 15 minutes until PbBr₂ is completely dissolved and the solution becomes transparent. The temperature was raised to 150 °C and 4 mL Cs-oleate solution was quickly injected. 10 s later, the mixture solution was cooled to room temperature by the ice-water bath. Finally, 8000 rpm of centrifugation for 4 minutes was used to isolate the aggregated PeNCs, and then n-hexane (crude solution: n-hexane = 1:3 by volume) was added to collect the precipitate again by centrifugation, and the precipitate was dispersed in n-hexane for storage.

Synthesis of TMOS composite: 0.69 g PbBr₂ and 50 mL ODE were loaded into 250 mL three-neck flask and dried under vacuum for 1 h at 120 °C. Dried 5 mL OLA and 5 mL OA were injected at 120 °C under N₂ and maintain for 15 minutes until PbBr₂ is completely dissolved and the solution becomes transparent, then the temperature was raised to 180°C and 4 mL Cs-oleate

solution was quickly injected. 5 seconds later, the mixture solution was cooled to room temperature by the ice-water bath. 8000 rpm for 4 minutes was used to isolate the aggregated PeNCs, and then ethylacetate (crude solution: ethylacetate = 1:3 by volume) was added to collect the precipitate again by centrifugation, and the precipitate was dispersed in toluene for storage. Then, 50 mL PeNCs–toluene solution (1 mg mL^{-1}) was added into a 100 mL beaker, and 500 μL TMOS was added to the beaker. After stirring for 36 hours, the precipitates were collected by centrifugation at 5000 rpm for 4 minutes. These precipitates were washed twice with toluene. The TMOS composite powders were obtained by vacuum drying for 36 h.

Synthesis of ILP composite: 0.69 g PbBr_2 and 50 mL ODE were loaded into 250 mL three-neck flask and dried under vacuum for 1 h at 120°C . Dried 5 mL APTES and 5 mL OA were injected at 120°C under N_2 and maintain for 15 minutes until PbBr_2 is completely dissolved and the solution becomes transparent, and then the temperature was raised to 180°C and 4-mL Cs-oleate solution was quickly injected. 10 seconds later, the mixture solution was cooled to room temperature by the ice-water bath. Then, the mixed solution was transferred to a 250 mL beaker, and 15 mL of PMHS was added to the beaker, and it was exposed to the air (21°C , 50 % humidity) and stirred for 6 hours. Finally, *n*-hexane is added to the beaker, and the quantum dots that had not formed complexes were washed and centrifuged until the supernatant becomes clear, this process took about 3 times. The precipitated complex needed to be dried in a vacuum drying oven for 36 hours.

Preparation of Sandwich-structured light conversion film: The PeNC composite powders were added into the commercialized glue (with the main ingredient was isobornyl acrylate, $C_{13}H_{20}O_2$) and stirred for 30 min until being evenly blended and the mixture was placed in a degassing bucket to expel the bubbles. Then the glue was coated on the bottom-PET polymer film with a thickness of 50 μm by doctor-blade coating and then the film was covered by a top-PET layer. The process was then followed by a UV curing for 5 min to obtain a fully solidified layer. Notably, the optical density of the light after light converting had been well modulated by the thickness of the ILP layer during the doctor-blading process. The final films after size-fixing are used directly as a light converting film and light scattering film between the blue-LED driven backlight and the LCD module. The same process was applied to prepare TMOS composite light conversion film and bare PeNCs light conversion film.

Measurement and characterizations: TEM images were recorded by an FEI Tecnai G2 F30 transmission electron microscope, and the acceleration voltage of 300 kV. PL QY and PL spectra were measured by Hamamatsu Quantaaurus-QY C11347-11 with an integrating sphere system and the PL QY value was determined by the photon numbers. The PL lifetimes were measured by the Edinburgh Instrument FLS980. XRD measurements were carried out by an X-ray diffractometer (Bruker Advance D8 Ew, Germany) with Cu $K\alpha$ radiation ($\lambda = 1.54178 \text{ \AA}$). The operation voltage and current were 40 kV and 25 mA, respectively. The 2-theta range was from 10° to 80° in steps of 0.02° . FTIR spectra were recorded on an ICAN 9 Fourier infrared spectrometer. XPS data were measured by Thermo ESCALAB 250XI. The spectra of the backlight modules were collected by EVERFINE ATA-500. Solid-state $^1\text{H-NMR}$ measurements (JNM-

ECZ600R, Japan) of the PeNCs-ILP are conducted at 14.01 T operating at a ^1H resonance frequency of 599.7 MHz. Liquid-state ^1H -NMR measurements (Advance Bruker 400M, Switzerland) of the APTES, PMHS, and PeNCs-APTES were conducted at 9.4 T operating at a ^1H resonance frequency of 400 MHz.

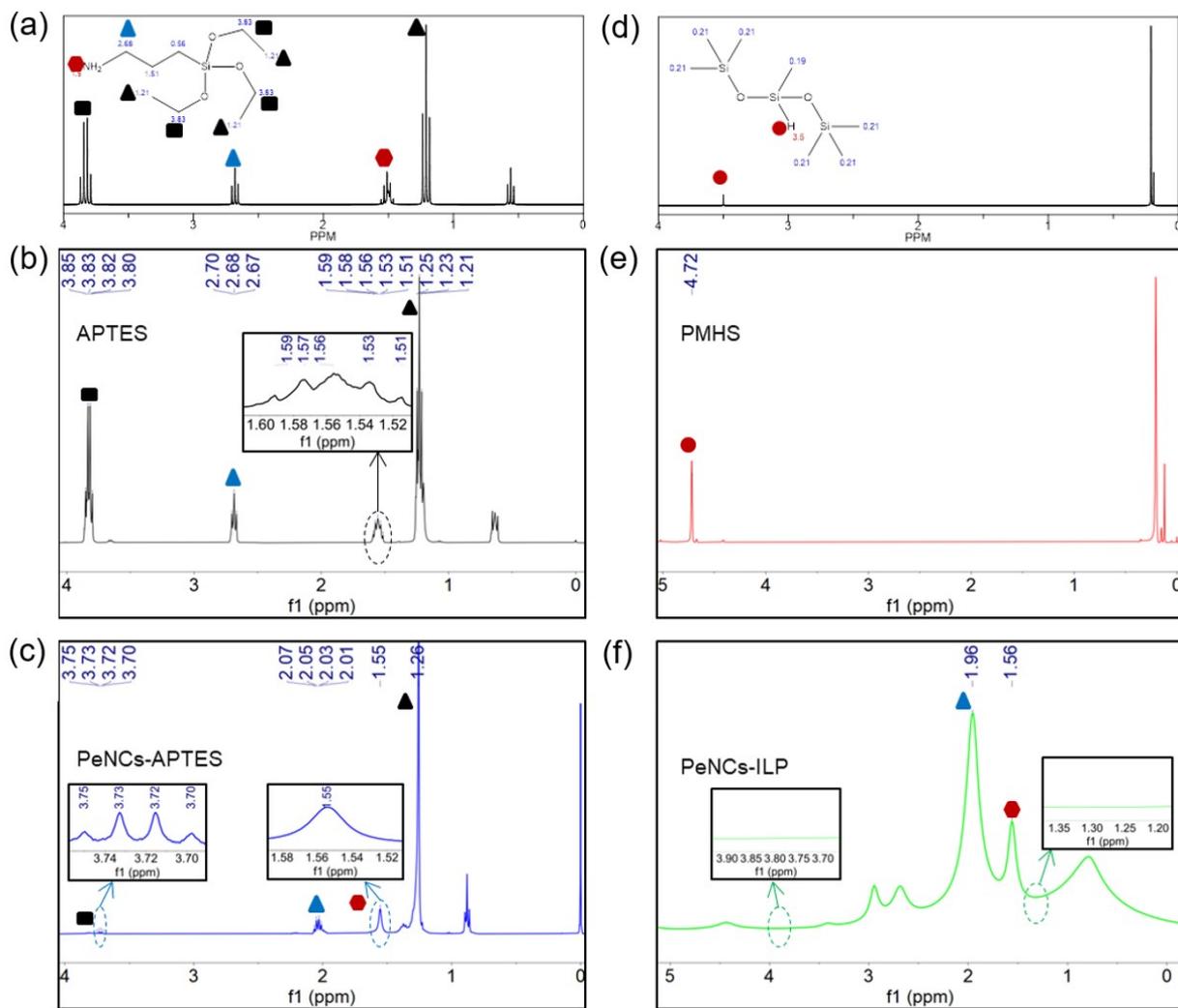


Fig. S1. (a) Theoretical and (b) as measured $^1\text{H-NMR}$ species of APTES. (c) $^1\text{H-NMR}$ result of PeNCs with APTES ligands. (d) Theoretical and (e) as measured PMHS species. (f) $^1\text{H-NMR}$ result of the final ILP composite.

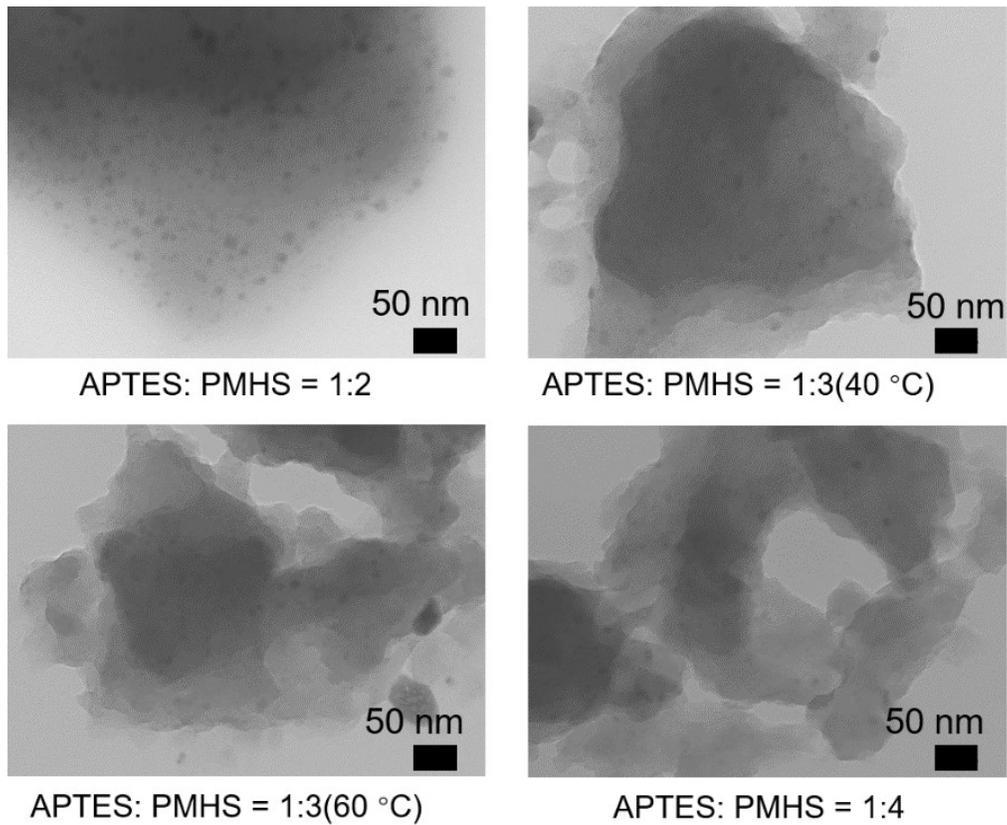


Fig. S2. TEM images of the ILP composites with different APTES: PMHS ratios used in the fabrication and of samples made at different temperatures for 1:3 composition.

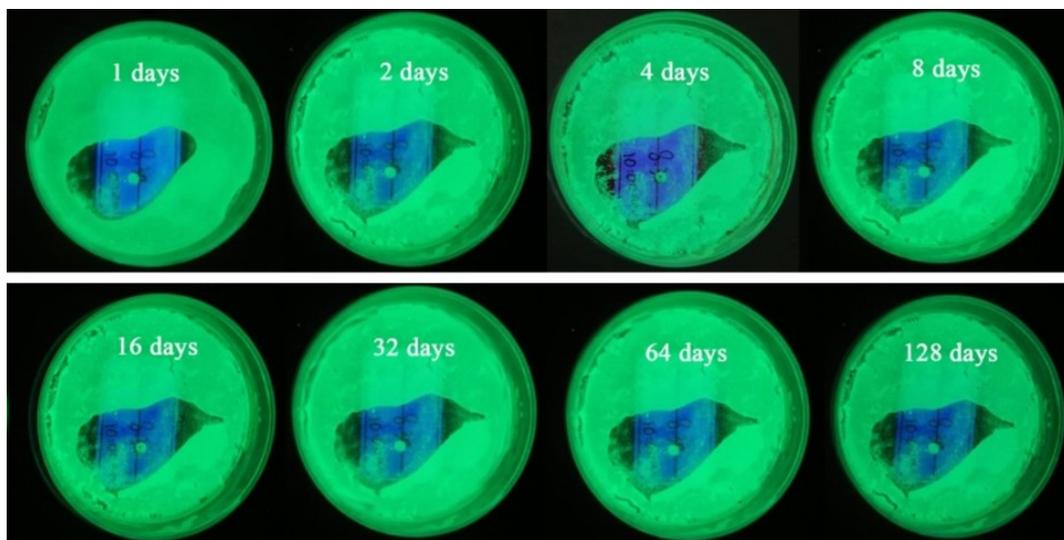


Fig. S3. PL evolution of the ILP composites (1:3) immersed in NaCl (0.1 g/ml) solution, measured under 365 nm UV light illumination.

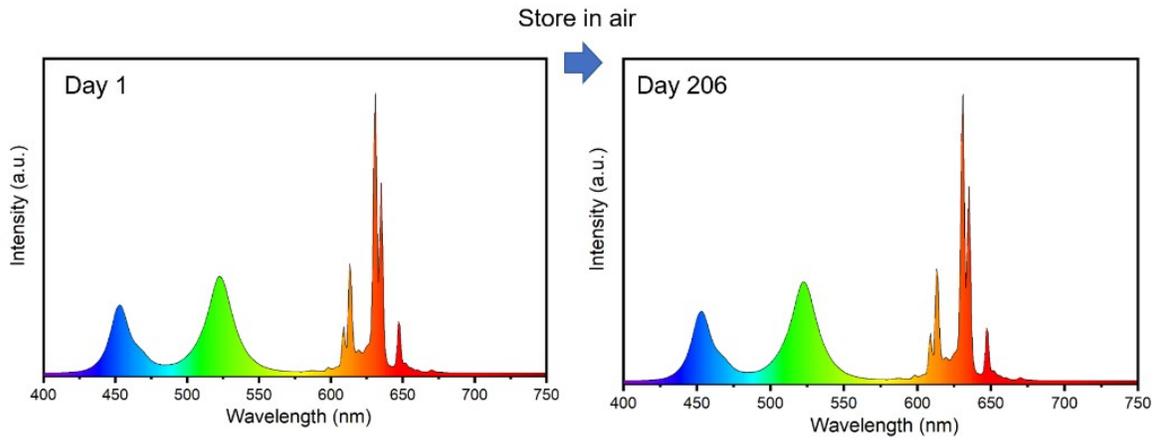


Fig. S4. Electroluminescence spectrum of the “on-chip” encapsulated WLED after preparation (day 1, left) and stored in air for 206 days (right).

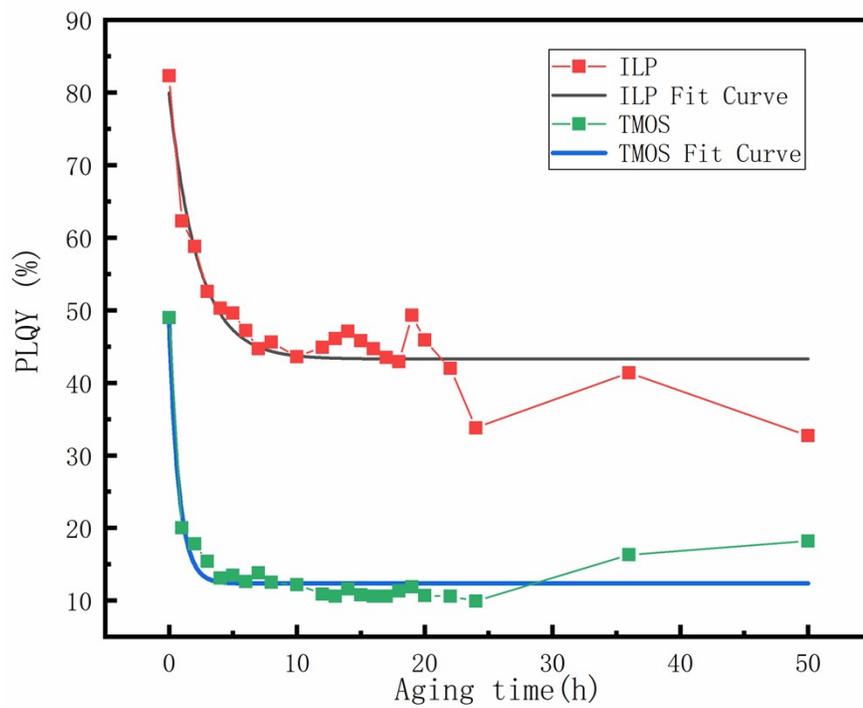


Fig. S5. PL QY tracking of the composite films based on ILP composite and TMOS composite respectively at 85°C and 85% humidity.

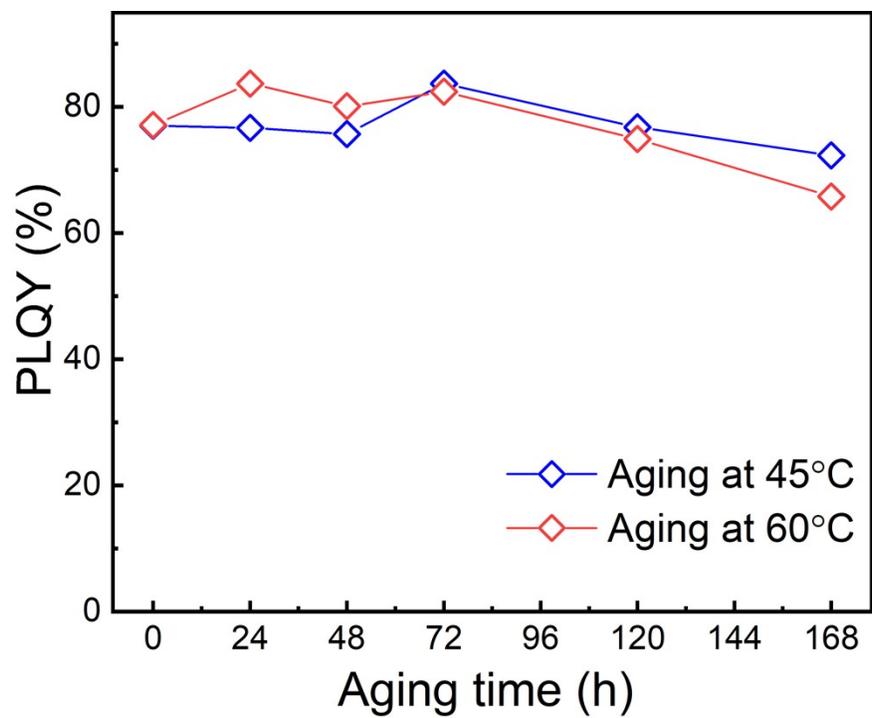


Fig. S6. ILP powders PL QY evolution under different aging condition, in which the powders remain 94% PL QY at 45 °C condition and 85% at 60 °C, after 168 hours storage.