Electronic Supplementary Information

Synthesis of highly white-fluorescent Cu-Ga-S quantum dots for solid-state lighting device

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Synthesis of Cu–Ga–S (CGS)/ZnS core/shell QDs

In a typical preparation of white CGS/ZnS QD emitters, 0.0625 mmol of Cu iodide, 0.5 mmol of 0.5 mmol of Ga iodide, 0.5 mL of 1-dodecanethiol (DDT) and 5 mL of oleylamine (OLA) were loaded in 50 mL of three-neck flask, and this mixture was degassed, N_2-purged at 120^{o}C and further heated to 180^{o}C. And then, a sulfur (S) stock solution, which was prepared beforehand by dissolving 2 mmol of S into 2 mL of 1-octadecence (ODE) at 190^{o}C for 20 min, was swiftly injected into the above cationic mixture at that temperature and the reaction for CGS core growth proceeded for 4 min. Consecutive ZnS shelling was performed in a two-step fashion as follows; the first ZnS stock mixture prepared by dissolving 4 mmol of Zn acetate, 2 mL of DDT, 2 mL of ODE and 4 mL of oleic acid (OA) at 190^{o}C was slowly added into CGS core growth solution and this shelling reaction was held at 220^{o}C for 30 min. And

the second ZnS stock solution consisting of 8 mmol of Zn stearate, 4 mL of DDT and 8 mL of ODE was sequentially introduced and the reaction proceeded at 250^{o}C for 1 h. As-synthesized CGS/ZnS QDs were repeatedly purified by a centrifugation with a solvent combination of hexane/ethanol and finally dispersed in chloroform.

Fabrication of white QD-LEDs

QD-LEDs were fabricated using a 50×50 mm^2 surface-mounted device (SMD) typed InGaN-based near-UV-emitting LED (λ=392 nm, Taewon Semiconductor, Korea). 3 mL of chloroform dispersion of CGS/ZnS QDs having an optical density (OD) of ~3.0 at 370 nm was thoroughly blended with thermo-curable epoxy resin/hardener (weight ratio of 1). After drying the above QD mixture at 60^{o}C in an oven for the removal of chloroform, the resulting QD paste was dispensed in the mold of LED. In addition, to vary the amount of QDs loaded inside LED mold, different volumes, *i.e.*, 0.5 (the lowest), 1.0, 1.5, and 2.0 mL (the highest QD-loading), of the above QD dispersion were taken for preparation of QD paste. Finally, the dispensed QD paste was then subjected to a two-step curing process of 70^{o}C for 30 min and 110^{o}C for 1 h.

Characterization

Absorption and photoluminescence (PL) spectra of QDs were obtained with UV–visible absorption spectroscopy (Shimadzu, UV-2450) and a 500 W Xe lamp-equipped spectrophotometer (PSI Co. Ltd., Darsa Pro-5200), respectively. Absolute PL quantum yield (QY) of QD dispersion was assessed in an integrating sphere with an absolute PL QY measurement system (C9920-02, Hamamatsu). An X-ray photoelectron spectrometer (XPS) (Thermo VG, U.K.) with Al K_{α} X-rays (E = 1486.6 eV) was used to analyze the surface

chemical compositions of core and core/shell QDs. The compositional analysis of as-synthesized QDs was performed with an energy dispersive spectroscopy (EDS) (EDAX Inc., Phoenix)-equipped scanning electron microscope operating at 15 kV. Powder x-ray diffraction (XRD) (Rigaku, Ultima IV) based on Cu K_α radiation was utilized for the structural information on QDs. High-resolution transmission electron microscopic (TEM) images were collected using JEOL JEM-4010 electron microscope operated at an accelerating voltage of 200 kV. Primary electroluminescent (EL) data including EL spectrum, luminous efficacy (LE), correlated color temperature (CCT), CIE color coordinates, and color rendering index (CRI) of the QD-LEDs were collected in an integrating sphere with a diode array rapid analyzer system (PSI Co. Ltd.).

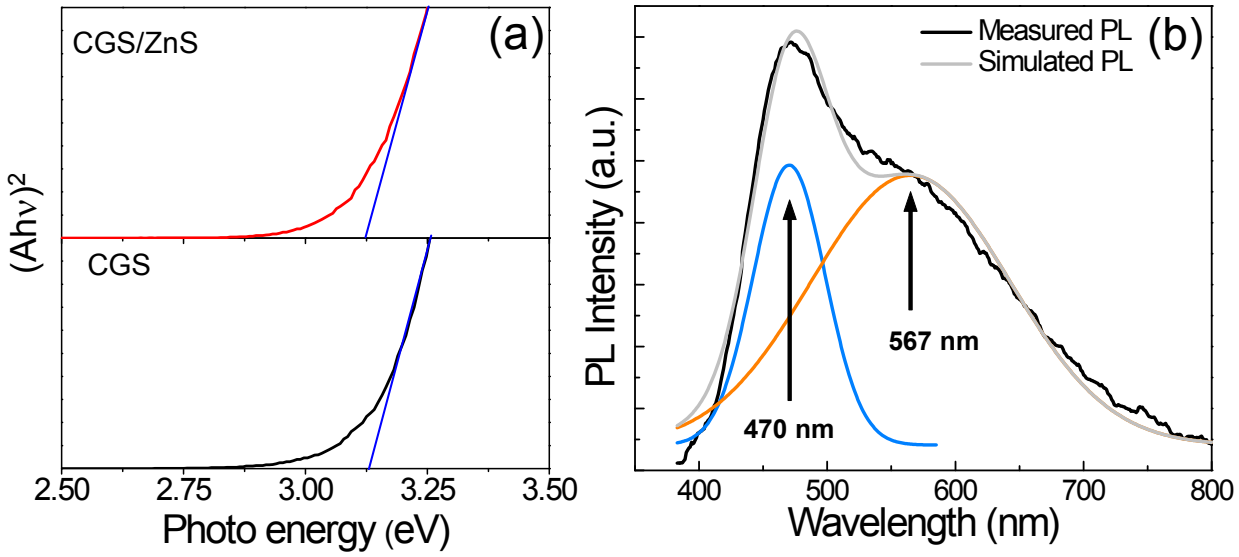

Fig. S1 (a) Tauc plots of CGS core and CGS/ZnS core/shell QDs and (b) measured *versus* simulated PL consisting of two resolved spectral components of CGS/ZnS QDs.

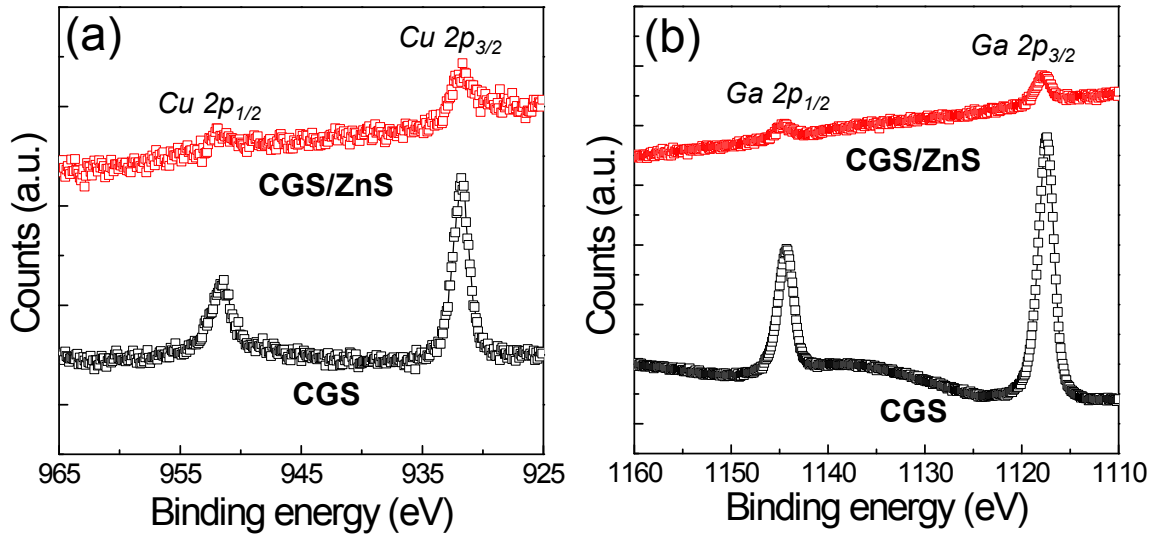

Fig. S2 Comparison of high-resolution XPS scans of (a) Cu 2p and (b) Ga 2p peaks of CGS and CGS/ZnS QDs.

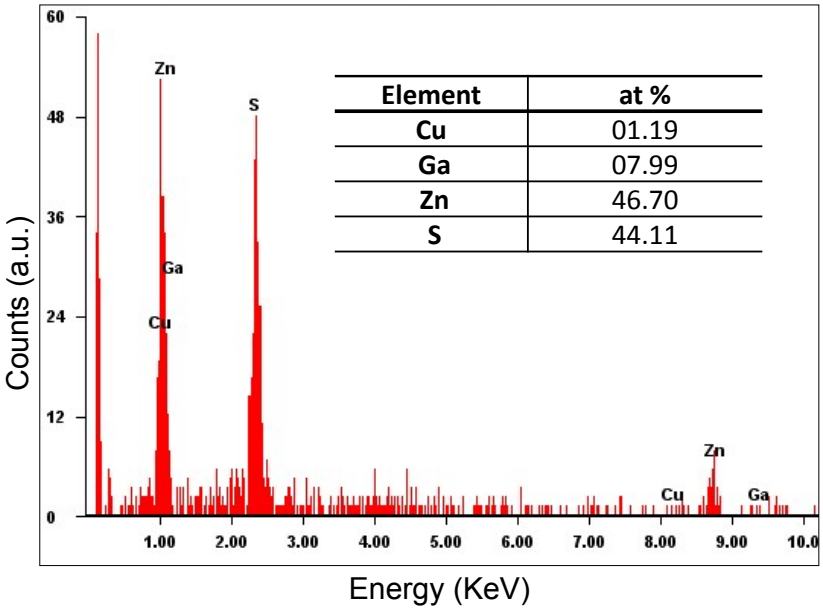

Fig. S3 EDS spectrum and chemical compositions of CGS/ZnS QDs.

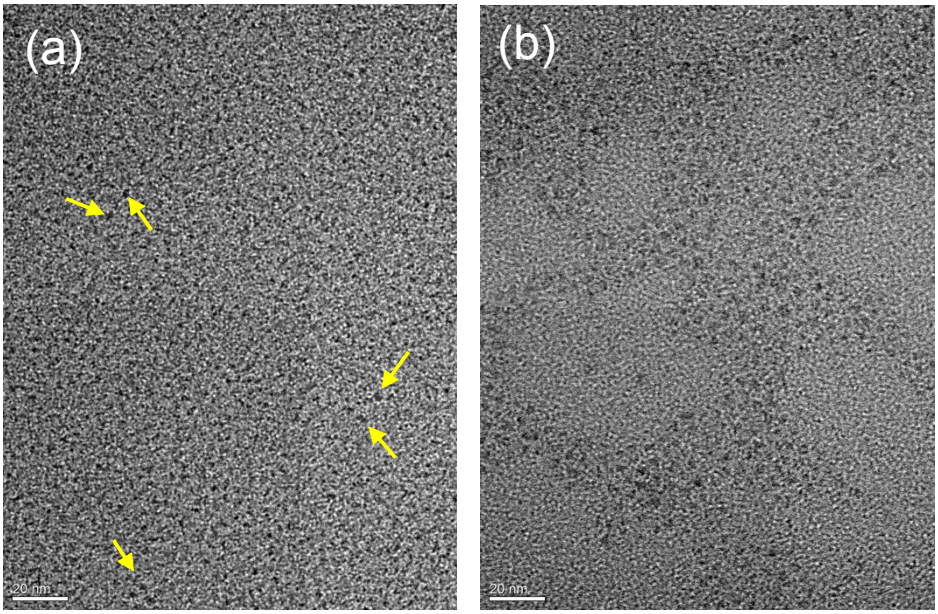

Fig. S4 Low-magnification TEM photographs of (a) CGS core and (b) CGS/ZnS core/shell QDs (scale bars: 20 nm).

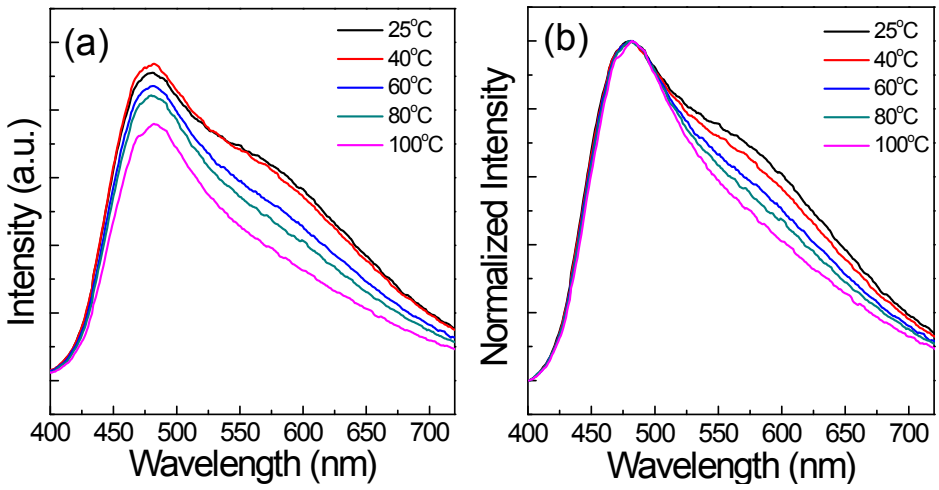

Fig. S5 Change in (a) as-recorded and (b) normalized PL intensity of a QD-epoxy resin mixture with increasing temperature from 25 to 100°C.

Table S1 Variations of CIE color coordinates, CRI, CCT, and luminous efficacy as a function of forward current.

Forward Current (mA)	CIE Color Coordinates	CRI	CCT (K)	Luminous Efficacy (lm/W)
10	(0.333, 0.391)	84	5527	20.8
20	(0.330, 0.390)	83	5556	20.4
50	(0.325, 0.387)	83	5784	16.3
70	(0.324, 0.389)	83	5817	14.2
100	(0.320, 0.388)	82	5919	11.1

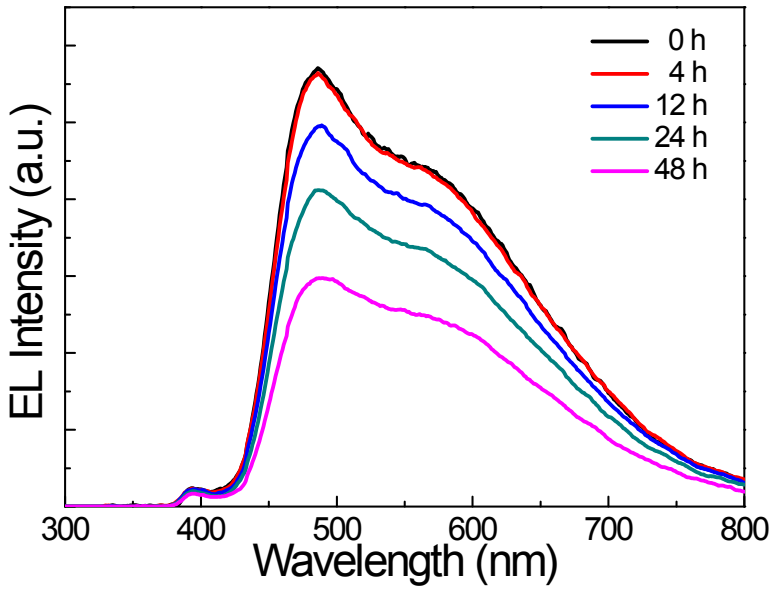

Fig. S6 Temporal evolution of EL spectra of white QD-LED operated at 20 mA for a prolonged duration up to 48 h.