Organic- Inorganic Hybrid Perovskite Quantum Dots with High PLQY and Enhanced Carrier Mobility through Crystallinity Control by Solvent Engineering and Solid State Ligand Exchange

Jin Woo Choi^{a,b} Hee Chul Woo^a, Xiaoguang Huang^c, Wan-Gil Jung^b, Bong-Joong Kim^b, Sie-Wook Jeon^d, Sang-Youp Yim^a, Jae-Suk Lee^{b,} and Chang-Lyoul Lee^{a,*}*

^{*a*}Advanced Photonics Research Institute (APRI), Gwangju Institute of Science and Technology (GIST), Gwangju 61005, Republic of Korea.

^bSchool of Material Science and Engineering, Gwangju Institute of Science and Technology (GIST), Gwangju 61005, Republic of Korea

^cKey Laboratory of Advanced Technology for Materials Synthesis and Processing, Wuhan University of Technology, Wuhan 430070, People's Republic of China

^dLighting Materials & Components Research Center, Korea Photonics Technology Institute (KOPTI), Gwangju 61007, Republic of Korea

E-mail: jslee@gist.ac.kr (Jae-Suk Lee), vsepr@gist.ac.kr (Chang-Lyoul Lee)



Fig. S1. Digital camera images of perovskite QDs solutions (**a**) in absence of UV light and (**b**) in presence of UV light. The benzene and butanol were used as solvents. When the polar solvent butanol was used, perovskite QDs decomposed due to their ionic properties.



Fig. S2. PLQY of perovskite QDs solutions with the precursor ratio (PbBr₂:CH₃NH₃Br).



Fig. S3. (a) and (b) X-ray diffraction (XRD) patterns of perovskite QDs with the precursor ratio (PbBr₂:CH₃NH₃Br).



Fig. S4. ¹H nuclear magnetic resonance (¹NMR) spectra of perovskite QDs solutions against the number of purification processes.



Fig. S5. (a) Digital camera images of perovskite QDs solutions (a) in absence of UV light and (b) in presence of UV light. Excess addition of OA and OAm in QDs solutions caused the perovskite QDs to decompose and lose their PL emission.



Fig. S6. PL emission of perovskite QDs solutions and films with the amount of added ligands (PLAP). PL emissions of perovskite QDs films were measured using Hamamatsu C9920-02.



Fig. S7. PL decay profiles of perovskite QDs films against the number of purification processes.



Fig. S8. Digital images of ligand exchanged perovskite QDs solutions. When the propyl or butyl acid and amine were used as short alkyl ligands, the precipitation occurred due to the poor solubility of ligands. For comparison, perovskite QDs solution passivated by OA and OAm was also displayed.



Fig. S9. UV-vis absorption spectra of perovskite QDs films before and after PLEP. The absorption intensities of each perovskite QDs films, directly related with film thickness, were controlled by spin-casting number of QDs solution before PLEP.



Fig. S10. PL emission of perovskite QDs films with different alkyl chain length ligands. PL emission of perovskite QDs films was measured using Hamamatsu C9920-02.



Fig. S11. The PLQY of perovskite QDs solutions with different alkyl chain length ligands through PLEP. The volume ratio of added ligands is $1 \times 10^{-4} \text{ v/v}$ %. The alkyl chain length of acid and amine is same.



Fig. S12. Log current density *vs*. log voltage (*J-V*) characteristics of the hole-only devices; (a) pristine (No PLEP), (b) propyl, (c) hexyl, (d) octyl and (d) dodecyl acid and amine.



Fig. S13. TEM image of perovskite QDs films with different alkyl chain length ligands; (a) butyl, (b) octyl, (c) decyl and (d) dodecyl acid and amine.



Fig. S14. External quantum efficiency (EQE) of perovskite QDs-LEDs with different alkyl chain length ligands

Table S1. PLQY and PL perovskite QDs solutions with different precursor ratio of $PbBr_2$ and CH_3NH_3Br . Here, the optical properties were measured by perovskite QDs solution without purification.

	1:1	1:0.9	1:0.8	1:0.7	1:0.6	1:0.5
PLQY (%)	73	78	85	92	91	88
PL (nm)	510	510	510	511	511	511

Table S2. PLQY, PL and FWHM of perovskite QDs solutions against the number of purification processes.

	As synthesized	Purification x1	Purification x2	Purification x3
PLQY (%)	92	84	81	78
PL (nm)	511	512	512	513
FWHM (nm)	35	33	31	31

Table S3. The PLQY of perovskite QDs in solutions and films with the amount of added ligands (PLAP).

					10 ⁻⁴ v/v%
Amount of ligands	0	5	10	20	30
PLQY (%) in solution	81	90	95	91	73
PLQY (%) in film	55	67	70	54	45

The PLQY in solution is relative PLQY, which was determined using Coumarine 500 (~47 %) as reference and PLQY at film is absolute PLQY, measured using Hamamatsu C9920-02.

Purification number	τ ₁ /ns (f ₁)	τ_2 /ns (f_2)	τ_3 /ns (f_3)	χ^2	$ au_{ m avr}/ m ns$
1st purification	2.22 (0.12)	4.55 (0.86)	13.84 (0.02)	0.934	4.46
2nd purification	1.55 (0.25)	4.53 (0.58)	17.25 (0.17)	0.989	5.95
3rd purification	1.08 (0.27)	4.57 (0.47)	22.20 (0.26)	1.022	8.21

Table S4. PL lifetime of perovskite QDs film against the number of purification processes.

Monitored wavelength was 510 nm. The PL decay curves were fitted by the tri-exponential function to investigate exciton dynamics of perovskite QDs. The intensity-weighted average exciton lifetime (τ_{avr}) was $f_I\tau_1 + f_2\tau_2 + f_3\tau_3$, where f_I , f_2 and f_3 are fractional intensities and τ_1 , τ_2 and τ_3 are lifetimes. Samples were prepared by dropping perovskite QDs solution on glass substrate to minimize the elimination of ligands during the spin-casting.

Table S5. The PLQY of perovskite QDs films with different alkyl chain length ligands.

	Pristine (No PLEP)	Propyl	Butyl	Hexyl	Octyl	Decyl	Dodecyl
PLQY(%) at film	55	60	72	82	78	73	68

Table S6. The PLQY of perovskite QDs solutions with different alkyl chain length ligands.

								10-	⁴ v/v%
	Pristine (No PLEP)	Acetic	Propyl	Butyl	Hexyl	Octyl	Decyl	Dodecyl	Oleic
PLQY(%) at solution	81	41	47	64	88	91	92	93	95

Table S7. Hole mobilities of perovskite QDs films with different alkyl chain length ligands.

Mobility	Pristine (No PLEP)	Propyl	Hexyl	Octyl	Dodecyl
cm ² ·V ⁻¹ ·s ⁻¹	2.0x10 ⁻⁴	2.4x10 ⁻⁴	6.2x10 ⁻³	1.3x10 ⁻³	9.0x10 ⁻⁴