Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.
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Electronic Supplementary Information

Highly-efficient and all-solution-processed red-emitting InP/ZnS-based quantum-dot light-emitting diodes enabled by compositional engineering of electron transport layers

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Figure S1

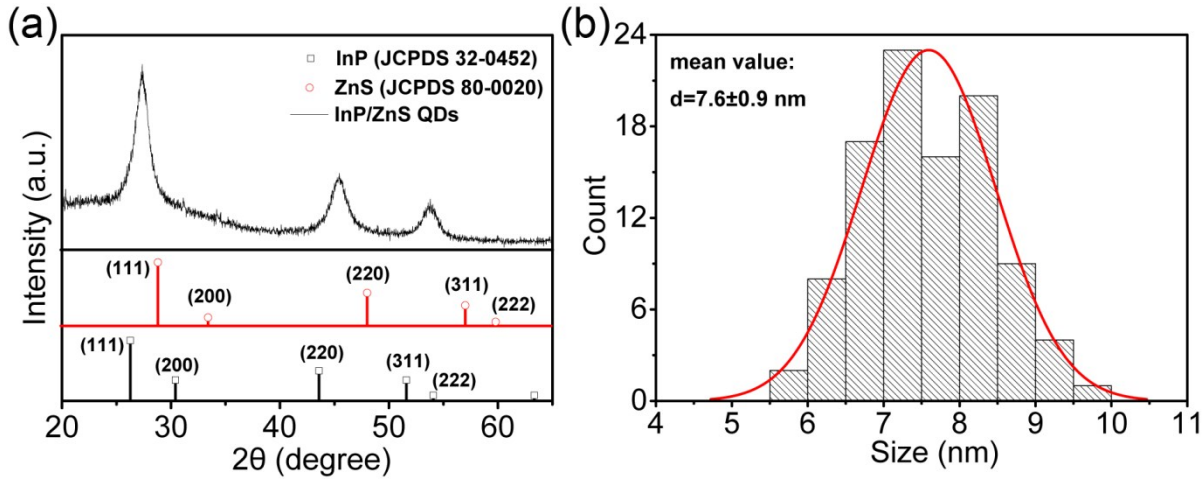

Figure S1. (a) XRD patterns of red-emitting InP/ZnS QDs, and the bottom lines represent the standard diffraction lines of zinc-blended InP (JCPDS No.32-0452) and ZnS (JCPDS No.80-0020) structure; (b) size histogram of 100 particles for red-emitting InP/ZnS QDs.

Figure S2

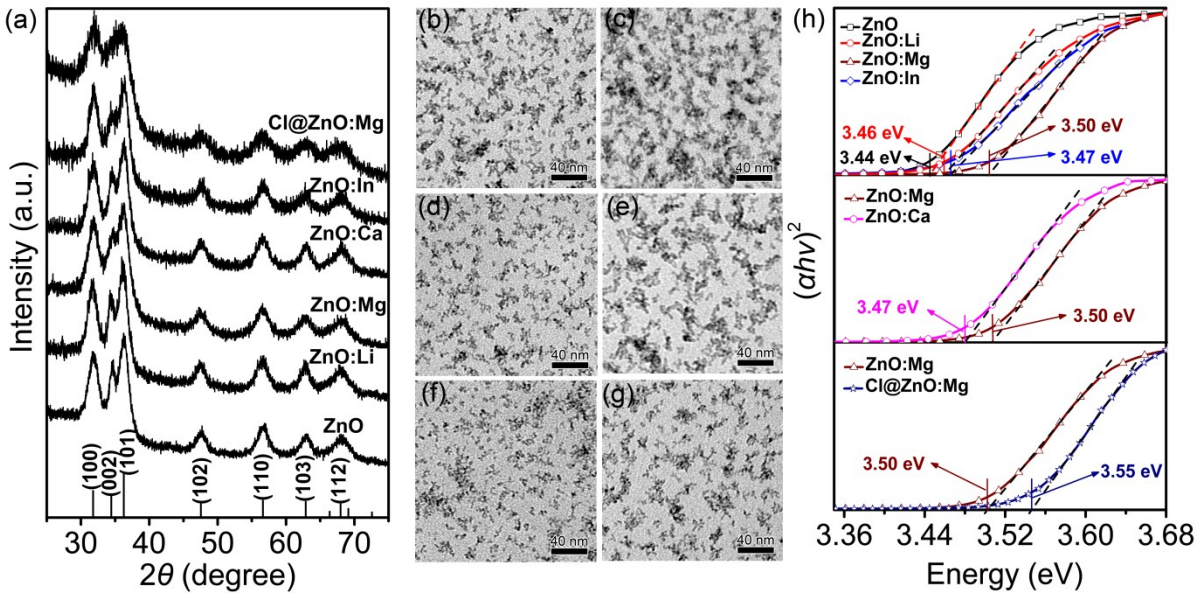

Figure S2. (a) XRD patterns of ZnO, ZnO:Li, ZnO:Mg, ZnO:Ca, ZnO:In and Cl@ZnO:Mg NPs, and the bottom lines represent the standard diffraction lines of wurtzite-type bulk ZnO (JCPDS card No. 36-1451); TEM images of (b) ZnO NPs, (c) ZnO:Li NPs, (d) ZnO:Mg NPs, (e) ZnO:Ca NPs, (f) ZnO:In NPs and (g) Cl@ZnO:Mg NPs; (h) the Tauc plots between $(\alpha h\nu)^2$ and photon energy for different types of ZnO NPs.

Figure S3

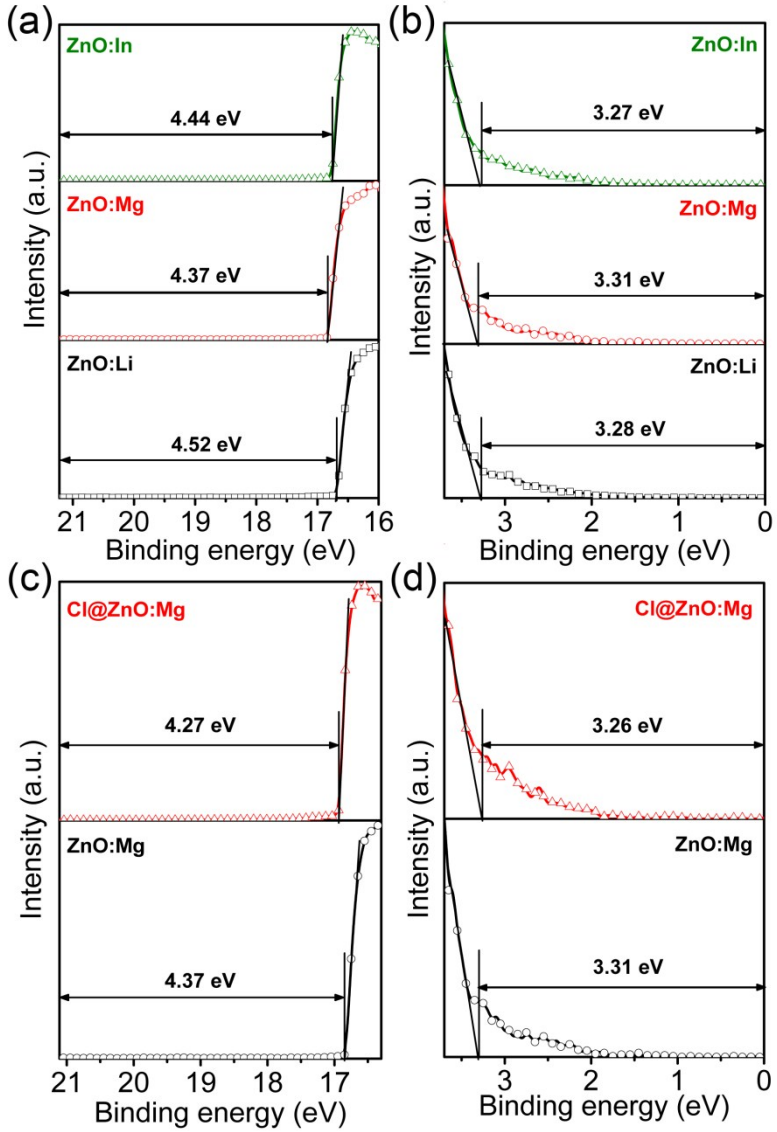

Figure S3. UPS spectra of (a) the high-binding energy secondary electron cut-off regions and (b) the valence-band edge regions of ZnO:Li, ZnO:Mg and ZnO:In NP films deposited on ITO substrates; UPS spectra of (c) the high-binding energy secondary electron cut-off regions and (d) the valence-band edge regions of ZnO:Mg and Cl@ZnO:Mg NP films deposited on ITO substrates. The valence-band maximum (VBM) levels can be calculated from the incident photon energy (21.22 eV), the high-binding cut-off energy ($E_{cut\text{-}off}$) and the onset energy in the valence-band region (E_{onset}) according to the equation of VBM=21.22-($E_{cut\text{-}off}$-E_{onset}). As a result, the VBM positions of ZnO:Li, ZnO:Mg, ZnO:In and Cl@ZnO:Mg NP films are estimated to be 7.80 eV, 7.68 eV, 7.71 eV and 7.53 eV below the vacuum level, respectively. By combining the VBM levels and optical band gaps, the corresponding conduction-band minimum (CBM) levels can be estimated to be 4.34 eV, 4.18 eV, 4.24 eV and 3.98 eV below the vacuum level, respectively.

Figure S4

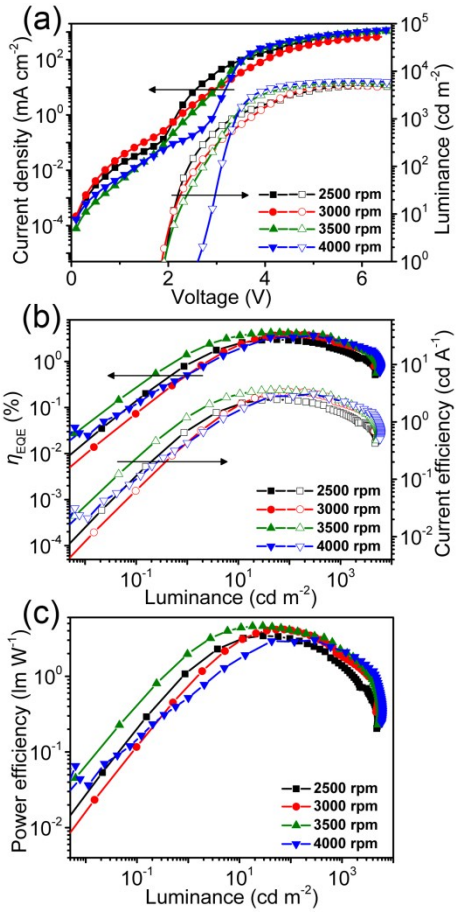

Figure S4. (a) The *J*-*L*-*V* characteristics and (b) η_{EQE} and current efficiency as a function of luminance for the red-light QD-LEDs with different thicknesses of QD layers; (c) the corresponding power efficiency as a function of luminance.

Figure S5

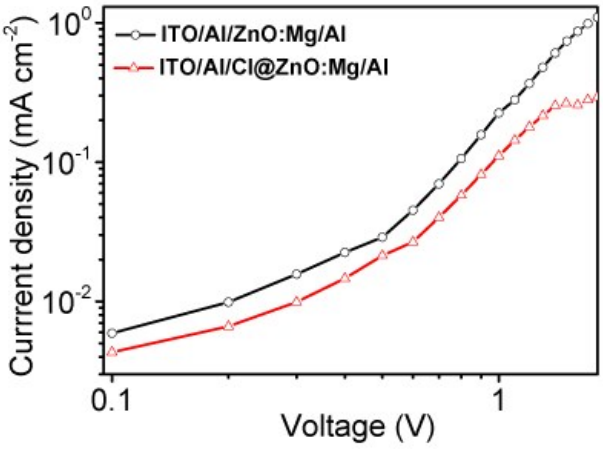

Figure S5. *J-V* characteristics of the electron-only devices

Table S1. Summary of UPS parameters of ZnO:Li, ZnO:Mg, ZnO:In and Cl@ZnO:Mg films obtained from UPS and absorption results.

Sample	$E_{cut\text{-}off}$ (eV)	E_{onset} (eV)	VBM (eV)	Band gap (eV)	CBM (eV)
ZnO:Li	16.70	3.28	7.80	3.46	4.34
ZnO:Mg	16.85	3.31	7.68	3.50	4.18
ZnO:In	16.78	3.27	7.71	3.47	4.24
Cl@ZnO:Mg	16.95	3.26	7.53	3.55	3.98

Table S2. Summary of device performance for all the QD-LEDs in this work.

ETL	L_{max} (cd m^{-2})	η_{EQE} (%)	η_A (cd A^{-1})	η_P (lm W^{-1})
ZnO	4687	2.42	2.00	2.17
ZnO:Li	4252	2.50	2.08	2.25
ZnO:Mg	5431	3.57	2.96	4.19
ZnO:Ca	5636	2.98	2.57	3.23
ZnO:In	4979	2.91	2.41	3.30
Cl@ZnO:Mg	5595	4.24	3.59	4.60

Table S3. Comparison of the device performance between this work and other red-emitting InP-based QD-LEDs reported previously.

Year	Device structure	EQE (%)	η_A (cd A^{-1})	V_{on} (V)	L_{max} (cd m^{-2})	Ref.
2015	ITO/PEDOT:PSS/poly-TPD/InP/ZnSe/ZnS/TPBi/Ca/Ag		1.8	3.4	1710	1
2016	ITO/PEDOT:PSS/TFB/InP/ZnSeS/ZnS/ZnO/Al	2.5	4.2	2.3	2849	2
2016	ITO/ZrO_2/InP/ZnS/TAPC/MoO_3/Al		1.0	> 5	530	3
2018	ITO/PEDOT:PSS/poly-TPD/InP/ZnSe/ZnS/TPBi/LiF/Al		0.85	3.7	230	4
2018	ITO/ZnO/InP/ZnSe/ZnS/CBP/HAT-CN/Al	6.6	13.6	2.0	1600	5
2019	ITO/PEDOT:PSS/TFB/InP/ZnS/Cl@ZnO:Mg/Al	4.24	3.59	2.0	5595	This work

Table S4. The fitting parameters of PL decay curves for Glass/QDs, Glass/QDs/ZnO, Glass/QDs/ZnO:Mg and Glass/QDs/Cl@ZnO:Mg films.

Sample	τ_1 (ns)	α_1 (%)	τ_2 (ns)	α_2 (%)	τ_3 (ns)	α_3 (%)	τ_{av} (ns)
Glass/QDs	0.75	27.65	12.74	30.47	130.07	41.87	58.55
Glass/QDs/ZnO	1.20	42.94	14.14	34.47	131.99	22.58	35.20
Glass/QDs/ZnO:Mg	1.62	30.15	14.96	42.15	145.86	27.70	47.20
Glass/QDs/Cl@ZnO:Mg	1.58	25.94	16.21	41.19	158.96	32.87	59.34

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