Supporting Information

Boosting Charge Transport in Lead Chalcogenide Quantum Dots with Short-Wave Infrared Band Gaps through Anion Composition Engineering

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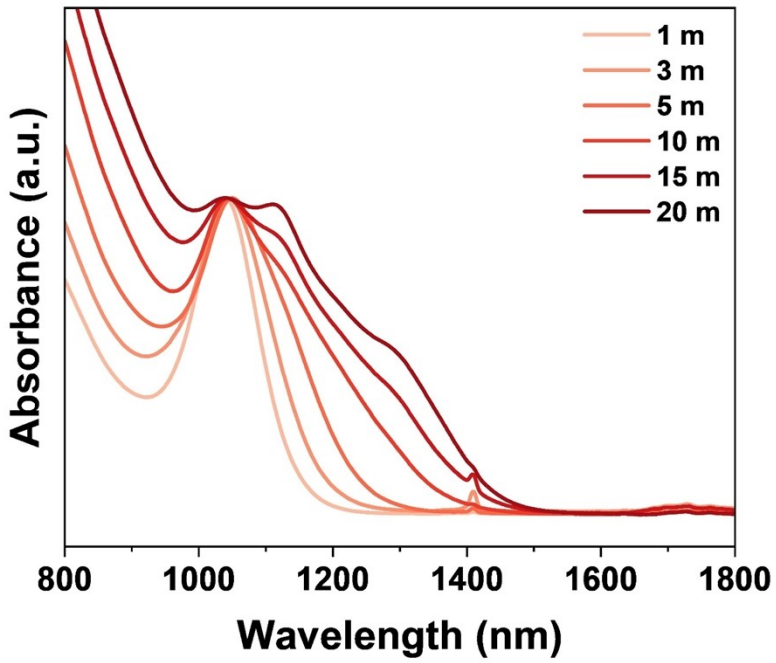


Figure S1. Absorption spectra of $PbSe_xS_{1-x}$ (x = 0.5) nanocrystals synthesized without DPP, recorded at different reaction times ranging from 1 to 20 minutes.

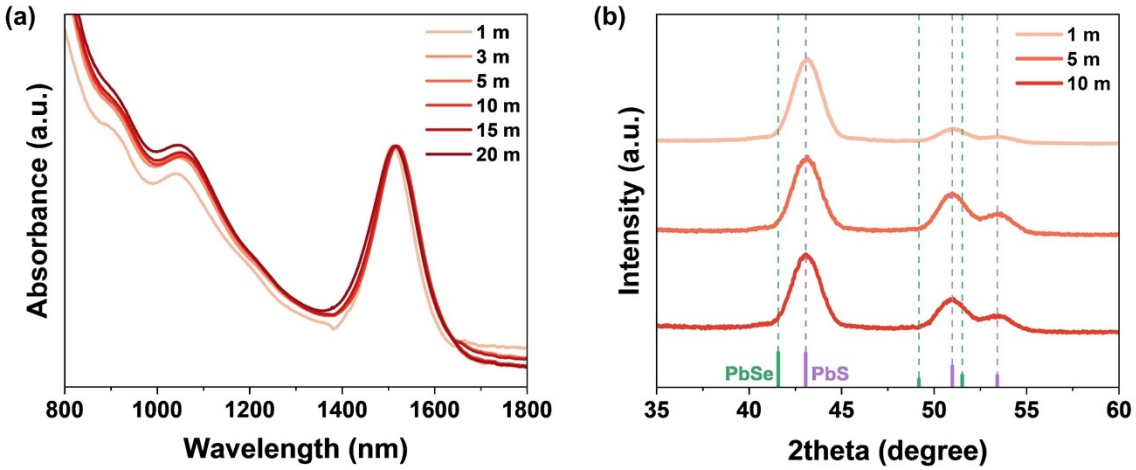


Figure S2. (a) Absorption spectra of $PbSe_xS_{1-x}$ (x = 0.75) nanocrystals synthesized without DPP, recorded at different reaction times ranging from 1 to 20 minutes. (b) XRD patterns of the nanocrystals synthesized at reaction times of 1, 5, and 10 minutes. The green and purple vertical lines indicate reference patterns of PbSe (PDF Card No.: 9000002) and PbS (PDF Card No.: 9008694), respectively.

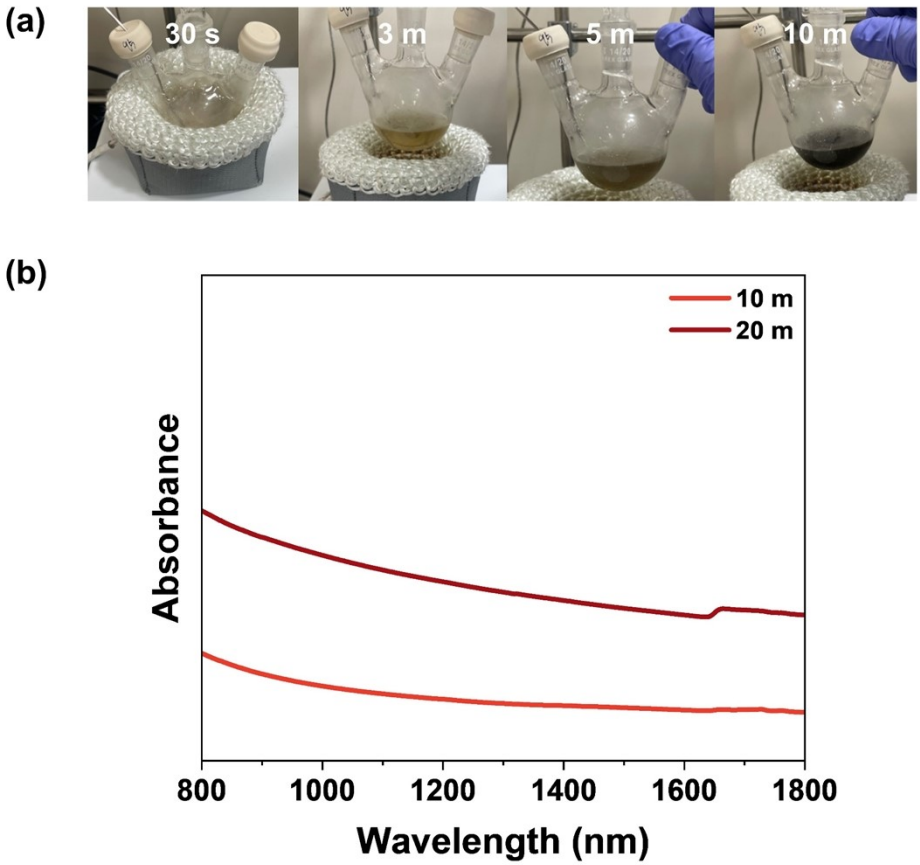


Figure S3. Synthesis results of $PbSe_xS_{1-x}$ (x = 0.95) nanoparticles without DPP. (a) Photographs of the reaction solution at different reaction times (30 seconds to 10 minutes). (b) The absorbance spectra of the solution reacted for 10 and 20 minutes. The baseline increase indicates scattering due to non-uniform particle sizes.

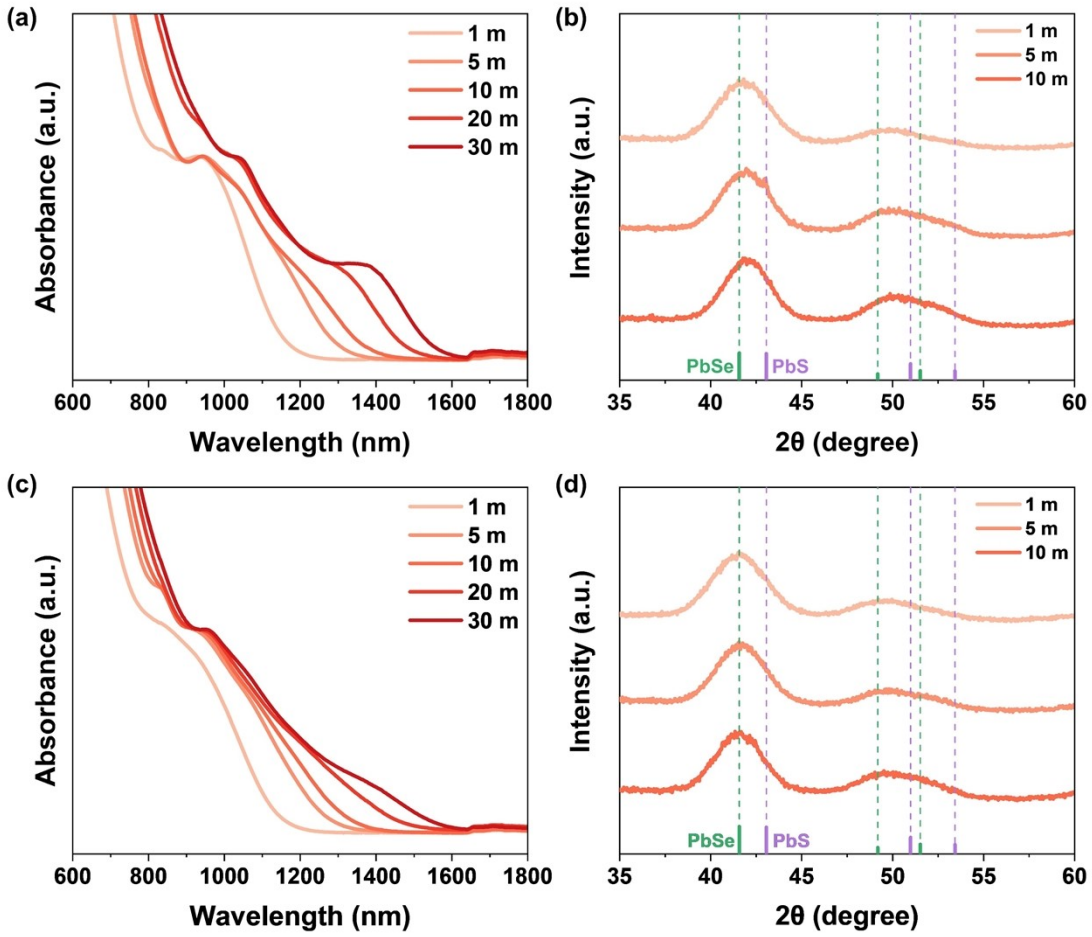


Figure S4. (a, b) Absorption spectra and X-ray diffraction (XRD) patterns of nanocrystals synthesized using TMS-S and TMS-Se precursors at a molar ratio of 0.5 : 0.5, with reaction times of 1, 5, 10, 20, and 30 minutes. (c, d) Absorption spectra and XRD patterns of nanocrystals synthesized at a TMS-S : TMS-Se molar ratio of 0.25 : 0.75, with the same reaction time conditions. The green and purple vertical lines in (b) and (d) indicate reference patterns of PbSe (PDF Card No.: 9000002) and PbS (PDF Card No.: 9008694), respectively.

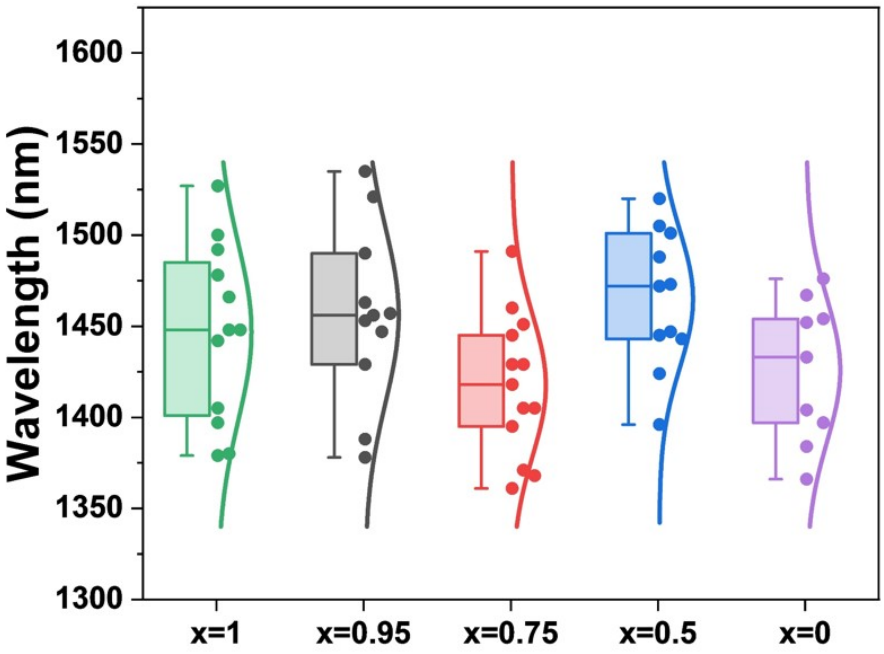


Figure S5. Reproducibility of the optical properties of $PbSe_xS_{1-x}$QDs with different compositions. Statistical distributions of (a) first excitonic absorption peak positions and (b) HWHM values obtained from multiple independently synthesized batches.

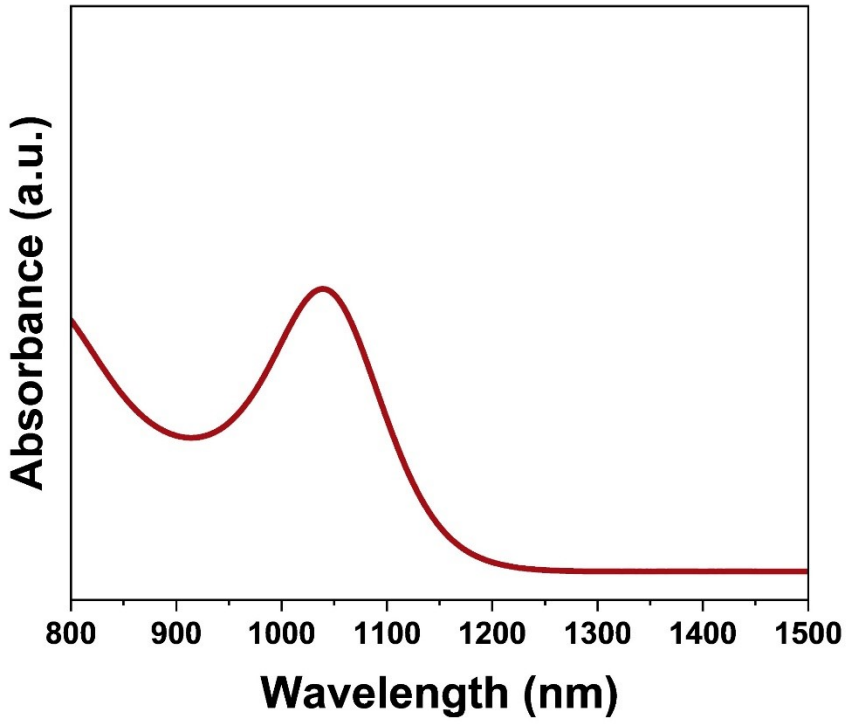


Figure S6. Absorption spectra of $PbSe_xS_{1-x}$ QDs (x=0.25) synthesized with DPP.

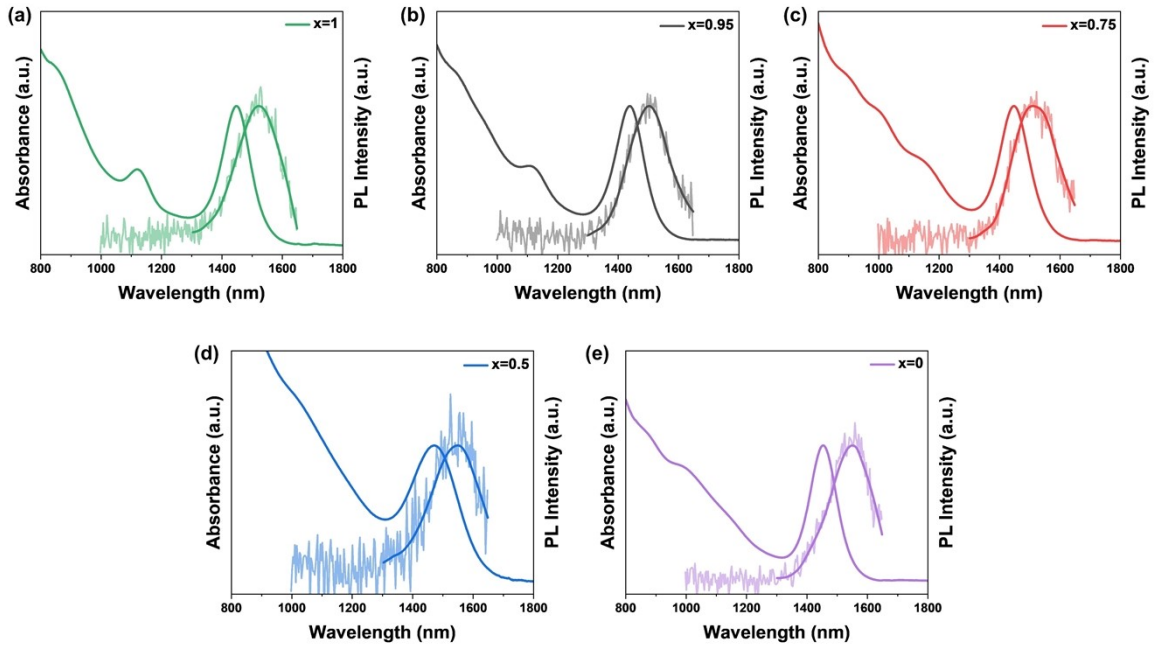


Figure S7. (a-e) Absorption spectra and PL spectra of PbSe, $PbSe_xS_{1-x}$, and PbS QDs QDs.

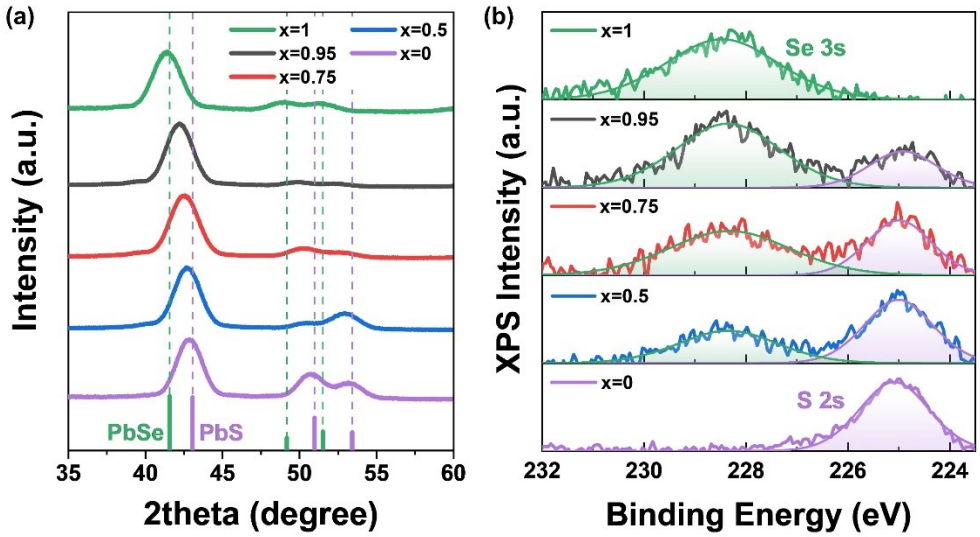


Figure S8. (a) XRD patterns of $PbSe_xS_{1-x}$ QDs compared to PbSe and PbS QDs. The green and purple dashed lines in (a) indicate the reference diffraction peaks for PbSe (PDF Card No.: 9000002) and PbS (PDF Card No.: 9008694), respectively. (b) XPS spectra of the corresponding samples, where the green and purple lines represent the deconvoluted core-level peaks of Se 3s and S 2s, respectively.

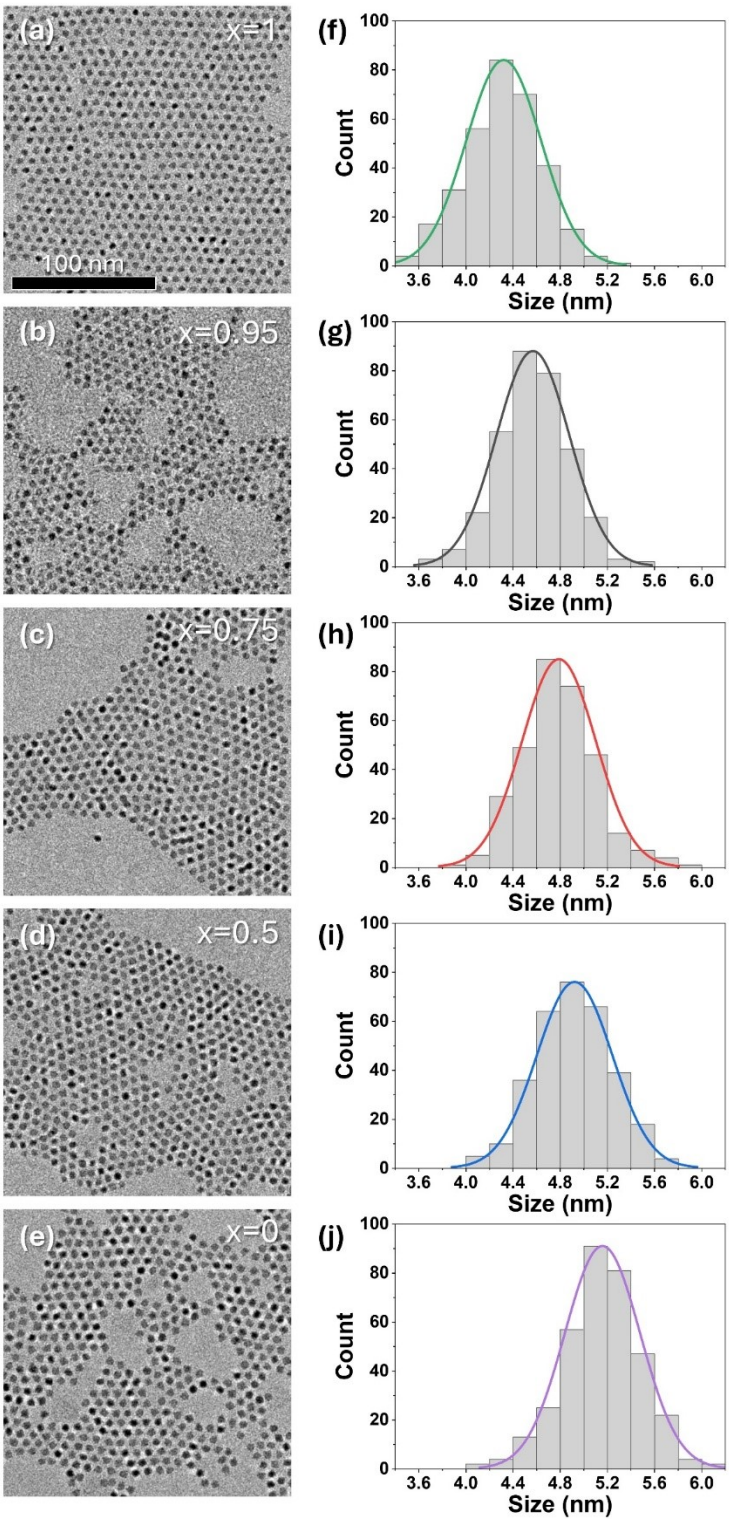


Figure S9. (a-e) Overview TEM images of $PbSe_xS_{1-x}$ QDs with different compositions and (f-g) their corresponding size distribution histograms.

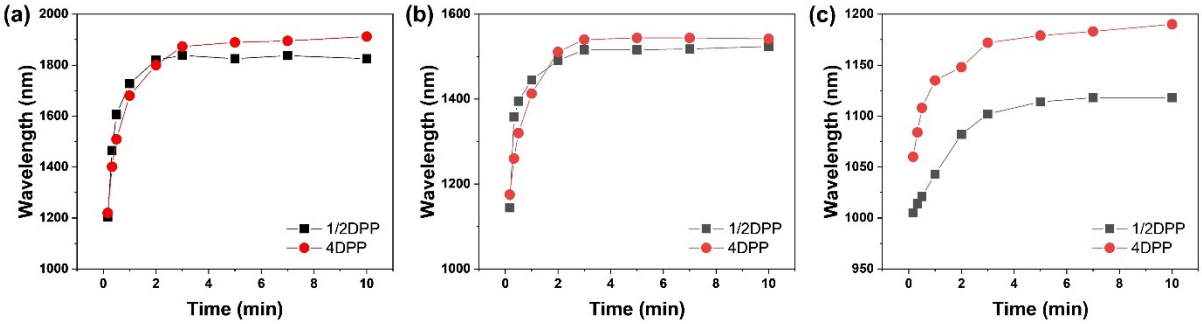


Figure S10. Time-dependent first excitonic absorption peak positions of $PbSe_xS_{1-x}$ QDs synthesized with different DPP amounts after a single injection. The DPP amount was varied to 1/2DPP and 4DPP relative to the original synthetic condition. The samples correspond to (a) x=0.95, (b) x=0.75, and (c) x=0.5, respectively.

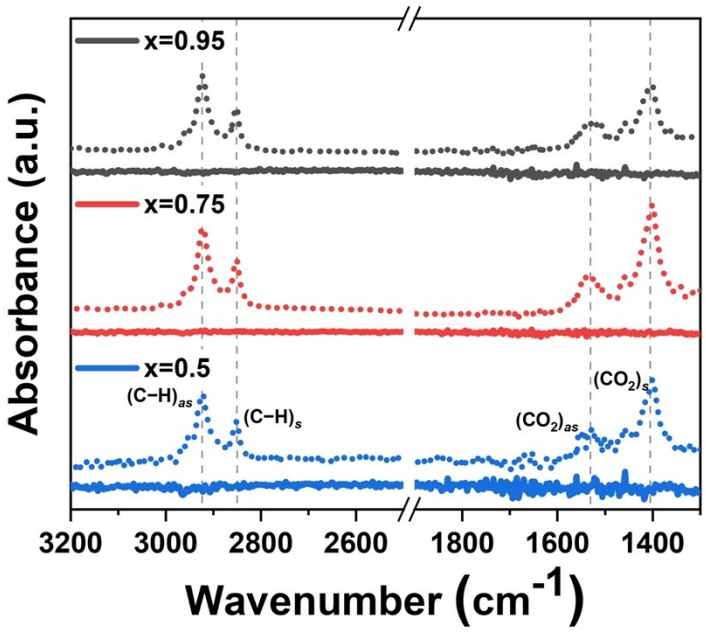


Figure S11. FTIR spectra of $PbSe_xS_{1-x}$ QDs with various compositions. Dashed lines represent $PbSe_xS_{1-x}$ QDs capped with OA ligands, while solid lines indicate with halide ligands. The disappearance of peaks corresponding to C-H and CO_2 bonding indicates a complete exchange from OA ligands to halide ligands.

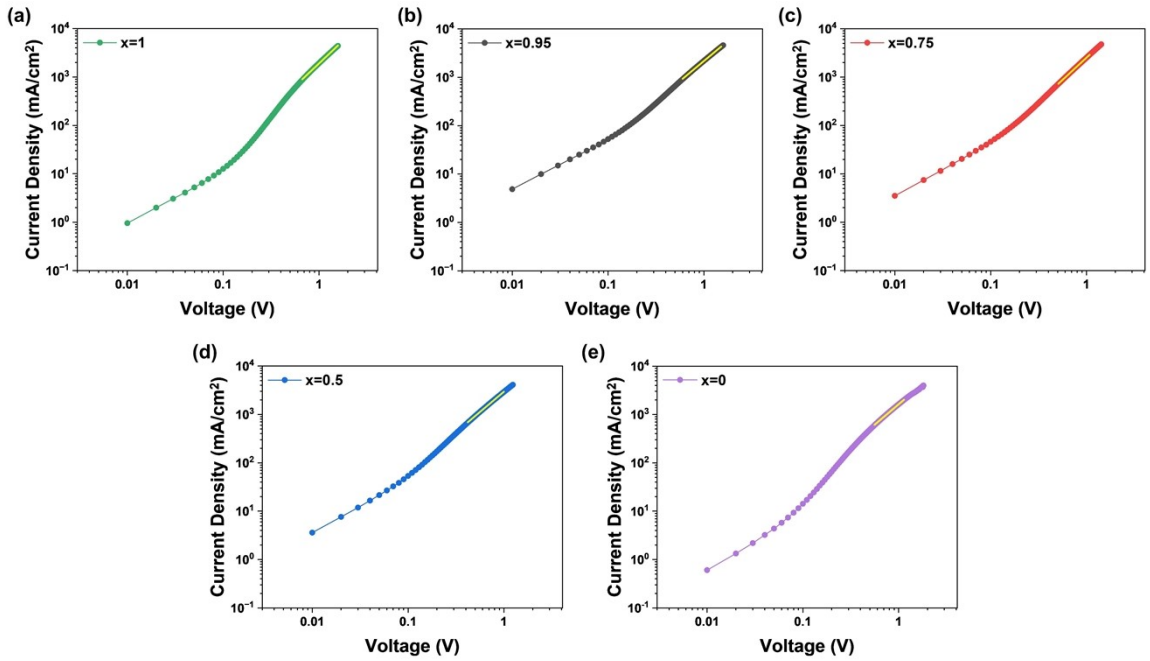
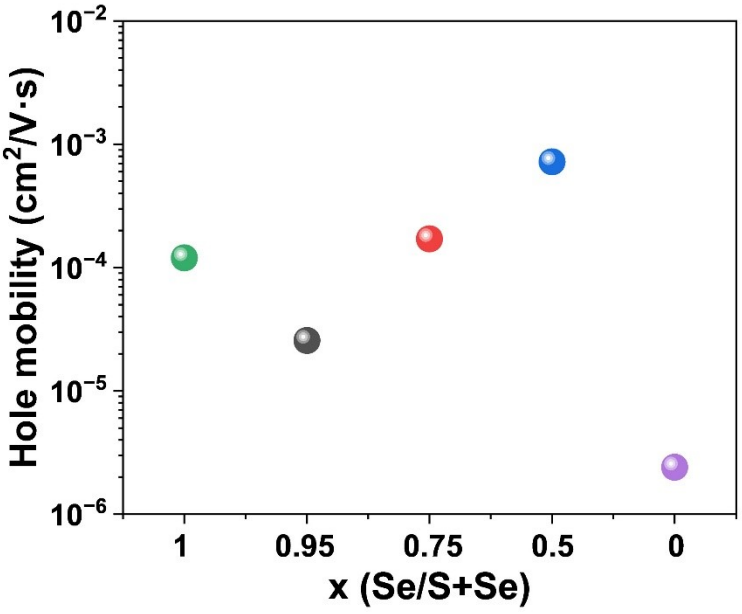


Figure S12. (a-e) Current density–voltage (J–V) characteristics of hole-only devices with the structure ITO/MoO_3/QD-I/MoO_3/Ag using PbSe, $PbSe_xS_{1-x}$, and PbS QDs. Measurements were performed under dark conditions and the yellow line represents the trap-free Child's region.

Figure S13. Hole mobility of PbSe, $PbSe_xS_{1-x}$, and PbS QDs extracted from FET measurements as a function of Se/(S+Se) composition.



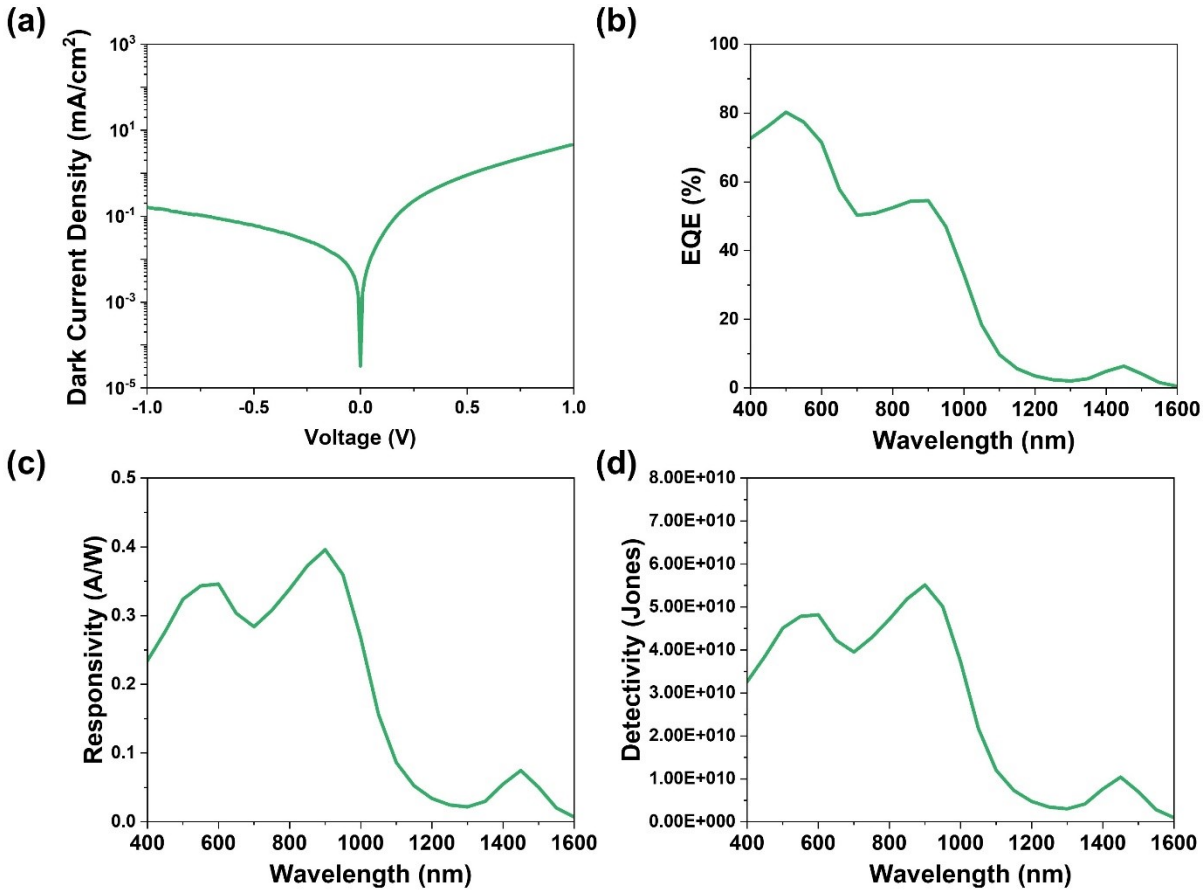


Figure S14. Photodetector characteristics of the representative $PbSe_xS_{1-x}$ QD device with x=0.5 fabricated with an ITO/ZnO/$PbSe_xS_{1-x}$ QD/PbS-EDT/Au/Ag structure. (a) Dark current density–voltage (J–V) characteristics measured under dark conditions. (b) External quantum efficiency (EQE) spectrum. (c) Spectral responsivity and (d) specific detectivity as a function of wavelength.

$PbSe_xS_{1-x}$	ε_r	μ_h ($cm^2/V \cdot s$)
x = 1	22.68	1.55×10^{-2}
x = 0.95	20.65	1.41×10^{-2}
x = 0.75	18.15	1.76×10^{-2}
x = 0.5	16.84	2.9×10^{-2}
x = 0	16.04	8.73×10^{-3}

Table S1. Dielectric constant (ε_r) and hole mobility (μ_h) for PbSe, $PbSe_xS_{1-x}$, and PbS QDs.

$PbSe_xS_{1-x}$	Pb:Chalcogen	Reaction time (s)	Optical density @400nm	Weight (mg)	Yield relative to Pb feed (%)
x = 1	2:1	30	0.48	41	5.35
x = 0.95	2:1	35	0.55	67.2	8.77
x = 0.75	2:1	90	1.2	164.1	21.42
x = 0.5	2:1.75	multi-injection	2.74	410.6	53.6

Table S2. Synthesis parameters, optical density at 400 nm, and the weight of QDs obtained