Supporting Information

Synergistic NH_4F and $ZnCl_2$-assisted surface engineering enables efficient red ZnSeTe quantum dots for QLEDs

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Experimental section

Materials

Zinc acetate ($Zn(OAc)_2$, 99.99%), sulfur powder (S, 99.999%), selenium powder (Se, 99.999%), trioctylphosphine (TOP, 90%), tellurium (Te, 99.999%), tri-n-octylamine (TOA, 97%), oleylamine (OLA, 80-90%), oleic acid (OA, 90%), 1-octadecene (ODE, 90%), n-hexane (AR), 1-dodecanethiol (98%), octane (99%), diphenylphosphine (DPP, 95%), and tri[2-(diphenylphosphino)ethyl]phosphine were purchased from Aladdin. Poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS), and poly(9,9-dioctylfluorene-co-N-(4-butylphenyl)diphenylamine) (TFB) were purchased from Aladdin Xi'an Polymer Light Technology. All of the above chemical raw materials were used without further purification.

Precursor Preparation

$Zn(OA)_2$ precursor: 56 mmol of $Zn(OAc)_2$ was dissolved in 80 mL of a solvent mixture containing 42 mL of OA, 19 mL of TOA, and 19 mL of TOP. The solution was then subjected to vacuum treatment at 120 °C for 1.5 h, followed by switching to a nitrogen atmosphere and heating to 300 °C for 1 h of reaction. Subsequently, a 0.7 M zinc precursor solution was obtained and cooled for later use.

$Zn(OLA)_2$ precursor: A mixture of 44 mmol of $Zn(OAc)_2$, 18 mL of OLA, and 90 mL of ODE was added to a 250 mL flask. The mixture was heated to 120 °C and degassed under reduced pressure for 1 h.

Te-TOP and Se-TOP: Appropriate amounts of Se and Te powders were weighed in a glovebox and mixed with TOP. The mixtures were stirred at room temperature until complete

dissolution, yielding TOP-Se (2 M) and TOP-Te (0.2 M) solutions. For another selenium precursor, an appropriate amount of Se powder was weighed, mixed with a suitable quantity of DPP, and stirred on a hotplate at about 120 °C for approximately 1 h until the Se powder dissolved, resulting in a DPP-Se (2 M) solution.

NH_4F precursor: The NH_4F precursor solution was prepared by dissolving 40 mmol of NH_4F in 20 mL of OLA. The mixture was heated to 120 °C under argon atmosphere and stirred until complete dissolution was achieved.

$ZnCl_2$ precursor: The $ZnCl_2$ precursor solution was prepared by dissolving 1 mmol of anhydrous $ZnCl_2$ in a mixture of 5 mL of OLA. The mixture was heated to 120 °C under argon atmosphere and stirred until complete dissolution was achieved.

Synthesis of ZnSeTe/ZnSeTe$_{inner}$/ZnSe/ZnS quantum dots

$Zn(OAc)_2$ (0.458 g, 2.5 mmol), oleic acid (OA, 2.5 mL), and 1-octadecene (ODE, 15 mL) were added to a 100 mL three-neck flask. The mixture was evacuated under vacuum at 120 °C for 30 min, then the temperature was raised to 240 °C under a nitrogen atmosphere. A rapid injection of 0.25 mL Se-DPP and 5 mL Te-TOP (0.2 M) was performed, followed by nucleation for 60 min. After nucleation, auxiliary additives (0.4 mL NH_4F (2 M) and/or 1 mL $ZnCl_2$ (0.5 M) were injected to obtain the corresponding samples QD-Pristine, QD-F, QD-Cl, and QD-F-Cl. The temperature was then raised to 260 °C, and 2 mL $Zn(OA)_2$ solution was injected. Subsequently, 0.6 mL Se-TOP (0.9 mL/h) and 1.2 mL Te-TOP (1.8 mL/h) were co-injected using a syringe pump. After the injection was complete, the reaction was maintained at 260 °C for 20 min. The temperature was further increased to 280 °C, and 12 mL $Zn(OA)_2$ solution was injected, followed by syringe-pump injection of 3 mL Se-TOP

(4.5 mL/h). The reaction was held at 280 °C for another 20 min. Next, the temperature was raised to 300 °C, and 4 mL $Zn(OA)_2$ solution was injected, after which 2 mL DDT was delivered via syringe pump (2 mL/h). The reaction was then cooled to 250 °C, and 4 mL $Zn(OLA)_2$ together with 4 mL octanethiol were injected and maintained for 60 min. After completion, the flask was cooled to room temperature. The crude quantum-dot solution was mixed with anhydrous ethanol in a 1:1 volume ratio, shaken thoroughly, and centrifuged at 4000 rpm for 4 min. The supernatant was discarded, and the precipitate was redispersed in 10 mL n-hexane for further characterization.

QLED fabrication

QLED devices were fabricated with the following structure: ITO/PEDOT:PSS/TFB/QD/ZnMgO/Al. Each layer except the Al cathode was deposited on ITO glass by spin-coating; the Al cathode was thermally evaporated under vacuum. The detailed fabrication procedure is as follows: ITO glass substrates were sequentially ultrasonically cleaned in deionized water, acetone, isopropanol, and ethanol for 15 min each. Subsequently, the cleaned ITO glass was treated in a UV-ozone cleaner for 15 min to enhance surface wettability and cleanliness. PEDOT:PSS was spin-coated onto the ITO glass at 3000 rpm for 40 s. The film was then annealed in air at 130 °C for 20 min. A TFB solution (8 mg/mL in chlorobenzene) was spin-coated at 3000 rpm for 40 s. The TFB layer was annealed at 130 °C for 20 min inside a glovebox. The QD solution (35 mg/mL in n-octane) was spin-coated at 3000 rpm for 40 s. The QD layer was annealed in the glovebox at 80 °C for 5 min. Then, a ZnMgO solution (25 mg/mL in ethanol) was spin-coated at 3000 rpm for 40 s and heated at 80 °C for 10 min inside the glovebox. Finally, the device was transferred to a

thermal evaporator. Under a vacuum of 5×10^{-4} Pa, an Al cathode was deposited at a rate of 2 nm/s.

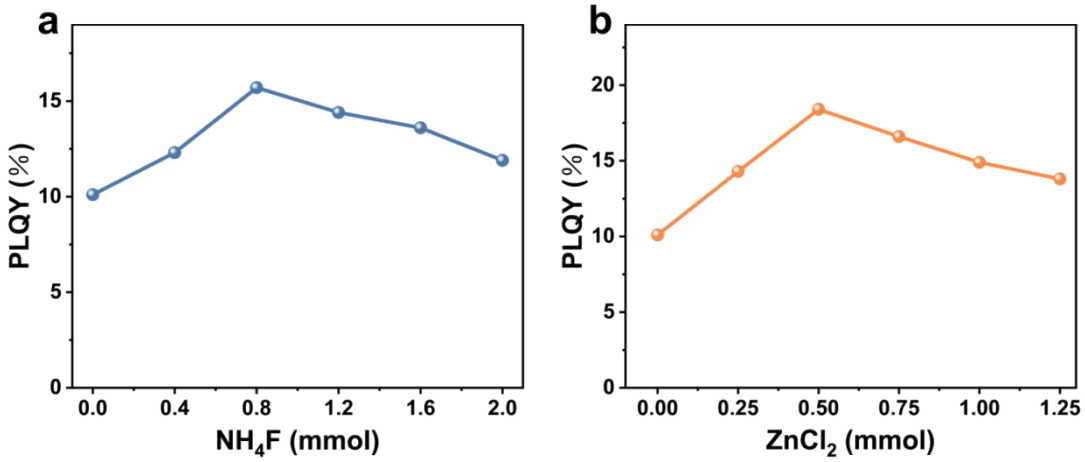

Fig. S1. PL QY of ZnSeTe/ZnSeTe$_{inner}$/ZnSe/ZnS core/shell QDs as a function of the amount of (a) NH_4F or (b) $ZnCl_2$ added during the core treatment step.

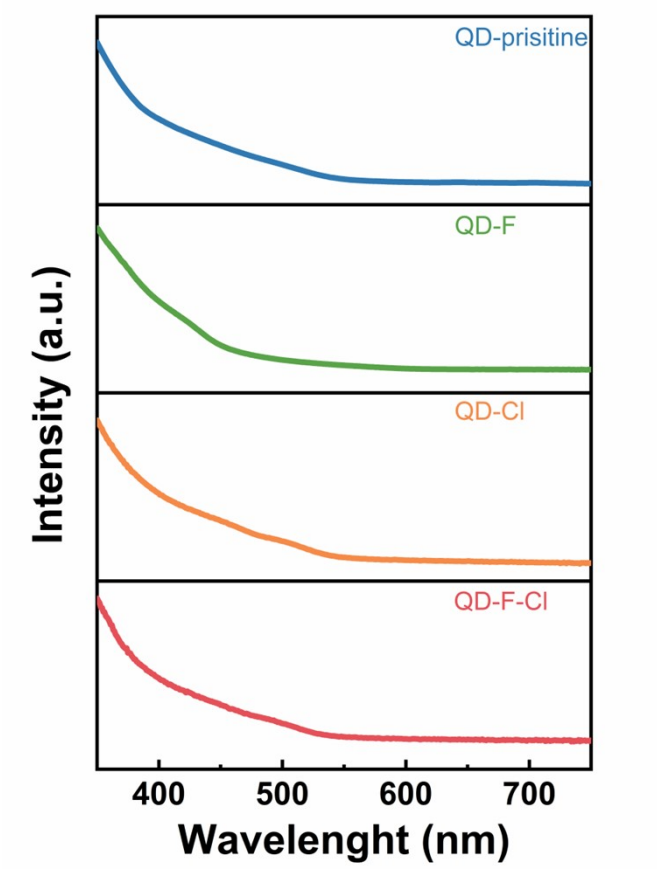

Fig. S2 | Absorption spectra of ZnSeTe QD cores for QD-pristine, QD-F, QD-Cl and QD-F-Cl.

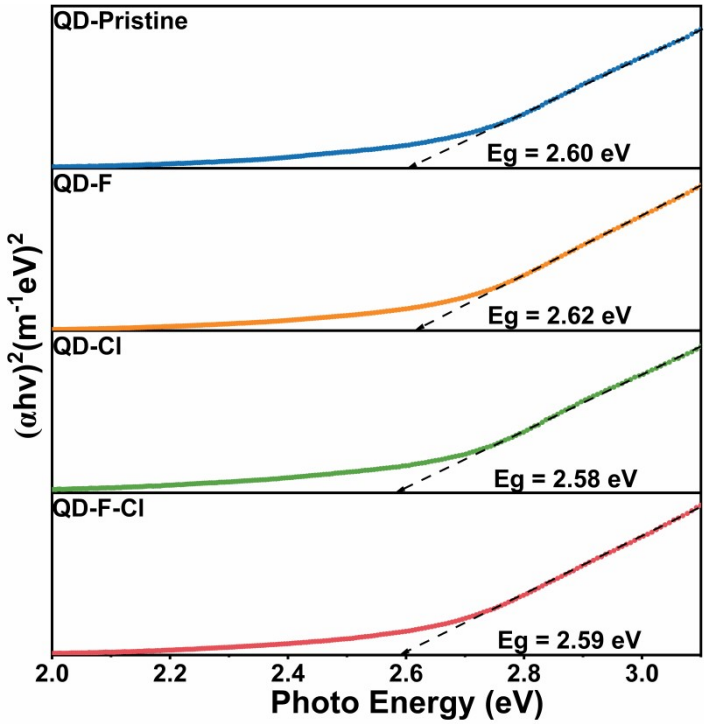

Fig. S3. Tauc plots of QD-Pristine, QD-F, QD-Cl, and QD-F-Cl.

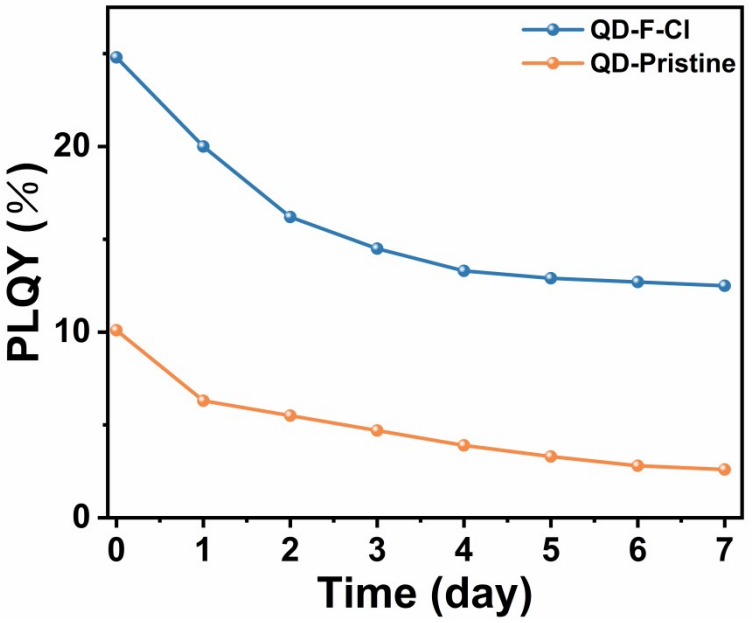

Fig. S4 | Stability test of QDs without passivation treatment (QD-Pristine) and with passivation treatment (QD-F-Cl).

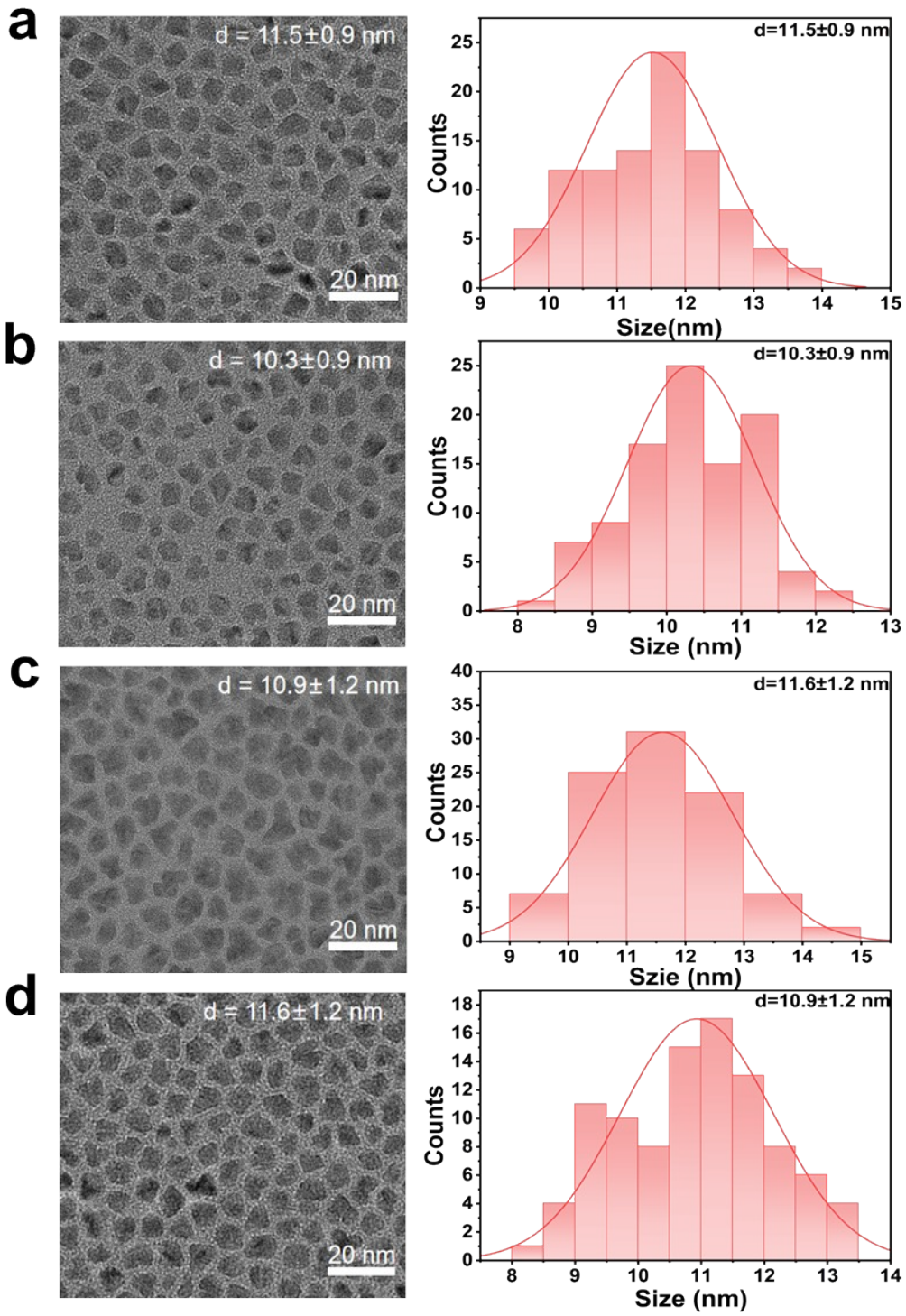

Fig. S5. TEM images and particle size distribution histograms of ZnSeTe QDs: (a) QD-Pristine, (b) QD-F, (c) QD-Cl, and (d) QD-F-Cl. Scale bars: 50 nm.

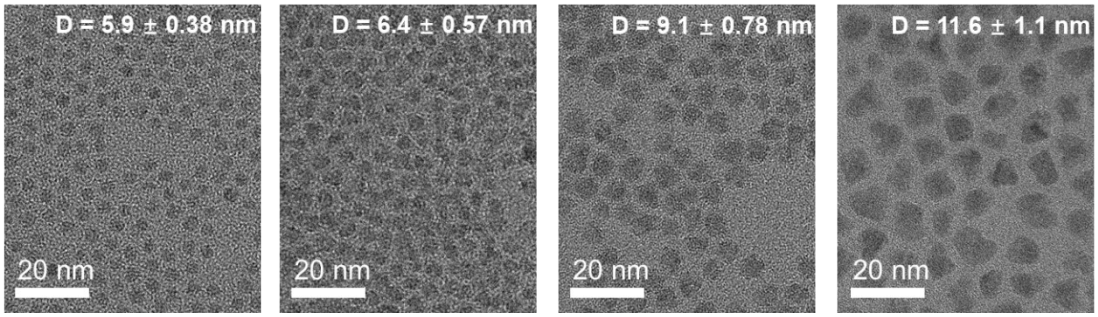

Fig. S6 | TEM images of QDs (from left to right): ZnSeTe, ZnSeTe/$ZnSeTe_{inner}$, ZnSeTe/$ZnSeTe_{inner}$/ZnSe, and ZnSeTe/$ZnSeTe_{inner}$/ZnSe/ZnS.

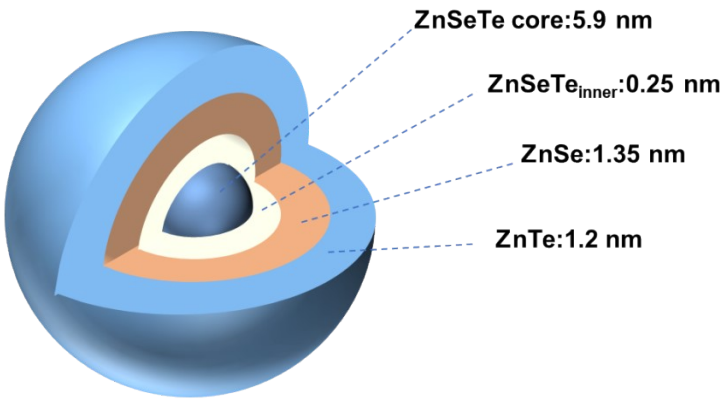

Fig. S7 | Thickness of each shell layer of ZnSeTe/ZnSeTe$_{inner}$/ZnSe/ZnS.

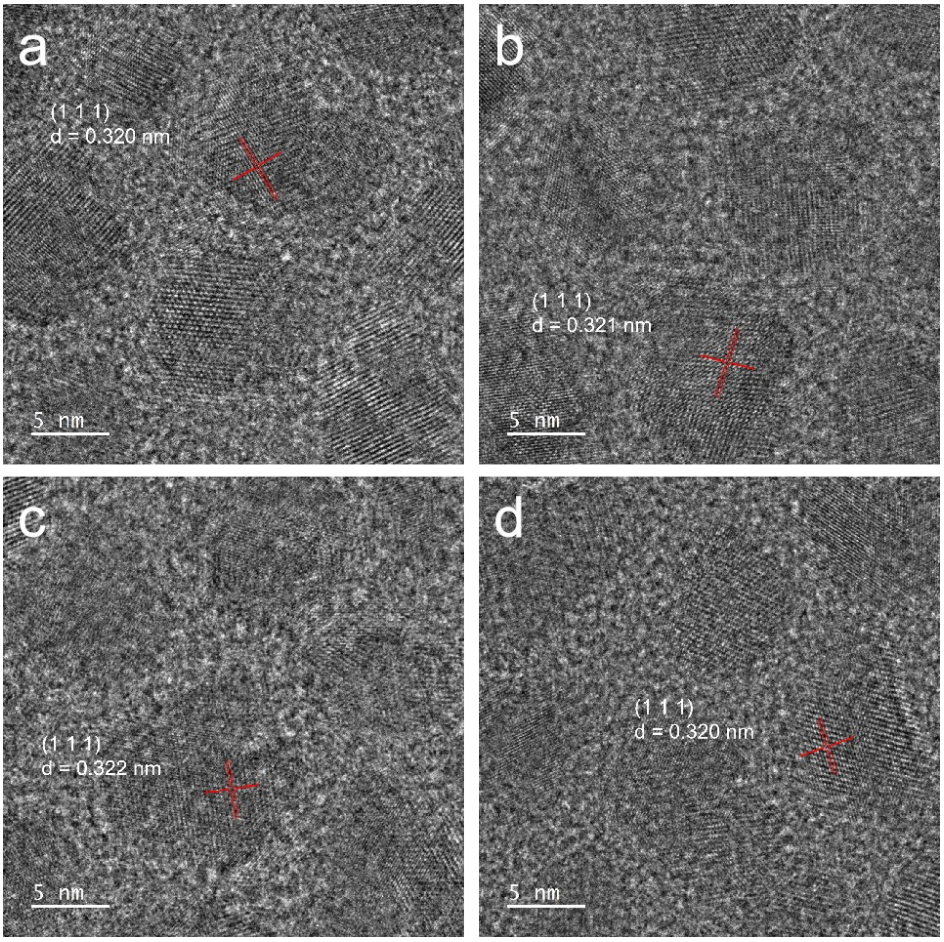

Fig. S8. High-resolution TEM images of ZnSeTe QDs: (a) QD-Pristine, (b) QD-F, (c) QD-Cl, and (d) QD-F-Cl. Scale bar: 5 nm.

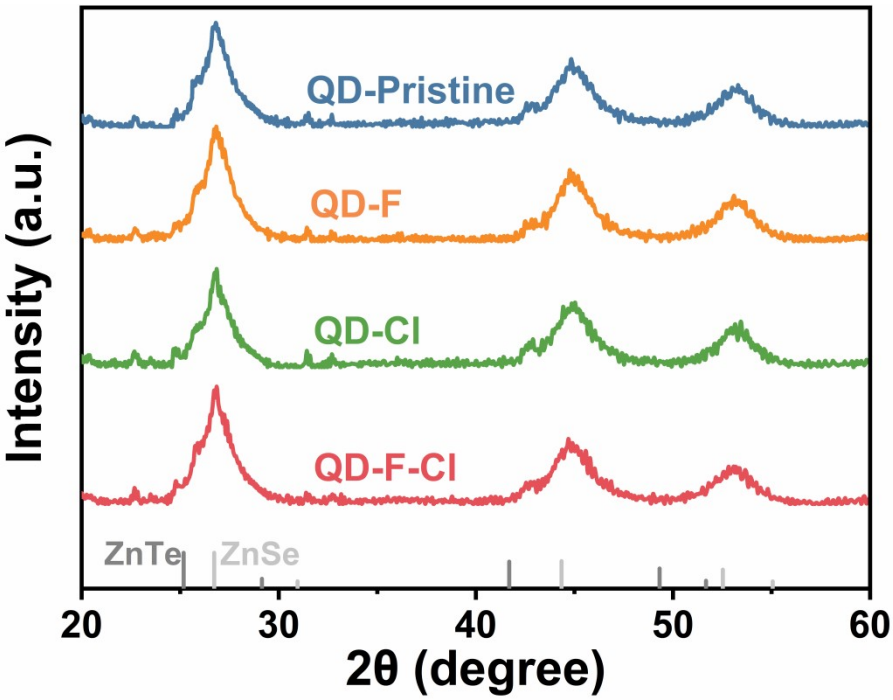

Fig. S9. XRD patterns of QD-Pristine, QD-F, QD-Cl, and QD-F-Cl.

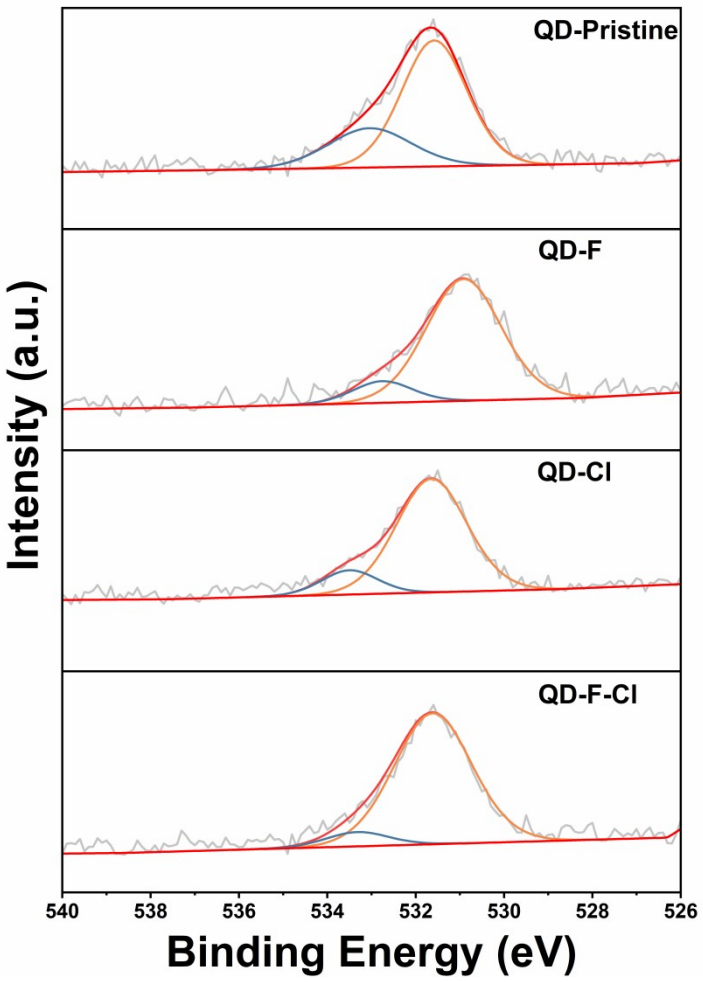

Fig. S10 | XPS spectra of O 1s for QDs treatment with different additives.

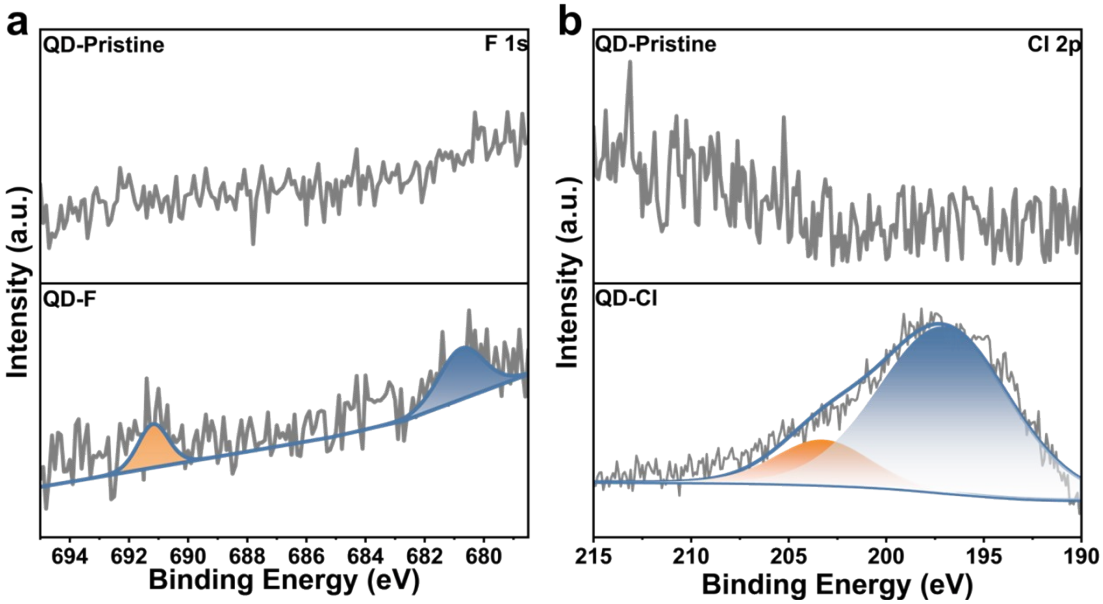

Fig. S11. (a) F 1s XPS spectra of QD-Pristine and QD-F; (b) Cl 2p XPS spectra of QD-Pristine and QD-Cl.

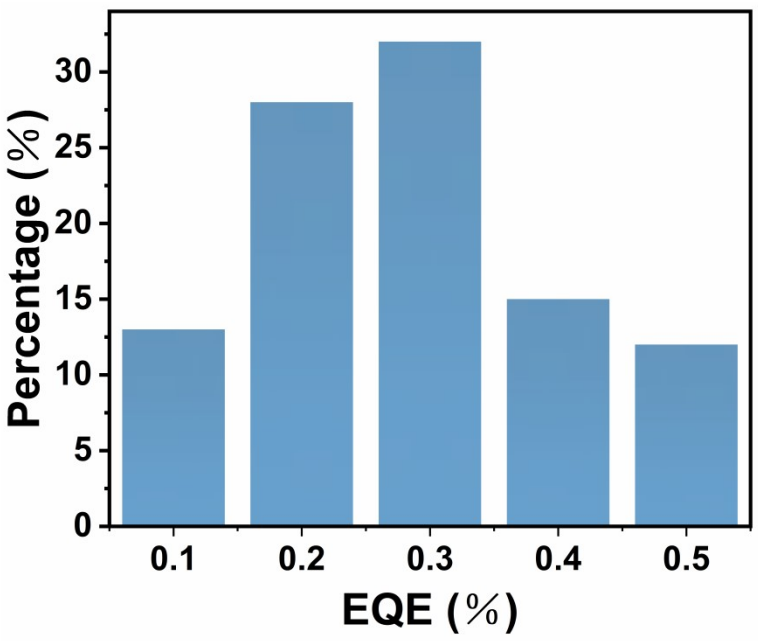

Fig. S12. Reproducibility of ZnSeTe/ZnSeTe$_{inner}$/ZnSe/ZnS based QLEDs.

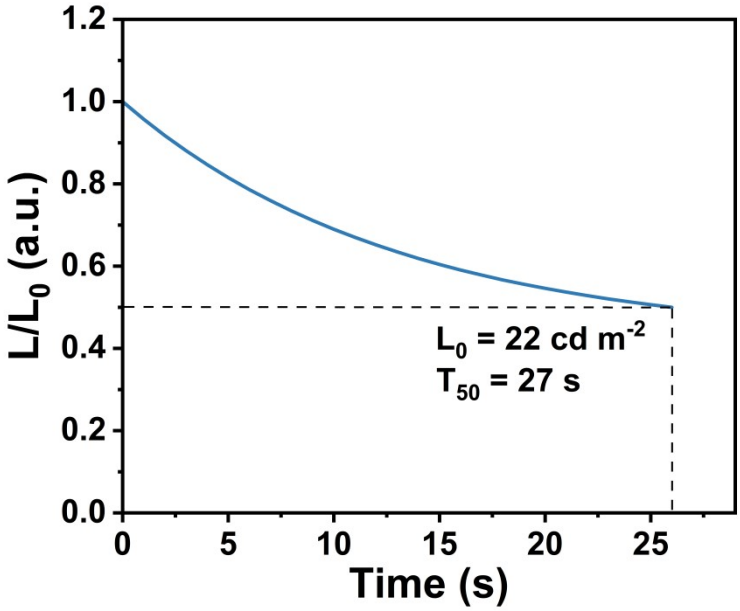

Fig. S13 | Operational stability of ZnSeTe/ZnSeTe$_{inner}$/ZnSe/ZnS based QLEDs.

Table S1. Parameters of TA lifetimes and decay components for QD-Pristine, QD-F, QD-Cl, and QD-F-Cl.

	A_1 (%)	τ_1 (ps)	A_2 (%)	τ_2 (ps)
QD-Pristine	61	77.6	38.6	764.3
QD-F	58.8	89.1	41.2	789.6
QD-Cl	57.7	96.4	42.3	835.5
QD-F-Cl	53.7	124.7	46.3	1123.9

Table S2. PL decay kinetics parameters for QD-Pristine, QD-F, QD-Cl, and QD-F-Cl.

	A_1 (%)	τ_1 (ns)	A_2 (%)	τ_2 (ns)	T_{av} (ns)
QD-Pristine	1	16.33	99	60.50	17.4
QD-F	3	28.06	97	122.73	40.1
QD-Cl	4	29.56	96	162.91	59.6
QD-F-Cl	10	35.41	90	165.95	73.7

The PL decay curves of ZnSeTe QDs were fitted using a double-exponential function:

$$I(t) = A_1 \times exp\left(-\frac{i}{\tau_1}\right) + A_2 \times exp\left(-\frac{i}{\tau_2}\right) \#(1)$$

Table S3. Radiative and non-radiative recombination rate parameters for QD-Pristine, QD-F, QD-Cl, and QD-F-Cl.

	PLQY (%)	T_{av} (ns)	K_{nr} (ns^{-1})
QD-Pristine	10.1	17.4	0.0516
QD-F	15.7	40.1	0.0210
QD-Cl	18.4	59.6	0.0136
QD-F-Cl	24.8	73.7	0.0102

The nonradiative recombination rates (k_{nr}) were calculated by the following relations:

$$PLQY = \frac{k_r}{k_r + k_{nr}} \#(2)$$

$$k_r + k_{nr} = \frac{1}{\tau_{av}} \#(3)$$

Table S4. Performance comparison of red-emitting ZnSeTe QDs and QLEDs.

Structure	PL Wavelength (nm)	PL QY (%)	EQE (%)	Ref
ZnSeTe/ZnSeTe$_{inner}$/ZnSe/ZnS	612	63	/	(1)
ZnSeTe/ZnSeTe$_{inner}$/ZnSe/ZnS	615	24.8	0.3	**This work**
ZnSeTe/ZnTe/ZnSe/ZnS	618	12	/	(2)

References

1. Y.-H. Kim, S.-Y. Yoon, Y.-J. Lee, D.-Y. Jo, H.-M. Kim, Y. Kim, S. M. Park, G. M. Park, Y. Kim, J. Kim and H. Yang, *ACS Appl. Nano Mater.*, 2023, **6**, 19947-19954.
2. X.Song, Y.Qin, C.Yu, J.Ning, J. Phys. Chem. Lett., 2025, 16(4): 1045-1050.