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

Supplementary information

Acid-Stimulated Bioassembly of High-Performance Quantum Dots in *Escherichia coli*

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This supporting information contains 16-page document, including 4 tables, 10 figures, references and this cover page.

Table S1. Emission lifetime measurements of freshly isolated Bio-QDs. Each curve was fitted with a three components exponential-decay function. τ_i are the lifetimes of each decay component and A_i their respective fractional intensity. χ^2 is the reduced chi-squared statistic. Average lifetimes were calculated using equation: $\tau=\sum A_i \times \tau_i^2 / \sum A_i \times \tau_i$.

Samples	τ_1 (ns)	τ_2 (ns)	τ_3 (ns)	A_1 (%)	A_2 (%)	A_3 (%)	Average lifetime (ns)	χ^2
pH 4.5 - 5 min	119±1.49	19±0.83	3.1±0.04	50±0.01	25±0.02	25±0.07	110	1.5
pH 4.5 - 2 h	130±0.62	31±0.23	3.6±0.05	61±0.01	29±0.03	10±0.1	119	2.0
pH 4.5 - 4.5 h	124±0.53	27±0.46	3.2±0.04	62±0.01	28±0.04	10±0.14	115	2.3
pH 4.5 - 7.5 h	125±0.56	27±0.48	3.4±0.04	62±0.01	28±0.04	10±0.13	116	2.4
pH 4.5 - 10.5 h	133±0.51	32±0.02	3.7±0.05	62±0.01	30±0.03	8±0.12	122	2.2
pH 3.5 - 4.5 h	41±1.39	0.91±0.06	0.53±0.06	19±0.01	19±0.1	62±0.03	38	1.4
pH 5.5 - 4.5 h	116±1.28	15.9±0.66	2.9±0.04	53±0.01	25±0.01	22±0.05	109	1.8
pH 6.5 - 4.5 h	109±1.46	12.3±0.67	2.8±0.04	49±0.01	28±0.02	23±0.05	102	1.5
pH 7.5 - 4.5 h	80±1.36	8.8±0.03	1.8±0.04	43±0.01	36±0.04	21±0.14	73	1.7
pH 8.5 - 4.5 h	89±2.08	9.0±0.40	2.2±0.04	34±0.01	41±0.02	25±0.06	79	1.4
pH 9.5 - 4.5 h	62±2.62	7.1±0.24	1.6±0.03	17±0.01	56±0.02	27±0.07	45	1.3
pH 10.5 - 4.5 h	41±1.11	5.8±0.11	1.14±0.02	16±0.01	60±0.07	24±0.26	28	1.5

Table S2. *Synthesis* rates and optical properties of Bio-QDs reported in literature and obtained in this study.

Organism	Material	Optical properties	Cell growth	Seleniumized cells	QDs growth	Ref.
Engineered *E. coli*	CdS	QY: 0.007 %	-	1 h 37 °C	3 h 37 °C	[1]
Rhodopseudomonas palustris	CdS	-	42 h 30 °C	-	72 h 30 °C	[2]
Stenotrophomonas maltophilia	CdS	QY: 0.3-2.08 %	12 h 37 °C	2 h 37 °C	5 h 37 °C - yellow	[3]
Engineered *E. coli*	CdSe/CdTe	-	-	-	6-12 h 37 °C	[4]
Engineered *E. coli*	CdTe	-	-	-	24 h 37 °C	[5]
E. coli	CdSe	-	10-14 h 37 °C	2 h 37 °C	32 h - yellow	[6]
S. cerevisiae	CdSe	-	24 h 30 °C	24 h 30 °C	24 h 30 °C - yellow	[7]
Engineered *S. cerevisiae*	CdSe	QY: 4.7%	24 h 30 °C	24 h 30 °C	24 h 30 °C	[8]
Helminthosporum solani	CdSe	QY: 1 %	96 h 37 °C	-	96 h 37 °C	[9]
Earthworm	CdTe	QY: 8.3% Lifetime: 4.54 ns	-	-	11 days 20 °C	[10]
Biomimetic synthesis	CdS	QY:15.8% (green), 14.1% (red)	-	-	Within days (37 °C); Within hours (80 °C)	[11]
E. coli	CdS_xSe_{1-x}	QY: 7.3% Lifetime: 133 ns	10 h 37 °C	5 min 37 °C	Yellow: within 0.5 h 37 °C	This work

Table S3. Se-*K* XANES edge fitted results of samples from different pH groups

Sample	CdSe	Se	Na_2SeO_3	SeMet	$SeCys_2$
pH 3.5	0	1	0	0	0
pH 4.5	0.567	0.16	0.024	0.0169	0.081
pH 5.5	0.189	0.127	0.16	0.528	0
pH 6.5	0	0	0.543	0.476	0
pH 7.5	0	0	0.874	0.143	0
pH 8.5	0	0	0.953	0.048	0
pH 9.5	0	0	0.954	0.064	0
pH 10.5	0	0	0.939	0.07	0

Table S4. Se *K*-edge EXAFS curve fitting parameters [a].

Sample	Shell	*N*	*R* (Å)	σ^2 (Å^2)	ΔE_0 (eV)	*R* (%)
Se[b]	Se-Se	**2**	2.38 (1)	0.004 (1)	6.9 (4)	0.1
CdSe[c]	Se-Cd	**4**	2.62 (1)	0.005 (1)	2.8 (4)	0.1
Na_2SeO_3[d]	Se-O	**3**	1.70 (1)	0.002 (3)	8.9 (5)	0.7
SeMet[e]	Se-C	**6**	1.96 (1)	0.002 (4)	8.7 (7)	0.9
pH 3.5[f]	Se-Se	2.6 (2)	2.35 (1)	0.004 (1)	7.3 (8)	0.1
pH 4.5[g]	Se-Se	1.0 (3)	2.33 (1)	0.004 (2)	3.2 (8)	0.1
	Se-Cd	2.2 (2)	2.59 (1)	0.006 (1)		
	Se-C	0.2 (2)	1.99 (1)	0.004 (4)		
	Se-O	0.2 (1)	1.49 (4)	0.007 (4)		
pH 5.5[h]	Se-Se	1.3 (3)	2.33 (1)	0.004 (1)	17.5 (8)	0.1
	Se-Cd	0.5 (3)	2.63 (5)	0.009 (1)		
	Se-C	1.5 (2)	1.97 (7)	0.002 (4)		
	Se-O	1.1 (1)	1.70 (1)	0.003 (1)		
pH 6.5[i]	Se-Se	0.7 (5)	2.38 (4)	0.002 (2)	13.8 (2)	0.9
	Se-Cd	0.3 (3)	2.78 (5)	0.003 (1)		
	Se-C	1.0 (6)	1.97 (5)	0.003 (1)		
	Se-O	2.1 (4)	1.70 (4)	0.003 (1)		
pH 7.5[j]	Se-C	0.5 (4)	2.15 (6)	0.002 (1)	10.0 (2)	0.7
	Se-O	2.8 (2)	1.70 (1)	0.002 (1)		
pH 8.5[k]	Se-O	3.1 (1)	1.71 (2)	0.002 (1)	12.4 (4)	0.7
pH 9.5[l]	Se-O	3.2 (3)	1.71 (2)	0.002 (1)	12.6 (4)	0.6
pH 10.5[m]	Se-O	3.2 (3)	1.71 (2)	0.002 (1)	13.3 (4)	0.6

[a] *N*, coordination number; *R*, distance between absorber and backscatter atoms; σ^2, Debye–Waller factor to account for both thermal and structural disorders; ΔE_0, inner potential correction; *R* factor (%) indicates the goodness of the fit. Errors are given in brackets. S_0^2 was fixed to 0.8 as determined from CdSe standard fitting. Bold numbers indicate fixed coordination number (*N*) according to the crystal structure. [b] Fitting range: $3 \le k$ (/Å) ≤ 14.2 and $1.7 \le R$ (Å) ≤ 2.5. [c] Fitting range: $3 \le k$ (/Å) ≤ 12.5 and Fitting range: $1.7 \le R$ (Å) ≤ 2.8. [d] Fitting range: $4.0 \le k$ (/Å) ≤ 13.3 and $1 \le R$ (Å) ≤ 1.7. [e] Fitting range: $3.5 \le k$ (/Å) ≤ 13.6 and $1.3 \le R$ (Å) ≤ 1.9. [f] Fitting range: $4.5 \le k$ (/Å) ≤ 13.5 and $1.3 \le R$ (Å) ≤ 2.5. [g] Fitting range: $2.5 \le k$ (/Å) ≤ 11.7 and $1.0 \le R$ (Å) ≤ 3.1. [h] Fitting range: $3.5 \le k$ (/Å) ≤ 12.0 and $1.0 \le R$ (Å) ≤ 2.7. [i] Fitting range: $2.8 \le k$ (/Å) ≤ 10.8 and $1.0 \le R$ (Å) ≤ 2.7. [j] Fitting range: $3.6 \le k$ (/Å) ≤ 12.2 and $1.0 \le R$ (Å) ≤ 2.2. [k] Fitting range: $3.6 \le k$ (/Å) ≤ 12.2 and $1.0 \le R$ (Å) ≤ 2.5. [l] Fitting range: $4.1 \le k$ (/Å) ≤ 12.2 and $1.0 \le R$ (Å) ≤ 1.7. [m] Fitting range: $4.1 \le k$ (/Å) ≤ 12.2 and $1.0 \le R$ (Å) ≤ 1.7.

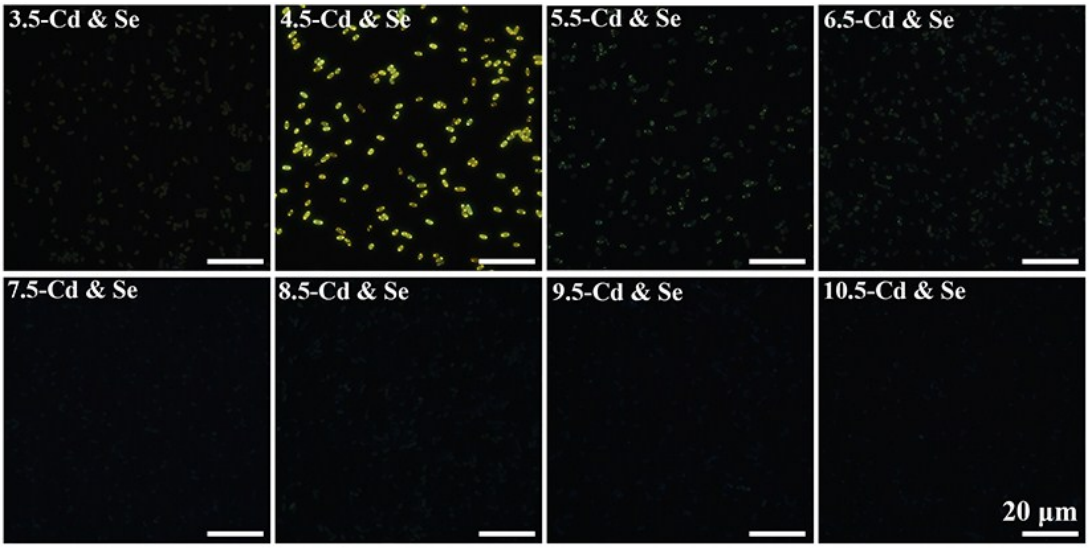

Fig. S1. Fluorescence images of the *E. coli* cells cultivated at different pHs. Only the Cd & Se group cells at pH 4.5 were lightened with bright yellow fluorescence.

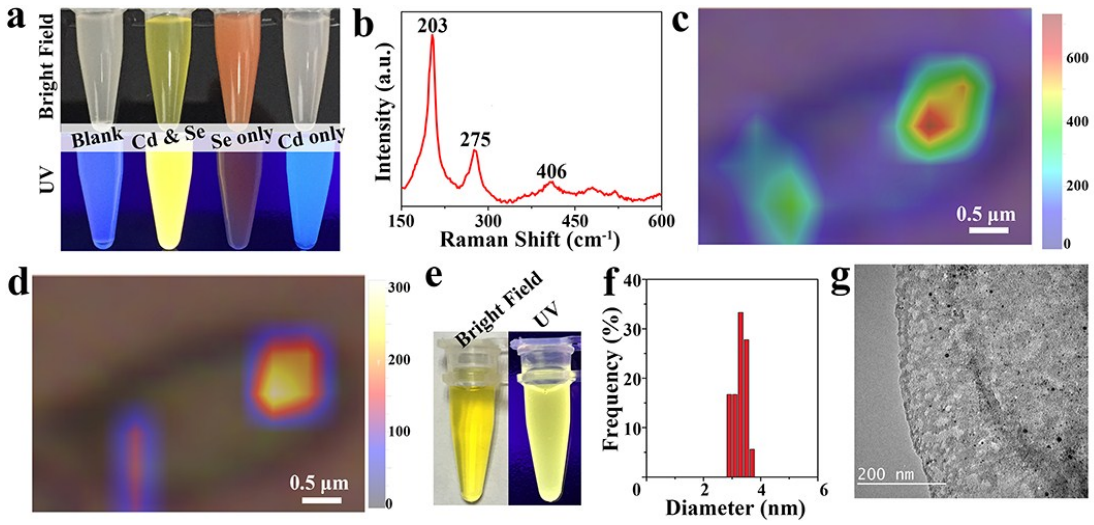

Fig. S2. Characteristics of the Bio-QDs synthesized at pH 4.5. (a) Images of *E. coli* cells cultivated at pH 4.5 under bright field light and UV light. Only the Cd & Se group cells showed bright yellow fluorescence. (b) The in-situ Raman spectrum of *E. coli*. Combined image of bright-field microscopy image with Raman mapping (c) at 203 cm^{-1} the characteristic peak of Cd-Se and (d) at 275 cm^{-1} the characteristic signal of Cd-S, respectively. (e) Bright field and UV irradiation images of the purified Bio-QDs. The extracted fluorophore from the cells still kept strong yellow fluorescence. (f) Size distribution and (g) surface chemical property of the Bio-QDs recorded by TEM image.

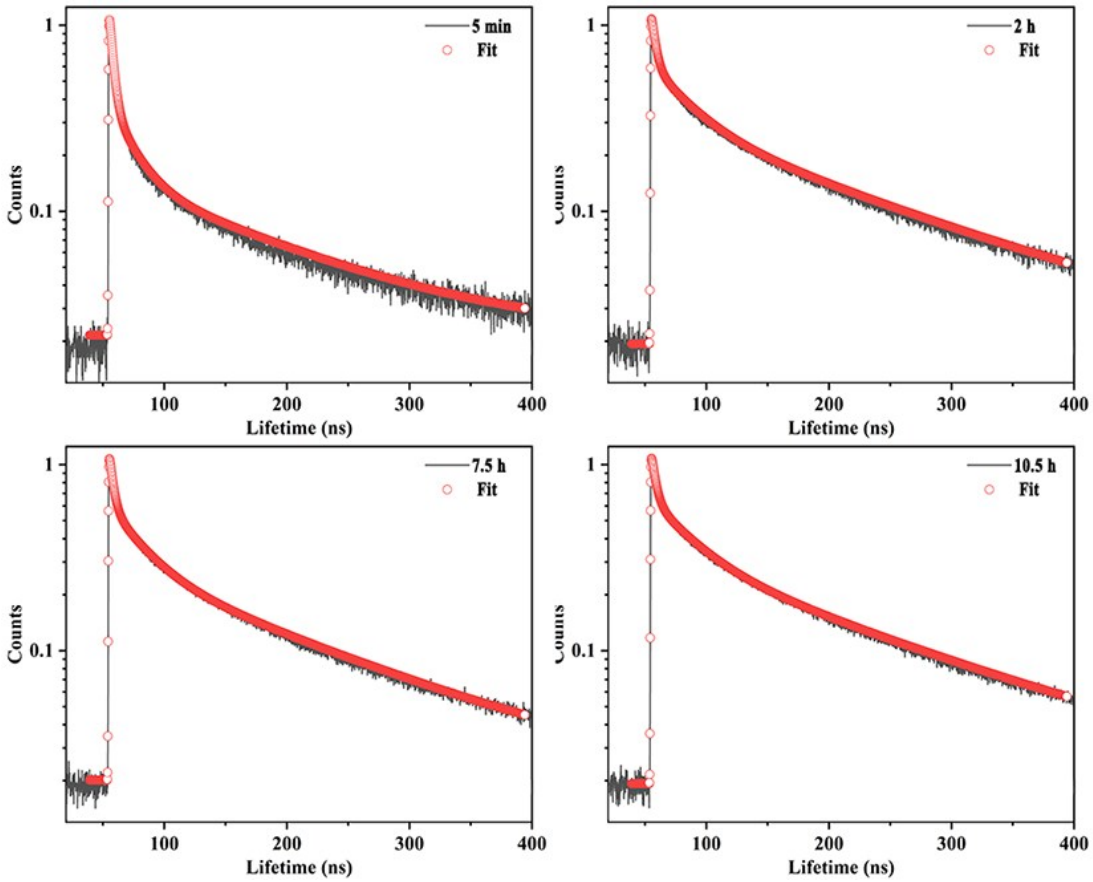

Fig. S3. Fitted results of the normalized emission lifetime of the Bio-QDs synthesized in pH 4.5 group. The fitted signal (red circle) superimposed on the experimental one (black line). Measured spectra matched well with the calculated results. The best-fit parameters are shown in Table S1.

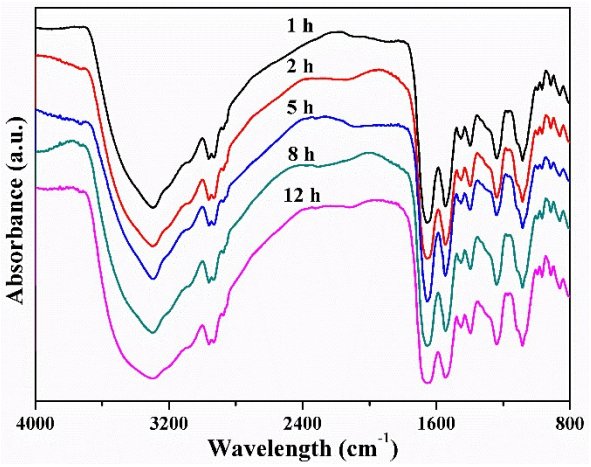

Fig. S4. Time-resloved FTIR spectrum of the synthesized Bio-QDs.

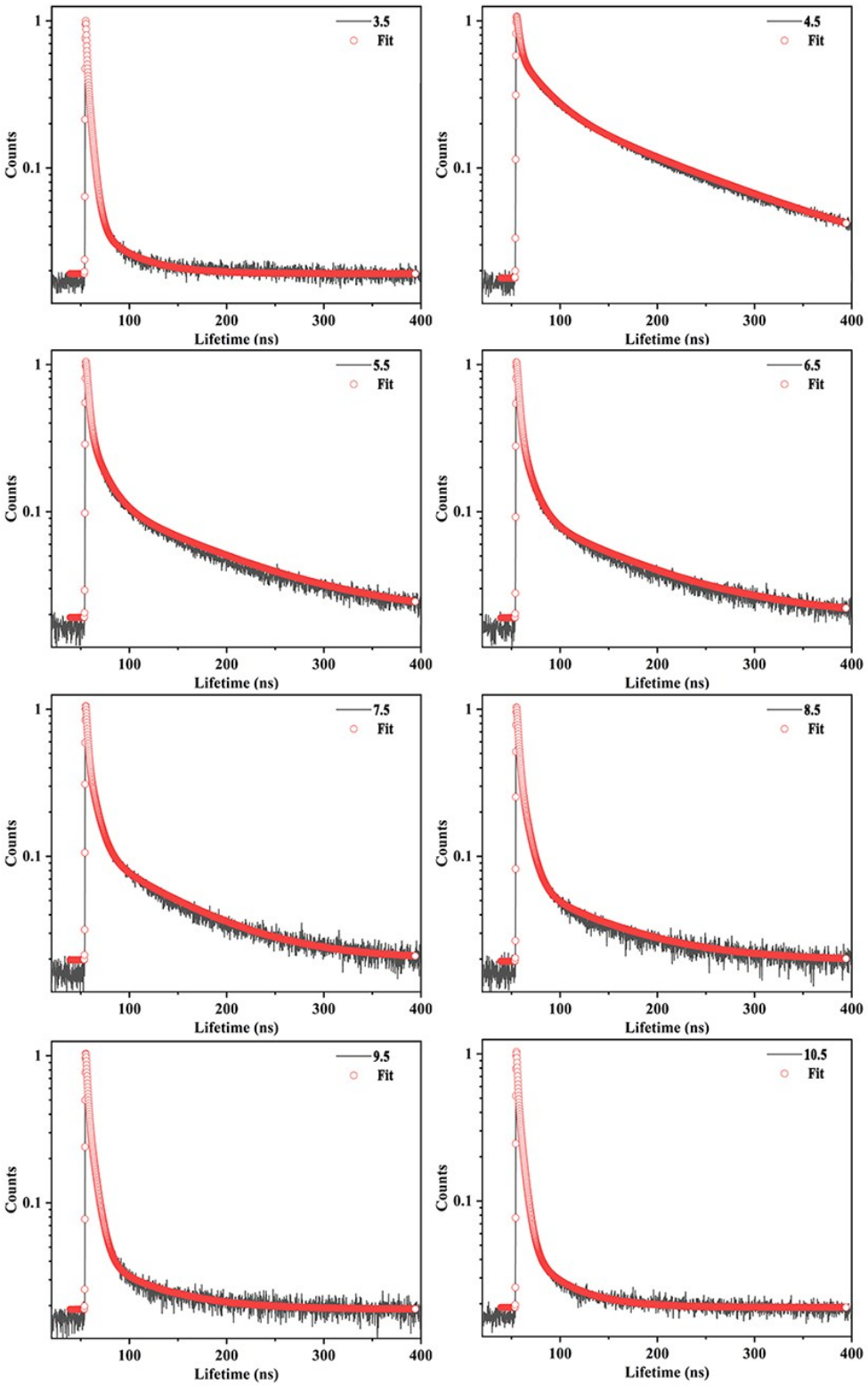

Fig. S5. Fitted results of the normalized emission lifetime of the Bio-QDs synthesized in different pH. The fitted signal (red circle) superimposed on the experimental one (black line). Measured spectra matched well with the calculated results. The best-fit parameters are shown in Table S1.

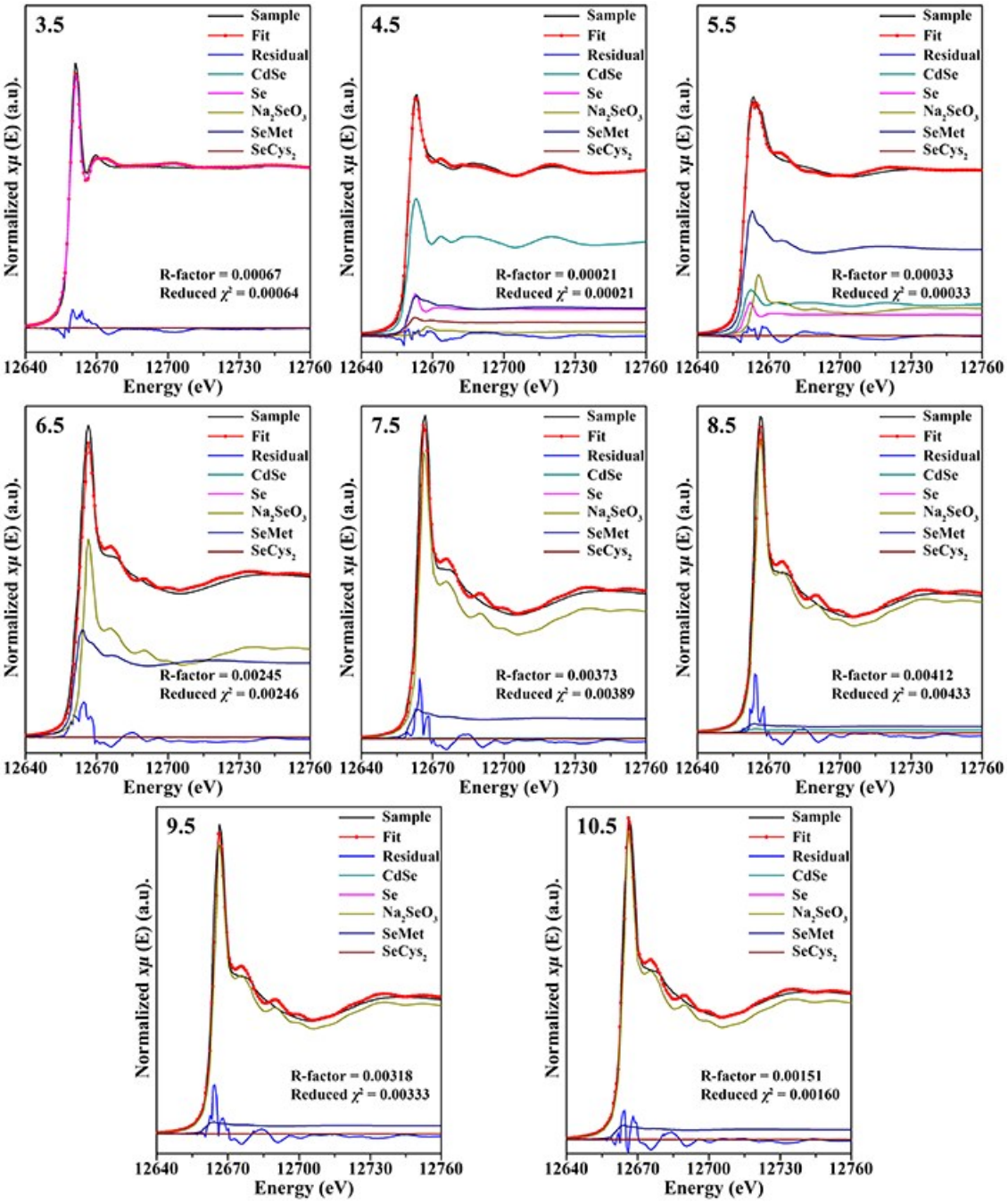

Fig. S6. LCF of the samples with standards based on Se *K*-edge XANES spectra.

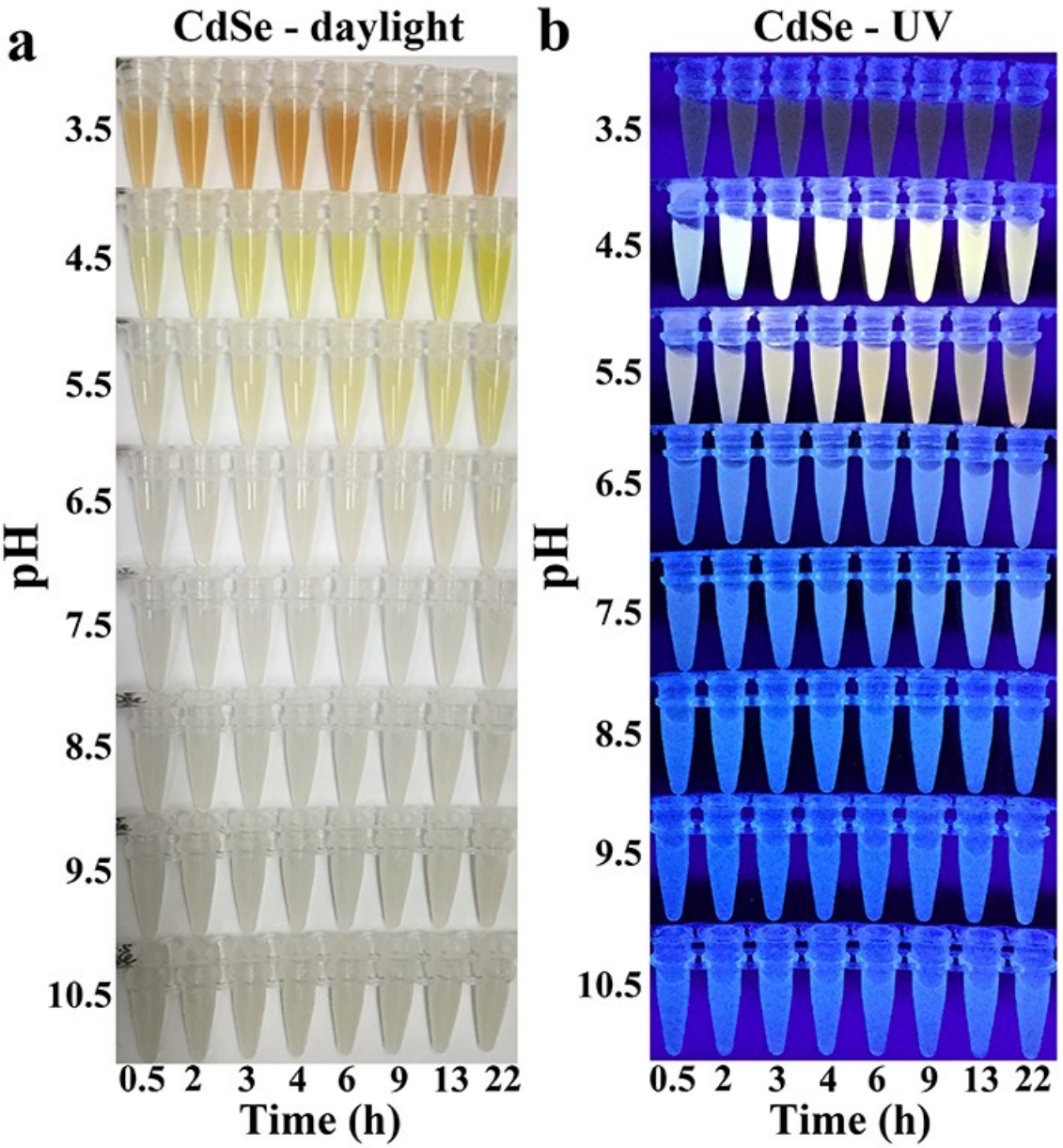

Fig. S7. Images of *E. coli* cells suspension collected at different incubation times under (a) bright field and (b) UV light. Cells were cultivated with 1mM Na_2SeO_3 and 6 mM $CdCl_2$ co-exposure at different pHs.

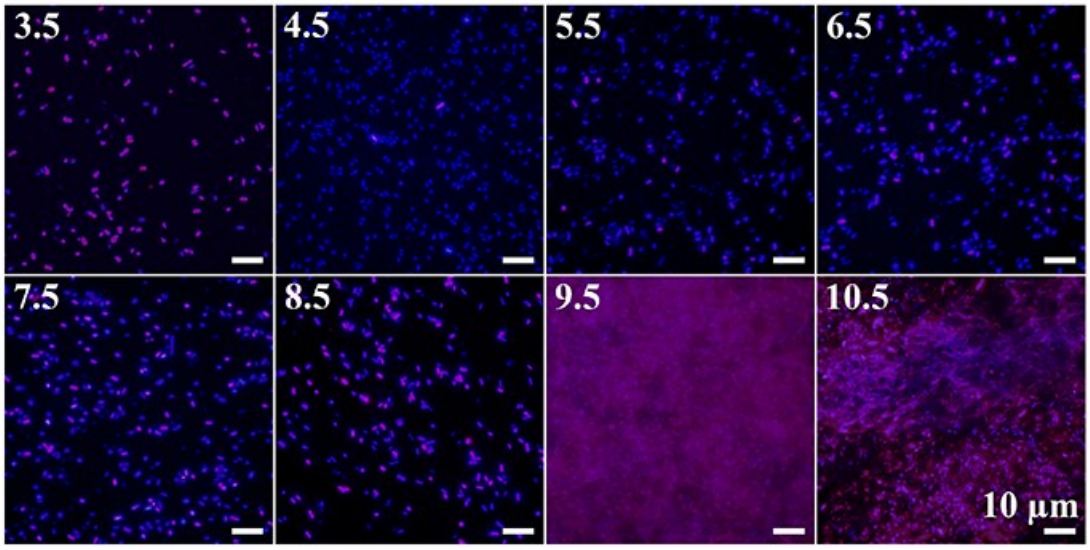

Fig. S8. Fluorescence images of cells after Se and Cd treatment for 3.5 h at different pHs. The blue signals represent the alive cells and the purple red signals represent the dead cells.

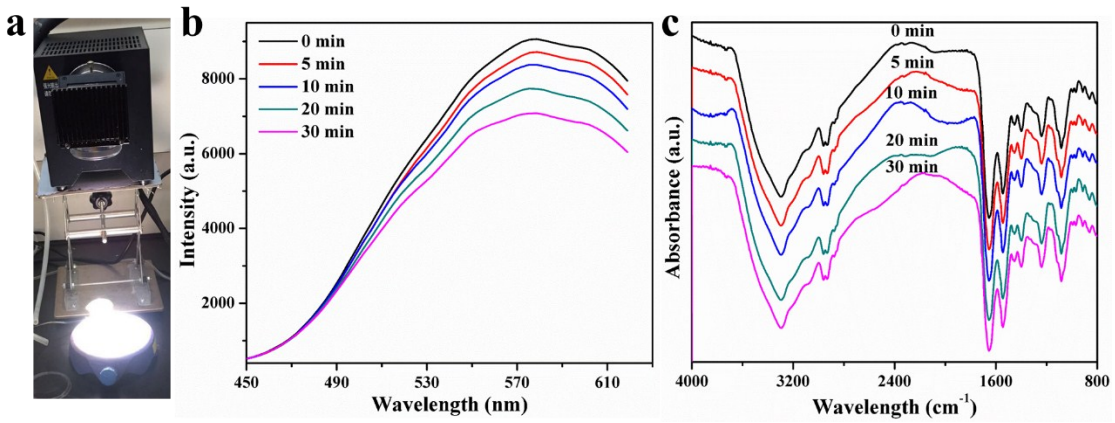

Fig. S9. (a) Photo of Bio-QDs under continuous illumination. Evolution in (b) fluorescence emission spectra and (c) FTIR spectra of the synthesized Bio-QDs with illumination time.

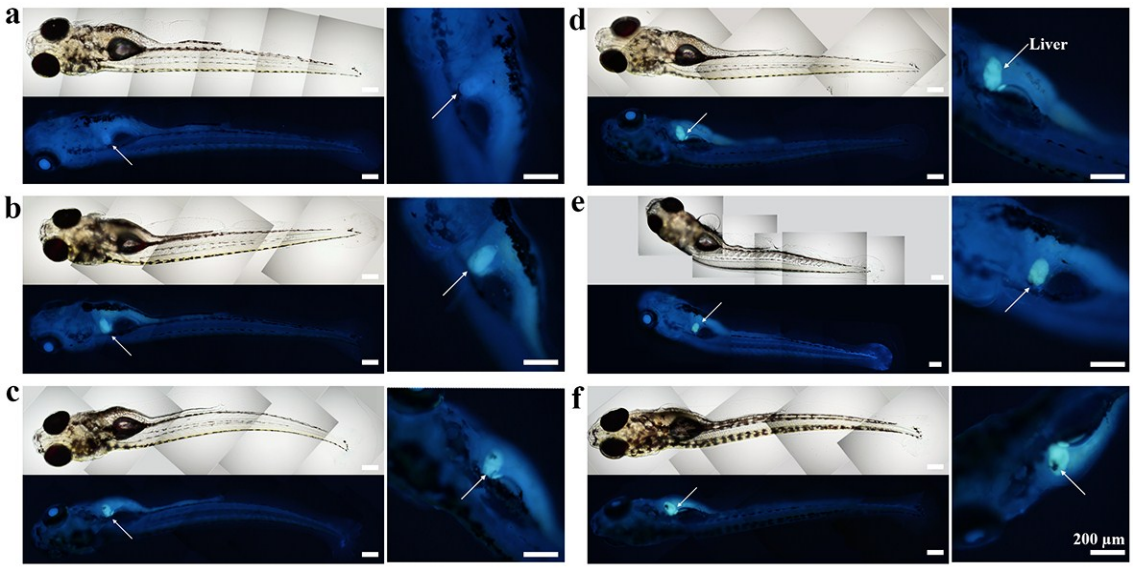

Fig. S10. Bright field and fluorescence images of the whole bodies (right) and the amplified parts (left) of larval zebrafish after 5-h soaking in Bio-QDs medium. The medium was spiked with different concentrations of Bio-QDs (a-f: 0, 2, 4, 6, 8, and 10 μg/mL).

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