

## Supplementary Information

### **One-pot synthesis of Ag–In–Ga–S nanocrystals embedded in a Ga<sub>2</sub>O<sub>3</sub> matrix and enhancement of band-edge emission by Na<sup>+</sup> doping**

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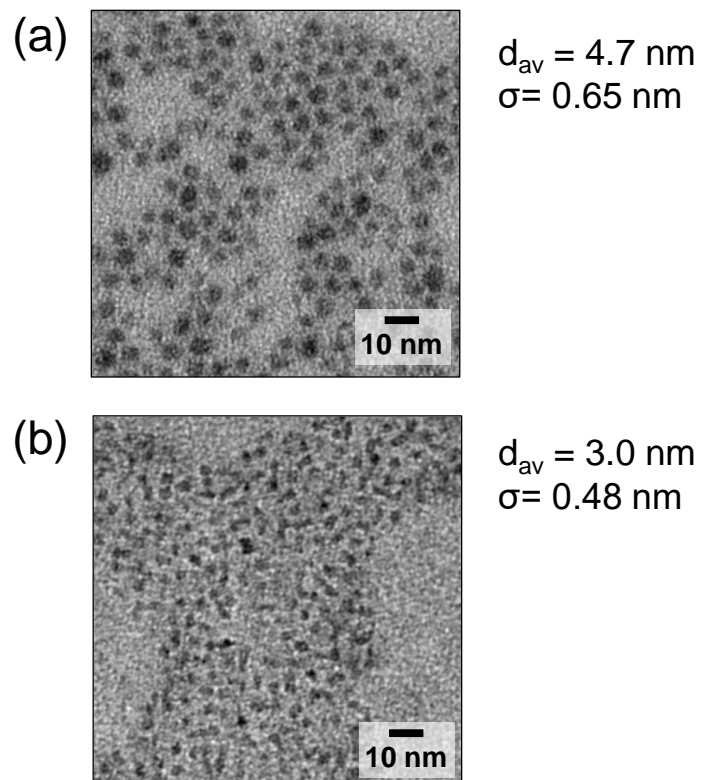
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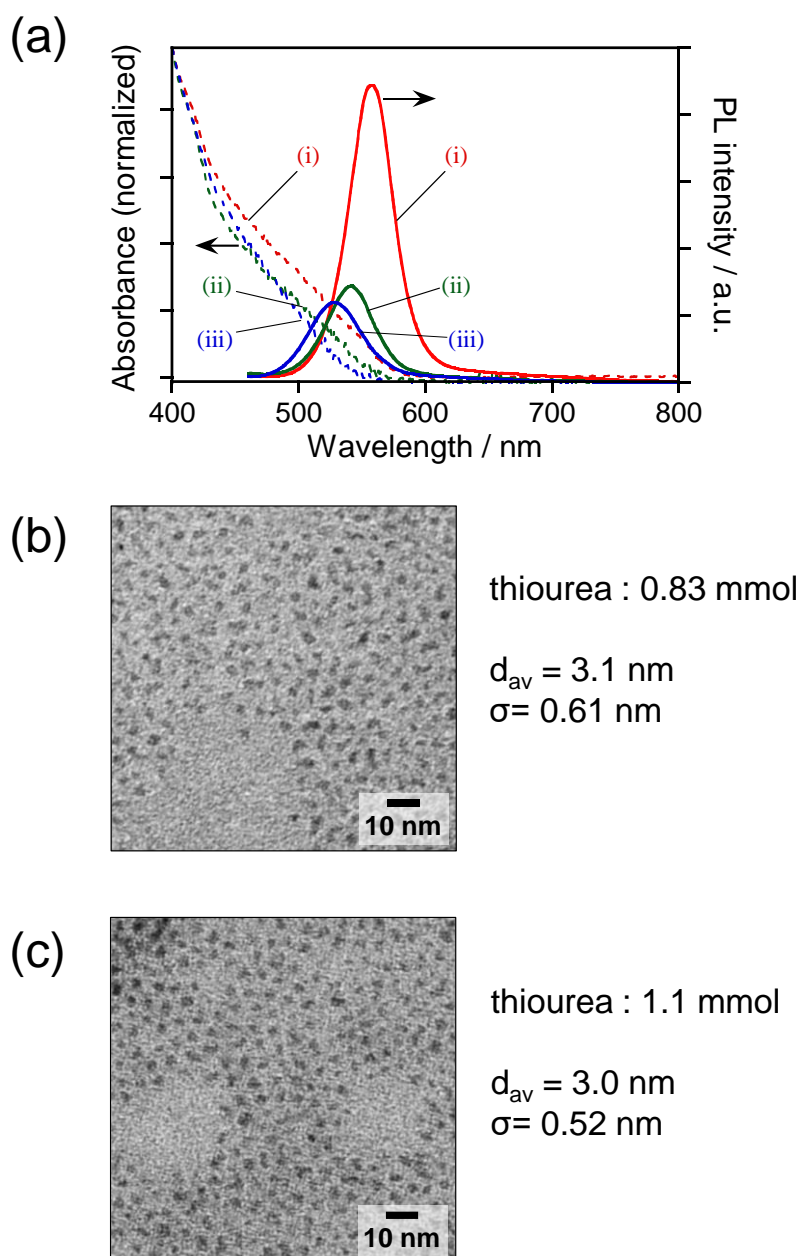
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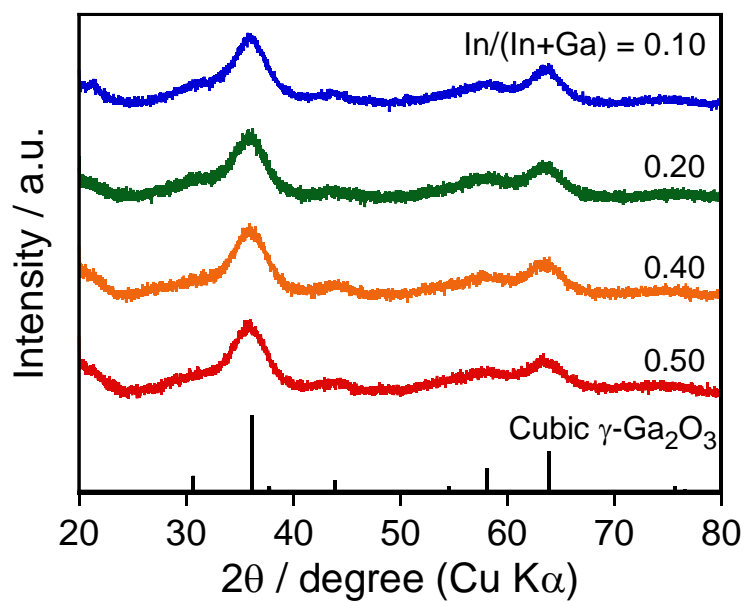
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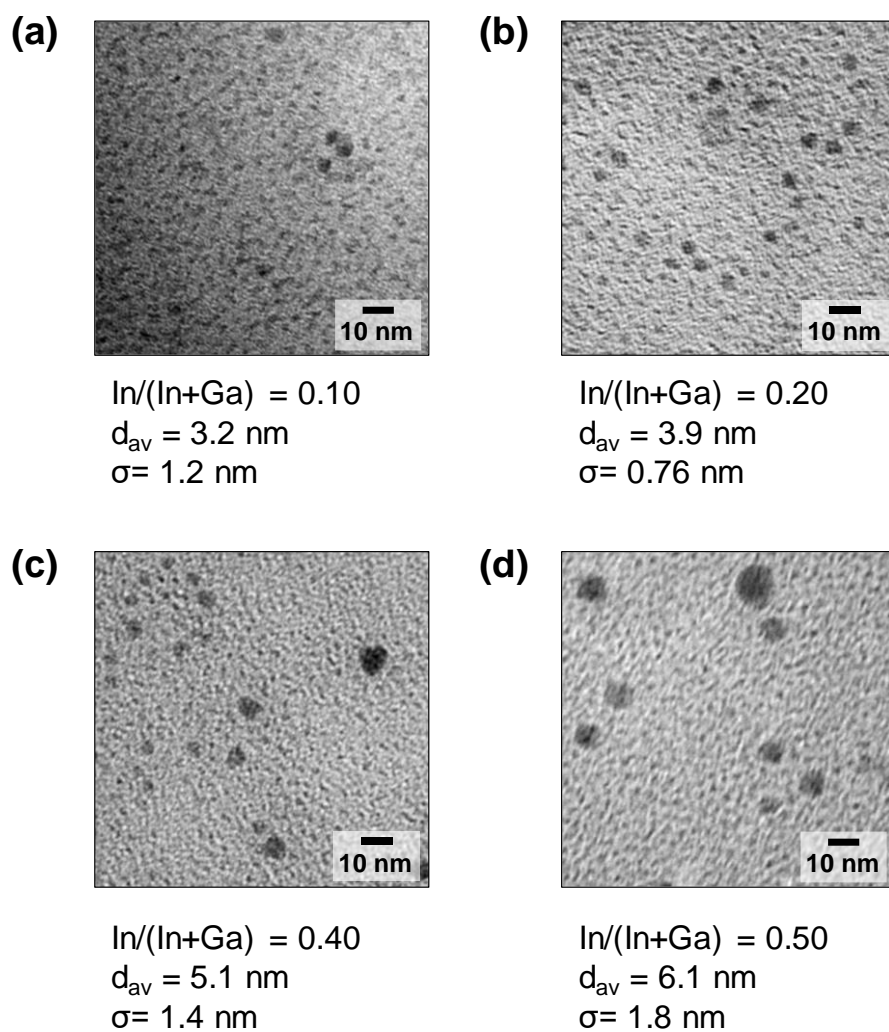
**Figure S1.** Wide-area TEM images of AIGS QDs prepared in the absence of  $\text{Na}^+$  ions. The samples were prepared with elemental sulfur (a) and thiourea (b) under the ratios of  $\text{Na}/(\text{Ag}+\text{Na})=0.30$  and  $\text{In}/(\text{In}+\text{Ga})=0.20$  in the precursors. The average size ( $d_{av}$ ) and standard deviation ( $\sigma$ ) of particles are shown beside the corresponding images.



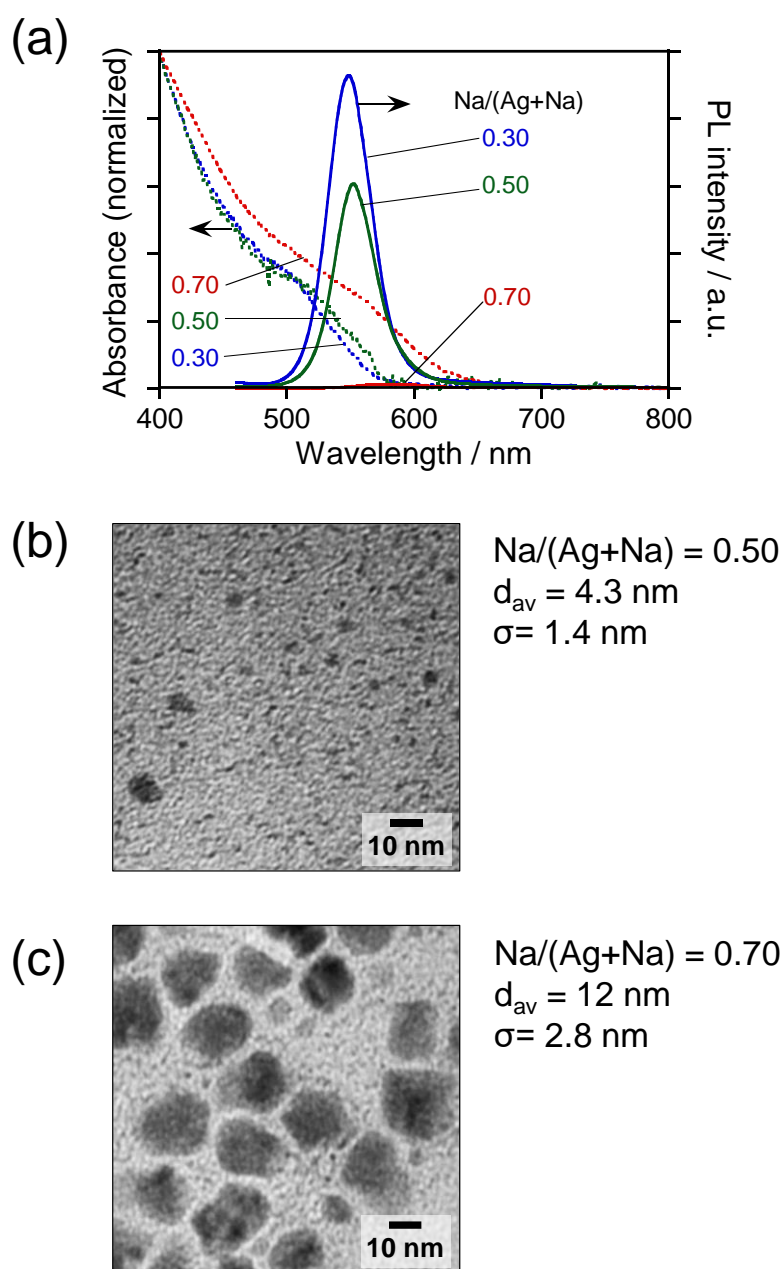
**Figure S2.** (a) Absorption and PL spectra of Na-AIGS(1.5)(tu) QDs prepared with various amount of thiourea. The QDs were prepared with the addition of (i) 0.55, (ii) 0.83, and (iii) 1.1 mmol thiourea under the fixed ratios of  $\text{Na}/(\text{Ag}+\text{Na}) = 0.30$  and  $\text{In}/(\text{In}+\text{Ga}) = 0.20$  in the precursors. The wavelength of excitation light for PL measurements was 365 nm. (b, c) TEM images of Na-AIGS(1.5)(tu) QDs obtained with (b) 0.83 and (c) 1.1 mmol thiourea in the precursors. The average size ( $d_{av}$ ) and standard deviation ( $\sigma$ ) of particles are shown beside the corresponding images.



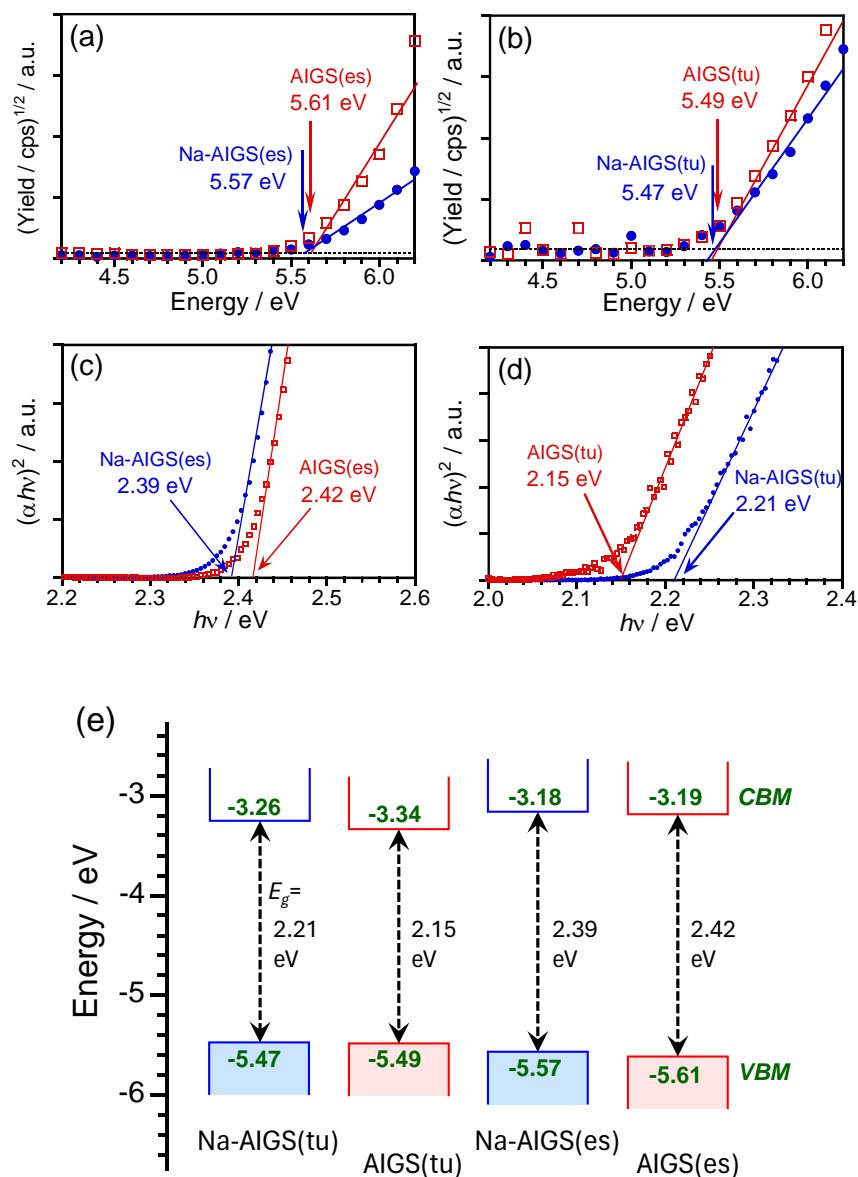
**Figure S3.** XRD patterns of Na-AIGS(1.0)(tu) QDs with different ratios of In/(In+Ga) in the precursors. The samples were prepared under the same conditions as those in Fig. 4.



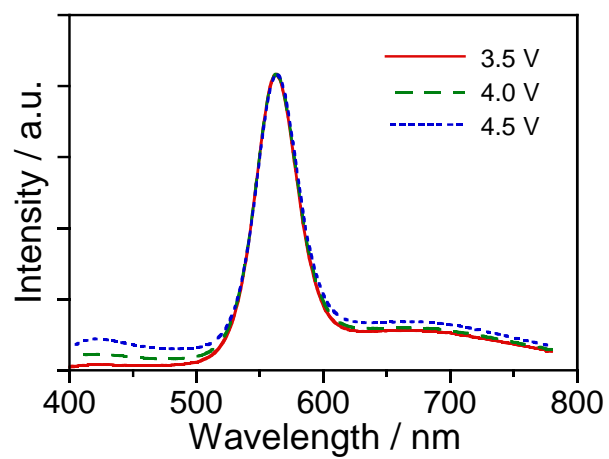
**Figure S4.** TEM images of Na-AIGS(1.0)(tu) QDs with different ratios of In/(In+Ga) in the precursors. The average size ( $d_{\text{av}}$ ) and standard deviation ( $\sigma$ ) of particles are shown below the corresponding images. The samples were prepared under the same conditions as those in Fig. 4.



**Figure S5.** (a) Absorption and PL spectra of Na-AIGS(1.0)(tu) QDs prepared with different Na/(Ag+Na) ratios in the precursors. The wavelength of excitation light for PL measurements was 365 nm. (b, c) TEM images of Na-AIGS(1.0)(tu) QDs with Na/(Ag+Na) of (b) 0.50 and (c) 0.70. The In/(In+Ga) ratio in the precursors was fixed to 0.20.



**Figure S6.** (a, b) Photoelectron yield spectra and (c, d) Tauc plots of AIGS(1.5) QDs prepared with and without Na<sup>+</sup> addition. The S precursors used for the preparation were thiourea (a, c) and elemental sulfur (b, d). (e) Electronic energy structures of AIGS and Na-AIGS QDs. The samples were prepared under the same conditions as those in Fig. 1.



**Figure S7.** EL spectra of the QD-LED device containing Na-AIGS(1.5)(tu) QDs at different applied voltages.



**Table S1.** Chemical compositions of Na-AIGS(1.5)(tu) QDs<sup>(a)</sup> prepared with different amount of thiourea in the precursors.

Amount of thiourea / mmol	Fraction (%)					Charge balance (anion/cation)	Metal ratios	
	Na	Ag	In	Ga	S		In/(In+Ga)	Na/metal
0.55	2.5	1.2	0.7	70	26	0.24	0.01	0.034
0.83	5.0	0.2	0.4	62	33	0.34	0.006	0.074
1.1	5.0	0.5	0.3	60	34	0.36	0.005	0.075

(a) The samples were prepared under the same conditions as those in Fig. S2.

**Table S2.** PL peak wavelengths and quantum yields of Na-AIGS(1.5)(tu) QDs<sup>(a)</sup> prepared with different amount of thiourea in the precursors.

Amount of thiourea / mmol	Peak wavelength / nm	PL QY / %
0.55	548	58
0.83	541	20
1.1	528	18

(a) The samples were prepared under the same conditions as those in Fig. S2.

**Table S3.** Chemical compositions of Na-AIGS(1.0)(tu) QDs<sup>(a)</sup> prepared with different In/(In+Ga) ratios in the precursors.

In/(In+Ga) in the precursors	Fraction (%)					Charge balance (anion/cation)	Metal ratio
	Na	Ag	In	Ga	S		In/(In+Ga)
0.10	5.7	0.2	0.2	65	29	0.29	0.002
0.20	4.4	1.3	0.7	63	30	0.30	0.011
0.40	4.4	1.3	1.2	62	31	0.32	0.019
0.50	3.1	0.8	1.7	62	32	0.33	0.027

(a) The samples were prepared under the same conditions as those in Fig. 4.

**Table S4.** PL peak wavelengths and quantum yields of Na-AIGS(1.0)(tu) QDs<sup>(a)</sup> prepared with different In/(In+Ga) ratios in the precursors.

In/(In+Ga) in the precursors	Peak wavelength / nm	PL QY / %
0.10	517	12
0.20	548	36
0.40	570	34
0.50	579	18

(a) The samples were prepared under the same conditions as those in Fig. 4.

**Table S5.** Chemical compositions of Na-AIGS(1.0)(tu) QDs<sup>(a)</sup> prepared with different Na/(Ag+Na) ratios in the precursors.

Na/(Ag+Na) in the precursors	Fraction (%)					Charge balance (anion/cation)	Metal ratios	
	Na	Ag	In	Ga	S		Na/(Ag+Na)	In/(In+Ga)
0.30	4.4	1.3	0.7	63	30	0.31	0.77	0.011
0.50	3.7	1.0	1.0	63	31	0.31	0.79	0.016
0.70	6.0	12	7.7	32	42	0.61	0.33	0.19

(a) The samples were prepared under the same conditions as those in Fig. S5.

**Table S6.** PL peak wavelengths and quantum yields of Na-AIGS(1.0)(tu) QDs<sup>(a)</sup> prepared with different Na/(Ag+Na) ratios in precursors.

Na/(Ag+Na) in the precursors	Peak wavelength / nm	PL QY / %
0.30	548	36
0.50	550	26
0.70	575	0.6

(a) The samples were prepared under the same conditions as those in Fig. S5.

**Table S7.** Parameters to fit PL decay profiles of AIGS(1.5) QDs.

Samples	$\tau_1$ (ns)	$A_1$ (%)	$\tau_2$ (ns)	$A_2$ (%)	$\tau_3$ (ns)	$A_3$ (%)	$\tau_{ave}$ (ns)	$\chi^2$
AIGS(es)	5.7	55	37	36	175	8.5	99	1.20
Na-AIGS(es)	5.1	57	33	35	127	7.7	68	1.12
AIGS(tu)	40	75	175	25	---	---	120	1.16
Na-AIGS(tu)	34	64	109	36	---	---	83	1.17

(a) The samples were prepared under the same conditions as those in Fig. 1.