Supporting Information

Solar Conversion Efficiency Performance of High Temperature Alloy over Low Temperature One: Comprehending Interfaces through *Excitonics* Study

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(A) Transmission electron microscopy:

Figure S1 illustrates the HRTEM images of G-300 CdZnSSe. As can be observed, the average diameter was 4.4±0.2 nm. These images are in consistency with the UV-Vis data, which reveals the uniform shape and size distribution.
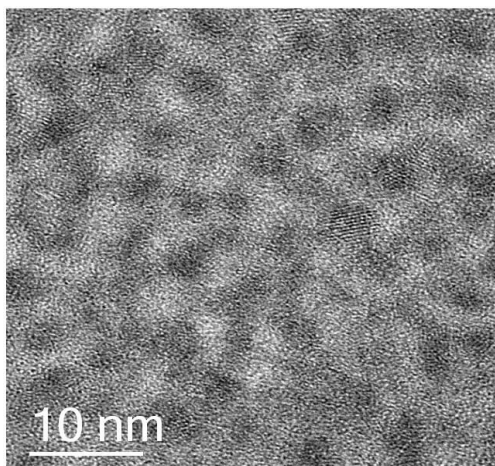

Figure S1. HRTEM image of CdZnSSe G-300 alloy NC.

(B) X-ray diffraction measurement:

In the present manuscript, XRD data was collected on Philips X-ray machine, model-PW 1710 with Ni-filtered Cu Kα radiation (λ=1.54178 Å), using silicon as an external standard. X-ray diffraction (XRD) analysis has been carried out to determine the chemical composition and morphological analysis of the synthesized alloy NCs.

(C) Energy dispersive X-ray (EDX) measurement:

EDX measurement has been done through scanning electron microscopy (JEOL JEC-3000FC; Tokyo, Japan). The nanoparticles were drop casted on silicon wafer and then dried overnight in open air (slow evaporation). The applied accelerating voltage was kept around 10.0–20.0 kV. Bruker signal processing unit was used for elemental analysis.
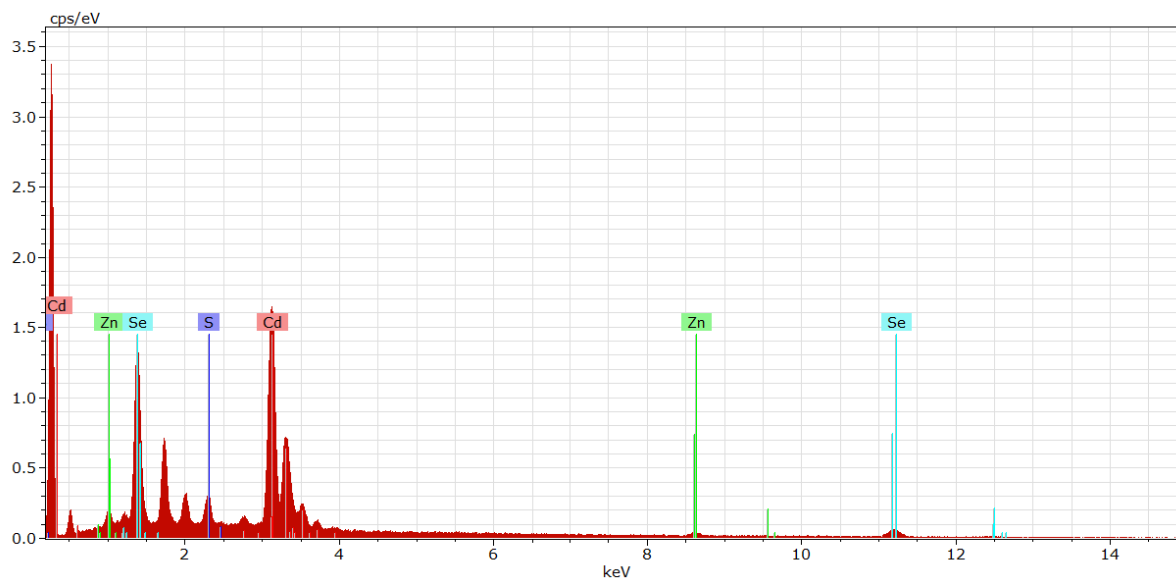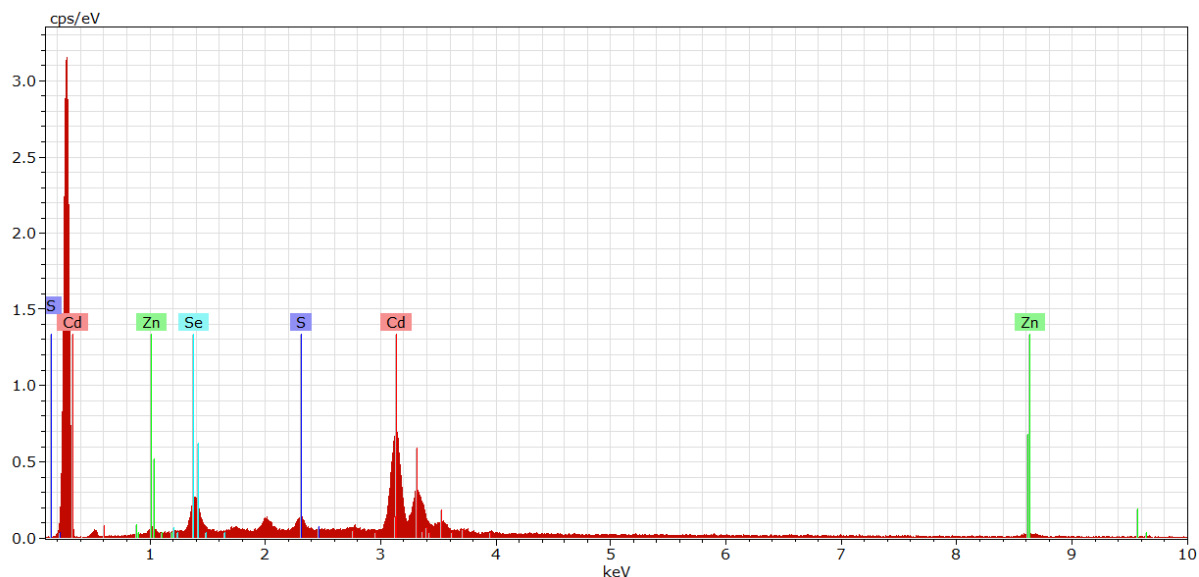
Figure S2. EDX spectra of (Upper) 'G-250' and (Lower) 'G-300' NCs.

Table S1. Elemental composition in G-250 as obtained from EDX spectra.

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Spectrum: Acquisition 2790.spx
El AN  Series   unn. C norm. C Atom. C Error (1 Sigma)
                [wt.%]   [wt.%]   [at.%]          [wt.%]
-------------------------------------------------------
Cd 48 L-series   50.43    71.70    59.40           1.59
Se 34 K-series   15.57    22.14    26.11           0.66
S  16 K-series    2.71     3.86    11.21           0.14
Zn 30 K-series    1.62     2.30     3.28           0.11
-------------------------------------------------------
        Total:   70.34   100.00   100.00
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Table S2. Elemental composition in G-250 as obtained from EDX spectra.

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Spectrum: Acquisition 2789.xls
El AN  Series   unn. C norm. C Atom. C Error (1 Sigma)
                [wt.%]   [wt.%]   [at.%]          [wt.%]
-------------------------------------------------------
Cd 48 L-series   35.71    71.12    58.55           1.17
Se 34 L-series    9.88    19.68    23.06           0.60
Zn 30 K-series    2.79     5.56     7.87           0.19
S  16 K-series    1.83     3.64    10.51           0.12
-------------------------------------------------------
        Total:   50.21   100.00   100.00
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(D) The variation of 1S exciton and PL maxima of gradient and homogeneous alloy as a function of reaction time:
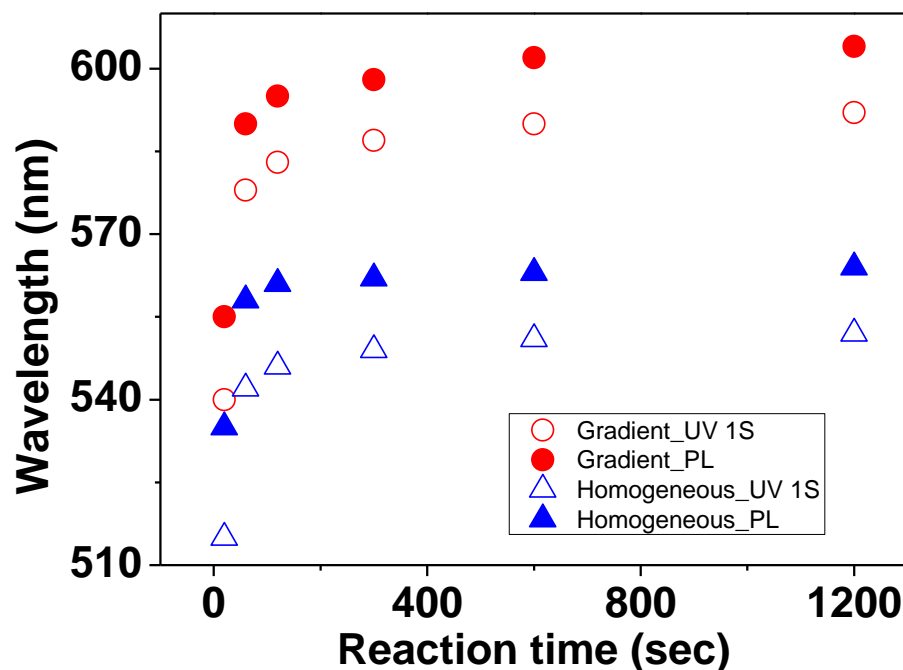

Figure S3. The variation of UV-vis absorption (1S exciton) and PL maixa of G-250 (red) and G-300 (blue) alloy NCs as a function of reaction time.

(E) Quantum yield Calculation:

The most widely accepted method of calculating Quantum yield is the relative method. Using solutions with comparable concentrations to maintain linearity such that the UV-Vis absorption never exceeds 0.1 and PL do not saturate. Also, a standard has to be chosen such that its absorption lies in or near the region our QD whose QY is to be calculated. For the present work Rhodamine B solution is prepared in acetonitrile which has a QY of 0.96 and has been chosen as the standard.

For quantum yield calculation, solutions were prepared to maintainUV-Vis absorption near 0.1. Fluorescence spectrum was recorded and PL integration was taken down for both the sample and the standard. Using the values the QY was calculated as,

$$\Phi_{SAMPLE} = (A_{REFERENCE}/ A_{SAMPLE}) * (F_{SAMPLE}/ F_{REFERENCE}) * \Phi_{REFERENCE}$$

Where Φ_{SAMPLE} and $\Phi_{REFERENCE}$ are the QY of the sample and the reference or standard; $A_{REFERENCE}$ and A_{SAMPLE} are the absorbance of the sample and the standard at the excitation wavelength; F_{SAMPLE} and $F_{REFERENCE}$ are the fluorescence integration areas of the sample and the standard respectively.

Quantum yields were calculated and found to vary as the reaction time proceeds. The general trend of QY shows the homogeneous alloy has higher QY than the gradient alloy NCs.
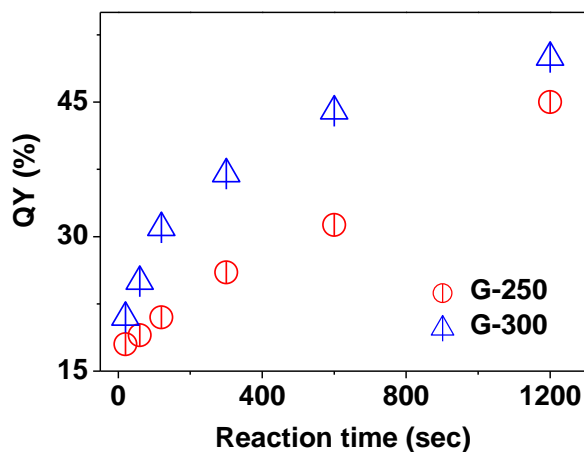

Figure S4. QY of G-250 and G-300 alloy NCs as a function of reaction time.

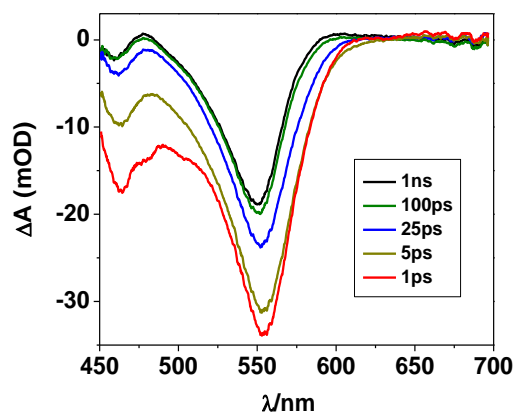**(F) Transient absorption spectrum of CdSe QD and G-250 alloy:**

Figure S5. Ultrafast transient absorption spectrum of CdSe QD, synthesized under similar condition, as a function of time delay after 400nm laser excitation. The spectrum shows clear 1S and 1P exciton peak at ~553 and ~464 nm respectively, however no clear 2S exciton was observed, as both 1S and 2S states are merged due to inhomogeneous broadening.
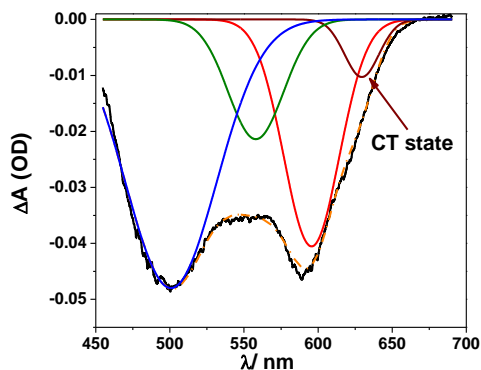

Figure S6. De-convoluted TA spectrum at 1 ps time delay of G-250 CdZnSSe alloy. The black line represents original data while the orange dotted line is fitted data. The red, olive and blue Gaussians are represented 1S, 2S and 1P states respectively while the dark red Gaussian is due to CT state. The evolution of CT state in the TA spectrum is consistence with the one observed in UV-vis absorption spectrum (see main text).

Table S3. Photovoltaic performance in terms of J-V measurement of G-250 and G-300 CdZnSSe alloy NCs for four-different device.

NC System	J_{sc} (mA/cm^2)		V_{oc} (Volt)		FF (%)		PCE (%)	
G-250	14.86	14.7±0.2	0.58	0.59±0.005	39	40±0.4	3.36	3.48±0.06
	15.08		0.58		41		3.6	
	14.13		0.60		40		3.4	
	14.75		0.60		40		3.59	
G-300	17.02	17.2±0.18	0.61	0.60±0.003	45	43±0.7	4.68	4.51±0.05
	17.24		0.60		42		4.43	
	17.66		0.60		42		4.47	
	16.82		0.61		43		4.46	