**Supporting Information**

# Greener Luminescent Solar Concentrators with High Loading Contents Based on In-Situ Cross-Linked Carbon Nano-Dots for Enhancing Solar-Energy Harvesting and Resisting Concentration-Induced Quenching

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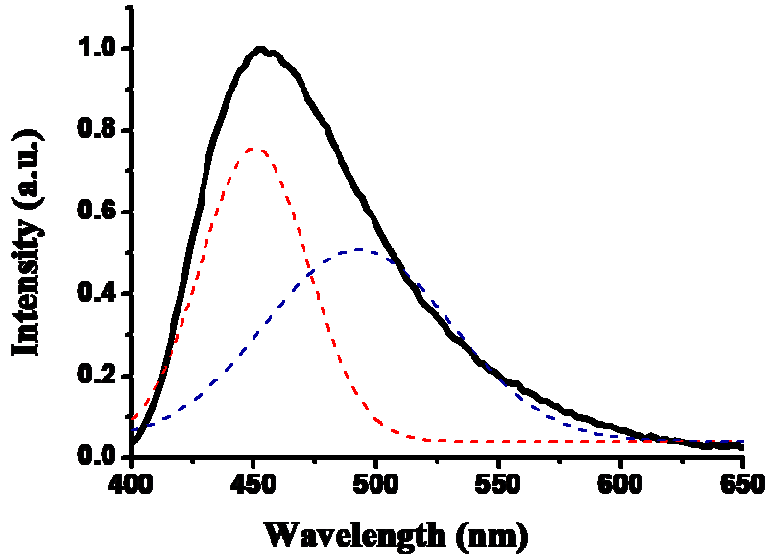


**Figure S1: PL spectrum fitted by two Gaussian functions for as-prepared, diluted Si-CNDs.**

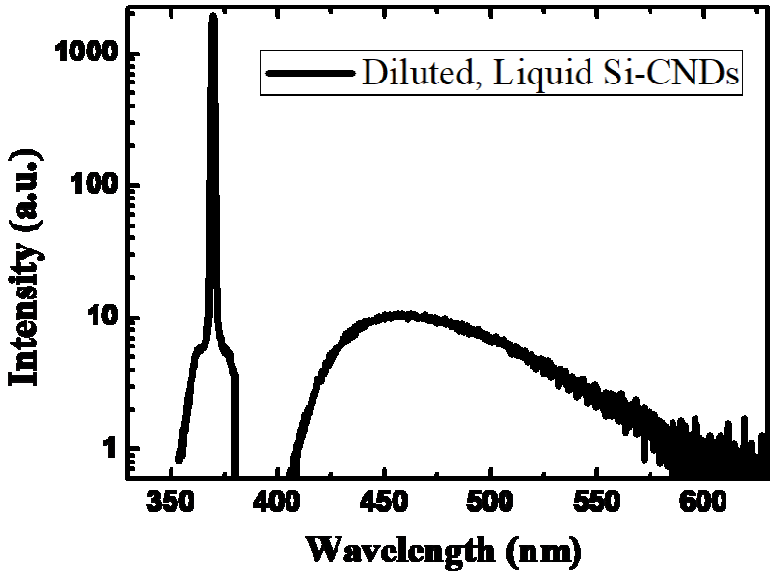


**Figure S2: The experimental data of absolute PL-QY measurement for as-prepared, diluted Si-CNDs.**

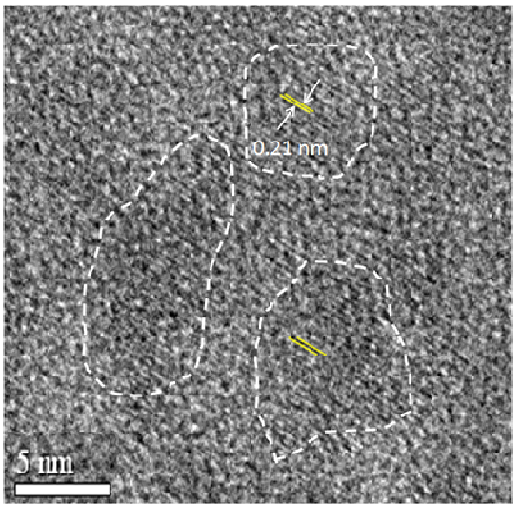


**Figure S3: High-resolution TEM imaging for Si-CNDs.**

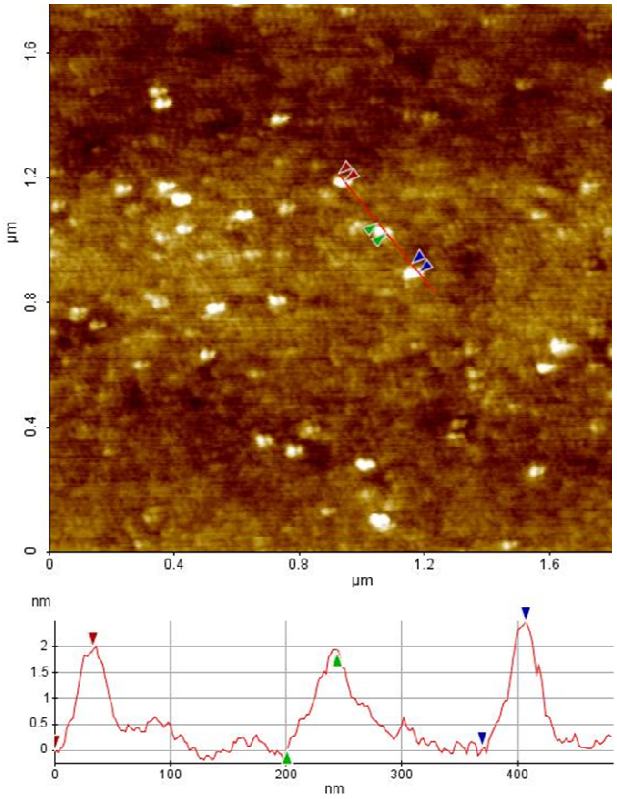


**Figure S4: The AFM imaging for Si-CNDs.**

The topographic heights of Si-CNDs were estimated based on several AFM images, which is 1.2 nm~6 nm.

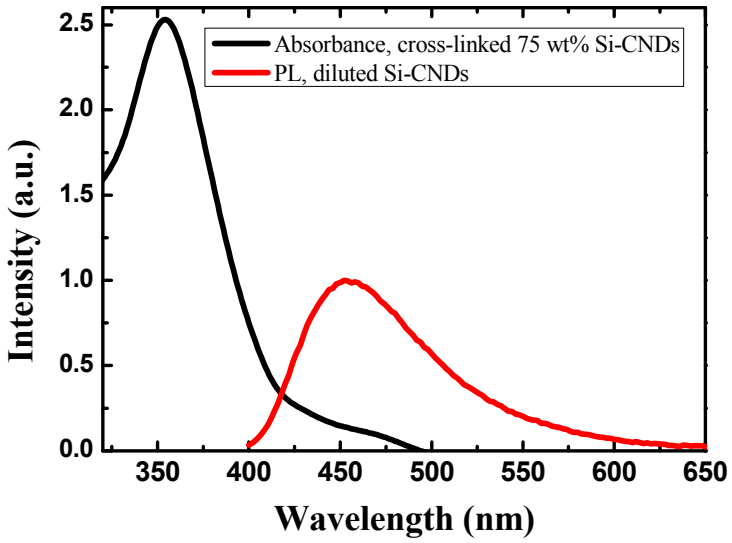


**Figure S5: Spectral overlap between optical absorption and PL emission.**

To highlight the reabsorption effect, the absorbance spectrum of 75 wt% loading Si-CNDs and PL emission of diluted Si-CNDs were shown together.

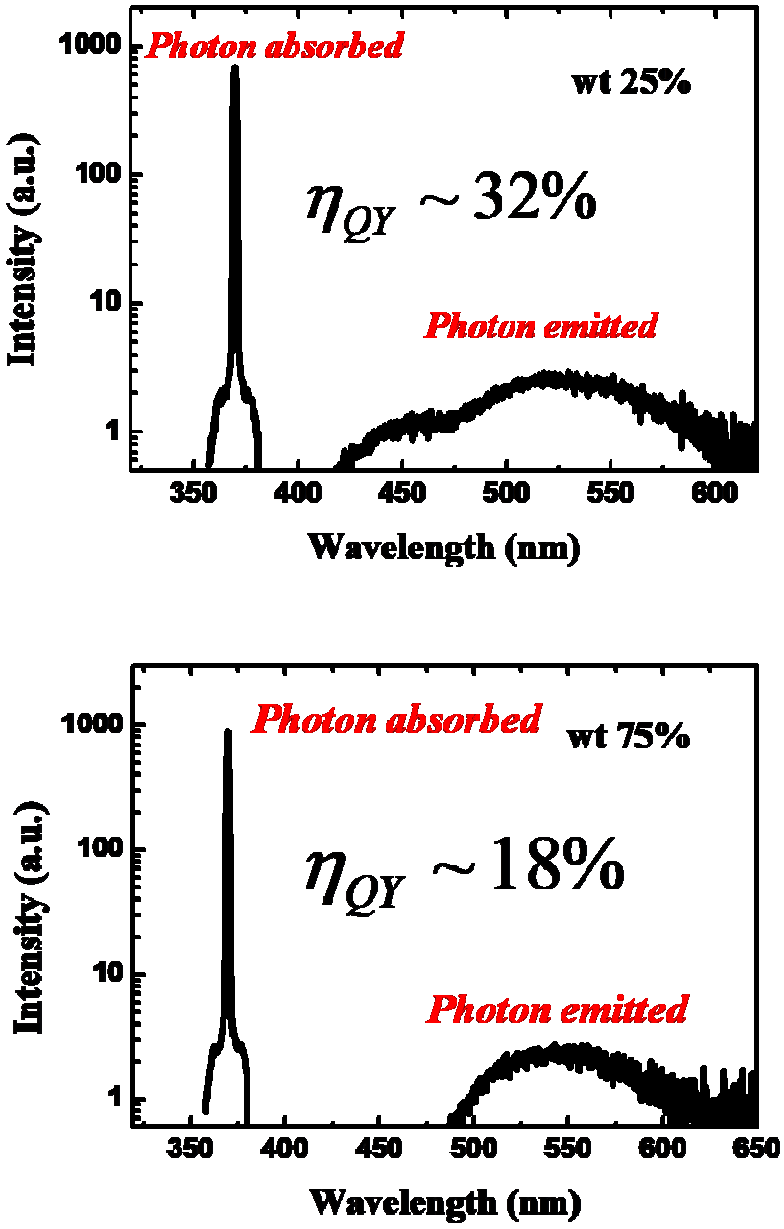


**Figure S6: Experimental data of absolute PL-QY measurement for cross-linked Si-CNDs.**

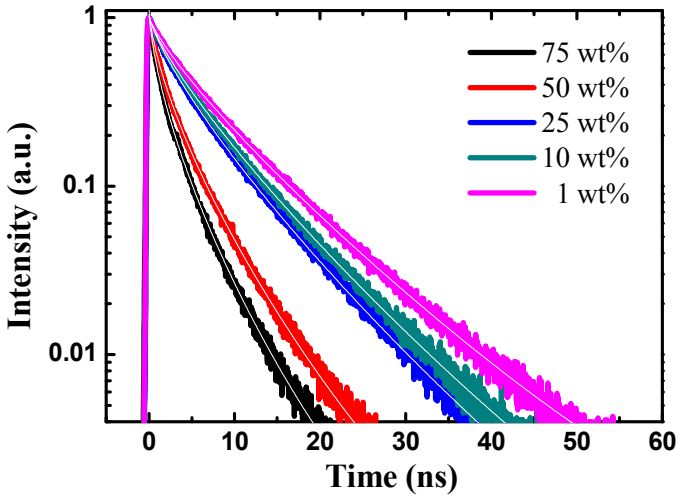


**Figure S7: Fitting results for cross-linked Si-CND thin films with different loadings**

To yield quantitative information on time-resolved PL measurements, stretched exponential function, $I(t) = I_0 \exp(-\frac{t}{\tau})^{\beta} + I_b$ was employed to fit the experimental data, where $I_0, \beta, \tau, I_b$ denote the PL intensity at initial time delay, dispersion parameter, characteristic lifetime, and background intensity, respectively. After yielding the fitting parameters, the average PL lifetimes can be derived using this equation, $\langle \tau \rangle = \frac{\tau}{\beta} \Gamma(\frac{1}{\beta})$, where $\Gamma$ is the gamma function. The average lifetimes for cross-linked Si-CNDs with different loadings are 6.5 ns (1 wt%), 5.4 ns (10 wt%), 4.8 ns (25 wt%), 2.4 ns (50 wt%), 1.8 ns (75 wt%), respectively.

| *Loadings* | *LSC PLQY(%)* | *Edge-emission efficiency (%)* | *IQE(%)* |
|---|---|---|---|
| 75 wt% | 18 | 51 | ~9 |
| 50 wt% | 22 | 60 | ~13 |
| 25 wt% | 32 | 68 | ~22 |
| 10 wt% | 40 | 70 | ~28 |

| Sample | Solid-state PL-QY (%) | Size ($cm^2$) | IQE (%) | EQE (%) | Supplementary | Ref |
|---|---|---|---|---|---|---|
| OLA-CNDs | 30 | 1.5 x 8 | 4 | 0.4 | | 1 |
| N-CNDs | NA | 2.5 x 1.6 | NA | 4.75 | 0.3 wt% loading | 2 |
| PbS/CdS QDs | 40 | 1.5 x 10 | 4.5 | 1.1 | | 3 |
| CdSe/CdS QDs | 45 | 21.5 x 1.3 | 10.2 | NA | 0.05 wt% loading | 4 |
| CuInS2/ZnS QDs | 81 | 2.2 x 2.2 | 26.5 | NA | | 5 |
| Perovskite QDs | 60 | 1.3 x 9 | NA | 2 | | 6 |
| CdSe/ZnS/ZnS QDs | 65 | 9x 10 | NA | 1.2 | 0.9 wt% loading | 7 |
| Si-CNDs | 45 | 3 x 3 | 22 | NA | 25 wt% loading | this work |

**Figure S8: The parameters for Si-CND LSCs with different loadings and the comparisons with other LSCs [1-7].**

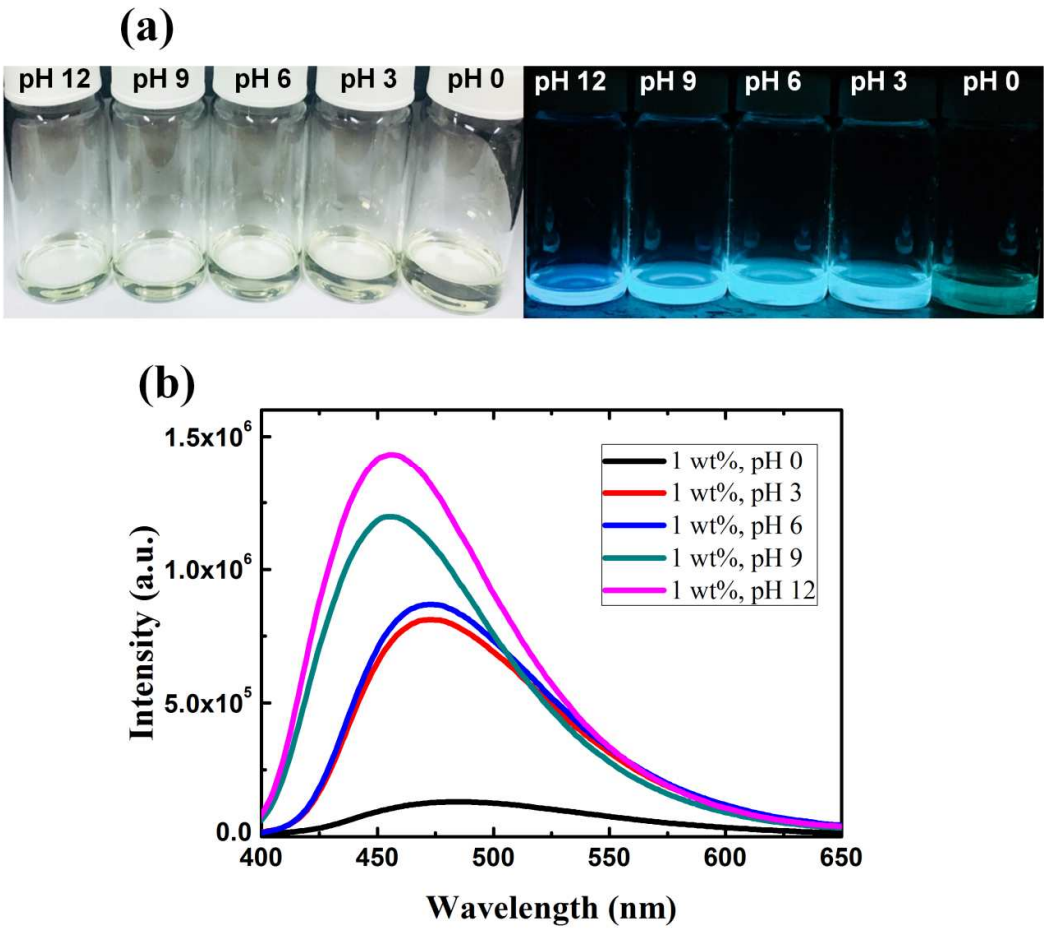


**Figure S9: (a) The photographs for Si-CNDs under ambient (left) and UV illumination (right) at different pH values and (b) PL spectrum for Si-CNDs at different pH values.**

## REFERENCES


(1) Zhou, Y.; Benetti, D.; Tong, X.; Jin, L.; Wang, Z. M.; Ma, D.; Zhao, H.; Rosei, F. Colloidal Carbon Dots based Highly Stable Luminescent Solar Concentrators. *Nano Energy* **2018**, *44*, 378-387.

(2) Li, Y.; Miao, P.; Zhou, W.; Gong, X.; Zhao, X. N-Doped Carbon-Dots for Luminescent Solar Concentrators. *J. Mater. Chem. A* **2017**, *5*, 21452-21459.

(3) Yufeng, Z.; Daniele, B.; Zhiyuan, F.; Haiguang, Z.; Dongling, M.; O., G. A.; Alberto, V.; Federico, R. Near Infrared, Highly Efficient Luminescent Solar Concentrators. *Adv. Energy Mater.* **2016**, *6*, 1501913.

(4) Meinardi, F.; Colombo, A.; Velizhanin, K. A.; Simonutti, R.; Lorenzon, M.; Beverina, L.; Viswanatha, R.; Klimov, V. I.; Brovelli, S. Large-Area Luminescent Solar Concentrators Based on /`Stokes-Shift-Engineered/' Nanocrystals in a Mass-Polymerized PMMA Matrix. *Nat. Photon.* **2014**, *8*, 392-399.

(5) Li, C.; Chen, W.; Wu, D.; Quan, D.; Zhou, Z.; Hao, J.; Qin, J.; Li, Y.; He, Z.; Wang, K. Large Stokes Shift and High Efficiency Luminescent Solar Concentrator Incorporated with CuInS2/ZnS Quantum Dots. *Sci. Rep.* **2015**, *5*, 17777.

(6) Zhao, H.; Zhou, Y.; Benetti, D.; Ma, D.; Rosei, F. Perovskite Quantum Dots Integrated in Large-Area Luminescent Solar Concentrators. *Nano Energy* **2017**, *37*, 214-223.

(7) Brennan, L. J.; Purcell-Milton, F.; McKenna, B.; Watson, Trystan M.; Gun'ko, Y. K.; Evans, R. C. Large Area Quantum Dot Luminescent Solar Concentrators for Use with Dye-Sensitised Solar Cells. *J. Mater. Chem. A* **2018**, *6*, 2671-2680.