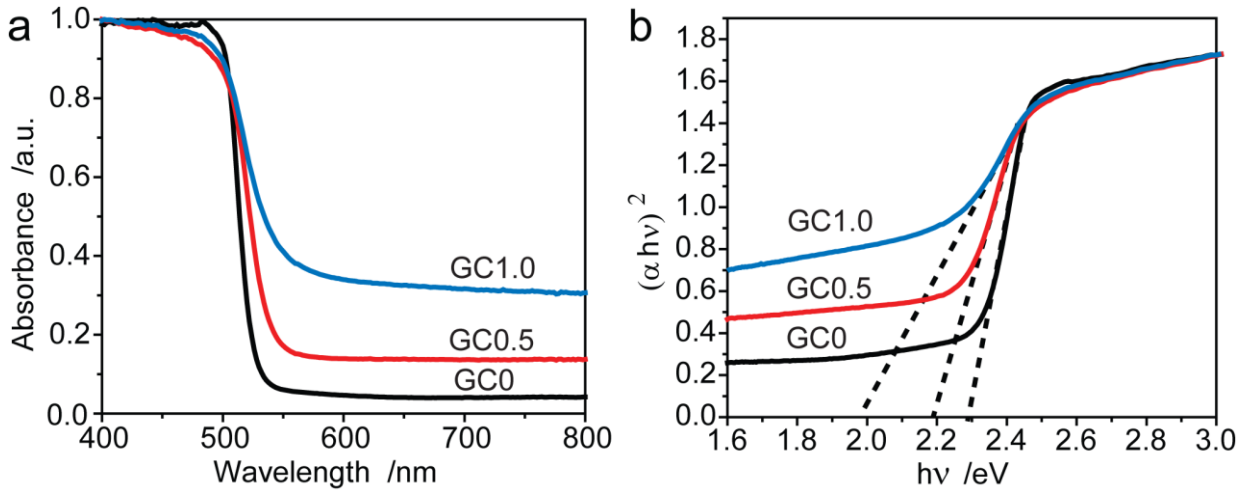


**Figure S1**. (a) UV-vis diffuse reflectance spectra and (b) the relationship between the transformed Kubelka-Munk function versus the light energy of $GC_X$ (X=0, 0.5, and 1.0)samples, and the obtained optical bandgap is estimated to be 2.29, 2.19, and 1.99 eV for GC0, GC0.5, and GC1.0 respectively.

**Theoretical calculation details**: The generalized gradient approximation (GGA)[S1] and the projector augmented wave (PAW)[S2] method are implemented. The electron-wave functions are expanded with the plane wave energy cutoff of $E_{cut}$ = 350 eV. Gamma point sampling is done for Brillouin zone integration due to the large supercell used.[S3] Since the phase of CdS in CdS/RGO composites is a mixture of hexagonal and cubic phase based on our experimental results, the CdS quantum dot (QD) has been constructed on a hexagonal and a cubic bulk CdS structure, respectively. Based on the smallest $(CdS)_{13}$ nanoparticle, the initial geometry of CdS QD is composed of 13 Cd and 13 S ions extracting from the hexagonal or the cubic CdS bulk structure. The distance between CdS QD is larger than 10 Å so that the interaction between them can be neglectable.

The graphene-CdS QD system contains the graphene supercell composed of seven unit cells of zigzag-edge graphene nanoribbon with width of ten zigzag lines and one CdS QD. The supercell length is set to 27 Å in the non-periodic direction. For the initial configuration, the equilibrium distance between CdS QD and graphene monolayer has been calculated. The result shown in Figure S2 indicates the 5 and 4 Å equilibrium distance for hexagonal and cubic CdS respectively. And then the electronic structures are obtained from the optimized initial geometries. For all the geometry optimization, the force acting on each atom is relaxed to less than 0.01 eV $Å^{-1}$.

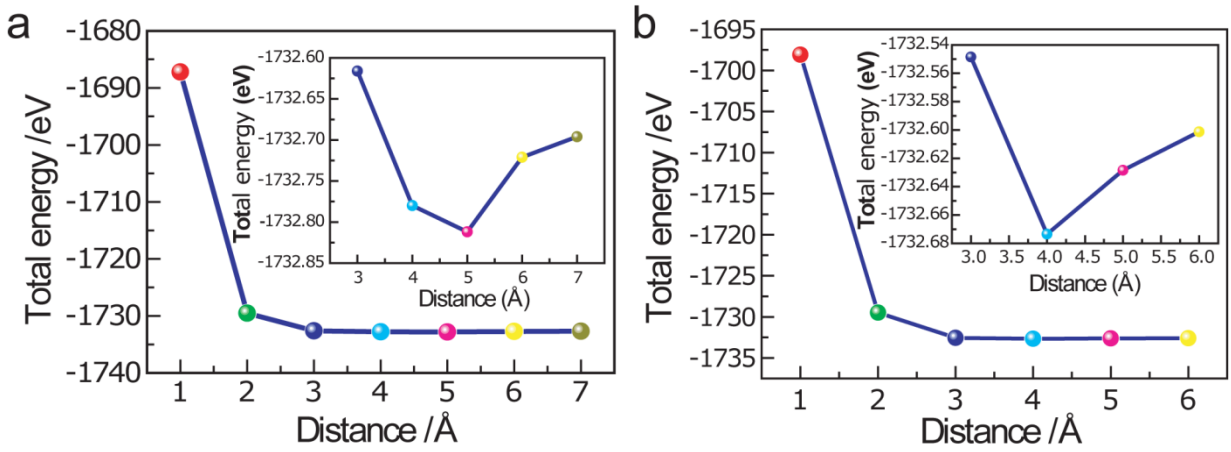


**Figure S2**. Total energy variation depending on (a) the graphene-hexagonal CdS QD lateral distance and the inset is enlarged plot for 3Å<d<7Å and (b) the graphene-cubic CdS QD lateral distance and the inset is enlarged plot for 3Å<d<6Å.

## References


[S1] J. P. Perdew, Y. Wang, *Phys. Rev. B* **1992**, *45*, 13244-13249.

[S2] A. Kasuya, R. Sivamohan, Y. A. Barnakov, I. M. Dmitruk, T. Nirasawa, V. R. Romanyuk, V. Kumar, S. V. Mamykin, K. Tohji, B. Jeyadevan, K. Shinoda, T. Kudo, O. Terasaki, Z. Liu, R. V. Belosludov, V. Sundararajan, Y. Kawazoe, *Nat. Mater.***2004**, *3*, 99-102.

[S3] a) W. Feng, S. Lei, Q. Li and A. Zhao, *J. Phys. Chem. C* **2011**, 115, 24858-24864; b) Y. Okamoto, *Chem. Phys. Lett.* **2006**, *420*, 382-386.