Supplementary Information for:

Organosilane-Functionalized Graphene Quantum Dots and Their Encapsulation into Bi-Layer Hollow Silica Spheres for Bioimaging Application

Ting Wen,[‡] Baocheng Yang,[‡] Yanzhen Guo, Jing Sun, Chunmei Zhao, Shouren Zhang, Miao Zhang and Yonggang Wang*

Institute of Nanostructured Functional Materials, Huanghe Science and Technology College, Zhengzhou, Henan 450006, China

- *Fig. S1* XRD pattens of the GOs, Graphenes and Si-GQDs. The lattice spacing of the {002} facets are 4.25 Å, 3.45 Å and 3.52 Å, respectively. Graphene were obtained by trating GOs at 100 °C for 24 h in a vacuum oven.
- *Fig. S2* C-NMR spectra of Si-GQDs. The peak with chemical shift of 166 ppm suggests the existence of C=ONH groups.
- *Fig. S3* (a, b) The optical photographs of Si-GQDs film on glass substrate and block under visible light and UV lamp with the wavelength of 365 nm; (c) PL spectra (above) and the normalized PL spectra (bottom) of the block (recorded from 320 to 460 nm in 20 nm increments).
- *Fig. S4* SEM and TEM images of the etched silica spheres without Si-GQDs obtained from solid silica spheres with 100 μ L (a, b) and 250 μ L (c, d) AEAPTMS, respectively.
- Fig. S5 (a-c) TEM images of the FHMSSs obtained from Si-GQDs embedded in solid silica spheres with AEAPTMS of 25 μL, 50 μL and 100 μL, respectively; (d-f) Normalized PL spectra of Si-GQDs embedded solid silica spheres and corresponding FHMSSs with AEAPTMS of 25 μL, 50 μL and 100 μL, respectively.
- *Fig. S6* (a) PL spectra and the normalized PL spectra (the inset) of FHMSSs (with $100 \,\mu\text{L}$ AEAPTMS) (recorded from 320 to 400 nm in 20 nm increments).



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The retained rate of PL intensity (R $_{PL retained}$) is defined with eq. 1 to represent the variation tendencies of retained fluorescent intensity after the etching process with AEAPTMS amount.

$$R_{PL retained} = \frac{A_{after}}{A_{before}} \times 100\%$$
Eq. 1

Where, A_{before} is the PL peak area of the Si-GQDs embedded solid silica spheres, A_{after} is the PL peak area of FHMSSs.



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