Supporting Information

Top-down fabrication of luminescent graphene quantum dots using self-assembled Au nanoparticles

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Supporting Information S1. Au nanoparticles formed on graphene

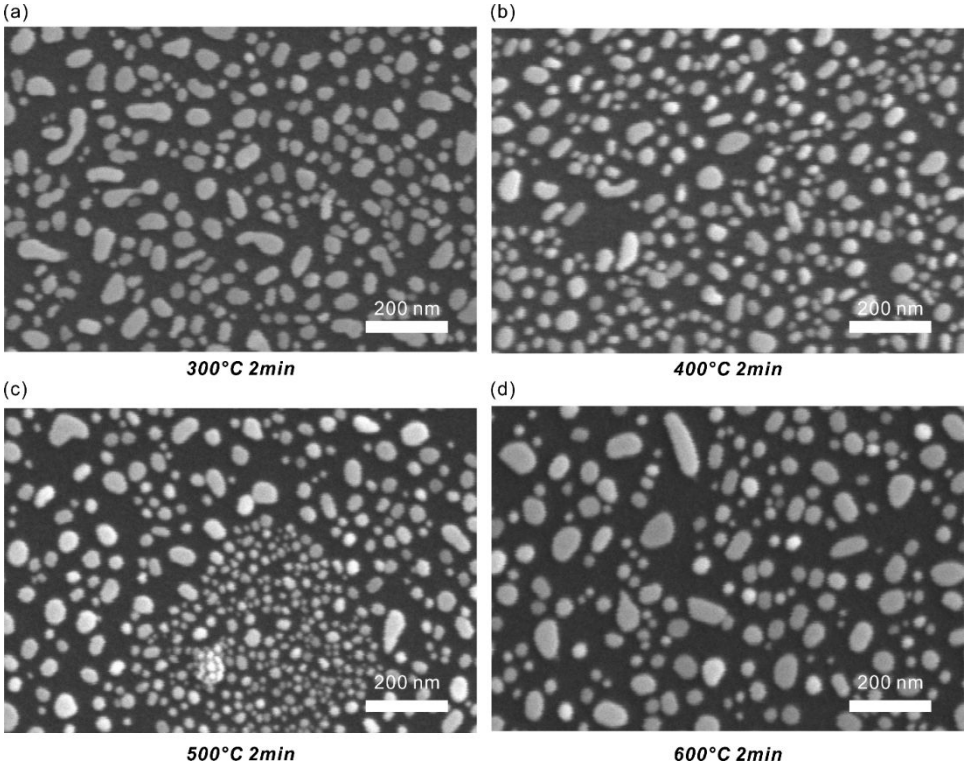

Figure S1. Scanning electron microscopy images of Au nanoparticles formed by thermal annealing of 5 nm-thick Au thin films for 2 min at different temperatures of (a) 300, (b) 400, (c) 500, and (d) 600 °C on native graphene layers. Regardless of the annealing temperatures, Au islands on graphene have irregular shapes and sizes.

Supporting Information S2. Lattice fringes

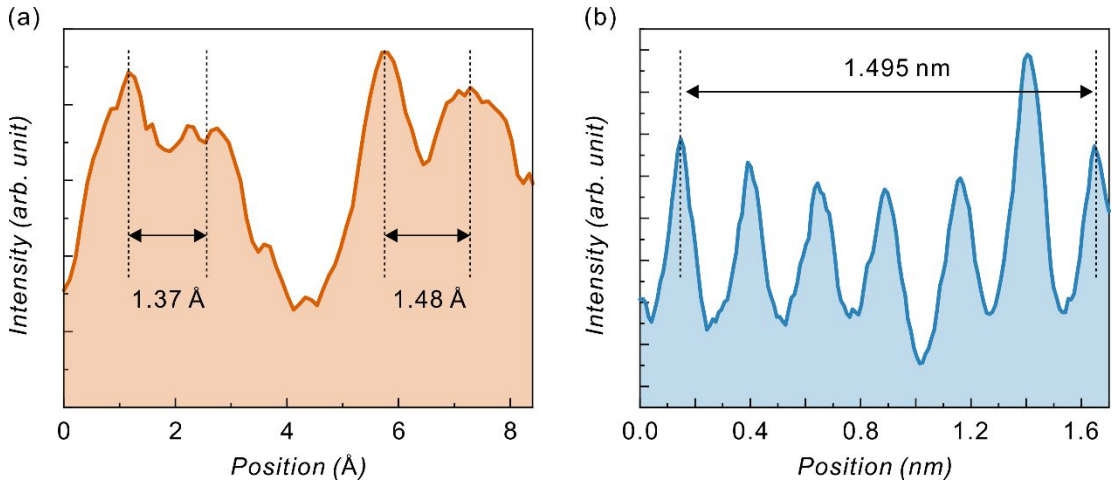

Figure S2. The intensity profiles of lattice fringes. (a) carbon-carbon bond length (averaged value: ~1.43 Å) and (b) lattice constant of graphene ($^{14.95}/_{6}$ Å = 2.49 Å)

Supporting Information S3. I_D/I_G ratio of GQDs

In general, I_D/I_G tends to decrease with the size of the GQDs owing to the decrease in the perimeter-to-area ratio. However, this trend is not valid for the as-fabricated GQDs, and this implies that the different edge structures of the fabricated GQDs depend on their sizes. The plasma-etching process can generate a complex edge structure composed of both zigzag and armchair structures. [M. M. Lucchese *et al.*, *Carbon* **2010**, *48*, 1592] The armchair structure is active for D-band Raman scattering, while the zigzag structure is inactive. [L. G. Cancado *et al.*, *Phys. Rev. Lett.* **2004**, *83*, 247401] Therefore, larger GQDs with stronger D-band intensities will have more dominant D-band-active (e.g., armchair) edge structures.

The significant decrease in the D-peak in 16 and 27 nm GQDs by thermal reduction may be attributed to the modification of the edge structure from D-band-active to D-band-inactive structures (e.g., zigzag). The slight change in the D-peak intensity from the 12 nm and 14 nm GQDs may indicate that the edge structure remains unchanged. From the long-wavelength emission characteristics shown in Figure 7, we expect that the zigzag structure could be dominant in the 12 and 14 nm GQDs both before and after thermal treatment.

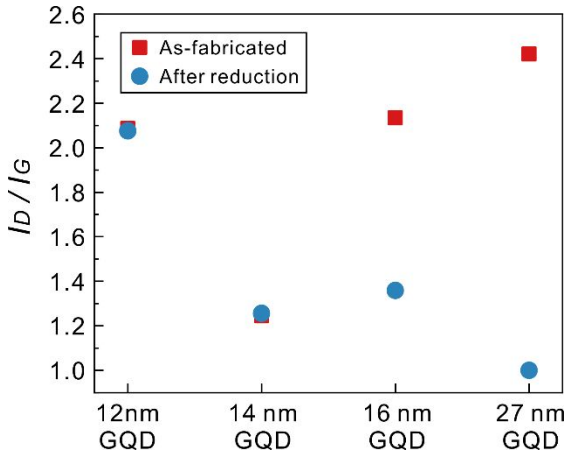

Figure S3. Intensity ratio of the D-band to the G-band (I_D/I_G) of the GQDs before and after thermal reduction.

Supporting Information S4. XPS analysis

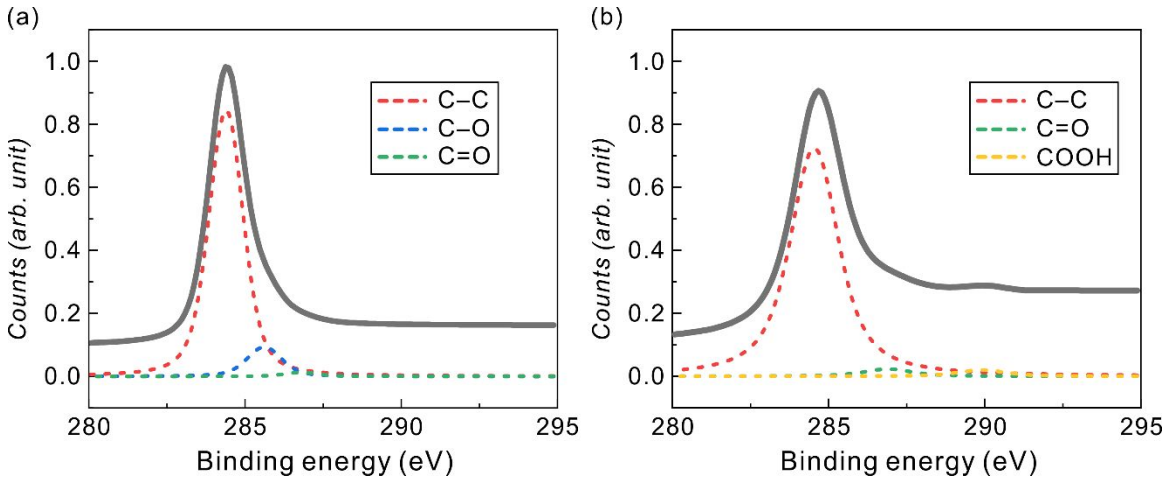

Figure S4. Deconvolution plot of the C 1s peak of GQDs (a) before and (b) after thermal reduction.

Supporting Information S5. Optical bandgap energy

The calculated optical bandgap energies of the 12, 14, and 16 nm GQDs were 3.92, 3.88, and 3.43 eV. Smaller bandgaps can also be identified in the graph, which is probably due to impurities or edge structure/passivation.

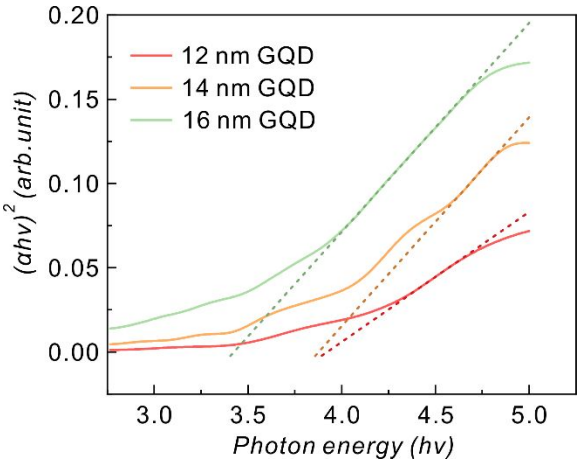

Figure S5. Tauc plot from the absorbance spectra of GQDs