Electronic Supplementary Material (ESI) for New Journal of Chemistry.

Supplementary Information

Generation of Graphene Quantum Dots from Oxidative Cleavage of Graphene Oxide Using Oxone Oxidant

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Experimental Section

Preparation of graphene quantum dots (GQDs) by sono oxidation: Graphene oxide (GO, 100 mg) was synthesized by Hummers method. It was completely dissolved in 100 mL DMF after which 500 mg of oxone was added and the solution was mixed. Sono oxidation of the mixture was performed in an ultrasonication bath (500 W) for 1 h at room temperature. The product suspension was filtered through a 200 nm nano-porous membrane to separate large graphene nanoparticles and GO. The final product solution was further dialyzed in a dialysis bag (retained molecular weight: 2000 Da) for 3 days to remove metal ions.

Preparation of GQDs by photo oxidation: GO (100 mg) was completely dissolved in 100 mL DMF. Five hundred mg of oxone was added followed by mixing. Photo oxidation was performed using UV lamp reactor equipment with an irradiation distance of 2 cm from the source (UV 100, Power Arc). UV lamp was placed axially in a quartz tube inside the reactor. The reaction was initiated by exposing the quartz tube to a mercury UV lamp (365 nm, 100 W) for 1 h at room temperature. The product suspension was filtered through a 200 nm nano-porous membrane to separate large graphene nanoparticles from GO. The final product solution was further dialyzed in dialysis bag (retained molecular weight: 2000 Da) for 3 days to remove metal ions.

Characterization: The morphologies of GQDs were measured by high-resolution transmission electron microscopy with the microscope operated at an accelerating voltage of 200 kV (JEOL JEM-2100 Field Emission Gun HR-TEM) and tapping-mode AFM images. Height profiles of GQDs and GO were obtained using a XE-100 AFM system (Park System, Inc., Korea). AFM samples were prepared by spin-coating a washed Si wafer at 4000 rpm. Raman spectroscopy measurements were taken using a confocal Raman system (WiTec, Alpha 300R) with a 532-nm laser. XPS measurements were taken in a SIGMA PROBE (ThermoVG) using a monochromatic Al-Kα X-ray source at 100 W. Surface analyses of GO were performed by Field-emission scanning electron microscopy (SEM) (JSM-6701F/INCA Energy, JEOL). All UV-vis absorption spectra were recorded using a 8453 UV-Vis spectrophotometer (Agilent Technologies, USA). Photoluminescence (PL) spectra were obtained on a Cary eclipse fluorescence spectrophotometer (Agilent Technologies, USA). Fluorescence lifetime decay was measured using a confocal microscope (MicroTime-200, Picoquant, Germany) with a 20x objective. A pulsed diode laser (375 nm with a pulse width of ~240 ps and an average power of ~1 μW) was used as an excitation source. An avalanche photodiode detector (PDM series, MPD) was used to collect whole emissions from the samples. Time-correlated single-photon counting (TCSPC) technique was used to count fluorescence photons.

Quantum yields (QYs) measurements:

Rhodamine B in water (QYs=0.31) was chosen as a standard. The quantum yields of GQDs in water were calculated according to:

$$\Phi = \Phi_r \times \frac{\mathrm{I}}{\mathrm{I}_r} \times \frac{\mathrm{A}_r}{\mathrm{A}} \times \frac{n^2}{n_r^2}$$

Where Φ is the quantum yield, I is the measured integrated emission intensity, n is the refractive index of the solvent, and A is the optical density. The subscript "r" refers to the reference standard with known quantum yield. In order to minimize re-absorption effects, absorbencies in the 10 mm fluorescence cuvette were kept under 0.1 at the excitation wavelength (340 nm).

Table S1 QYs of the GQDs from sono and photo oxidation by using Rhodamine B as a reference.

Sample	Integrated emission intensity *(I)*	Abs. at 340 nm *(A)*	Refractive index of solvent *(η)*	Quantum Yield *(φ)*
Rhodamine B	11562	0.072	1.33	0.31
GQDs by Sono oxidaiton	2120	0.071	1.33	0.056
GQDs by Photo oxidaiton	1762	0.068	1.33	0.049

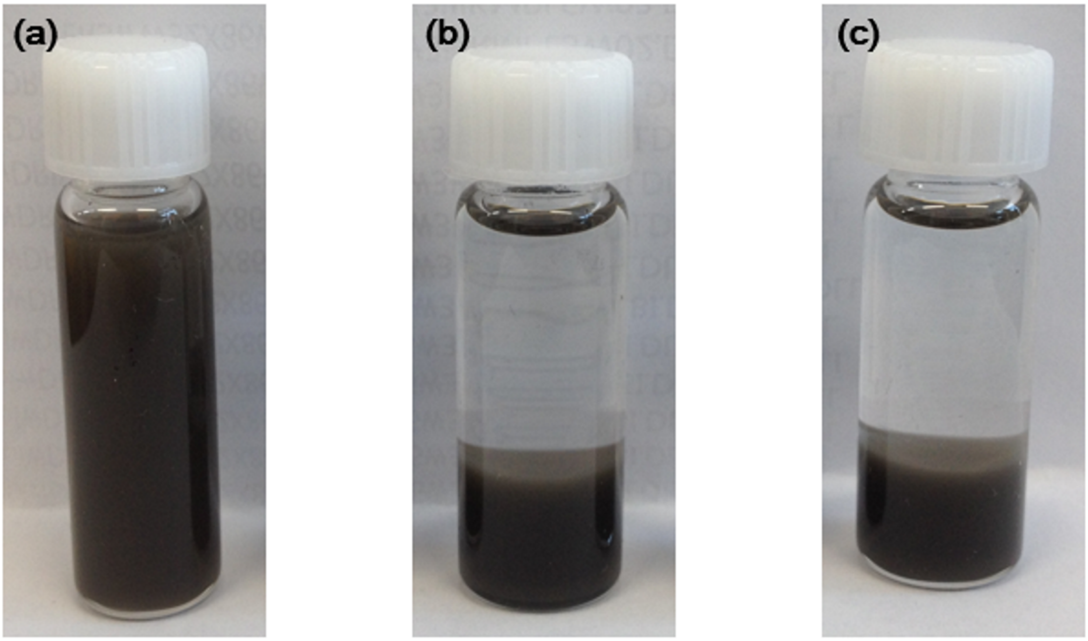

Figure S1. Photographs of (a) before the reaction (b) after sono oxidation reaction (c) after photo oxidation reaction.

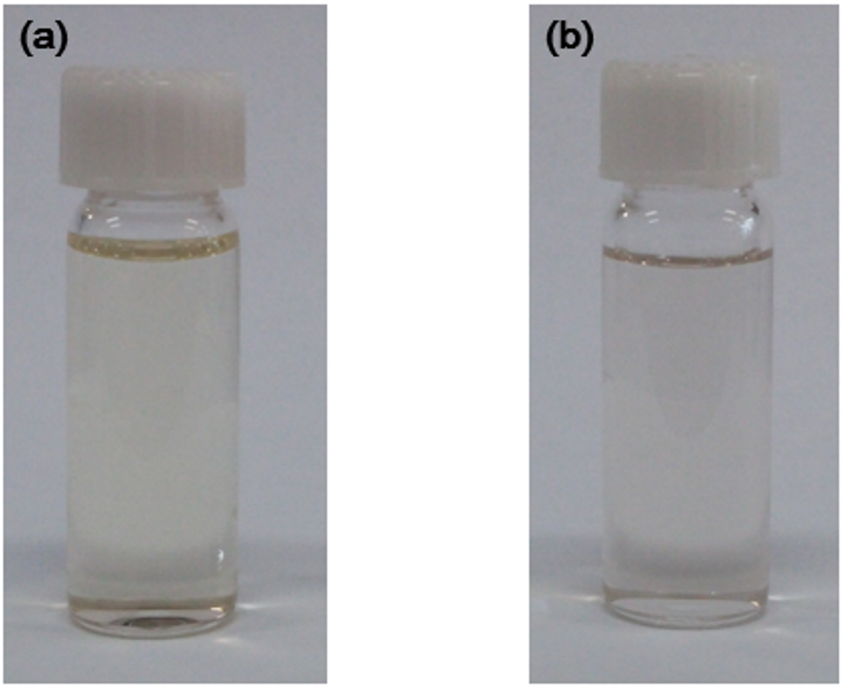

Figure S2. Photograph of synthesized GQDs solution (a) by sono oxidation reaction (b) by photo oxidation reaction.

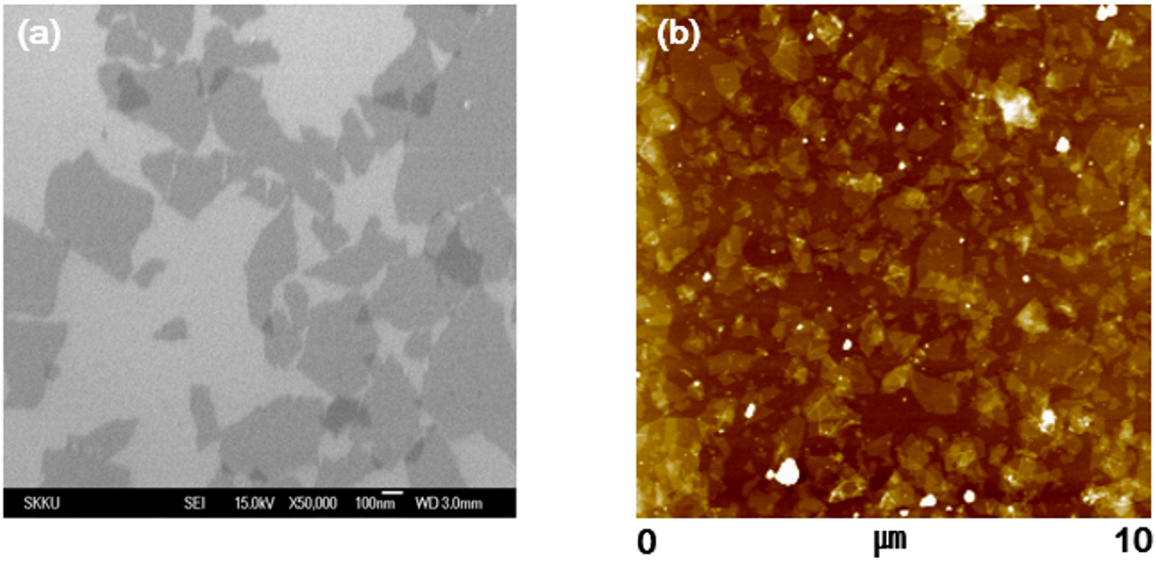

Figure S3. GO as a starting materials (a) SEM image (b) AFM image.

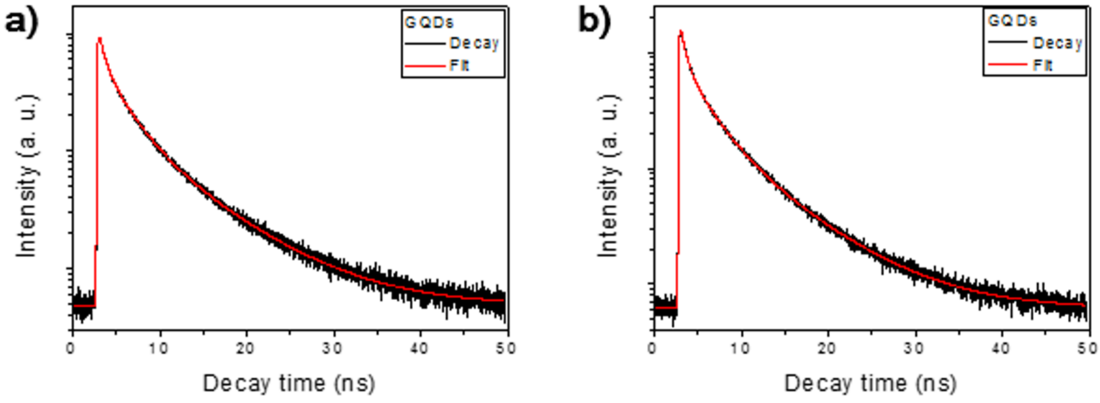

Fig. S4 Time-resolved PL decay profiles of synthesized GQDs (a) by sono oxidation reaction (b) by photo oxidation reaction.

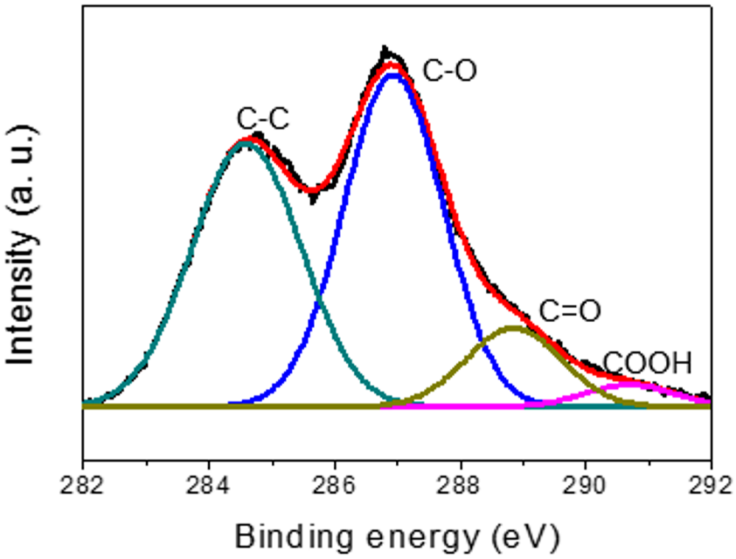

Figure S5. High resolution XPS spectra of synthesized GO.

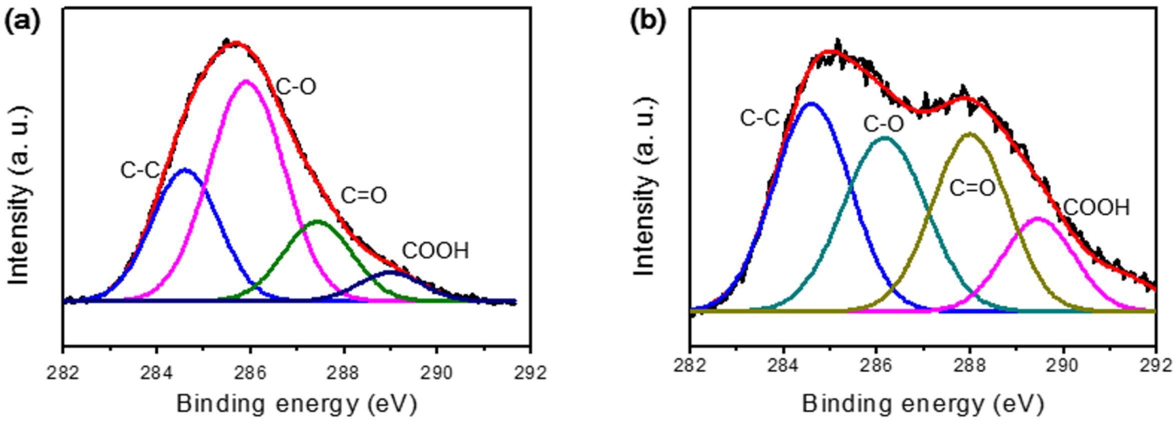

Figure S6. High resolution XPS spectra of synthesized GQDs (a) by sono oxidation reaction (b) by photo oxidation reaction.