

Structural and optical properties of N-doped graphene quantum dots

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Abstract

Graphene quantum dots (GQDs) are atomically-thick, conductive nanosheets of sp^2 hybridized carbons, which exhibit size-, shape-, and edge-dependent electrical/optical properties. [1-3] Therefore, it is essential to be able to engineer the size and edge state of GQDs by their functionalization because this can alter the energy states of GQDs as that of graphene, thereby controlling their properties. Nitrogen (N) atom, having a comparable atomic size and five valence electrons for bonding with carbon atoms, has been widely used for chemical doping of carbon nanomaterials. For instance, doping of graphene with substituent N heteroatoms could effectively modulate the band gap of graphene, thereby exhibiting novel properties for device applications. [4-5] In this work, we studied the effect of N atoms on the structural and optical properties of N-doped GQDs (NGQDs) prepared by hydrothermal cutting of graphene sheets and subsequent hydrazine treatment.

Figs. 1(a) and (b) show typical transmission electron microscopy (TEM) images of GQDs and NGQDs. Inset of Fig. 1(a) shows a high-resolution TEM (HRTEM) image of a typical GQD, indicating high crystalline structure with a lattice parameter of 0.244 nm, corresponding to (1120) lattice fringes of graphene. [3,6] It is also possible to see the hexagonal unit cell of GQDs in the magnified HRTEM image, [7] demonstrating that the GQDs consist of graphene. The average sizes of GQDs and NGQDs analyzed from the TEM images are estimated to be 5.03 ± 0.05 and 5.55 ± 0.05 nm, respectively. The NGQDs are not only n-type but also larger than undoped GQDs while the edge-related defects are reduced, resulting from the bonding of the increased pyridinic N atoms with C atoms at the edge of GQDs, as confirmed by Raman spectroscopy and x-ray photoelectron spectroscopy.

Fig. 2(a) and (b) show absorption and photoluminescence (PL) spectra of GQDs and NGQDs in the UV to visible ranges. The absorption band of GQDs is peaked at ~ 300 nm, and after doping they show an additional absorption peak at ~ 354 nm, originating from the impurity energy level formed by the N doping, as confirmed by electron energy loss spectroscopy. The GQDs emit bright blue PL peaked at 418 nm, and after hydrazine treatment the PL emission is redshifted by ~ 15 nm and its decay becomes slower, resulting from the N-related impurity level.

References

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Figures

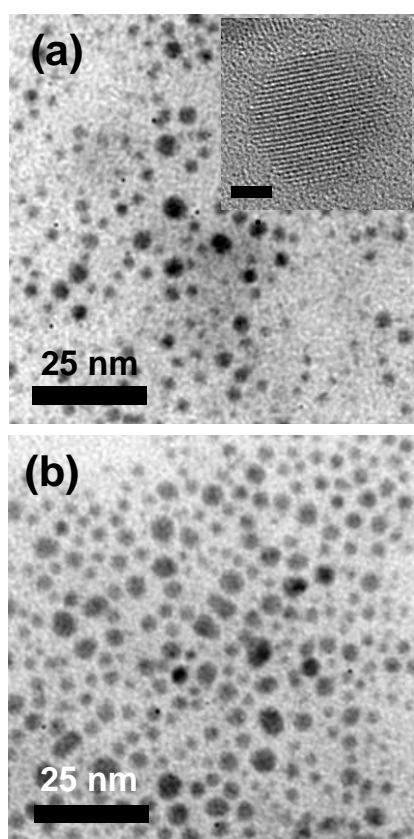


Fig. 1. Transmission electron microscopy images of (a) GQDs and (b) NGQDs.

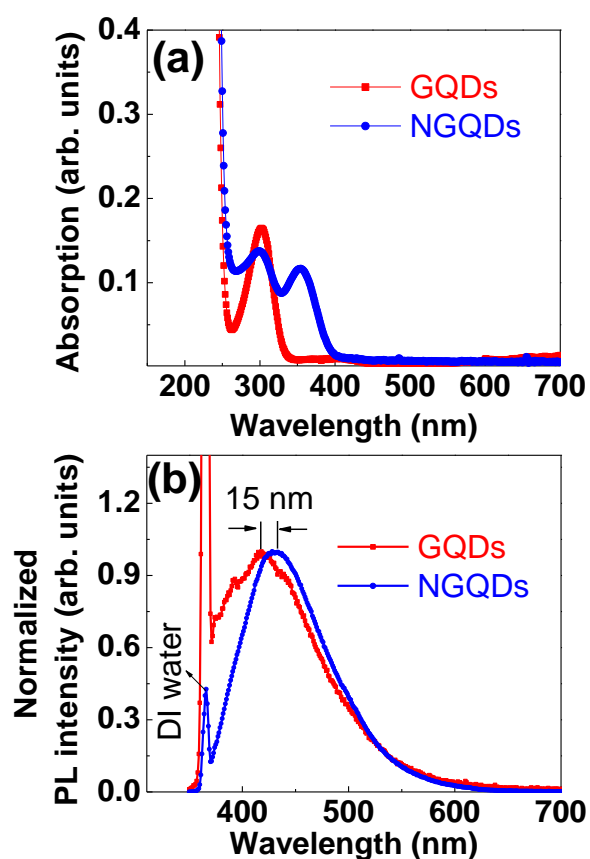


Fig. 2. (a) Absorption and (b) photoluminescence spectra of GQDs and NGQDs in the UV to visible ranges.