Green, simple and large scale synthesis of N-doped graphene quantum dots with uniform edge groups by electrochemical bottom-up synthesis

Linfan Tian [1, 2], Siwei Yang [3], Zhongyang Wang [1, 2]

1. Shanghai Advanced Research Institute, Chinese Academy of Science, Shanghai, China
2. School of Physical Science and Technology, ShanghaiTech University, Shanghai, China
3. Shanghai Institute of Microsystem and Information Technology, Chinese Academy of Science, Shanghai, China.

Corresponding author: wangzy@sari.ac.cn

Abstract

As a new kind of photoluminescence (PL) material, graphene quantum dots (GQDs) have drawn extensive attention of researchers because of their remarkable properties. It deserves to note that the specific PL mechanism of GQDs is the fundamental issue for its development. It has been demonstrated that the size, heteroatoms, edge carbon atoms, defects and edge groups all obviously affect the PL progress of GQDs [1, 2]. As the inevitable constituent part of GQDs, edge groups have a significant influence on the PL spectra. However, by now, the real role of edge groups in PL process is unclear. This can be due to the unavoidable different edge groups in most reported GQDs [1]. Thus, it is very necessary for the development of new preparation approach for GQDs with uniform edge groups.

We developed an electrochemical bottom-up synthesis of N-doped GQDs (N-GQDs) with large amount of well-defined edge groups for the first time [5]. This progress is green (both atom utilization and yield are higher than 95 %, without by-products), simple and suitable for large scale synthesis. The exclusive edge group of the obtained N-GQDs is -NH_2 because of the amino oxidative coupling of o-phenylenediamine, which results in unique excitation wavelength independence behavior. The high quantum yield is believed to originate from the localized π state due to the graphitic doping of nitrogen [3, 4]. The edge groups have little influence on the spectral resolved PL lifetime as well as the size distribution [5].

References

[1] J. Sun, S. W. Yang, Z. Y. Wang, G. Q. Ding, et.al. Part. Part. Syst. Charact., **32** (2014), 434.
[2] D. Y. Pan, J. C. Zhang, Z. Li, et.al. Chem. Comm. **46** (2010), 3681.
[3] T. Kondo, S. Casolo, T. Suzuki, T. Shikano, J. N. Nakamura, et.al. Physical review B, **86** (2012), 035436.
[4]L. Y. Zhao, R. He, K. T. Rim, T. Schiros, A. N. Pasupathy et.al. Science, **333** (2011), 999.
[5]L. F. Tian, S. W. Yang, Z. Y. Wang, et.al. RSC. Adv., 107 (2015), 241905.

Figures

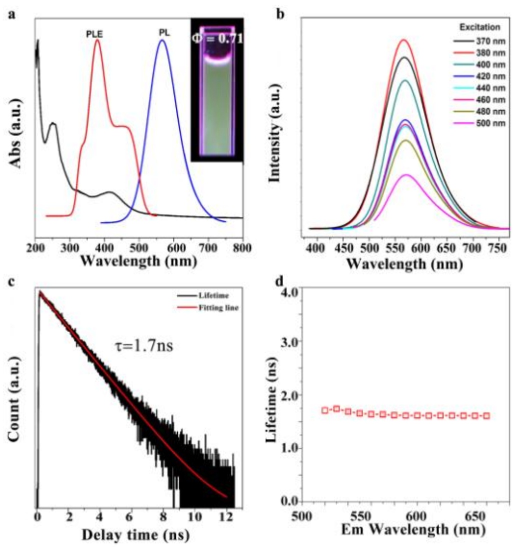

Figure 3 (a) UV-vis absorption, PL (λ_{ex}=420 nm) and PLE (λ_{em}=569 nm) spectra of N-GQDs aqueous solution, inset: the photograph of N-GQDs aqueous solution under 365 nm UV light. (b) PL spectra of N-GQDs aqueous solution at different excitation wavelengths (370-500 nm). (c) The lifetime of N-GQDs measured at 25 °C (λ_{ex} =420 nm and λ_{em}=570 nm). (d) PL decay curves of N-GQDs tested at 25 °C with 10 nm increments of progressive emission wavelength (λ_{ex}=420 nm).