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Full Length Article

Pulsed laser ablation based synthetic route for nitrogen-doped graphene quantum dots using graphite flakes

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ABSTRACT

Graphene quantum dots (GQDs) prepared by pulsed laser ablation in liquid (PLAL) process face barriers to commercialization due to their poor optical properties and the use of expensive carbon precursors. In this work, we report a one-step route for the preparation of N-doped graphene quantum dots (NGQDs) with excellent optical properties using a low-cost carbon precursor. The as-prepared NGQDs exhibited excellent optical properties and high quantum yield compared to pristine graphene quantum dots (0.8% → 9.1%) due to the presence of the N atoms. A possible recombination mechanism of NGQDs was investigated by time-resolved photoluminescence spectroscopy. The increase of N atoms incorporated in the GQDs resulted in an increased fraction of the short recombination lifetime from the intrinsic state. We also report a possible mechanism for the formation of the N atoms in the GQDs structure during the PLAL process, which is explained based on the plasma plume, cavitation collapse, and nitrogen precursor decomposition model.

1. Introduction

Quantum-sized particles have attracted considerable attention from researchers working on various applications such as those related to the optoelectronic and biomedical fields. In recent years, inorganic semiconductor quantum dots (e.g., Cd and Zn based quantum dots) have been intensively studied as promising optoelectronic materials owing to their extraordinary and unique properties such as bandgaps and narrow bandwidths that are easily controllable by controlling the size of the Quantum dots (QDs) during the synthesis process [1,2]. Nevertheless, the practical application of QDs remains severely limited due to their highly toxic and harmful effects on human cells [3–5].

Recently, graphene quantum dots (GQDs) have received much attention as the most suitable alternative due to their low toxicity, low cost, strong luminescence, and facile synthesis. Typically, GQDs have been fabricated mainly by a wet chemical route using expensive carbon precursors such as graphene, graphene oxide (GO), carbon nanotube (CNT), and carbon nanofiber (CNF) [6–8]. In addition, the wet chemical routes are usually performed under strong acidic conditions, which require complex, time-consuming washing steps [9–13]. Thus, it is necessary to develop a simple and cost-effective method for the fabrication of GQDs.

An alternative to the wet chemical method is pulsed laser ablation in liquid (PLAL) for the synthesis of GQDs. The PLAL method is facile, simple, and environmentally friendly because it does not require a harmful acidic chemical reactant and post-purification steps. Previously, our group proposed a PLAL process for preparing GQDs using coal as an abundantly available and inexpensive carbon precursor [14]. However, the as-prepared GQDs showed poorer optical properties (e.g., UV–Vis and photoluminescence (PL) intensity) compared to those produced by the chemical method. Hence, to overcome such issues, it is essential to develop a synthetic method that enhanced the optical properties of GQDs using the PLAL process.

From the perspective of the pulsed laser process, the nanoparticles prepared by pulsed laser ablation are highly dependent on the initial states of the precursor (e.g., solid target and/or dispersion in liquid medium) [15]. Thus, many researchers have attempted to modify the PLAL process to improve the optical properties of the prepared nanoparticles. Sasaki and co-workers proposed the synthesis of Au and ZnO nanoparticles via the PLAL process with ultrasonic waves [16]. The Au and ZnO nanoparticles exhibited enhanced absorbance due to the application of the ultrasonic waves during PLAL. Also, Hamad and co-

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