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Carbon dots derived from rose flowers for tetracycline sensing

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ABSTRACT

Herein, an innovative and simple method for synthesizing carbon dots (CDs) with satisfactory fluorescence has been successfully established while rose flowers served as carbon source for the first time. Meanwhile, the fluorescence (FL) mechanism of current CDs was elucidated in detail by fluorescence, UV–vis, HR-TEM, and FTIR-based analyses. Subsequently, this type of CDs was employed for detecting tetracycline (TC) on the basis of the interactions between TC and CDs, and allowed quenching their fluorescence. Moreover, the proposed analytical strategy permitted detecting TC in a linear range of $1.0 \times 10^{-8} - 1.0 \times 10^{-4}$ mol/L with a detection limit of 3.3×10^{-9} mol/L at a signal-to-noise ratio of 3. Significantly, the CDs described here were further applied for fluorescent coding, demonstrating their promising future towards various applications in analytic science.

1. Introduction

Carbon nanomaterials, mainly including carbon nanotubes, fullerenes, graphene, and carbon nanofilms, are promising scaffolds with fantastic sizes less than 10 nm [1], and have been playing critical roles in various fields such as biological markers [2], biochemistry and biomedicine [3]. Recently, appearances of fluorescent carbon dots (CDs) have exhibited tremendous impact on the advancement of various fields including electronics, photonics, energy, catalysis, and medicine [4], and attracted increasing interests owing to their less-toxicity, biocompatibility [5], photostability [5–9] and ease of preparation [10,11]. Thereby, CDs have been considered as a promising choice in potential applications for biosensing [12], catalysis [13], and imaging [14–16]. For example, CDs were utilized for conventional bioimaging of Ehrlich ascites carcinoma cells [17]. Meanwhile, a variety of methods for preparing CDs have been demonstrated in the past decades on account of their advanced characteristics, including electrochemical oxidation processes [18,19], arc discharge [20], hydrothermal cutting strategies [21], chemical oxidation methods [1,22,23], combustion/thermal, and so on. Nevertheless, some of the proposed methods above required high costs, tedious steps, or special equipment, thus developing simple and rapid methods for synthesizing CDs are still meaningful.

Tetracyclines (TCs), as effective antibiotics, have been widely used in the therapy of human and animal infections by Gram-positive, Gram-negative bacteria and show effective activity for rickettsia, viral infection on the basis of their broad spectrum activity against pathogenic microorganisms, nice oral absorption, and relatively less toxicity and low cost [24,25]. However, irrational abusing of TCs may lead to increasing potential risk by residues in human body. To date, TCs abuse exist in our daily food such as milk [26] and honey [27]. Worse still, a large amount of data has demonstrated that long-term and repeated intake of TCs may affect the growth and formation of teeth. For the purpose of monitoring TCs, diverse techniques such as high performance liquid chromatography (HPLC) [28], chemiluminescence [29], capillary electrophoresis (CE) [30], and dipstick colorimetric methods [31] have been developed. Howbeit, most of these approaches exhibited more or less defects of sophisticated and costly instruments or complicated processes. Thus, the new and efficient methods for analyzing TCs remain necessary.

Hereby, a simple and efficient method for synthesizing CDs has been built up by using rose flowers as carbon source for the first time (Fig. 1), which showed blue fluorescence together with a quantum yield of 13.45%. Subsequently, we applied this type of CDs for sensitively and selectively assaying TC based on the mechanism that the energies of CDs were captured for forming new bonds between the surface groups of TC and CDs [32]. In addition, the synthesized CDs were employed for detecting TC residues in human urine samples, suggesting their practicability. In short, we have broadened a new road for sensing and fluorescent staining.

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2. Experimental

2.1. Chemicals and materials

The rose flowers were bought from flower market (Chongqing, China). Phosphorus pentoxide (P_2O_5), TCs including tetracycline (TC), oxytetracycline (OTC), aureomycin, and doxycycline (DOXC), streptomycin, lincomycin, cysteine, histidine, glutathione, ibuprofen, ammonium thiocyanate, glucose, p-aminobenzoic acid, procaine were obtained from Shanghai Sangon Biotechnology Co., Ltd. (Shanghai, China). Ascorbic acid, disodium hydrogen phosphate (Na_2HPO_4) and sodium dihydrogen phosphate (NaH_2PO_4), sodium chloride (NaCl) were purchased from Dingguo Changsheng Biotechnology Co., Ltd. (Beijing, China). Ultrapure water, 18.25 MΩ, produced with an Aquapro AWL-0502-P ultrapure water system (Chongqing, China) was applied to all the following experiments.

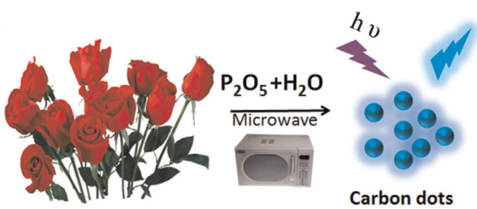

Fig. 1. Schematic illustration of synthesizing CDs based on rose flowers.

2.2. Instrumentation

All fluorescence data were performed on a Hitachi F-7000 fluorescence spectrophotometer (Tokyo, Japan) with excitation slit set at 5 nm band pass and emission at 5 nm band pass in 1 cm × 1 cm quartz cell. Meanwhile, UV/vis spectra were recorded by a Shimadzu UV-2450 spectrophotometer (Tokyo, Japan). The high resolution transmission electron microscopy (HR-TEM) images were taken by a TECNAI G2 F20 microscope (Portland, America) at 200 kV and Fourier Transform Infrared Spectroscopy (FTIR) spectra were recorded by SHIMADZU IRprestige-21 spectrometer (Tokyo, Japan). Elemental and functional groups analyses were obtained by ESCALAB 250 X-ray photoelectron spectrometer. The quantum yields were obtained by using Absolute PL quantum yield spectrometer C11347 (Hamamatsu, Japan). The powder of CDs was obtained by lyophilisation in PiloFD8-4.3V (California, USA). The thermostatic water bath (DF-101s) was purchased from Gongyi Instrument Co., Ltd. (Gongyi, China). A Fangzhong pHS-3C digital pH meter (Chengdu, China) was used to measure the pH values of the aqueous solutions and a vortex mixer QL-901 (Haimen, China) was used to blend the solution.

2.3. Synthesis of CDs

The rose flowers as the carbon resource were first applied to synthesize CDs here. Briefly, rose flowers were on baking in the oven with 60° until the petals can be ground into powder. Then, 10 mL ultrapure water was added into the mixture of 10 mg petals

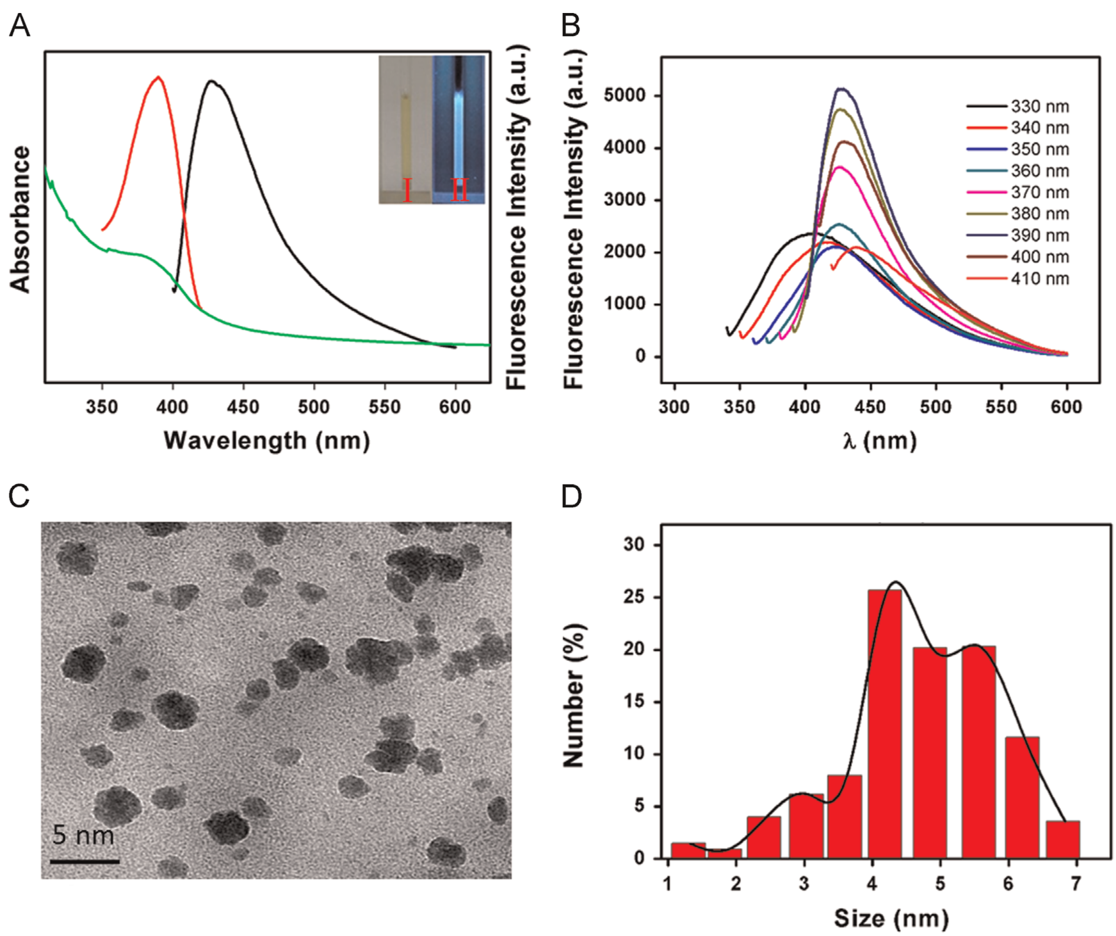

Fig. 2. (A) Fluorescence excitation and emission spectra and UV–vis spectrum of CDs. Inset: photographs of CDs solution (I,II); (B) emission spectra of CDs for varying excitation wavelengths; (C) HR-TEM images of CDs; (D) size distribution analysis of CDs. (For interpretation of the references to color in this figure, the reader is referred to the web version of this article.)