

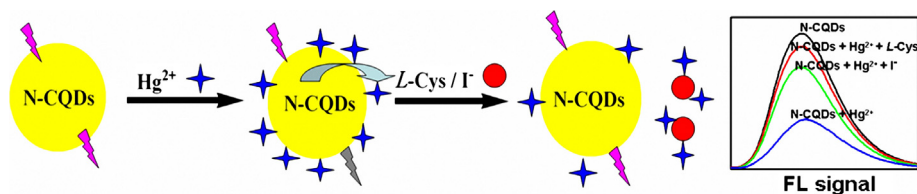
Short Communication

Nitrogen-doped carbon quantum dots as fluorescent probe for “off-on” detection of mercury ions, L-cysteine and iodide ions

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GRAPHICAL ABSTRACT

An “off-on” method for highly sensitive and selective detection of Hg^{2+} and L-cysteine (L-Cys) or Hg^{2+} and I^- using home-made nitrogen-doped carbon quantum dots (N-CQDs) as fluorescent probe was reported.



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ABSTRACT

The application of fluorescent nanoparticles to the detection of inorganic ions and organic compounds has been attracted wide attention recently. In this paper, an “off-on” method for highly sensitive and selective detection of Hg^{2+} and L-cysteine (L-Cys) or Hg^{2+} and I^- using home-made nitrogen-doped carbon quantum dots (N-CQDs) as fluorescent probe was reported. The N-CQDs with a fluorescence quantum yield of 42.2% were prepared using tartaric acid, citric acid and ethanediamine as the precursors in the oleic acid media. The fluorescence of the obtained N-CQDs could be quenched selectively and sensitively by the addition of Hg^{2+} (turn-off) with a detection limit of 83.5 nM. When L-Cys or I^- was added into the N-CQDs- Hg^{2+} system, the fluorescence was recovered effectively (turn-on). This process could be used to the detection of L-Cys or I^- with a detection limit of 45.8 and 92.3 nM, respectively.

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1. Introduction

The pollution of heavy metal ions has become a serious worldwide problem due to their severe risks to human health and the environment. As one of the most toxic heavy metals, mercury with the feature of strong toxicity and bioaccumulation can cause seri-

ous human health problems even at very low concentration [1,2]. On the other hand, L-cysteine, as a small-molecular-weight biological thiol-containing amino acid, is essential in maintaining biological redox homeostasis. An abnormal level of L-Cys has been linked to many diseases, such as slow growth, liver damage, skin lesion, Alzheimer's disease and cardiovascular disease [3–6]. It is also well-known that the content of iodine is directly related to human health [7]. Iodine deficiency could lead to some serious problems such as intellectual disability. Iodine excess could also cause the disease. It is of great necessity to develop rapid and user-friendly

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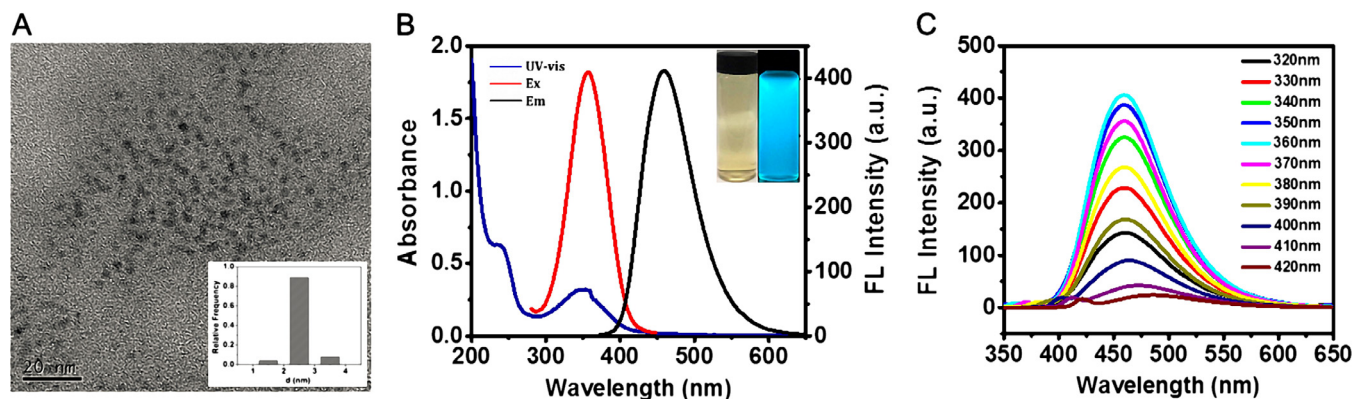


Fig. 1. The TEM image (A), the UV-vis and photoluminescence spectrum (B), and the excitation-dependent emission spectrum (C) of the N-CQDs. Inset of A, the size distribution of the N-CQDs. Inset of B, the photographs under illumination of white (left) and UV (365 nm, right) light.

method for the detection of mercury ions, L-cysteine and iodide ions.

Recently, the application of fluorescent nanoparticles to the detection of inorganic ions and organic compounds, temperature sensing, bioimaging and nanomedicine has been attracted wide attention [8–12]. Various nanomaterials including fluorescently doped silicas and sol-gels, hydrophilic polymers (hydrogels), hydrophobic organic polymers, semiconducting polymers dots, quantum dots, carbon dots, other carbonaceous nanomaterials, upconversion nanoparticles, noble metal nanoparticles (mainly gold and silver), etc. have been investigated for these applications [13–16]. Among them, carbon dots have received particular concern because of their excellent optical properties, good biocompatibility, great aqueous solubility, and simple synthesis [17]. Heteroatom-doped carbon dots which often possess high fluorescent quantum yield have even become the focus.

Xu et al. developed a new fluorescent probe based on ensemble of gold nanoclusters and polymer protected gold nanoparticles for turn-on sensing of L-cysteine [18]. Li et al. developed a simple and distinctive method for the ultrasensitive detection of Cu^{2+} and Hg^{2+} based on surface-enhanced Raman scattering using cysteine-functionalized silver nanoparticles attached with Raman-labeling molecules [19]. Amjadi et al. investigated a carbon dots-silver nanoparticles fluorescence resonance energy transfer system as a novel turn-on fluorescent probe for selective determination of cysteine [20]. Lin et al. prepared europium-decorated graphene quantum dots as a novel “off-on” fluorescent probe for the label-free determination of Cu^{2+} and L-cysteine [21]. Developing convenient methods based on novel nanomaterials for the detection of metal ions and biological active substances is still of great significance.

In this report, water-soluble N-CQDs prepared from tartaric acid, citric acid and ethanediamine by a facile one-pot solvothermal method were used to the “off-on” detection of Hg^{2+} , L-Cys and I^- with high sensitivity and selectivity. The N-CQDs could be obtained conveniently, and the established method has a promising prospect for the detection of real samples.

2. Experimental

2.1. Chemicals

Citric acid monohydrate (CA), tartaric acid, ethanediamine and oleic acid were all analytical reagents. They were purchased from Shanghai Sinopharm Chemical Reagent Co. Ltd. (China). Quinine sulfate (AR) was purchased from Aladdin Industrial Corporation (Shanghai, China). Ultrapure water was prepared with a Milli-Q system (Millipore, Bedford, MA, USA). Other reagents were all analytical and were used without further purification.

2.2. Characterization of the N-CQDs

The fluorescence spectra were obtained by a Varian Cary Eclipse Fluorescence Spectrophotometer. Transmission electron microscopy (TEM) and high-resolution TEM (HRTEM) images were recorded using FEI Tecnai G2 F20 instruments. The X-ray photoelectron spectroscopy (XPS) spectra were recorded using a Thermo ESCALAB 250Xi multifunctional imaging electron spectrometer. A UV-2550 spectrophotometer was used to record the UV-vis spectra. A Magna-IR 750 Fourier transform infrared spectrometer (Nicolet) was used to record the FTIR spectra. The fluorescence decay experiments were performed using an Edinburgh Instruments FLS920 fluorescence spectrometer.

2.3. Preparation of the N-CQDs

The N-CQDs were prepared according to a simple one-step solvothermal method reported by our group [22,23]. Typically, CA monohydrate (1.5 g) and tartaric acid (1.5 g) were placed in a three neck flask, and then ethanediamine (5 mL) and oleic acid (30 mL) were added. The mixture was heated at 220 °C for 30 min under vigorous magnetic stirring. The colorless solution changed to a clear brownish black solution as the reaction progressed. After the reaction, the solution was cooled at room temperature, and then black solid precipitate was obtained directly. The precipitate was washed sufficiently with *n*-hexane, dispersed in ultrapure water and centrifuged at 6000 rpm for 30 min to remove large particles product.

3. Results and discussion

The N-CQDs were prepared by a facile solvothermal method with oleic acid as the reaction media. The product was then characterized by TEM, XPS, FTIR, UV-vis and photoluminescence spectrum. From the TEM image (Fig. 1A) we can find that the resulting product has good dispersion with a narrow size distribution in the range of 1 to 4 nm (inset of Fig. 1A). The average diameter of the N-CQDs is about 2.66 nm. The obtained N-CQDs are amorphous as the HRTEM images do not show any discernible lattice fringes.

There are two distinct absorbance bands centered at 239 and 351 nm (Fig. 1B). The peak at 239 nm can be ascribed to the π - π^* transitions of the aromatic $\text{C}=\text{C}$ sp^2 domains which cannot produce observed fluorescence signal [24,25]. The other absorption peak at 351 nm can be ascribed to the trapping of excited state energy of the surface states, which can lead to strong fluorescence [26,27]. The fluorescent spectrum for the obtained N-CQDs conformed to the UV-vis absorption features. Fig. 1C shows that the emission peak remains essentially unchanged with the variation

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