



ICESD 2014: February 19-21, Singapore

Determination of Hypochlorous Acid in Tap Water Using Highly Fluorescent Graphene Oxide

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Abstract

We report on the effect of hypochlorous acid (HClO) in the fluorescence emission of a fluorescent graphene oxide modified by hexylenediamine (GO-C₆NH₂). The synthesis process and the optical property of GO-C₆NH₂ were also introduced in detail. We found a strong quenching effect by HClO on the fluorescence emission of the fluorescent graphene oxide. The fluorescent quenching efficiency of GO-C₆NH₂ establish as a function of the amount of HClO, which the fluorescence intensity ratio is proportional to the different concentrations of HClO employed for the measurement. The results show that this fluorescent naomaterials are a promising tool for sensing trace HClO in tap water.

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Selection and peer review under responsibility of Asia-Pacific Chemical, Biological & Environmental Engineering Society

Keywords: Fluorescent graphene oxide, Hypochlorous acid, Quenching fluorescence

1. Introduction

As we know, hypochlorous acid (HClO) is playing an important role in water treatment and living organisms [1]. Endogenous HClO is essential to life and shows vital antibacterial properties. Normally, Hypochlorite is used in the concentration range of 10⁻⁵–10⁻² M [2], since concentrated HClO is a potential health hazard to human and animals [3]. It has been reported that excessive or misplaced instake of HClO was damaged to host tissues (particularly proteins), observed in and led to numerous diseases (e.g., atherosclerosis, kidney disease, and some cancers) [4], which may relate to the reaction of hypochlorite with

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DNA, RNA, fatty acids, cholesterol, and proteins *in vivo*. On the other hand, it is not fully understood about the exact interaction mechanism of hypochlorite to destroy invading microorganisms and cause these diseases. Therefore, a rapid and sensitive method for the determination of hypochlorite is desirably needed to investigate the functions of HClO in water.

It is a vital task for detection of trace amount of HClO in water with a broad range of implications from national security to the cleanup of demilitarized installations. So far, there are many methods available for the hypochlorite determination, such as the normalized and well-known iodometric titration, many colorimetric methods based on reaction of hypochlorite with organic reagents [5]. However, these strategies require cumbersome preparative procedures of sensing molecule and/or unwieldy sample preparations. Furthermore, they had to be carried out either in organic medium or organic/water medium, directly limiting their use in real-life situations. Therefore, nanotechnology is ideally suited to serve the Nation needs by providing new materials and methods that can be employed for trace HClO detection. Fluorescent nanomaterials, like nanoscaled graphene oxide, have the potential to be applied in trace HClO detection in a number of drinking water environments. The fluorescent graphene oxide has recently received increasing attraction due to its special surface properties and high stability. The fluorescence quantum yield (QY) of graphene oxide through the functional modification with alkylamines [6-9] has been reported to be greatly enhanced. In our work, we report here the addition of HClO shows great effect on the fluorescence emission of the fluorescent graphene oxide. It is proposed that the fluorescent graphene oxide can react with HClO, leading to the quenching fluorescence.

2. Experimental section

2.1. Materials

Natural graphite flakes, N,N-dimethylformamide (DMF), dichlorosulfoxide, tetrahydrofuran (THF), 1, 6-hexylenediamine, were purchased from Shanghai Chemical Reagent Co. The pH of solution was adjusted by phosphate buffer. The phosphate buffer was prepared by disodium hydrogen phosphate dodecahydrate ($\text{Na}_2\text{HPO}_4 \cdot 12\text{H}_2\text{O}$) and sodium dihydrogen phosphate dihydrate ($\text{NaH}_2\text{PO}_4 \cdot 2\text{H}_2\text{O}$). Aqueous solutions were all prepared using ultrapure water ($18.2 \text{ M}\Omega \cdot \text{cm}$) from a Millipore water purification system.

2.2. Synthesis of fluorescence graphene oxide

The starting graphene oxide powder was obtained from natural graphite by a modified Hummers method [10]. The fluorescent graphene oxide ($\text{GO-C}_6\text{NH}_2$) with the modification of hexylenediamine was prepared by the following procedure. About 20 mg of dried graphene oxide was dispersed in DMF (5 mL), then, refluxed in 20 mL of SOCl_2 at 80°C for 24 hours. The supernatant was discarded and the remaining solid was washed with anhydrous THF two times to remove excess SOCl_2 and DMF after the centrifugation at 10000 rpm for 10 min. Then, the activated GO acyl chloride (GO-COCl) was stirred at 80°C for 48 hours following by the addition of hexylenediamine. The reaction solution then dispersed in ethanol (20 mL). A light yellow supernatant was obtained after the mixture was vacuum filtered. The suspension shows bright blue fluorescence under UV illumination and the maximum fluorescence intensity is located at 440 nm with a 365 nm excitation wavelength.

3. Results and discussion

It is shown in figure 1 that highly fluorescent GO nanosheets were successfully synthesized through the

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