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Fluorescence quenching capillary analysis for determining trace-level nitrite in food based on the citric acid/ethylenediamine nanodots/nitrite reaction

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ABSTRACT

We found that nitrite after protonation can react with amine radical on citric acid/ethylenediamine carbon nanodots (CA/EDA-CDs) to form nitrosamines, and fluorescence quenching of CA/EDA-CDs occurred during this process. Using the reaction mechanism a fluorescence quenching capillary analysis (FQCA) was developed. After optimized reaction conditions, the following results were obtained: the required concentration of CA/EDA-CDs was 12 mg/L, HCl concentration was 32 mmol/L, and the reaction conducted in room temperature for 20 min. Under optimized conditions, FQCA has a linear response in 20–500 μg/L in which RSD was less 4.5% (n = 11), the detection limit was 6.5 μg/L and the recovery was in 95–105%. The measured results were consistent with the national standard method. FQCA has been used for determining nitrite in foods and nature waters. The capillary in FQCA was used as the container for CA/EDA-CDs/NO_2^- reaction and NO_2^- determination, and realized trace-level analysis for micro-volume samples (< 10 μL/time).

1. Introduction

Nitrite exists in a variety of foods and natural waters and can transform into various nitrogen oxides after entering the human body (Lundberg, Weitzberg, & Gladwin, 2008). Presently, there is controversy on whether nitrite affects health (Erkekoglü & Baydar, 2010; Bryan, Alexander, Coughlin, Milkowski, & Boffetta, 2012; Habermeyer et al., 2015). One of them is the advantage that nitrite can transform into nitric oxide which has a preventive and therapeutic effect on many diseases in vivo (Habermeyer et al., 2015; Bryan et al., 2007), and the another disadvantage is that nitrite may react with amines in acidic medium to form nitrosamines which has carcinogenic risks in vivo (Erkekoglü & Baydar, 2010). Questionless, the excessive intake of nitrite has a significant correlation with occurrence of methemoglobin (Bryan et al., 2007), so a regulation from the World Health Organization (WHO) is that the nitrite content in drinking water should be controlled below 3 mg/L (WHO, 2017). Besides, the excessive nitrite content in natural water will cause serious impact on biology (Bulushi, Poole, Deeth, & Dykes, 2009).

At present, in the latest reports on determining trace amount of nitrite, there are electrochemical, photometric, fluorescent and chemluminescent methods, etc.

Mani, Prakash, and Chen (2012) reported an amperometric nitrite sensor which was a glassy carbon electrode (GCE) modified by reduced graphene oxide (rGO) which was reduced from graphene oxide (GO) by using hydrazine, Ag/AgCl was used as a reference electrode, and platinum wire was used as a counter electrode in their method. Employing rGO not only increased the electro-active surface but also the catalytic oxidation current of GCE. By using the catalytic activity of the rGO/GCE and the oxidation potential change from nitrite to nitrate, they determined nitrite in water samples in range 0.41–7.68 mg/L.

In order to enhance the electron-transfer ability of rGO/GCE, Radhakrishnan, Krishnamoorthy, Sekar, Wilson, and Kim (2014) modified the Fe_2O_3/rGO on GCE to form a Fe_2O_3/rGO modified electrode which was used to detect nitrite in tap water. So based on the catalysis of Fe_2O_3/rGO, the electrode in this method extended the detect range (0.0023–36 mg/L). Haldorai, Hwang et al. (2016), Haldorai, Kim et al. (2016) proposed another nitrite sensor, in which Fe_2O_3 was replaced by Co_3O_4, and then applying the cyclic voltammetry and the chrono-amperometry, the Co_3O_4/rGO electrode was used for detecting nitrite in water in the range of 0.046–17 mg/L.

Yang and Li (2014) utilized GO and hexadecyl trimethyl ammonium bromide (CATB) to synthesize the CTAB-GO complex under ultrasonic agitation, and formed CTAB-GO/multi-walled carbon nanotubes (MWCNT) complex in chitosan/acetic acid solution. This CTAB-GO/MWCNT complex was modified on a GCE to fabricate a nitrite sensor which was used for the detection of nitrite in urine in range 0.23–37 mg/L.

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Haldorai et al. immobilized Cytochrome-c (Cyt-c) on surface of MWCNT-TiN to form a modified electrode sensor for testing nitrite (Haldorai, Hwang et al., 2016; Haldorai, Kim et al., 2016). The role of TiN on the electrode was to increase the electric activity and to facilitate the immobilization effect of Cyt-c, which can further improve the electro-catalytic activity on the electrode, so the sensitivity of their method has been greatly improved (0.046–92 mg/L).

The single-walled carbon nanotube (SWCNT) was used for nitrite detection. For example, the synthetic myoglobin/SWCNT complex and nafion solution were spread on the surface of graphite electrode to form the G/My-SWCNT/Nafion electrode (Turdean & Szabo, 2015). The nitrite in the range 23–230 mg/L can be quantified according to the current changes produced by translating nitrite into nitrate under the catalysis of iron ions in myoglobin on the electrode.

There are also many reports on the use of nanoparticles for indirectly determining nitrite. Ning, Zhang, and Zheng (2014) used sodium borohydride to reduce silver nitrate into silver nanoparticles (AgNPs) which formed the dendrimer with PAMAM in methanol solution. And then the dendrimer was modified on GCE to form a nitrite sensor which has been used to detect nitrite content in the range 0.18–66 mg/L in tap water and milk. The purpose of using AgNPs mainly was to improve the electrocatalytic activity and the surface area of this electrode.

Li et al. (2014) mixed graphite powder, TiO_2 and 1-butylpyridine six fluorophosphates, and then which was added into Teflon holder of electrode to made a nano-TiO_2/ionic liquid electrode which has a good electro-catalytic activity. TiO_2 on the sensor surface improved its antifouling performance and increased its conductivity and surface area, thus the electro-catalytic activity of the sensor was improved. At last they detected nitrite in the sausage in the range 0.023–690 mg/L.

Some chromogenic methods have also been reported for detection of nitrites. Apyari and others developed a method for testing nitrite in combination with diffuse reflectance spectroscopy. In this method, polyurethane foam (PUF) was used as diazotization reagent, 3-hydroxy-7,8-benzo-1,2,3,4-tetrahydroquinoline (HBTHQ) was used as azo reagent, and a digital camera and a computer were used as a data processing device (Apyari, Dmitrienko, Ostrovskaya, Anaev, & Zolotov, 2008; Apyari, Dmitrienko, & Zolotov, 2011).

Using the diffuse reflectance spectroscopy, Luiz, Pezza, and Pezza (2012) determined nitrite in the range 0.3–5.0 mg/L by detecting the colored product on the filter-paper where the diazotization reaction was taken place among dapsone 4,4′-diamino-diphenyl sulphone and ethylenediamine hydrochloride with nitrite in acid medium. Khana Vilab and Tubino (2012) detected nitrite in cheese and meat based on a reaction between nitrite, sulfadiazine and α-naphthol to produce a coloured product, the light emitting diode (LED) was used as the light source and photosensitive resistors was used as the detector. Making use of nitrite to catalyze the reaction between potassium bromate and methyl red which was modified by 1-butyl-3-methimidazole six fluorophosphates ionic liquid, Zhang et al. (2014) quantified nitrite in food and river water in the range 4.0–192 μg/L according to its fading degree. Noor, Tan, Lee, Kwok, and Tajuddin (2016) immobilized safranine-O on butyl acrylate microspheres, and utilized an optical fiber reflectometer to detect nitrite in the range 10–100 μg/L based on the diazotization reaction between safranine-O and nitrite to produce the blue compounds in acidic media.

The fluorescence methods for determine nitrite were also reported. Guo, Zhang, Shangguang, and Zhen (2013) used nitrite to diazotize with an amine group on the o-phenylenediamin under acid conditions, a stronger fluorescent benzotriazole was generated after the covalent bond formed between diazotization group and the adjacent amino group, the hydroxypropyl-β cyclodextrin was complexed with benzotriazole to increase the sensitivity of testing nitrite. The linear range of the method was in 40–800 μg/L and the detection limit was 13.6 μg/L. Liu, Yang, Abdel-Halim, and Zhu (2013) employed nitrite to quench the fluorescence of gold nanoclusters to indirectly detect nitrite in the range 0.92–2300 μg/L. Based on a reaction which nitrite can enhance the fluorescence of pyridine derivative formed by 2-hydro-pyridine derivative in acidic medium, Wang et al. (2016) have also determined nitrite in the range 9.2–207 μg/L.

Carbon nanodots (CDs), which is a new kind of nanomaterial, has been widely used for its excellent luminescence property, low toxicity, biocompatibility, low cost and easy preparation (Lim, Shen, & Gao, 2015). Lin, Xue, Chen, and Lin (2011) synthesized CDs by using glycerol, polyethylene glycol and serine through microwave heating and constructed a NO_2^-/H_2O_2/CDs chemiluminescence system which was applied to detect nitrite (4.6–460 μg/L) in pond water, river water and milk. Subsequently, based on this system, Lin, Dou, Li, Ma, and Lin (2015) further improved the sensitivity to detect nitrite by adding Na_2CO_3.

Besides, carbon nanodots formed by ethylenediamine (EDA) was also focused (Zhai et al., 2012) due to high fluorescence yield and amino acid content. It was found that under acidic condition, nitrite had observably quenching effect on fluorescence of EDA/CA-CDs, so in this research, a novel method for trace-level nitrite test was developed by us on the basis of the reaction system and the fluorescence capillary analysis (FCA) (Gao, Li, & Jiang, 2006; Li & Gao, 2007; Li, Liu, Gao, Chen, & Li, 2008; Zhao et al., 2008; Li, Du, Chen, & Gao, 2010; Li, Li, & Gao, 2015; Li, Li, Yang, & Gao, 2017), which is low cost, having green chemistry property, and suitable for the nitrite determination in microvolume biological samples.

2. Experiment

2.1. Reagents and instruments

The main reagents used in this research were sodium nitrite, ethylenediamine ($C_2H_8N_2$) and citric acid ($C_6H_8O_7{\cdot}H_2O$, Chengdu, Kelong Chemical), and the related material was glass capillary (45 mm × 0.7 mm, West China glassware factory, Sichuan University). Water used in the experiment was ultrapure water (0.065 μS/cm), and the reagents used were all analytical purity.

The main instruments used in this experiment were a RF-5031PC fluorescence spectrophotometer (Shimadzu, Japan), an AB204-N electronic analytical balance (Mettler, Toledo), a WP800T microwave (Galanz, Chengdu), a TP3001 thermometer (Waytop, Nanjing), a CM-230 water meter (Aike, Chengdu), a HI1290 pH (Hanna, Italy) and a K30 thermostat (Orsus, Hangzhou).

2.2. Reagent preparation

2.2.1. Standard stock solution of nitrite

0.150 g sodium nitrite was weighed and dissolved by water, and then was transferred into a 100-mL volumetric flask to dilute to the tick mark, and stored in 4 °C. When used, it was diluted to the required concentration.

2.2.2. Preparation of CA/EDA-CDs

2.10 g of $C_6H_8O_7{\cdot}H_2O$ was dissolved in 20 mL pure water in a 100-mL beaker, and 2.0 g of ethylenediamine was dropwise added in stirring. The beaker was put into one 800 W microwave oven after full blending to heat at the medium scale of the microwave oven, and stopped after about 4 min (the solution in the beaker become brown and yellow sticky matter). And then the beaker was put in a vacuum dryer at 60 °C for one hour to remove the residual small molecules therein. Finally, 500 mL deionized water was added into the beaker to dissolve the sticky matter (Hu et al., 2014; Qu, Wang, Lu, Liu, & Wang, 2012). 1.5 mL of EDA/CA-CDs solution was transferred in 500-mL volumetric flask to volume with hydrochloric acid (pH1.5).

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