Sensors and Actuators B 255 (2018) 1577–1581

Contents lists available at ScienceDirect

Sensors and Actuators B: Chemical

journal homepage: www.elsevier.com/locate/snb

Research paper

A "turn-on" fluorescence sensor for Pb^{2+} detection based on graphene quantum dots and gold nanoparticles

Xiaofang Niu, Yuanbo Zhong, Rui Chen, Fei Wang, Yanjun Liu, Dan Luo*

Department of Electrical and Electronic Engineering, Southern University of Science and Technology, Xueyuan Road 1088, Nanshan District, Shenzhen, Guangdong, 518055, PR China

ARTICLE INFO

Article history:
Received 5 March 2017
Received in revised form 4 August 2017
Accepted 22 August 2017
Available online 26 August 2017

Keywords:
Lead
Fluorescence sensor
Graphene quantum dots
Gold nanoparticles

ABSTRACT

Heavy metal, such as Pb^{2+}, detection technologies are quite important in environment monitoring and human health protection. However, most existing technologies are often time consuming, expensive with sophisticated equipment, and requirement of complicated sample pre-treatment, which limit the useful range of real-time application. Here, we report the development of a "turn-on" fluorescence sensor for Pb^{2+} detection based on graphene quantum dots and gold nanoparticles. We achieved an extremely broad detection range of Pb^{2+} from 50 nM to 4 μM, with a detection limit of 16.7 nM. This sensing system is highly sensitive and selective for determination of Pb^{2+}. The proposed strategy is expected to provide considerable implication for other heavy metal, antigen, or DNAs by modifying sensing molecules, and fast examination in chemical and biological applications.

© 2017 Elsevier B.V. All rights reserved.

1. Introduction

Lead is one of the major environmental pollutants origins from gasoline, batteries and industrial pigments, accompanying by severe human health risks including muscle paralysis, memory loss, anemia, cardiovascular dysfunction and mental troubles [1–3]. Therefore, quantitative and sensitive detection is important, particularly in aqueous environments. Many technologies have been developed for the detection of lead ions (Pb^{2+}), such as atomic absorption spectroscopy (AAS) [4], plasma atomic emission spectroscopy [5], surface enhanced Raman spectroscopy [6], anodic stripping voltammetry [7], colorimetric [8], biochemical [9], and electrochemical techniques [10], but these methods are time consuming, expensive with sophisticated equipment, and may require complicated sample pre-treatment. Therefore, developing novel method for Pb^{2+} real-time sampling is highly desirable. Recently, florescence spectrometry has attracted considerable interests due to its simplicity, low-cost and high sensitivity and selectivity [11]. The naked-eye fluorescence sensor for Pb^{2+} detection is a promising method in fields of clinical toxicology, environmental monitoring and industry process monitoring.

Grapheme quantum dots (GQDs) [12–15] have attracted considerable interest for developing various applications in biological imaging [16], sensor [17], phototransistors [18] and photovoltaic devices [19] in recent years. Comparing to conventional fluorescence probes such as organic dyes and inorganic semiconductor quantum dots, which are hindered from drawbacks as such poor photostability, easy photobleaching, and short lifetime, GQDs exhibit great advantage due to their intrinsic superiorities such as high photoluminescence, excellent biocompatibility, good resistance to photobleaching, and stable emission [20–23]. Therefore, GQDs are expected to be excellent alternatives to organic dyes and semiconductor QDs for fluorescence sensor both in biological system and heavy metal detection.

Herein, for the first time, we develop a "turn on" fluorescent sensor for detection of Pb^{2+} based on GQDs and gold nanoparticles (AuNPs). The mechanism of detection is due to the fluorescence recovery from the separation of GQDs and AuNPs induced by adding Pb^{2+}. The proposed fluorescence sensor possesses an extremely broad detection range with a low detection limit, which achieves simply and inexpensively quantitative analysis of Pb^{2+}.

DNAzyme (catalytic strand) are main members of functional nucleic acids which can either bind to a target molecule or perform catalytic reactions with the ability to recognize metal ion [24]. In 2003 Brown reported a DNAzyme, which could be binding Pb^{2+} and result in catalytic hydrolysis of the oligonucleotide [25]. Then a series of reports have been presented based on the DNAzyme for detection of Pb^{2+}. Tang et al. developed an enzyme-free electrochemical sensor for detection of Pb^{2+} base on the particular DNA strands [26]. Zhang et al. reported a photoelectrochemical

* Corresponding author.
E-mail address: luo.d@sustc.edu.cn (D. Luo).

http://dx.doi.org/10.1016/j.snb.2017.08.167
0925-4005/© 2017 Elsevier B.V. All rights reserved.

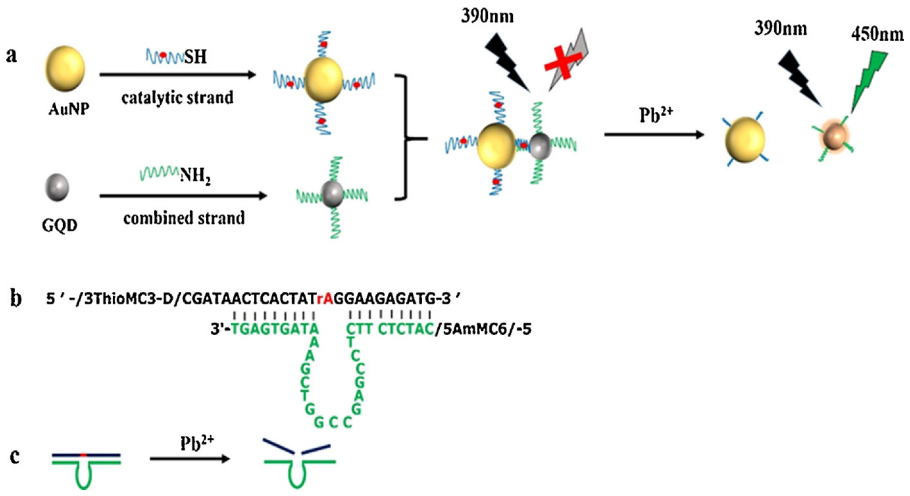

Fig. 1. (a)Schematic illustration of Pb^{2+} detection based on fluorescence resonance energy transfer between GQDs and AuNPs. (b) Structure formula of combined GQDs and AuNPs. (c) Schematic of catalytically cleave of the DNA molecule.

DNAzyme sensor based on ZnO nanoflower for detection of Pb^{2+} by the DNA strands [27]. In this study, catalytic strand and its combined strand were used as the mediums to connect GQDs and AuNPs. Based on strong fluorescence of GQDs, fluorescence quenching between GQDs and AuNPs and fluorescence recovery with the present of lead ion are used to achieve quantitative analysis of Pb^{2+}. Fig. 1(a) shows the schematic illustration for Pb^{2+} detection based on fluorescence resonance energy transfer (FRET) between GQDs and AuNPs [28]. Here, the GQDs were immobilized with amine modified combined strand by 1-Ethyl-3-(3-dimethylaminopropyl) carbodiimide (EDC). Then the AuNPs were modified with catalytic strand by sulfhydryl groups. After mixing each other of the modified particles, a complementary base-pairing reaction took place, leading to the connection of GQDs and AuNPs by catalytic strand (DNAzyme) and combined strand, which was single oligopeptide and modified on GQDs and AuNPs, respectively, (the structural formula is as shown in Fig. 1(b)). Substantial fluorescence quenching will be triggered in the system at the excitation wavelength of 390 nm due to the FRET from GQDs to AuNPs. In appearance of Pb^{2+}, the GQDs and AuNPs started to disassemble by catalytically cleaving of DNA molecule due to catalytic activity of catalytic strand [29,30]. The DNA molecule will be catalytically cleaved at the "rA" site, releasing a AuNPs-linked oligonucleotide fragment, a related long oligonuleotide fragment labeled with AuNPs, and a DNAzyme strand with GODs, as shown in Fig. 1(c). The fluorescence will recovery at wavelength of 450 nm.

2. Materials and methods

In our experiment, Gold (III) chloride trihydrate ($HAuCl_4{\cdot}3H_2O$), sodium citrate solution, phosphate buffer solution, EDC, $MnCl_2$, FeCl3, KCl, $HgCl_2$, $CuCl_2$, $MgCl_2$, $CaCl_2$, $ZnCl_2$, $CdCl_2$, $AgNO_3$, and $Pb(NO3)_2$ were all purchased from Sigma-Aldrich (St. Louis, MO, USA). Graphene quantum dot (GQD) solution was purchased from Nanjing XFNANO Materials Tech Co., Ltd. (Nanjing, China). The catalytic strand (5′-/3ThioMC3-D/CGATAACTCACTATrAGGAAGAGATG-3′) and combined strand (5′-/5AmMC6/CATCTCTTCTCCGAGCCGGTCGA-AATAGTGAGT-3′) were purchased from Shenggong Bioengineering Ltd. Company (Shanghai, China). PBS buffer was used for this reaction. The role of this mixture was indicating the preparation process of PBS buffer, which was mixed by 0.1 M NaCl, 5 mM NaHPO4 and 5 mM Na2HPO4. The pH of the buffer was 7.4 and the catalytic strand and AuNPs were added in the PBS buffer in centrifuge tube. All aqueous solutions were prepared using double-deionized water obtained from a Millipore Ultra-Pure Reagent Water System (Millipore, Continental Water Systems, ElPaso, TX, USA). The morphology and size of GQDs and AuNPs were characterized using a Tecnai F30 transmission electron microscopy (TEM). A UV–visible spectrophotometer (Metash, UV-5200PC) was used to measure the absorption spectrum of AuNPs. A fluoroSENS-9003 analytical spectrophotometer was used to measure the emission spectrum of GQDs.

To prepare AuNPs conjugation with combined strand, 1 ml of 24 mM $HAuCl_4$ was firstly mixed with 98 ml of H_2O and then heated to 110 °C for 5 min, then 10 ml of 14.55 mM trisodium citrate solution was rapidly injected into the solution. The boiling solution was stirred rapidly and refluxed for 20 min. After the color of the solution gradually changed from colourless to wine red, the solution was kept stirring at room temperature for 30 min and then filtered by a 0.22 μm filter. The average size of prepared AuNPs was around 18 nm 500 μl of AuNP solution prepared was diluted 10 times by adding water and then mixed with the thiol-modified catalytic strant (5′-/3ThioMC3-D/CGATAACTCACTATrAGGAAGAGATG-3′, 100 μM). Next, PBS buffer was slowly added in and left for 16-h standing. The product was centrifuged at 10,000 rpm for 10 min to remove the supernatant unreacted reagents, and the remaining red precipitate was collected at the bottom. The process of GQDs-ssDNA conjugation was following. Firstly, the GQD (1 mg/mL) functionalized with carboxylic acid group was sonicated for 10 min and added by 27 mM EDC. The solution was shaking for 2 min and then sonicated for 30 s. After that, the amine-modified combined strand (5′-/5AmMC6/CATCTCTTCTCCGAGCCGGTCGAAATAGTGAGT-3′, 100 μM) was added into the solution at, the solution was incubated for 1 h at room temperature.

3. Results and discussion

The AuNPs were synthesized by the citrate reduction method and modified with oligonucleotides strand. A TEM experiment was performed to characterize the morphology and sizes of AuNPs. The average diameter of the monodispersed AuNPs and GQDs was about 18 nm (Fig. 2a) and 5 nm (Fig. 2b), respectively. It can be seen that both AuNPs and GQDs were well dispersed and separated. When the AuNPs and GQDs were conjugated, the distance between GQDs and AuNPs was only several nanometers, as shown in Fig. 2c.

Download English Version:

https://daneshyari.com/en/article/7141888

Download Persian Version:

https://daneshyari.com/article/7141888

Daneshyari.com