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Research paper

An ultrasensitive and ultraselective chemiluminescence aptasensor for dopamine detection based on aptamers modified magnetic mesoporous silica @ graphite oxide polymers

Yuanling Sun, Yanna Lin, Chaofan Ding, Weiyan Sun, Yuxue Dai, Xiaodong Zhu, Hao Liu, Chuannan Luo*

Key Laboratory of Interfacial Reaction & Sensing Analysis in Universities of Shandong, School of Chemistry and Chemical Engineering, University of Jinan, Jinan 250022, PR China

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ABSTRACT

In this work, an ultrasensitive and ultraselective chemiluminescence (CL) aptasensor was prepared for dopamine (DA) detection based on aptamers modified magnetic mesoporous silica @ graphite oxide polymers (Fe_3O_4·mSiO_2·nSiO_2@GO). Firstly, Fe_3O_4·mSiO_2·nSiO_2@GO and Au nanoparticles (AuNPs) were prepared and characterizated by transmission electron microscopy (TEM), scanning electron microscope (SEM), fourier transform infrared (FTIR), X-ray powder diffraction (XRD) and others. Then, dopamine aptamer (D-Apt) was immobilized on the surface of Fe_3O_4·mSiO_2·nSiO_2@GO while AuNPs was modified by ssDNA (a single stranded DNA partially complementary to D-Apt). The immobilization properties of Fe_3O_4·mSiO_2·nSiO_2@GO to D-Apt and the adsorption properties of Fe_3O_4·mSiO_2·nSiO_2@GO@D-Apt to AuNPs@ssDNA were researched sequentially. When DA existed, AuNPs@ssDNA was desorbed from the surface of Fe_3O_4·mSiO_2·nSiO_2@GO@D-Atp and catalyzed luminescence. After that, under optimized CL conditions, DA could be measured with the linear concentration range of 2.5×10^{-13} to 2.0×10^{-9} mol/L. The detection limit was 3.9×10^{-14} mol/L (3δ) while the relative standard deviation (RSD) was 2.6%. Finally, the Fe_3O_4·mSiO_2·nSiO_2@GO@D-Apt/AuNPs@ssDNA – CL aptasensor was used for the determination of DA in practical samples and recoveries ranged from 98.0% to 102.0%. Those satisfactory results clarified the proposed CL aptasensor achieved ultraselective, ultrasensitive and reliable detection of DA and revealed potential application in monitoring and diagnosis of human neurological diseases.

1. Introduction

Dopamine (DA), as a kind catecholamine neurotransmitter, has aroused researchers' widespread concern. DA has a variety of functions in the central nervous system. It is mainly responsible for the brain's lust or feeling and the information transmission of excitement or happiness [1]. In addition, many addictive behaviors are related to DA [2]. DA has become a marker of related neurological disease, such as Alzheimer's disease [3] and Parkinson's disease [4]. Therefore, it is of great important to measure DA in clinical diagnosis and study the mechanism of related diseases. At present, various methods have been reported for DA detection, including colorimetric [5], high performance liquid chromatography (HPLC) [6], electrochemistry [7,8], chemiluminescence [9] and electrochemiluminescence [10] et al. However, these are still some problems with these methods, such as procedures sophisticated, equipment expensive and time-consuming et al. Hence, seeking for a simple, cost-effective, sensitive and selective method to detect DA is of great significance.

Chemiluminescence (CL) technique is a kind detection method associated with CL intensity (I) in chemical reactions, in which the optical signal emitted from CL reagents that transformed from the excited state back to the ground state [11,12]. I produced by single chemical reactions is relatively weak. So CL enhancers, as catalysts, are introduced in CL analysis method. The common catalysts are a variety of metal ions and various enzymes [13,14]. Among them, gold nanoparticles (AuNPs) have been used widely as CL catalysts widely. Li [15] found that AuNPs with different sizes could enhance the CL of the luminal-$NaIO_4$ system in alkaline solution. Gold colloids with nanoparticles of different sizes were found to enhance the CL of the luminal-H_2O_2 system [16]. Because its advantages of no background interference, high sensitivity, easy operation,

* Corresponding author.
E-mail address: chm_sunyl@126.com (C. Luo).

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fast analysis and easy to realize automation, CL technique has been widely used in numerous fields, such as pharmacology [17], environmentology [18], biomedicine [19] and enzyme immunoassay [20]. However, its applications are usually restricted by low selectivity which could be improved by introducing some specific recognizable materials in the CL system [21–23].

Aptamers (Apt), a kind alternative to antibodies, has been regarded as a new kind combined molecule due to its remarkable performances [24,25]. Apt has the advantages of low molecular weight, simple structure, easy synthesis or modification and long-term preservation. In addition, it can bind various targets tightly and specifically, such as proteins, amino acids, inorganic ions, even intact viruses and cells. Therefore, aptamers-based sensing devices have been widely developed, especially in the construction of a variety of biosensors, such as electrochemistry biosensor [26], colorimetric biosensor [27], fluorescence biosensor [28] and chemiluminescence biosensor [29]. Huo [27] developed a sensitive and selective colorimetric biosensor for ATP detection, in which ATP-binding aptamers act as the identification element and AuNPs-unmodified as the probe. An ultrasensitive chemiluminescence aptasensor was prepared for thrombin detection based on iron porphyrin catalyzing luminescence desorbed from chitosan modified magnetic oxide graphene composite (CS@Fe_3O_4@GO) [29].

Silica materials are a class of industrial raw materials and usually used as a reinforcing agent for rubber, ceramic and other materials [30]. Moreover, because of its non-toxic, tasteless and good biocompatibility, silica material can be used as the carriers of drug or protein [31]. Mesoporous silica materials also have both characteristics of mesoporous materials and nanomaterials. They have been regarded as new inorganic biological materials because of its ultra-high specific surface area, large pore volume, controllable morphology and size. Besed on its high chemical stability, biocompatibility, convenient synthesis and low cost, mesoporous silica materials has aroused widespread concern by researchers in the field of biomedical and bioanalysis [32–34]. Lai [35] successfully synthesized a thionine-doped mesoporous silica nanosphere/polydopamine nanocomposite (MSN-TH/PDA) for developing a new signal transduction strategy of electrochemical immunoassay and successfully used in the detection of human IgG with wide linear range and low detection limit. The hierarchical mesoporous silica nanoparticles (HMSNs) were fabricated by a two-step synthetic process and a feasible strategy to tailor drug release behaviors was proposed based on its specific hierarchical mesoporous structures [36].

Graphene oxide (GO) is a new type of carbon materials with excellent properties of high surface area and rich surface functional groups [37]. It has been generally accepted that hydroxyl and epoxy groups are randomly distributed on monolithic films of GO while carboxyl and carbonyl groups at the edges of films. Based on this, the surface of the GO is easily modified to synthesize GO composite materials [38,39], such as magnetic graphene oxide composites (MGO). MGO has both advantages of high adsorption properties of nanomaterials and easy separation of magnetic materials, which makes MGO widely used in many fields, such as electrons [40], drug delivery [41], water purification [42] and so on, especially in biosensors. Jin [43] developed an ECL biosensor for thrombin detection. Ir (III) complex has been conjugated to aptamers and adsorbed on MGO in the biosensor. When thrombin presences, the aptamers will release into solution and result in the increases of ECL intensity. Lin [44] proposed a reusable biosensor based on a MGO-modified Au electrode to detect vascular endothelial growth factor (VEGF) in human plasma for cancer diagnosis, in which Avastin was used as the specific bio-recognition element and MGO as the carrier for Avastin loading.

In this study, Fe_3O_4·$mSiO_2$·$nSiO_2$@GO and AuNPs were synthesized and characterized. The surface of Fe_3O_4·$mSiO_2$·$nSiO_2$@GO was immobilized by D-Apt to form Fe_3O_4·$mSiO_2$·$nSiO_2$@GO@D-Apt. Then AuNPs@ssDNA was modified on the surface of Fe_3O_4·$mSiO_2$·$nSiO_2$@GO@D-Apt and the final polymers – Fe_3O_4·$mSiO_2$·$nSiO_2$@GO@D-Apt/AuNPs@ssDNA were synthesized successfully. After that, the polymers were applied to the CL and were used to improve the selectivity of DA detection. AuNPs@ssDNA was released from the surface of Fe_3O_4·$mSiO_2$·$nSiO_2$@GO@D-Apt when DA existed in solutions and induced changes of ΔI. Ultimately, an ultrasensitive and ultraselective and simple Fe_3O_4·$mSiO_2$·$nSiO_2$@GO@D-Apt/AuNPs@ssDNA – CL aptasensor was constructed for the detection of DA.

2. Experimental

2.1. Materials

DA aptamer: 5′-AGAGAACCTGGGGGAGTATTGCGGAGGAAG GTTTTTTT –3′ and ssDNA: 5′-CAAAAAAAACCTGGGGGAGTATTG-$(CH_2)_3$-SH-3′ were purchased from Sangon Biotech Company (Shanghai, China). DA (98%), glycine, cystine and other biological interferences were purchased from Solebo Biological Technology Co. Ltd. (Beijing, China). Graphite powder was purchased from Hongyan Chemical Reagent Factory (Tianjin, China). Chloroauric acid was supplied by Shenyang gold Jiuqi Chemical Co. Ltd (Jilin, China). Ferrous chloride and ferric chloride were supplied by Sinopharm Chemical Reagent Co., Ltd (Beijing, China). 1- (3-dimethylaminopropyl) –3-ethylcarbodiimide Luminol (EDC) and N-hydroxysuccinimide (NHS) were supplied by Sass chemical technology Co., Ltd (Shanghai, China). Ethyl orthosilicate (TEOS), Cetyltrimethylammonium bromide (CTAB) and Aminopropyltriethoxysilane (APTs) were purchased from Mclean biochemistry Co., Ltd (Shanghai, China). Sodium hydroxide, Acetic acid, Ethanol, Potassium permanganate and all other chemicals unless specified were analytical reagent grade and used without further purification. Redistilled water was used throughout the work. Phosphate buffer (PBS, pH = 7.4, 0.01 mol/L) solution was used to prepare all DA solutions in all the experiment.

2.2. Apparatus

The IFFM-E flow injection CL analyzer (Xi'an Remex Electronic instrument High-Tech Ltd, China) was equipped with an automatic injection system and a detection system. A certain amount of Fe_3O_4·$mSiO_2$·$nSiO_2$@GO@D-Apt/AuNPs @ssDNA dispersed and placed in cup with magnet was placed in bottom to achieve on-line magnetic separation as shown in Fig. 1.

2.3. Preparation of GO

GO was prepared from nature graphite powders by a modified Hummers method described in previous literatures [45,46]. Briefly, 1.0 g of graphite powder and 200 mL of mixed acid (V_{H2SO4}:V_{HNO3} = 8:1) were added into a 500 mL of three mouth flask. Then 6.0 g of potassium permanganate was added and the diluted suspension was stirred for 12 h at 90 °C. Afterwards, 30% H_2O_2 was dropped till no reaction. Finally, the mixture was centrifuged and washed by 0.2 mol/L HCl and water, respectively. The obtained product was vacuum dried at 60 °C.

2.4. Preparation of Fe_3O_4·$mSiO_2$·$nSiO_2$

0.02 mol of $FeCl_3$ and 0.01 mol of $FeCl_2$ were mixed and dissolved in 100 mL of water. 10 mL of ammonia was added and the mixture was stirred for 60 min under the atmosphere of N_2 at 90° C. The product was isolated by the external magnetic field and washed.