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Efficient lucigenin/thiourea dioxide chemiluminescence system and its application for selective and sensitive dopamine detection

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ABSTRACT

Thiourea dioxide, a well-known eco-friendly, stable and cost-effective industrial reductant, has been used as the coreactant of lucigenin chemiluminescence for the first time. This chemiluminescence system is highly efficient, and its chemiluminescence peak intensity is about 75 times higher than that of the famous lucigenin/H_2O_2 system. Interestingly, dopamine dramatically suppresses the chemiluminescence of lucigenin/thiourea dioxide system. Based on this newly-developed system, highly sensitive detection of dopamine, lucigenin, and thiourea dioxide was achieved. The linear ranges are 20–800 nM, 20 nM–0.1 mM, and 0.01–10 mM for dopamine, lucigenin, and thiourea dioxide, respectively. The detection limits are 14.7 nM, 8.0 nM, and 2.4 μM for dopamine, lucigenin, and thiourea dioxide, respectively. Moreover, this method shows excellent selectivity for the detection of dopamine against many compounds, such as ascorbic acid, uric acid, amino acids and sugars. This study suggests that the newly-found user-friendly lucigenin/thiourea dioxide system is a promising chemiluminescence system with broad applications.

1. Introduction

Chemiluminescence (CL) analysis is attractive because of its simple, fast, sensitive feature and has been widely used in many fields [1–3]. Lucigenin (*N*,*N*′-dimethylbiacridinium dinitrate) is one of the most popular CL luminophores. Its CL properties were first observed by Gleu and Petsch in 1935 [4]. Since that time many researchers have investigated the mechanism of the CL reaction. Lucigenin emit chemiluminescent light in alkaline medium in the presence of oxidizing agents (e.g., hydrogen peroxide) [5]. The emission reaction could be enhanced by metal ions [6], and different metal ions may have different enhancement effects on the lucigenin CL [7]. These properties expand the application range of lucigenin CL. However, the application of lucigenin in CL analysis is usually limited by lack of specificity [7]. So it is highly desired to develop new lucigenin CL systems, particularly new lucigenin CL systems with high selectivity and good stability for bioassays in which lucigenin is often used as label.

Thiourea dioxide (TD) is a well-known industrial reducing agent [8,9]. It is eco-friendly, low-cost, facile and stable, thus it has been vastly used in paper, textile and leather-processing industries. A unique property of this reductant is that it can decompose to generate oxygen [10,11]. It implies that TD may also react with lucigenin to generate CL. In addition, it is necessary to develop new methods for TD detection because of the broad applications of TD.

Dopamine (DA) is an important neurotransmitter and plays a significant role in the function of human metabolism, central nervous, renal and hormonal systems [12]. Abnormal concentrations of DA could cause neurological disorders, Parkinson's disease, and schizophrenia [13]. Therefore, it is of great importance to develop effective, selective and sensitive approaches to detect DA. Up to now, various analytical methods have been exploited for the detection of DA, such as chromatography coupled with spectroscopy (e.g., high-pressure liquid chromatography-mass spectrometry (HPLC–MS)) [14], electrochemistry [15,16], spectrophotometry [17,18], fluorescence [19], chemiluminescence [20,21] and electrochemiluminescence [22–24]. These methods, however, have some limitations. For instance, chromatographic methods are time-consuming, labor intensive, and expensive with complicated procedures. Similarly, the synthesis of fluorescent or colorimetric probes for DA detection involves complicated and

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time-consuming procedures. Electrochemical methods usually suffer from the interferences of uric acid (UA) and ascorbic acid (AA). Although some CL methods have also been reported to detect DA, they involve the use of oxidizing agents and metal ions, making the detection methods less selective [21].

In this study, TD has been developed as the coreactant of lucigenin CL for the first time and the effect of DA on this new CL system was investigated. DA dramatically suppresses the CL of lucigenin/TD. The lucigenin/TD system was used to detect lucigenin, TD and DA with excellent sensitivity. The inhibition mechanism of DA on the CL of lucigenin/TD is discussed and the sensitive detection of DA based on this inhibition mechanism has been demonstrated. This DA detection method is simple, fast and shows excellent selectivity against many compounds, such as ascorbic acid, uric acid, amino acids and sugars.

2. Experimental section

2.1. Materials and apparatus

Ascorbic acid and hydrogen peroxide were purchased from Beijing Chemical Reagent Company (Beijing, China). Lucigenin was purchased from TCI (Shanghai, China). TD was obtained from Aladdin (Shanghai, China). Lysine, aspartic acid, alanine, arginine, uric acid, glucose and sucrose were purchased from Sinopharm Chemical Reagent Co. Ltd. (Beijing, China). Lucigenin stock solution (1.0 mM) was prepared by dissolving 0.0510 g lucigenin in 100 mL water. All the chemicals were analytical-reagent grade and were used without further purification. Doubly distilled water was used throughout all experiments.

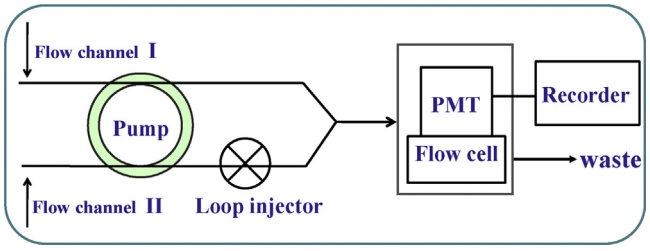
The CL was measured by a flow injection CL system consisting of a Biophysics Chemiluminescence (BPCL) ultra-weak luminescence analyzer (the Institute of Biophysics, Chinese Academic of Sciences), an intelligent flow injection sampler (IFIS-C mode) (ReMax Inc., Xi'an, China) and a home-made flow cell. The flow cell was put in a light-tight box of the luminescent analyzer. The loop injector was equipped with an injection loop of 50 μL.

2.2. Detection procedure for TD

Scheme 1 shows the schematic diagram of the flow system for TD detection. 10 μM lucigenin in water and 0.5 M NaOH solution were pumped into the flow cell through channels I and II at a flow rate of 2.0 mL/min, respectively. Different concentrations of TD in water were injected through the loop injector.

2.3. Detection procedure for lucigenin

10 mM TD in water and 0.5 M NaOH solution were pumped into the flow cell through channels I and II at a flow rate of 2.0 mL/min, respectively. Different concentrations of lucigenin in water were injected through the loop injector.



Scheme 1. A schematic diagram of the flow system for this CL system.

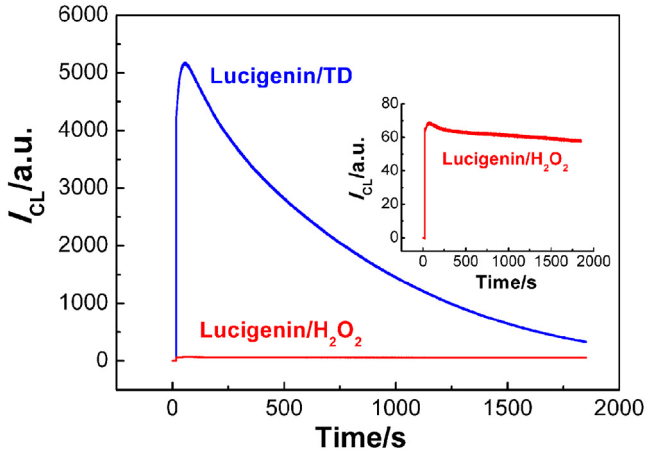


Fig. 1. The CL intensity-time curves for the lucigenin/H_2O_2 (red line) and lucigenin/TD systems (blue line). Inset: enlarged CL intensity–time curve for the lucigenin/H_2O_2 system. c(lucigenin): 10.0 μM; c(TD): 1.0 mM; c(H_2O_2): 1.0 mM; c(NaOH): 0.5 M; photomultiplier tube voltage: 700 V. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.).

2.4. Detection procedure for dopamine

10 μM Lucigenin in water and 0.5 M NaOH solution were pumped into the flow cell through channels I and II at a flow rate of 2.0 mL/min, respectively. Different concentrations of dopamine were mixed with 1.0 mM TD first, and then the mixture were injected through the loop injector.

3. Results and discussion

3.1. Chemiluminescence of lucigenin/TD system

Fig. 1 shows the CL intensity-time curves of lucigenin/TD system and lucigenin/H_2O_2 system. By comparison, the CL peak intensity of lucigenin/TD system is about 75 times higher than that of lucigenin/H_2O_2 system. It indicates that TD is an effective coreactant for lucigenin CL. The CL spectrum of this new system was measured by using various band pass filters at wavelengths of 400 nm, 425 nm, 440 nm, 460 nm, 490 nm, 535 nm, 555 nm, 575 nm, 620 nm, and 640 nm. As shown in Fig. 2, the maximum emission wavelength is about 490 nm, which is consistent with the typical

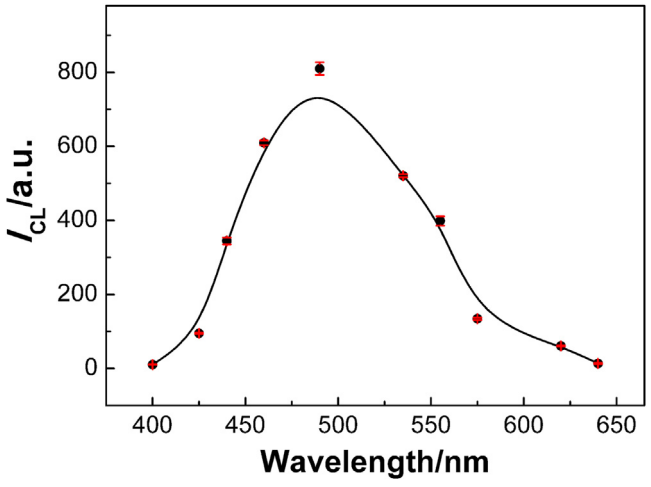


Fig. 2. CL spectrum of lucigenin/TD system. c(lucigenin): 10.0 μM; c(TD): 1.0 mM; c(NaOH): 0.5 M; photomultiplier tube voltage: 1000 V.