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Hailemariam Barkae Tesfaye

Ibrahim Halawa Mohamed

Haile Fereja Tadesse

Addisu Kitte Shimeles

Ma Xian-Gui

Chen Ye-Quan

Xu Guo-Bao

1. State Key Laboratory of Electroanalytical Chemistry, Changchun Institute of Applied Chemistry, Chinese Academy of Sciences, 5625 Renmin Street, Changchun 130022, Jilin,China,, guobaoxu@ciac.ac.cn

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Luminol/Sulfamic Acid Electrochemiluminescence and Its Application for Dopamine Detection

Tesfaye Hailemariam Barkae[1,2,3], Mohamed Ibrahim Halawa[1,4,5], Tadesse Haile Fereja[1,6], Shimeles Addisu Kitte[1,7], Xian-Gui Ma[1,2], Ye-Quan Chen[1], Guo-Bao Xu[1*]

(1. *State Key Laboratory of Electroanalytical Chemistry, Changchun Institute of Applied Chemistry, Chinese Academy of Sciences, 5625 Renmin Street, Changchun 130022, Jilin,China*; 2. *School of Applied chemistry and Engineering, University of Science and Technology of China, Hefei 230026, Anhui, China; 3. Wolkite university, College of Natural & Computational Science, Department of Chemistry, P.O Box 07, Wolkite, Ethiopia*; 4. *Department of Pharmaceutical Analytical Chemistry, Faculty of Pharmacy, Mansoura University, 35516, Mansoura, Egypt*; 5. *College of Chemistry and Environmental Engineering and College of Physics and Optoelectronic Engineering, Shenzhen University, Shenzhen 518060, Guangdong, China*; 6. *Ambo University, College of Medicine and Health Sciences, Department of Pharmacy, P. O. Box 19, Ambo, Ethiopia*; 7. *Jimma University, College of Natural Sciences, Department of Chemistry, P. O. Box 378, Jimma, Ethiopia*)

Abstract: Herein, sulfamic acid (SA) was utilized, for the first time, to enhance significantly the luminol electrochemiluminescence (ECL). With the SA concentration increased from 0.1 μmol·L^{-1} to 500 μmol·L^{-1} the ECL intensity increased proportionally. The developed luminol/SA ECL system was employed to detect dopamine (DA) based on its prominent quenching effect. The Stern-Volmer equation of $I_0/I = 1+K_{sv}$[DA] could be applied to express well the quenching mechanism of DA in the luminol/SA ECL system. The calibration plot showed that the increase in the DA concentration from 0.5 to 20 μmol·L^{-1} decreased linearly the ECL intensity with a detection limit of 30 nmol·L^{-1}. The luminol/SA ECL system was successfully carried out for DA detection in urine real sample by employing the standard addition method with the excellent recoveries of 103% ~ 105%. Selectivity of the as-developed ECL system was also investigated by using uric acid, ascorbic acid, sugars and amino acids. The results indicated that the ECL intensities changed negligibly in the presence of other substances.

Key words: electrochemiluminescence; sulfamic acid; luminol; dopamine

1 Introduction

Electrogenerated chemiluminescence (ECL) is generated from electrochemical reactions[1,2]. ECL is a technique that combined both the merit of electrochemistry and chemiluminescence. ECL has acquired much attention recently because of its simple format, high sensitivity and rapid response. Typical features of low price, low oxidation potential, and high emission yield have made luminol ECL to be used in clinical, environmental, diagnostic and food analysis[2,3]. The excited 3-aminophthalate is the key emitter of luminol ECL and can be created by using various coreactants, including superoxide radical, hypobromite or hydrogen peroxide[4,5]. For instance, H_2O_2 is the most commonly used coreactant for luminol ECL, however, its serious drawbacks, such as low stability, easy decomposition and very poor selectivity, restrict its application in bioassays[6]. Hence, it is still impor-

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tant to explore new coreactants which are distinguished by stability, selectivity and sensitivity for luminol ECL.

Sulfamic acid (SA) is an inorganic dry acid and monoamide of sulfuric acid[7]. It has been widely used as acid cleaning agent, catalyst during esterification, fixative, chloride stabilizer in paper and pulp industry, anticorrosive agent and nitrite scavenger[8-12].

Dopamine (DA), as a catecholamine neurotransmitter in the central nervous system, plays pivotal physiological functions, such as locomotors activity, secretion of other neuroendocrines, behavioral control besides the cognitive functions such as emotions, euphoria and attention[13-15]. The abnormal low level of DA in human blood is linked with Alzheimer's, and schizophrenia, Parkinson's and depression are mental dysfunctions[16,17]. While high level of DA is linked with challenges in energy metabolism and lead death[18]. Hence, it is important to maintain the proper amount of DA for proper function of our body, and develop effective, highly sensitive and selective methods to detect DA. Different analytical techniques have been used so far for the detection of DA including fluorescence[19-21], electrochemiluminescence[22-24], electrochemical method[25-28], HPLC[29,30], colorimetry[31,32], surface enhanced Raman Scattering[33,34]. These reported techniques have several challenges and limitations including time consumption in sample preparation, electrode modification, low sensitivity in addition to the sophisticated and high cost instrumentation.

Herein, we exploited SA as an efficient coreactant of luminol ECL. We have also developed new methods for the detections of SA and DA. The mechanism of luminol-SA ECL and the quenching mechanism of DA on luminol-SA ECL are discussed.

2 Experimental Section

Luminol and ascorbic acid were bought from Beijing Chemical Reagent Company (Beijing, China). SA was bought from Aldrich. Aspartic acid, lysine, arginine, alanine, glucose, sucrose and uric acid were obtained from Sinopharm Chemical Reagent Co. Ltd. (Beijing, China). Superoxide dismutase (SOD) was obtained from HWRK Chem. Co. Ltd. (China) and sodium azide (NaN_3) was bought from Fuchen Chemical Reagents Factory (China). Mannitol and thiourea were obtained from Chemical Reagent of Sinopharm Co. Ltd. (Shanghai, China). 10 mmol·L^{-1} of luminol was dissolved in 0.1 mol·L^{-1} NaOH, while 10 mmol·L^{-1} SA in CBS (0.1 mol·L^{-1}) solution, and the desired pH was adjusted by sodium carbonate and disodium carbonate. Deionized water was used throughout the experiment.

2.1 Apparatus

CHI 660B potentiostat (CHI instrument, Shanghai, China) was used for electrochemical measurements. Glassy carbon electrode (GCE), Ag/AgCl and gold electrode were used as the working, reference and auxiliary electrodes, respectively. Alumina powder of 0.3 μm was used to polish the working electrode before running each ECL experiment. Fresh solution was used in each experiment. BPCL ultra-weak luminescence analyzer provided by Institute of Biophysics, Chinese Academy of Sciences was used to measure intensities of ECL. The light tight box was used to keep the electrochemical cell where controls were made outside the box. The cell was directed to the face of the PMT detector at 900 V.

2.2 ECL Detections of SA

ECL of SA detection was carried out by mixing 200 μL of 0.1 mol·L^{-1} CBS (pH 11.5) with 100 μL of luminol (1 mmol·L^{-1}) containing different concentrations of SA on GCE. ECL intensity peak used for the detection purpose was recorded by scanning potential from 0.0 V to 1.5 V at 100 mV·s^{-1}. PMT was biased at 900 V.

2.3 ECL Detections of DA

Briefly, DA detection was performed by mixing 400 μL of 0.1 mol·L^{-1} CBS solution (pH 11.5) with 100 μL of SA (10 mmol·L^{-1}) and 100 μL luminol (1 mmol·L^{-1}) containing different concentrations of DA. ECL peak intensity recorded in the potential range from 0.0 V to 1.5 V at 100 mV·s^{-1} was considered for the quantification of DA. PMT voltage was biased at 900 V.

2.4 Real Sample Procedure

Performance and practicability of the developed

method was investigated by applying the ECL method to detect DA in real sample. The healthy urine sample (diluted 200 times) was obtained. The samples with the known concentration of DA ranged from 0.6 μmol·L^{-1} to 1.2 μmol·L^{-1} were used for the spiking and performing the recovery experiment. Concentration of DA was estimated based on the ECL response and calibration curve. PMT voltage was biased at 900 V.

3 Results and Discussion

3.1 ECL Behaviors of Luminol-Sulfamic Acid System

The electrochemical responses accompanied with the ECL curves of luminol in the presence and absence of SA are shown in Figure 1. The buffer (black color) and SA (red color) solutions displayed no ECL emission; whereas the luminol solution (blue color) gave very weak ECL signal. When SA was added into the luminol solution (green color), a notable ECL intensity at about 1.36 V was obtained (with ~12-fold increment), indicating that SA is a good coreactant of luminol ECL. The electrochemical behaviors of SA, luminal, and the mixture of SA and luminol were also studied using CV. As shown in Figure 1(B), the oxidation peak of luminol appeared at about 0.4 V and the oxidation of SA occurred at the potential higher than 1 V. The comparisons of CVs in Figure 1(B) and ECL curves in Figure 1(A) indicate that the oxidation of SA is necessary for the enhancement of luminol ECL[5].

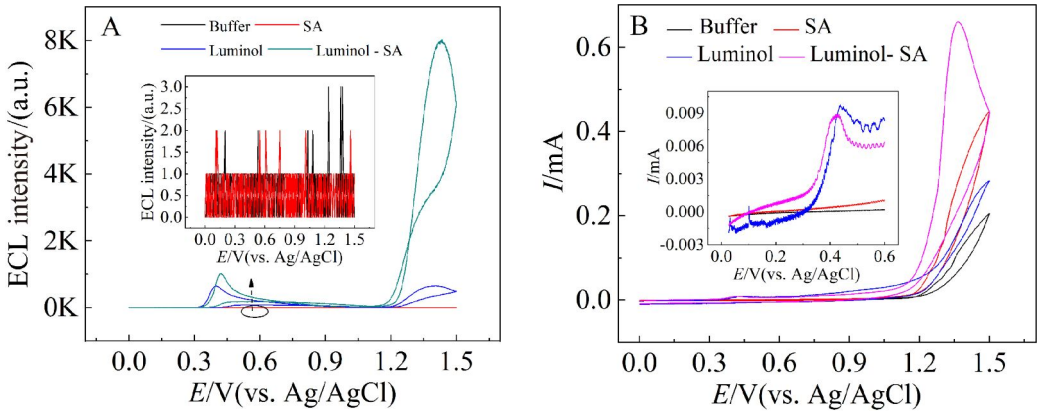

Figure 1 (A) ECL signal vs potential profile and (B) cyclic voltammograms in CBS (0.1 mol·L^{-1}, pH 11.5): Buffer solution alone (black color), 0.5 mmol·L^{-1} SA (Red color), 100 μmol·L^{-1} luminol (blue color), a mixture of 0.5 mmol·L^{-1} SA and 100 μmol·L^{-1} luminol (green color); scan rate at 100 mV·s^{-1}, PMT at 900 V (color on line)

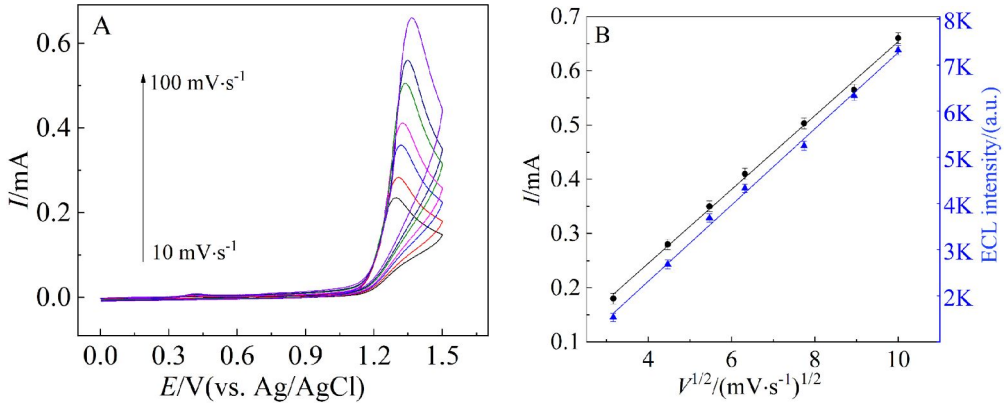

Figure 2 (A) Effect of scan rate on cyclic voltammograms and (B) variations of peak current and ECL intensity with the square root of scan rate ($v^{1/2}$) in 0.5 mmol·L^{-1} SA and 100 μmol·L^{-1} luminol (color on line)

3.2 ECL Mechanism

To discover the ECL mechanism, the effect of scan rate on ECL was examined. As indicated in Figure 2(A) and (B), linear variation was observed between anodic current and ECL signal with the square root of scan rates ($\upsilon^{1/2}$), confirming that the ECL system is controlled by diffusion of both luminol and SA[35-37]. To further reveal the mechanism, the ECL spectra of luminol/SA and the influences of superoxide dismutase, sodium azide[38], mannitol, and thiourea on luminol/SA ECL were studied. The luminol/SA ECL spectrum was collected by using 10 pieces of different filters glass at the wavelengths of 400, 425, 440, 460, 490, 535, 555, 575, 620 and 640 nm as illustrated in Figure 3. The maximum ECL was observed at around 450 nm, confirming that 3-aminophthalate (AP^{2-}*) is responsible for light emission[39,40]. Figure S1 presents the influences of superoxide dismutase[41], sodium azide[38], mannitol[42], thiourea[43] on ECL intensity. Both thiourea and sodium azide had little effect on their ECL intensities, while superoxide dismutase and mannitol decreased their ECL intensities, indicating that $O_2^{\bullet-}$ generated from the oxidation of hydroxyl ion also contributes to ECL. Accordingly, ECL mechanism of luminol/SA is proposed as follows. Deprotonated luminol (L^-) and SA undergo oxidation to produce luminol anion radical ($L^{\bullet-}$) (Eq. 1) and SA* radical (Eq. 2). Then SA* reacts with $L^{\bullet-}$ to produce 3-aminophthalate (AP^{2-}*) (Eq. 3) and gives ECL upon jumping to ground state (Eq. 6). CL is also partly generated by the reaction (Eq. 5) of luminol anion radical ($L^{\bullet-}$) with $O_2^{\bullet-}$ produced from the oxidation of hydroxyl ion (Eq. 4).

$$L^- - e \rightarrow L^{\bullet-} \quad (1)$$

$$SA - e \rightarrow SA^{\bullet} \quad (2)$$

$$L^- + SA^{\bullet} \rightarrow AP^{2-}* \quad (3)$$

$$OH^- - e \rightarrow O_2^{\bullet-} + 2H^+ \quad (4)$$

$$L^- + O_2^{\bullet-} \rightarrow AP^{2-}* \quad (5)$$

$$AP^{2-}* \rightarrow AP^{2-} + h\upsilon \quad (6)$$

3.3 pH Optimization

As shown in Figure 4, ECL intensity is found to be highly dependant on pH of the solution. ECL intensity was slightly increased at pH lower than 10. A great enhancement in ECL intensity was observed upon increasing pH from 10 to 11.5. The enhancement in ECL intensity could be ascribed to the easier oxidations of luminol and SA as a result of deprotonation at higher pH. However, further increasing pH led to a decline in ECL intensity. The decrease in ECL intensity probably resulted from side reactions. Therfore, a pH of 11.5 was chosen for the subsequent experiments[5].

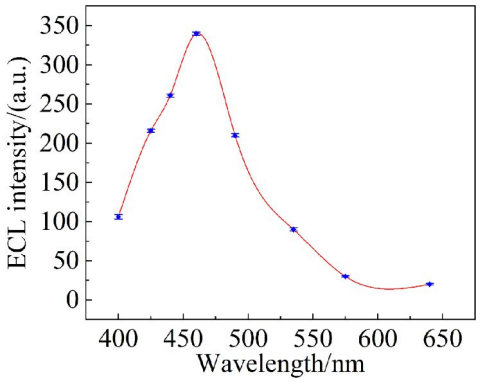

Figure 3 Emission spectrum of 100 μmol·L^{-1} luminol and 0.5 mmol·L^{-1} SA system. 0.1 mol·L^{-1} CBS of pH = 11.5 and PMT was biased at 900 V. (color on line)

3.4 Detection of SA

SA has been extensively used. However, its detection methods have seldom been reported. In view of the importance of SA detection for its application, the detection of SA by luminol ECL was tested. Figure 5 clearly shows a good linear relationship between ECL intensity and SA concentration ranging

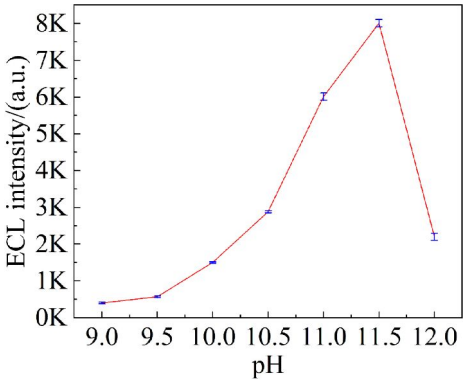

Figure 4 Effect of pH on ECL intensity using 0.1 mol·L^{-1} CBS, 0.5 mmol·L^{-1} SA, and 100 μmol·L^{-1} luminol. PMT was biased at 900 V. (color on line)

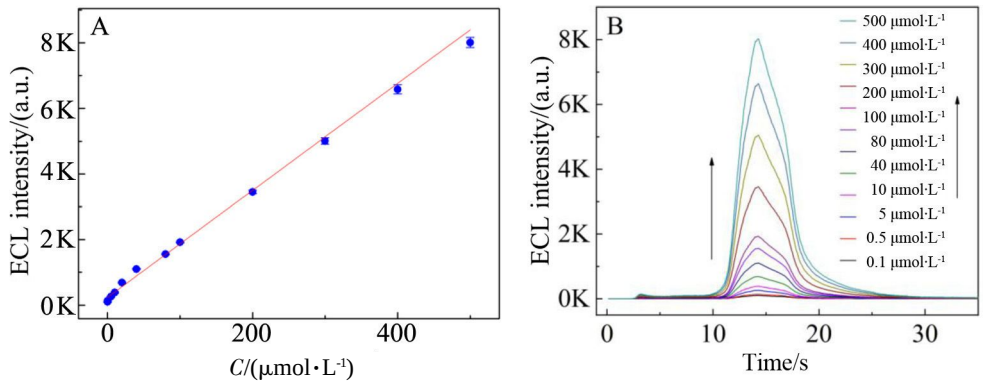

Figure 5 (A) SA calibration curve and (B) ECL intensity vs. time at different concentrations of SA. 100 μmol·L^{-1} luminol in 0.1 mol·L^{-1} pH 11.5 CBS. (color on line)

from 0.1 μmol·L^{-1} to 500 μmol·L^{-1} (Slope = 16.31; intercept = 235.01; correlation coefficient = 0.99; n = 3). Limit of detection (0.02 μmol·L^{-1}) was achieved at a signal-to-noise ratio of three. Our method is more sensitive than the titrimetric method as previously reported[44].

3.5 Detection of Luminol

The developed method was also applied to detect luminol. Figure 6 shows a linear relationship of ECL intensity with the varied concentrations of luminol. The calbration plot illustrates that the luminol-SA ECL platform under the proper optimazation enabled the quantification of luminol from 0.1 μmol·L^{-1} to 100 μmol·L^{-1} (Figure S2). The regression equation is I_{ECL} = 72.613$C_{Luminol}$ + 727.71, where the correlation coefficient of (R^2) = 0.998 is acieved. The detection limit was 0.043 μmol·L^{-1} at a signal-to-noise ratio of three. For 0.5 μmol·L^{-1} luminol the relative standard deviation was 1.05%.

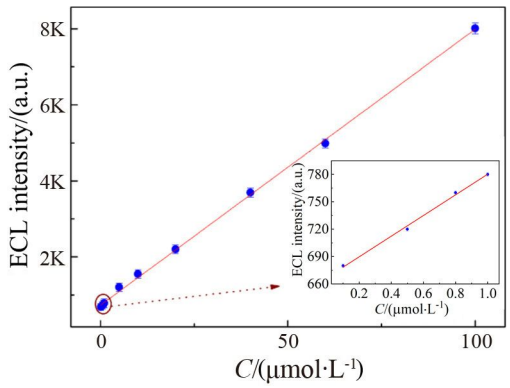

Figure 6 ECL intensity vs. concentration of luminol. 0.5 mmol·L^{-1} SA in 0.1 mol·L^{-1} pH 11.5 CBS. The inset shows the plot at low concentration (≤ 1.0 μmol·L^{-1}). (color on line)

3.6 Detection of DA

Figure 7 depicts that ECL intensity decreased gradually as the DA concentration varied from 0.5 to 20 μmol·L^{-1}. Under the optimized condition a linear relation could be achieved with the correlation coefficient of (R^2) being 0.997. Stern-Volmer equation has involved with the quenching trend with equation: $I_0/I = 1 + K_{sv}$[DA][45]. Regression equation is calculated to be I_0/I = 0.989 + 0.036C with 0.036 as the quenching constant (Figure 7 (A)). The detection limit has been found to be 30 nmol·L^{-1} at a signal-to-noise ratio of three. At 0.5 μmol·L^{-1} of DA the RSD% has been found to be 3.99% for nine replications. Dynamic quenching is involved where the collision between DA and AP^{2-}* happened during excitation and avoided ECL[46,47].

3.7 Selectivity of the Method for DA Detection

Selectivity of the as-developed ECL platform has been validated using common interfering and competing compounds like UA, AA, sugars and amino acids. As indicated in Figure 8, 20 times the concentration of DA (0.5 μmol·L^{-1}) was used for other interfering compounds (10 μmol·L^{-1}). Up on addition of 0.5 μmol·L^{-1} of DA, decrement in ECL intensity was observed. Other interfering substances had little effect on ECL intensity, indicating good selectivity.

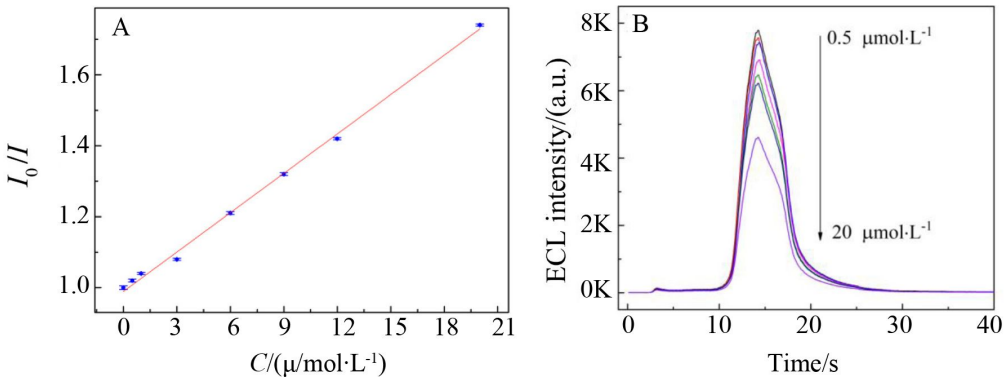

Figure 7 ECL quenching vs. DA concentration increment (color on line)

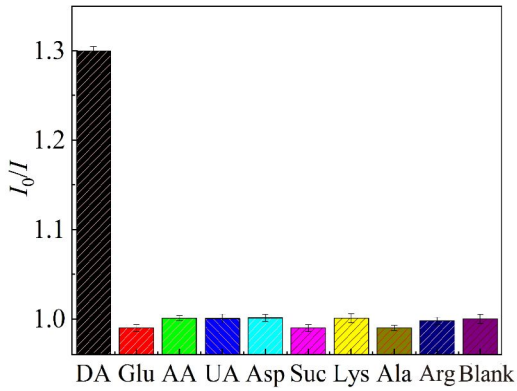

Figure 8 Detection of DA against other interfering substances. DA concentration used was 0.5 μmol·L^{-1}, while the concentrations of other substances used were 10 μmol·L^{-1}. I denotes ECL intensity of the system with other substance and DA, and I_0 denotes ECL intensity of the blank. luminol: 100 μmol·L^{-1}; SA: 0.5 mmol·L^{-1}; 0.1 mol·L^{-1} pH 11.5 CBS; PMT at 900 V. (Suc, sucrose; Glc, glucose; Arg, arginine Asp, aspartic acid; Lys, lysine; Ala, alanine). (color on line)

3.8 Quantification of DA in Urine Samples

With the ECL platform developed, further investigation for the quantification of DA in real sample of urine was made with standard addition method-based recovery experiments. Urine samples collected from healthy volunteers were mixed and diluted 200 times with buffer solution. Standard solutions of three known concentrations of DA solution were spiked into urine samples (Table 1). The recoveries fell in the range of 103% to 105% after spiking 0.6, 0.9 and 1.2 μmol·L^{-1} DA in urine samples. For nine replications RSD% has been found to be 2.9 ~ 3.7, indicating good reproducibility.

4 Conclusions

SA was successfully used as an effective coreactant of luminol ECL system. The developed ECL system has been found to be simple, sensitive and selective for the quantification of DA in urine sample. The possible ECL mechanism has been discussed. The quenching mechanism of DA followed the known Stern-Volmer equation, $I_0/I = 1 + K_{sv}$[DA]. Moreover, the system was applied for practical detection of DA in urine sample and good recovery was achieved. The relative standard deviation (RSD) for nine replicate detections of DA at 0.5 μmol·L^{-1} was 3.99%. Dynamic quenching was involved where collision between DA and AP^{2-}* happened during excitation.

Supporting Information:

Additional information as noted in text. This

Table 1 Performances of luminol-SA ECL for the quantification of DA in human urine samples

Sample	DA added/(μmol·L^{-1})	DA obtanied/(μmol·L^{-1})	Recovery/%	R.S.D./%
Urine 1	0.6	0.63	105	3.4
Urine 2	0.9	0.93	103	2.9
Urine 3	1.2	1.25	104	3.7

material is available free of charge via the internet at http://electrochem.xmu.edu.cn.

Acknowledgements

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鲁米诺/氨基磺酸电化学发光及其多巴胺检测应用

Tesfaye Hailemariam Barkae[1,2,3], Mohamed Ibrahim Halawa[1,4,5], Tadesse Haile Fereja[1,6], Shimeles Addisu Kitte[1,7], 马显贵[1,2], 陈业权[1], 徐国宝[1*]

(1. 中国科学院长春应用化学研究所电分析化学国家重点实验室, 吉林 长春 130022; 2. 中国科学技术大学应用化学与工程学院, 安徽 合肥 230026; 3. Wolkite university, College of natural & computational science, Department of chemistry, P.O Box 07, Wolkite, Ethiopia; 4. Department of Pharmaceutical Analytical Chemistry, Faculty of Pharmacy, Mansoura University, 35516, Mansoura, Egypt; 5. 深圳大学化学与环境工程学院及物理与光电工程学院, 深圳 广东 518000; 6. Ambo University, College of Medicine and Health Sciences, Department of Pharmacy, P. O. Box 19, Ambo, Ethiopia; 7. Jimma University, College of Natural Sciences, Department of Chemistry, P. O. Box 378, Jimma, Ethiopia)

摘要: 在本文中, 我们首次观察到氨基磺酸可以显著增强鲁米诺电化学发光, 而且鲁米诺电化学发光的强度随着氨基磺酸浓度在 0.1 μmol·L^{-1} 至 500 μmol·L^{-1} 范围增加而线性增加。同时, 我们观察到多巴胺可以显著猝灭鲁米诺-氨基磺酸电化学发光。基于该猝灭现象, 我们建立了多巴胺的电化学发光分析方法, 该方法的线性范围为 0.5 至 20 μmol·L^{-1}, 检出限为 30 nmol·L^{-1}。该方法具有较好的选择性, 尿酸、抗坏血酸、糖和一些氨基酸对电化学发光影响较小。采用标准加入法, 成功地将鲁米诺 - 氨基磺酸体系用于尿液中多巴胺的电化学发光测定, 回收率为 103% ~ 105%。另外, 我们还考察了多巴胺的猝灭机理, 并用 Stern-Volmer 方程计算了的动态猝灭常数。

关键词: 电化学发光; 氨基磺酸; 鲁米诺; 多巴胺