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A novel solid-state electrochemiluminescence detector for capillary electrophoresis based on tris(2,2′-bipyridyl)ruthenium(II) immobilized in Nafion/PTC-NH_2 composite film

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ABSTRACT

A new electrochemiluminescence (ECL) detector for capillary electrophoresis (CE) based on tris(2,2′-bipyridyl)ruthenium(II) ($Ru(bpy)_3^{2+}$) immobilized in Nafion/PTC-NH_2 (an ammonolysis product of 3,4,9,10-perylenetetracarboxylic dianhydride (PTCDA)) composite film was presented for the first time. The Nafion/PTC-NH_2 composite film could effectively immobilize tris(2,2′-bipyridyl)ruthenium(II) via ion-exchange and electrostatic interaction. Cyclic voltammetric and ECL behavior of Nafion/PTC-NH_2/Ru composite film was investigated compared to Nafion/Ru composite. The Nafion/PTC-NH_2/Ru composite film exhibited good ECL stability and simple operability. Then the CE with solid-state ECL detector system was successfully used to detect sophora – a quinolizidine type – alkaloids as sophoridine (SR) and matrine (MT). The CE-ECL parameters that affected separation and detection were optimized. Under the optimized conditions, the linear range was from 2.5×10^{-8} to 2×10^{-6} mol/L for SR, 1.0×10^{-8} to 1.0×10^{-6} mol/L for MT. The detection limit (S/N = 3) was estimated to be 5×10^{-9} and 10^{-9} mol/L for SR and MT, respectively. It was shown that the CE coupling with solid-state ECL detector system exhibited satisfying sensitivity of analysis.

1. Introduction

Tris(2,2′-bipyridyl)ruthenium(II) ($Ru(bpy)_3^{2+}$)-based electrochemiluminescence (ECL), since its oxidation–reduction reaction mechanism was postulated by Rubinstein and Bard [1], has attracted great attention because of its inherent high sensitivity, selectivity, and stability. Meanwhile, it has been widely investigated in flow injection analysis (FIA), high performance liquid chromatography (HPLC), and capillary electrophoresis (CE) for separation and detection [2–9]. Moreover, it is believed that $Ru(bpy)_3^{2+}$-ECL is a promising detector for CE. CE, owing to better resolution, shorter analysis time, and lower sample consumption, has been an attractive tool for the separation of biochemical and medical analytes with high performance. And the CE-$Ru(bpy)_3^{2+}$ ECL technique has proven to be a sensitive, efficient analytical technique and been successfully applied in analysis of proteins [10], amines [11], alkaloids [12], quinoline [13], etc. because of its advantages of both high-efficient separation of CE and broad applicability of $Ru(bpy)_3^{2+}$-ECL. Coupling CE with reagentless ECL detectors is significative, however, the fabrication of efficient and durable solid-state ECL detectors for CE is limited and needs further investigation.

The immobilization of $Ru(bpy)_3^{2+}$ and its derivatives on an electrode surface has been focused extensively due to its low cost, super-sensitivity, simple operability, etc. and kinds of approaches, including sol–gel entrapment [14], Langmuir–Blodgett technique [15,16], and layer-by-layer self-assembly [17], have been adopted for immobilizing $Ru(bpy)_3^{2+}$. Furthermore, new materials such as nanoparticles, cation exchange polymer are developed for more sensitive response and better stability of the solid-state ECL detector [18,19]. Nafion, a kind of cation-exchange polymer whose fluorocarbon skeleton is strongly resistant to chemical attack [20], has gained much attention as $Ru(bpy)_3^{2+}$ can be easily incorporated into the film via an ion-exchange process and hydrophobic interactions [21,22]. However, the use of pure Nafion is confined owing to the slow rate of charge transfer through the film and the migration of $Ru(bpy)_3^{2+}$ into the electro-inactive hydrophobic regions. In recent years, many researches have been done to fight this existent drawback by doping new materials like nanoparticles into the Nation film [23] and no doubt that the Nafion-contained composite films still require deeper studies. One kind of nano-organic compound PTC-NH_2, synthesized by ammonolysis of 3,4,9,10-perylenetetracarboxylic dianhydride (PTCDA) which is a kind of π-stacking organic molecules owning the desirable organic electronic properties, optical properties and interspaces on the surface, has advantages of porous structure, electrical conductivity and thermal stability [24–26]. Meanwhile, PTC-NH_2 has amino groups

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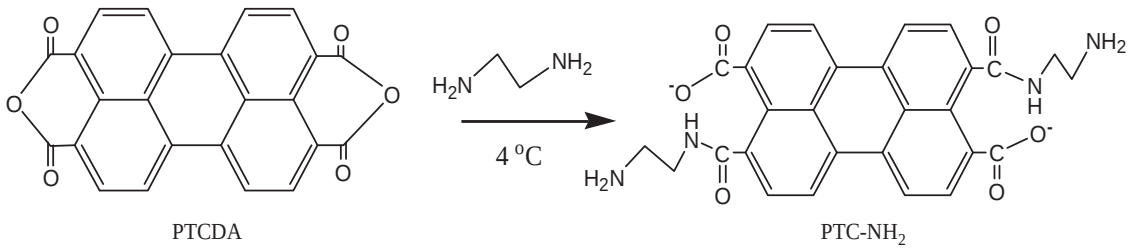

Fig. 1. The schematic diagram of the synthesis of PTC-NH_2.

and is negatively charged in liquid solution. Thus, after doping PTC-NH_2 into Nafion, Nafion/PTC-NH_2 composite film can not only immobilize more $Ru(bpy)_3^{2+}$ but also improve the electrical current so that it can enhance the ECL response as well as sensitivity compared to Nafion/Ru composite.

Quinolizidine alkaloids, as the largest single group of legume alkaloids, are important due to their toxicity and some of them especially sophora alkaloids exhibit potentially high pharmacological activities such as anti-inflammatory, antiviral, antiarrhythmic and antitumor activities [27]. No doubt sensitive as well as rapid methods for analyzing quinolizidine alkaloids are of interest. Several analytical methods involving thin-layer chromatography (TLC) [28], gas chromatography (GC) [29], HPLC [30] and CE [31], have been employed to analyze quinolizidine alkaloids. However, reports about establishing a novel solid-state ECL detector coupling to CE for sensitively detecting SR and MT are few. In this paper, we constructed a novel solid-state ECL detector based on $Ru(bpy)_3^{2+}$ immobilized in Nafion/PTC-NH_2 composite film-modified 500 μm Pt disk electrode and the Nafion/PTC-NH_2/Ru composite film was characterized by scanning electron microscopy (SEM) and electrochemical experiments. Then, the present solid-state ECL detector was applied to CE for separation and detection of SR and MT, showing excellent sensitivity of analysis compared with other analytical methods [32–34].

2. Experimental

2.1. Reagents and materials

$Ru(bpy)_3Cl_2{\cdot}6H_2O$, Nafion (5 wt%) were purchased from Sigma–Aldrich Co. (St. Louis, MO, USA). PTCDA was purchased from Lian Gang Dyestuff Chemical Industry Co. Ltd. (Liaoning, China). Matrine (>98% purity) was obtained from Angsheng Biological Reagent Co. Ltd. (Shaanxi, China). Sophoridine (>98% purity) was obtained from Nanjing Zenlang Medical Technology Co. Ltd. (Nanjing, China). Nafion was diluted to 2 wt% with ethanol solution. Phosphate buffer solutions (PBS) with various pHs and concentrations were prepared by mixing standard stock solutions of 0.1 M K_2HPO_4, 0.1 M NaH_2PO_4, 0.1 M KCl and adjusting the pH with 0.1 M H_3PO_4 or NaOH, then diluted with doubly distilled water. All chemicals were analytical grade and used without further purification. All solutions were prepared with doubly distilled water and stored in the refrigerator (4 °C). All standard solutions and phosphate buffers were prepared and filtered through 0.45 μm cellulose acetate membrane filters (Xingya Purification Material Factory, Shanghai, China) prior to injection. Fresh human urine sample was collected from a healthy volunteer. Before analysis, urine samples were filtered through 0.45 μm membrane and then diluted with pure water by 40-fold to decrease the interference of the ionic strength of the sample matrix.

2.2. Apparatus

Cyclic voltammetric (CV) measurements were carried out with a CHI 610A electrochemistry workstation (Shanghai CH Instruments, China). The morphologies of PTC-NH_2 film and Nafion/PTC-NH_2/Ru composite film were characterized by scanning electron microscopy (SEM, S-4800, Hitachi, Tokyo, Japan) at an acceleration voltage of 20 kV. CE separation and ECL detection were carried out on a MPI-A CE-ECL Analyzer (Xi'an Remax Electronic Science-Tech, Xi'an, China) equipped with a high-voltage power supply (0–20 kV) for performing electrokinetic sample injection and electrophoretic separation, an electrochemical potentiostat, a multifunction chemiluminescence detector, and a multichannel data collection analyzer. An uncoated fused silica capillary (40 cm × 50 μm i.d.) purchased from Yongnian Reafine Chromatography (Hebei, China) was used for CE separation. The new capillary was rinsed with 0.1 mol/L NaOH solution overnight, then flushed with water (5 min) and sequentially equilibrated with the running buffer for 10 min to maintain an active and reproducible inner surface before use. The end-column ECL detection cell was assembled with a three-electrodes system to trigger ECL, which comprised a Pt wire as the auxiliary electrode, an Ag/AgCl (KCl saturated) electrode as the reference electrode and a 500 μm Pt disk electrode as the working electrode.

The surface of the working electrode was polished sequentially with 0.3 and 0.05 mm α-Al_2O_3, and sonicated in ethanol and double-distilled water, respectively, for 5 min. Then the working electrode was scanned from −0.2 to +1.5 V (versus Ag/AgCl) at a rate of 300 mV/s for 5 min in 1.0 mol/L H_2SO_4 to clean and activate the surface of the working electrode.

2.3. Fabrication of the modified Pt electrode

1.0 g PTCDA were dissolved in 5 mL acetone under continual stirring and then excessive ethylenediamine was added to the PTCDA solution drop by drop at 4 °C, and then PTC-NH_2 was obtained by washing the precipitate of above mixture with acetone and ethanol. Consequently, PTC-NH_2 was dispersed homogeneously in ethanol. And the synthesis of PTC-NH_2 is shown in Fig. 1.

Typically, 0.5 mg/mL PTC-NH_2–2.0 wt% Nafion mixture was made and dispersed in ultrasonic bath for 10 min to obtain a homogeneous, well-dispersed solution. Firstly, the prepared Pt electrode was coated with 2 μL Nafion/PTC-NH_2 suspension, and allowed to evaporate in air. Secondly, the modified Pt electrode was then immersed in 50 mM $Ru(bpy)_3^{2+}$ aqueous solution for incorporation of $Ru(bpy)_3^{2+}$ for 2 h (referred as Nafion/PTC-NH_2/Ru). Subsequently, the electrode was removed from the solution and rinsed thoroughly with deionized water, then it was dried in air for further characterization and application and ready for use.

Fig. 2 illustrates the SEM images of PTC-NH_2 film (A) and Nafion/PTC-NH_2/Ru composite film (B), from which we can find that the PTC-NH_2 film grows in irregularly quadrateshaped molecular islands distributed relatively homogeneously over the surface (A) and $Ru(bpy)_3^{2+}$ is proved to overcast the Nafion/PTC-NH_2 composite film (B).

And the prepared Nafion/PTC-NH_2 composite film-modified Pt disk electrode was ready for assembling, adjusted and fixed by

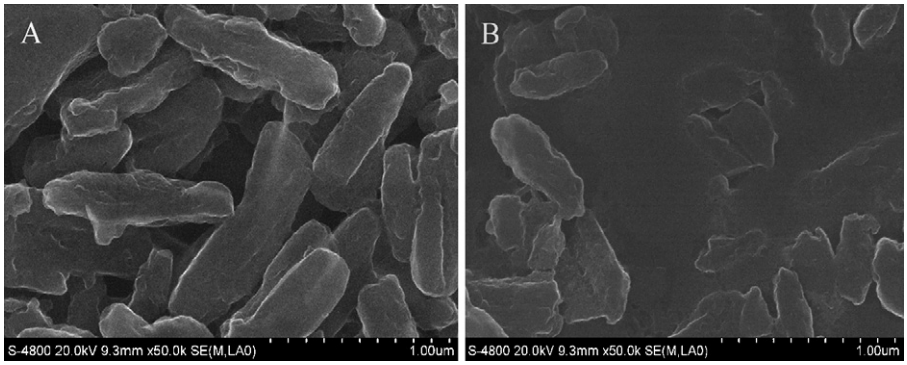

Fig. 2. SEM images of PTC-NH_2 (A) and Nafion/PTC-NH_2/Ru composite film (B).

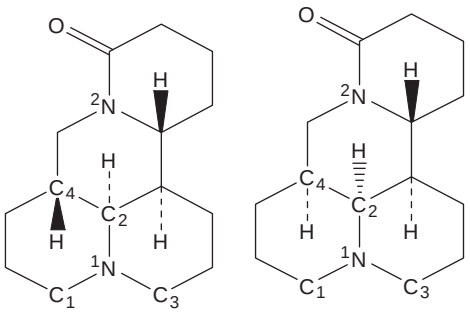

Fig. 3. Chemical structures of the two quinolizidine alkaloids.

three screws from three directions to align with the capillary tip. The distance between the working electrode and the capillary tip was set at 130 ± 10 μm with the aid of an optical microscope. Then, about 350 μL of 0.1 M PBS was added into the cell. The ECL emission from the detection cell was collected with a photomultipliertube set at −800 V, which was sensitive to photons with a wavelength range of 200–800 nm. Electrokinetic injections were performed at 10 kV for 10 s. The inlet end of the capillary was held at a positive potential and the outlet end was maintained at ground.

SR and MT standard solutions were prepared with doubly distilled water and stored in the refrigerator (4 °C). As seen in Fig. 3, chemical structures of the two quinolizidine alkaloids are shown. SR and MT, as isomeric compounds, differ only in the configuration of hydrogen atoms in C_4 position.

3. Results and discussion

3.1. CVs and ECL of Nafion/PTC-NH_2/Ru composite film

To investigate the cyclic voltammograms (CVs) and ECL intensity of Nafion/PTC-NH_2/Ru composite film, another Pt electrode was coated with the same volume of 2.0 wt% pure Nafion solution, then allowed to evaporate in air, and immersed in 50 mM $Ru(bpy)_3^{2+}$ aqueous solution for 2 h (referred as Nafion/Ru), then dried in air and ready for use. CVs and ECL studies were taken from 0.2 to 1.25 V (versus Ag/AgCl) in 0.1 M PBS (pH 8.5) at a rate of 100 mV/s at room temperature. CVs and corresponding ECL intensity–potential curves of $Ru(bpy)_3^{2+}$ immobilized in pure Nafion film (Nafion/Ru) and Nafion/PTC-NH_2 composite film (Nafion/PTC-NH_2/Ru), are shown in Fig. 4.

The electrocatalytic current of Nafion/PTC-NH_2/Ru film is much larger than that of Nafion/Ru film, which may be attributed to the faster partitioning of reagents into the film through pores that exist in the network and the increasing electrical conductivity after doping PTC-NH_2 into Nafion film. In addition, the ECL intensity in Nafion/Ru composite film (Fig. 4B, curve a) is also significantly lower than that observed in the Nafion/PTC-NH_2/Ru composite (Fig. 4B, curve b).

3.2. Optimization of experiment conditions

To obtain the optimal amount of PTC-NH_2 doped in Nafion film for maximal ECL intensity, the amount of PTC-NH_2 nanoparticles added to 2.0 wt% Nafion solution (prepared by mixing 5 wt% Nafion and alcohol with the volume ratio of 2:3) was varied from

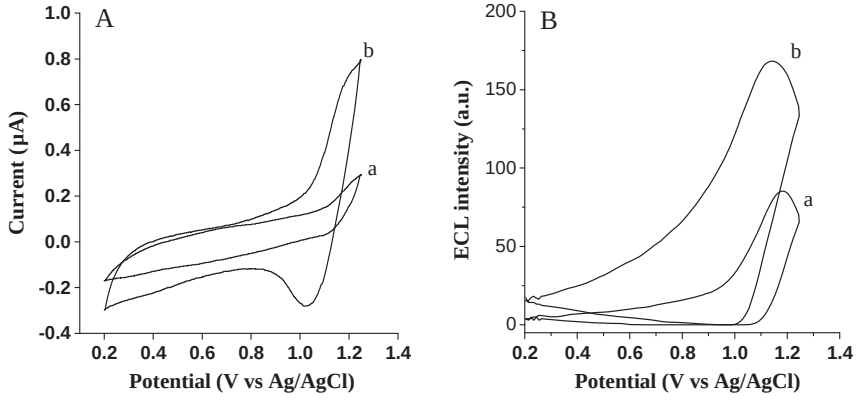

Fig. 4. CVs (A) and corresponding ECL intensity–potential curves (B) of $Ru(bpy)_3^{2+}$ immobilized in pure Nafion film (a) and Nafion/PTC-NH_2 composite film (b) in 0.1 M PBS (pH 8.5) at a scan rate of 100 mV/s.

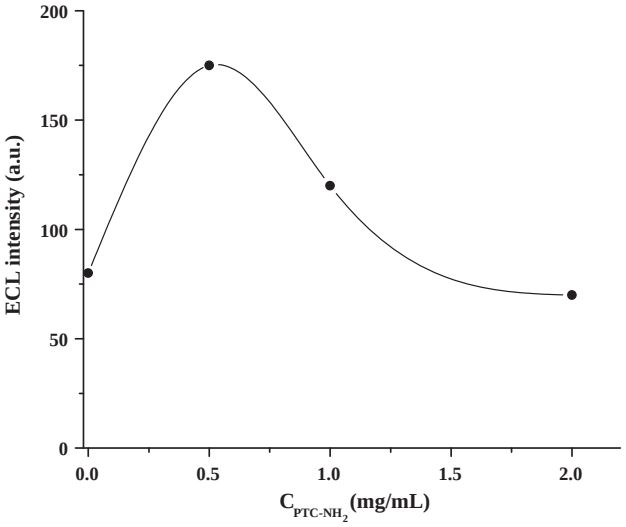

Fig. 5. Dependence of ECL intensity on the concentration of PTC-NH_2 doped into Nafion film at a scan rate of 100 mV/s.

0 to 2 mg/mL to change the concentration of PTC-NH_2 in Nafion solution.

And the effect of the amount of PTC-NH_2 doped in Nafion film on the ECL intensity is shown in Fig. 5, from which we select 0.5 mg/mL PTC-NH_2–2.0 wt% Nafion mixture as the optimum proportion.

Fig. 6 shows the stability of the solid-state ECL detector under consecutive cyclic voltammetric scans for 8 cycles in 0.1 M PBS (pH 8.5) at a scan rate of 100 mV/s. The mechanical stability of the sensor is due to enhanced ion-exchange and electrostatic interactions after doping PTC-NH_2 into Nafion.

3.3. Application

SR and MT were employed to evaluate characterization of this present CE with solid-state ECL detector system. The cathodic buffer reservoir (which is also the ECL detection cell) contained 350 μL of 0.1 M PBS (pH 8.5).

The effect of detection potential on ECL intensity was investigated over the range from +1.05 to +1.30 V (vs. Ag/AgCl) and the results were shown in Fig. 7. It can be seen that the highest ECL intensity was achieved at +1.20 V for both SR and MT. So, +1.20 V was selected as the optimal potential applied on the working electrode for the detection of SR and MT.

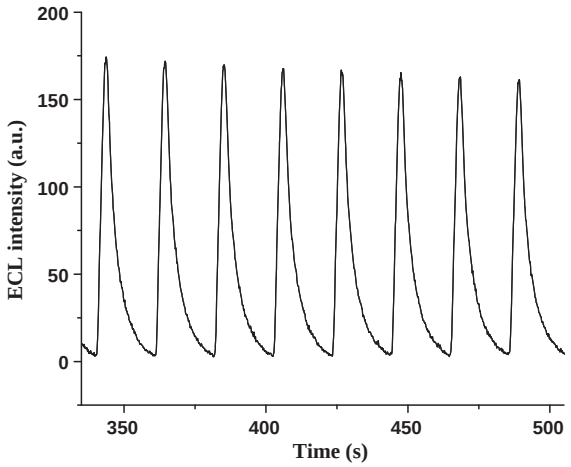

Fig. 6. ECL intensity of Nafion/PTC-NH_2/Ru composite film modified-electrode in 0.1 M PBS (pH 8.5) under continuous CVs for 8 cycles with the scan rate of 100 mV/s.

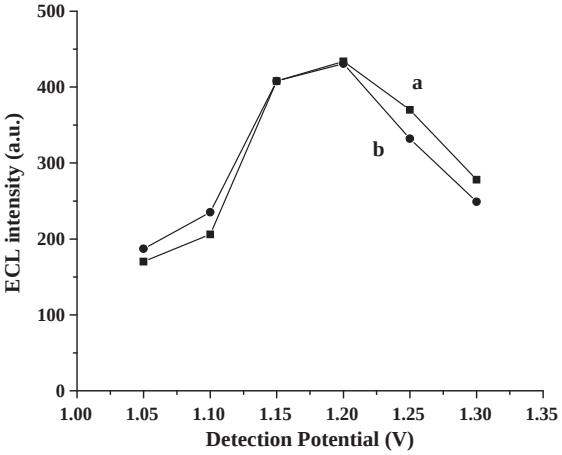

Fig. 7. Effect of detection potential on ECL intensity of the analytes. (a) ECL intensity of 1.0×10^{-6} M SR and (b) ECL intensity of 1.0×10^{-6} M MT.

The pH value of the buffers showed obvious effect on the ECL response since the ECL reaction of $Ru(bpy)_3^{2+}$ with SR and MT was a pH-dependent process with the maximum emission occurred in basic medium. The dependence of pH on ECL intensity for SR and MT was investigated from pH 6.0 to 9.5. In the detailed investigation, 8.5 was adopted as the optimal pH value since it gave the highest signal intensity (Fig. 8). The concentration of PBS also affected the ECL signal and separation efficiency, and the best response was obtained at buffer concentrations of 50 and 100 mmol/L for running buffer and ECL detection cell buffer, respectively.

3.4. Linearity, detection limit and reproducibility

Under the optimal conditions: detection potential 1.2 V, electrokinetic injection 10 kV for 10 s, separation voltage 15 kV, electrophoretic buffer 50 mmol/L PBS at pH 8.5, 100 mmol/L PBS at pH 8.5 in the detection cell, baseline separation of SR and MT was achieved within 7 min and the typical electropherogram of standard solution is illustrated in Fig. 9.

The ECL intensities for the detection of SR and MT as a function of concentrations were found to be linear from 2.5×10^{-8} to 2×10^{-6} mol/L for SR, 1.0×10^{-8} to 1.0×10^{-6} mol/L for MT with the detection limit (S/N = 3) of 5×10^{-9} mol/L for SR and 1.0×10^{-9} mol/L for MT, respectively. The regression equations of ECL peak intensity (counts) versus the concentration (10^{-8} mol/L)

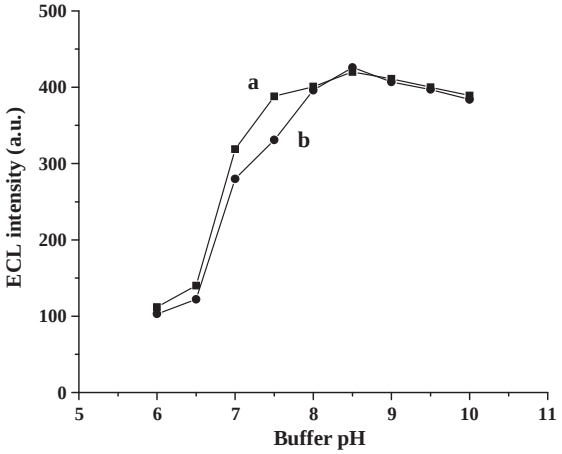

Fig. 8. Effect of buffer pH on ECL intensity of the analytes. (a) ECL intensity of 1.0×10^{-6} M SR and (b) ECL intensity of 1.0×10^{-6} M MT.

Table 1
Recoveries of SR and MT in urine samples at different spiked levels.

Analytes	Sophoridine			Matrine		
Spiked ($\times 10^{-8}$ mol/L)	10	50	100	10	50	100
Found ($\times 10^{-8}$ mol/L)	8.6	46	89	8.7	51	90
RSD (%)	4.3	3.2	3.6	4.5	3.0	3.7
Recovery (%)	86	92	89	87	102	90

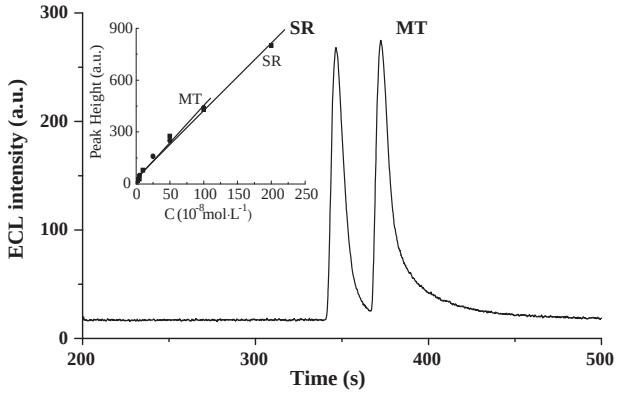

Fig. 9. Electropherograms of two standard mixtures: SR (5.0×10^{-7} M) and MT (5.0×10^{-7} M).

are $I_{ECL} = 32.521 + 3.9099C$ with correlation coefficient of 0.9923 for SR, $I_{ECL} = 25.259 + 4.2695C$ with correlation coefficient of 0.9891 for MT.

And with five repeated injections of 2×10^{-6} mol/L SR and 1.0×10^{-6} mol/L MT, the relative standard deviation of peak heights was 5.03% for SR and 4.40% for MT. The reproducibility was satisfying on the whole.

3.5. Application to human urine samples

The proposed strategy was also applied to the detection of SR and MT in human urine samples. The urine samples were filtered through 0.45 μm cellulose acetate membrane and then diluted with pure water by 40-fold before CE-ECL analysis. Fig. 10 illustrates the electropherograms of blank urine sample (a) urine sample spiked with 5.0×10^{-7} M SR and 5.0×10^{-7} M MT (b). The two alkaloids were not detected in blank urine sample, while they could be separated and detected in urine samples spiked. Besides, there were also several unknown small peaks from unknown compounds in the urine sample matrix, but they hardly interfere with separation

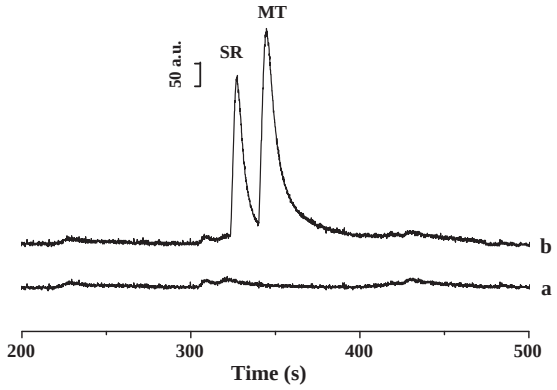

Fig. 10. Electropherograms of the blank urine sample (a) and urine sample spiked with 5.0×10^{-7} M SR and 5.0×10^{-7} M MT (b).

and detection of the two analytes. Recoveries of SR and MT in urine samples at different spiked level were listed in Table 1. It showed acceptable recoveries of 86–102%. Thus, the results indicated that the proposed method was efficient and reliable.

4. Conclusions

The Nafion/PTC-NH_2 composite film has proven to be effective matrices for the immobilization of $Ru(bpy)_3^{2+}$ on an electrode surface to prepare a solid-state ECL detector for CE. The coupling of organic cation exchange polymer (Nafion) with nano organic compound PTC-NH_2 provides an effective approach for immobilizing ECL reagent-$Ru(bpy)_3^{2+}$. Herein, both Nafion and PTC-NH_2 can incorporate $Ru(bpy)_3^{2+}$ by ion-exchange and electrostatic interactions. Compared with the pure Nafion film, the interfusion of PTC-NH_2 into Nafion takes advantages of immobilizing more $Ru(bpy)_3^{2+}$ and improving the electrical current due to porous structure, electrical conductivity as well as negative charges of PTC-NH_2. This solid-state ECL detector coupled with CE is cost-effective with good sensitivity for detecting SR and MT. Moreover, this solid-state ECL detector for CE has the potential of detecting other medications.

Acknowledgments

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