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A novel chemiluminescence method for the determination of ergometrine maleate in serum sample without chemiluminescence reagent

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ABSTRACT

In this paper, a novel flow injection-chemiluminescence (FI-CL) method was proposed for the determination of ergometrine maleate in serum. The new CL reaction was based on the direct oxidation of ergometrine maleate by the complex of metal chelate diperiodatocuprate(III) ($K_5[Cu(HIO_6)_2]$) in an alkaline medium. The CL intensity was enhanced in the presence of ascorbic acid. Hereby under the optimum conditions, ergometrine maleate was determined over the range of 4.0×10^{-9} g mL^{-1} to 4.0×10^{-7} g mL^{-1} with a limit of detection (3σ) of 1.1×10^{-9} g mL^{-1}. The relative standard deviation (R.S.D.) was 2.1% for 8.0×10^{-9} g mL^{-1} ergometrine maleate ($n = 7$). The sensitive method was successfully applied to the direct determination of ergometrine maleate (ng mL^{-1}) in pharmaceutical injection and serum samples. The mechanism of the reactions was also discussed.

1. Introduction

Chemiluminescence (CL) is known to be a popular analytical method because of the higher sensitivity, lower detection limit, wider linear range, which can be achieved with simpler instrument. It has been widely applied in many fields such as clinical research, biotechnology, pharmacology, and environmental chemistry [1,2]. The oxidants in CL system such as H_2O_2, $K_3Fe(CN)_6$, $NaIO_4$, $KMnO_4$, and BrO^- have attracted more attention in luminol-based CL system [3–5]. Recently, the transition metals in uncommon oxidation state such as Ag(III) and Cu(III) have been reported, which were almost used in luminol-based CL system as a valid oxidant. Transition metals in the higher oxidation state generally can be stabilized by chelation with suitable polydentate ligands. It is fairly stable in alkaline media [6–8]. The structure of the Cu(III) [9] is illustrated in Scheme 1. The researches related to the transition metals in uncommon oxidation state mainly focused on the kinetic and mechanistic of oxidant reaction [10,11]. The Ag(III) based CL methods have been reported for the determination of cortisol [12] and epinephrine [13] based on the reaction of Ag(III) complex(bis(hydrogenperiodato)argentate(III), $[Ag(HIO_6)_2]^{5-}$) with luminol. The application of Cu(III) complex in CL system was also reported to propose a sensitive and selective CL method for H_2O_2 analysis for its unique catalysis effect [14]. It was based on the reaction of luminol–H_2O_2. But it has not been reported as a single oxidant applied in CL system.

Ergometrine maleate ((8s)-9,10-didehydro-N-[(s)-2-hyd-roxy-1-methylethyl]-6-methylergoline-8-carboxamide monomaleate, EM) is a naturally occurring, water-soluble amino alkaloid which can be isolated from ergot. It has no α-adrenergic blocking activity. It has been used in obstetrics as an orally active oxytocic drug for its direct stimulating action on smooth muscle, especially that of blood vessels and the uterus, and antiserotonin effects [15]. The usage of EM can prolong uterine contractions in the later stages of labour and to check post-partum haemorrhaging. The methods for the determination of EM have been reported such as HPLC [16,17], immunoassay [18], spectrophotometry [19], amperometry [20], oscillopolarographic [21], fluorescence [22,23], and chemiluminescence [24,25]. The fluorescent detection has been used in quantitative analysis for its fluorescence property [26]. The HPLC method required specific instrument, which added the cost and complexity of the assay. In these methods, some require more expensive instruments (such as fluorescence analysis or HPLC method); some are not sensitive enough to finish determination of EM in real sample (such as serum). The goal of the present work was to develop a direct CL method coupled with flow injection for the determination of EM in commercial pharmaceutical products and serum.

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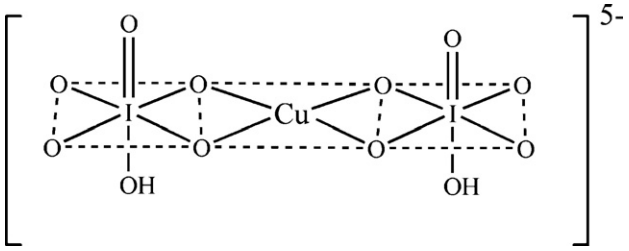

Scheme 1. Structure of copper(III) complex.

In this paper, a novel oxidant trivalent copper (Cu(III)) complex was used for the directly oxidization without any other CL reagents. Cu(III) was generally regarded as an uncommon oxidation state and not often be used directly in CL reaction for unstable in aqueous solution. While, copper metal chelate diperiodatocuprate(III) ($K_5[Cu(HIO_6)_2]$, DPC) was obtained by complexation of trivalent oxidation of copper and periodate in strong alkaline medium. The CL reaction was based on the directly oxidant of EM by DPC in the presence of ascorbic acid in alkaline medium. The ascorbic acid acted as sensitizer in the CL system. So a novel CL system of DPC-EM-ascorbic acid was used for the quantitative analysis of EM. The usage of ascorbic acid could eliminate the interference of endogenous ascorbic acid in real sample (such as serum). The flow injection analysis (FIA) is one of the most popular techniques for the analysis coupled with other detection technique for its inherent analytical advantages, such as high speed of response and favorable reproducible and realizable automation [27]. Hereby, a novel more sensitive chemiluminescence method has been developed for the determination of EM by coupling FIA techniques. It had been applied to directly determine trace amount of EM in serum without any special pretreatment of sample. Based on the electrochemical reaction and kinetics curve of the CL reaction experiment, the mechanism of the reactions was also discussed. To the best of our knowledge, it was the first time that the Cu(III) complex was employed as an individual oxidant in CL systems without any other chemiluminescence reagents.

2. Experiment

2.1. Reagents and chemical

Potassium periodate (KIO_4) was purchased from Shanghai Chemical Reagent Company (Shanghai, China). Potassium persulfate ($Na_2S_2O_8$) was purchased from Shanghai Aijian Chemical Reagent Company (Shanghai, China). Cupric sulfate ($CuSO_4{\cdot}5H_2O$), potassium hydroxide (KOH) and ascorbic acid (Vc) were purchased from Chongqing Chemical Reagent Company (Chongqing, China). The standard medicine of EM was purchased from Chongqing Institute for Drug Control (Chongqing, China). The EM injection was purchased from Hefeng Pharmaceutical Company of limited liability (Shanghai, China). All chemicals were of analytical reagent grade and used without further purification. Doubly distilled water was used throughout the work.

The 0.01 mol L^{-1} DPC stock solution was prepared by oxidizing Cu(II) in the alkaline medium according to the known method [28]. In briefly, KIO_4 (0.23 g), $CuSO_4{\cdot}5H_2O$ (0.125 g), $Na_2S_2O_8$ (0.14 g) and KOH (0.8 g) were added in 30 mL water. The mixture was heated to boiling for about 20 min on a hot plate with constant stirring. The boiling mixture turned intensely red and the boiling mixture was continued for another 20 min for the completion for the reaction. The mixture was then cooled and diluted to 50 mL with distilled water. The stock solution obtained was stored under refrigeration which was found fairly stable for several months, and DPC solutions were freshly prepared before use. The complex was confirmed at 415 nm by UV/Visible spectrum.

2.2. Apparatus

The CL-FIA system used in this work was shown in Fig. 1. Two peristaltic pumps (HL-2, Shanghai Huxi, China) were used to deliver all the chemicals at a flow rate of 2 mL min^{-1}. The polytetrafluorothylene (PTFE) flow tubes (0.8 mm i.d.) were used to connect all the components in the system. Injection was made by using a sixteen-port injection valve (Hanzhou, China) equipped with a loop of 75 μL. The CL signal was monitored by a BPCL ultra-weak luminescence analyzer (Institute of Biophysics, Chinese Academy of Science, Beijing, China) consisting of a flat coil glass flow cell facing the window of the photomultiplier tube (PMT). Data acquisition and treatment were performed with BPCL software running under Windows 98. The UV-absorbance was detected with the UV-Vis-2001 spectrophotometer (Hitachi Ltd., Japan). CHI832 electrochemical workstation (ChenHua Ltd., Shanghai, China) was used to test the electrochemical characteristics of copper metal chelate complex.

2.3. Procedure

The flow injection system is easy to operate. As shown in Fig. 1, the peristaltic pump propelled the DPC solution, analyte (a standard EM solution or a sample containing EM), ascorbic acid solution and carrier solution through the system at 2.0 mL min^{-1}, respectively. When the injection valve was set to the sampling position, the analyte solution and ascorbic acid solution were mixed at point M_1 and ran through the reagent loop until the loop was full of the mixture solution; the stream of mixture of DPC solution and carrier solution ran the whole system until a stable baseline was recorded. When the injection valve was switched to the injecting position, the DPC stream was merged with mixture solution at point M_2 by the carrier stream, which bypassed the reagent loop (75 μL mixture solution) and ran directly through the flow cell, producing CL emission. And the CL signal was then recorded simultaneously. The PMT was operated at −950 V. The relative CL intensity ΔI (defined as the difference of CL intensity between the presence and absence of analyte, respectively) was proportional to corresponding concentration of EM.

3. Results and discussion

3.1. Kinetics curve of the CL reaction of EM–DPC–ascorbic acid

In the primary experiment, the emitted CL was based on the oxidation reaction of EM by DPC in the presence of ascorbic acid. In the batch mode, a typical response curve (intensity versus times) is used to describe the CL emission. The response curve depends on the experimental factors such as pH and reagent concentration. So the experimental parameters are kept constant, the intensity–time curve of EM–ascorbic acid in DPC solution was recorded to study the kinetic characteristic of the CL reaction in Fig. 2. The CL intensity peak appeared within 2 s since the mixture solution of EM–ascorbic acid was injected. The CL signals would decrease to baseline within 100 s. As seen from Fig. 2, the CL reaction of DPC–EM–Vc is a quick reaction obviously. The kinetic curve indicated the CL system is rapid and sensitive enough and suitable for the analysis of EM.

3.2. Optimization of experiment procedure

3.2.1. The effect of DPC solution concentration

The CL was emitted from the oxidation reaction of EM by DPC in the presence of ascorbic acid. As the only oxidant, the

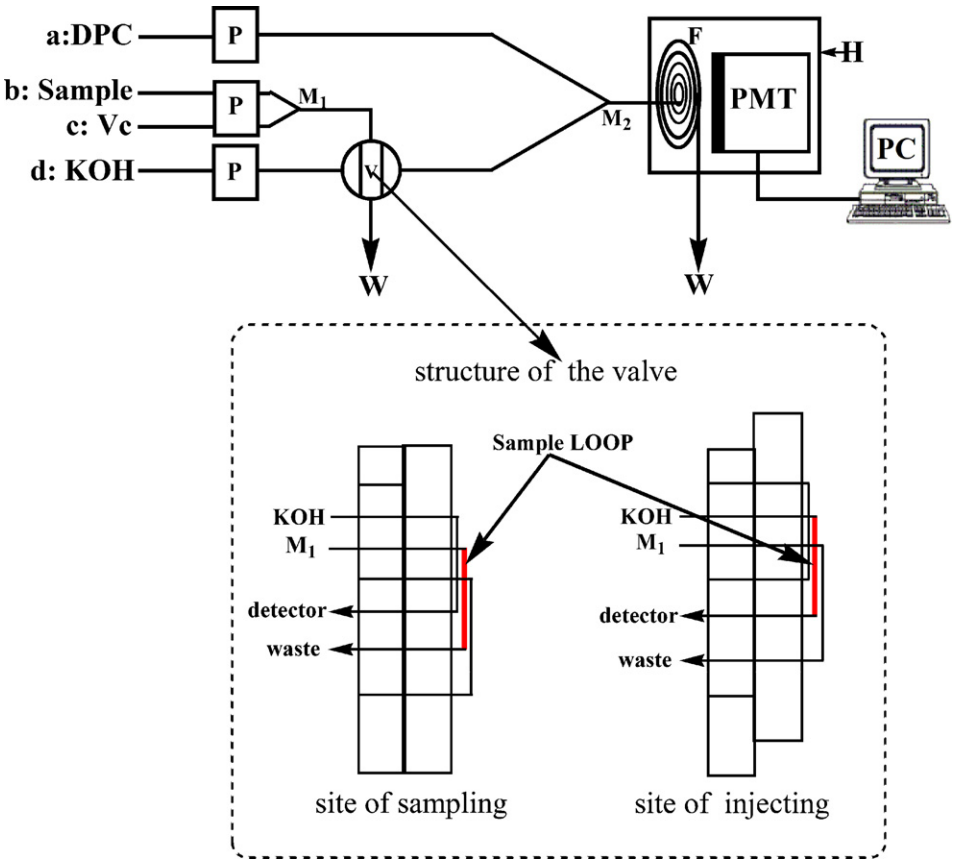

Fig. 1. Schematic diagram of the CL-FIA system: M_1, M_2: mixed position; V: injection valve; F: spiral glass flow cell; PMT: photomultiplier tube; PC: personal computer; H: light tight house; W: waste; P: peristaltic pump. (a) DPC solution; (b) analyte solution; (c) ascorbic acid solution; (d) carrier (KOH solution).

concentration of DPC is important. To test the effect of DPC solution, a range of concentrations of DPC (5×10^{-5} mol L^{-1} to 1×10^{-3} mol L^{-1}) was investigated. Seen from Fig. 3, the CL intensity was increased along with the increase of the DPC concentration in a low-concentration range, and reached the maximum at 5×10^{-4} mol L^{-1}. Above 5×10^{-4} mol L^{-1}, the CL intensity decreased probably because higher concentration of DPC caused self-absorption. So the optimal concentration was 5×10^{-4} mol L^{-1}.

3.2.2. The effect of carrier solution concentration

It is known that DPC was synthesized in strong alkaline solution (KOH solution). Additional, the pH conditions were obtained by the use of KOH solution. KOH was regarded as carrier solution, which bypassed the sample loop. The effect of KOH solution concentration (0.01–1 mol L^{-1}) was plotted in Fig. 4, which showed the optimal concentration of KOH solution was 0.1 mol L^{-1}. Above 0.1 mol L^{-1}, the CL base line uplifted and CL intensity decreased.

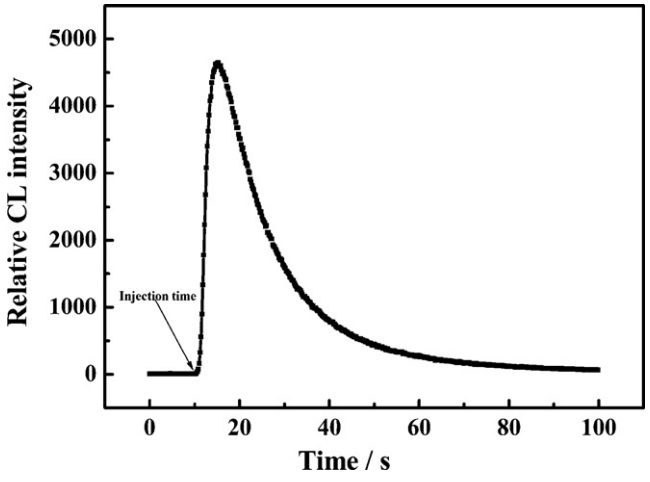

Fig. 2. The kinetics curves of DPC–EM–Vc. DPC, 5×10^{-4} mol L^{-1}; KOH, 0.1 mol L^{-1}; the present EM concentration was 4×10^{-7} g mL^{-1} (Vc: 1×10^{-5} g mL^{-1}).

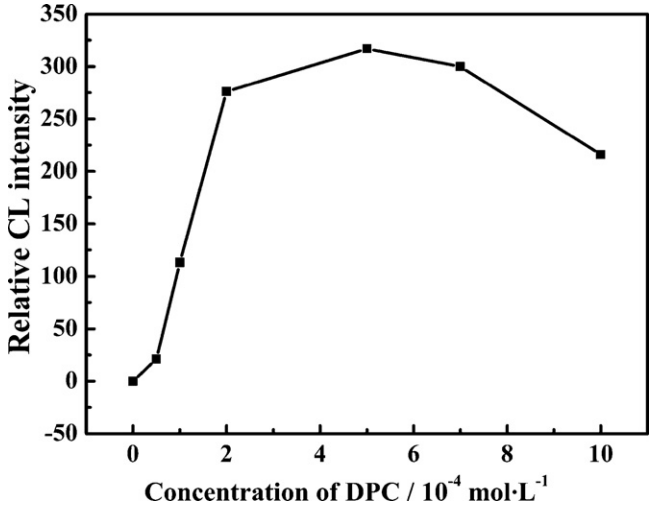

Fig. 3. The effect of the DPC concentration. KOH, 0.1 mol L^{-1}; Vc solution: 5×10^{-6} g mL^{-1}; EM, 4×10^{-8} g mL^{-1}.

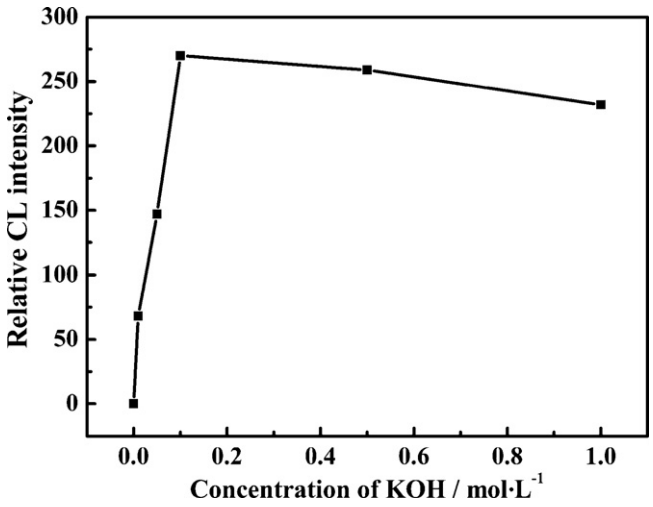

Fig. 4. The effect of KOH solution concentration.
DPC solution, 5×10^{-4} mol L^{-1}; Vc solution: 5×10^{-6} g mL^{-1}; EM: 4×10^{-8} g mL^{-1}.

For obtaining the highest sensitivity and accuracy, the concentration of 0.1 mol L^{-1} of KOH solution was selected as optimum carrier solution.

3.2.3. The effect of ascorbic acid solution concentration

It was found that the relative CL intensity of DPC–EM enhanced in the presence of the solution of ascorbic acid, which acted as sensitizer. The effect of ascorbic acid solution concentration was compared in the range of 1×10^{-6} g mL^{-1} to 1×10^{-4} g mL^{-1} in Fig. 5. The results showed that the optimal concentration of ascorbic acid solution was 1×10^{-5} g mL^{-1}. Above 1×10^{-5} g mL^{-1}, the relative CL intensity decreased for the change of pH. For obtaining the highest sensitivity and accuracy, the concentration 1×10^{-5} g mL^{-1} of ascorbic acid solution was selected as optimum.

3.3. The analytical characteristic of the FI-CL method

Under the optimum experimental conditions, the calibration graph of change of CL intensity, ΔI, against EM concentration was linear in the range of 4.0×10^{-9} g mL^{-1} to 4.0×10^{-7} g mL^{-1} and the detection limit was 1.1×10^{-9} g mL^{-1} (3σ). The linear chart of CL intensity against EM concentration was shown in Fig. 6. The linear calibration range was 4.0×10^{-9} g mL^{-1} to 4.0×10^{-7} g mL^{-1} with the regression equation $\Delta I = 10.02C + 108.63$ (C was the EM concentration at ng mL^{-1} level). The relative standard deviation for 8.0×10^{-9} g mL^{-1} EM solution was 2.1% ($n = 7$).

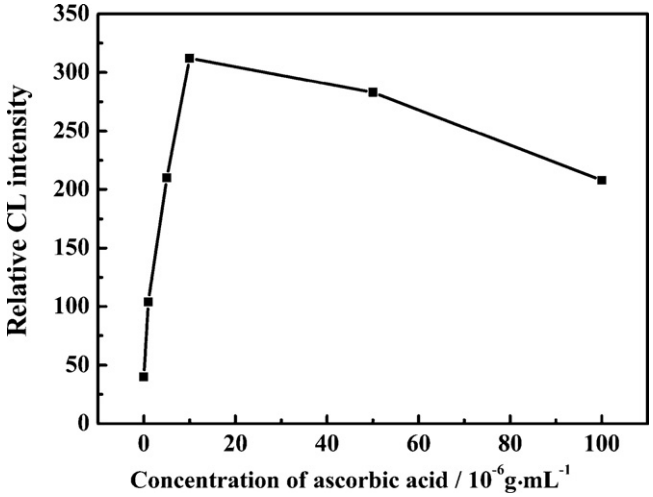

Fig. 5. The effect of ascorbic acid solution concentration. DPC solution, 5×10^{-4} mol L^{-1}; EM, 4×10^{-9} g mL^{-1}; KOH, 0.1 mol L^{-1}.

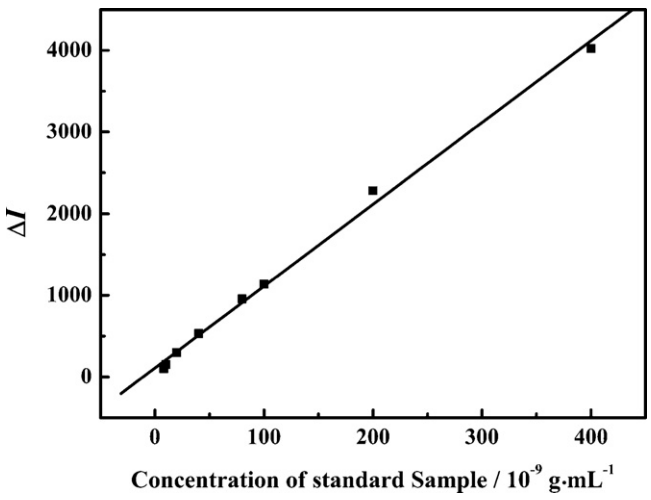

Fig. 6. The linear chart of CL intensity versus EM concentration.

Table 1
The tolerable ratio of coexisting foreign interfering substance.

Coexisting foreign interfering substance	Tolerable ratio
K^+, Na^+, Ca^{2+}, Mg^{2+}, Al^{3+}, NH_4^+, Mn^{2+}, Cl^-, Br^-, NO_3^-, SO_4^{2-}, IO_4^-, SeO_3^{2-}	1000
Fe^{3+}, $C_2O_4^{2-}$, Fe^{2+}	200
Citric acid, lactic acid	100
Lactose, glucose, sugarcane and uric acid	10

3.4. Influence of coexisting foreign species

The interference studied was conducted by analyzing a standard solution of 5×10^{-8} g mL^{-1} EM, to which varying amounts of possible interfering substances were added. The tolerated limit for each foreign specie was taken as a relative error not greater than ±5%. The tolerable ratio for foreign species was listed in Table 1. The interference of protein in human serum could be ignored when human serum was ultrafiltered, thus implying that the present method may be directly applied to the determination of EM in human serum.

3.5. Analytical applications of the flow CL method

Following the procedure described above, the proposed method was applied to the determination of EM in pharmaceutical injection and serum samples.

3.5.1. Pharmaceutical injections

The EM injection (0.2 mg mL^{-1} per tube) was diluted to different concentrations with double-distilled water. The results are listed in Table 2, which agreed well with those obtained by the fluorescence analysis method.

3.5.2. Serum sample

The proposed method was applied to determine EM in human's serum sample. 1 mL of fresh serum was diluted to 100 mL with PBS solution. A known amount of EM standard solution was added to propose the recovery test. The results of the recovery test are showed in Table 3. As can be seen from Table 3, the recoveries was from 98.5% to 104.0% and t-test assumed that there was no signifi-

Table 2
Results for the determination of EM in pharmaceutical injection samples.

Sample	Measured value (10^{-7} g mL^{-1})[a]		RE (%)[b]
	Proposed method (A)	Fluorescence method [23] (B)	
No. 1	2.15	2.08	3.4
No. 2	3.56	3.63	1.9
No. 3	7.92	8.05	1.6

[a] Average value of the measure ($n = 3$).
[b] $((A - B)/B) \times 100\%$.

cant difference between recoveries and 100% at confidence level of 95%.

3.6. Comparison with other CL systems

Based on the reaction of EM and DPC on present of ascorbic acid, a novel CL method was proposed for the analysis of EM in serum. Although several CL method [24,25] have been reported for the determination of EM, the method reported in Ref. [24] was only applied to the determination of EM in pharmaceutical preparations for its poor selectivity. Ref. [25] reported that the DL was 2.6 μg L^{-1}. It lay particular emphasis on the set-up of CL sensor, but not on the CL system self. While the proposed novel method can be applied to the determination of EM in human serum for its higher sensitivity and selectivity (Table 3).

3.7. Mechanistic studies of the CL reaction

3.7.1. The oxidant property of copper metal chelate

The electrochemical oxidation process is helpful to explain the oxidant characteristic of DPC. The copper(II) metal chelate was prepared in strong alkaline solution, which contained copper(II) and metaperiodate anion. An obvious oxidation peak was observed at 0.42 V on the cyclic voltammetry curve of the solution of copper(II) metal chelate on the platinum electrode in 0.1 mol L^{-1} potassium hydroxide solution, in the potential range 0–0.7 V (vs. SCE; Fig. 7). Seen from Fig. 7 the electrochemical process was quasi-reversible. ΔE (as the difference of oxidation potential and reduction potential) was 88 mV. The electron transfer process was a single electron process. The copper(II) could be oxidized to copper(III). While the oxidation potential was lower than that of other oxidant (such as $KMnO_4$, Ce(IV)). Compared with these oxidant, copper(III) metal chelate was a weak oxidant. DPC was the stable oxidation products of copper(II) metal chelate by chelating with polydentate ligands (IO_4^-), which could probable applied in the CL system for its oxidation.

3.7.2. Kinetics curve of the CL reaction of EM–DPC

We also found that EM was directly oxidized by DPC in basic solution, which exhibits an intense luminescence. So a typical response curve (intensity versus times) of EM in DPC solution was

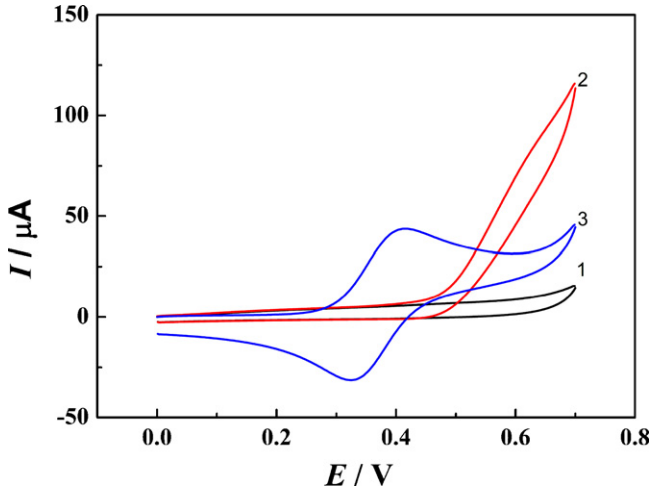

Fig. 7. Cyclic voltammetry curve of copper(II) metal chelate on the platinum electrode in strong alkaline solution (KOH). Line 1: KIO_4 + KOH; line 2: Cu(II) + KOH; line 3: Cu(II) + KIO_4 + KOH. [Cu^{2+}] = 0.01 mol L^{-1}; [IO_4^-] = 0.03 mol L^{-1}; scan rate (V/s) = 0.001; segment = 2; sample interval (V) = 0.001; quiet time (s) = 2; sensitivity (A/V) = 1×10^{-4}.

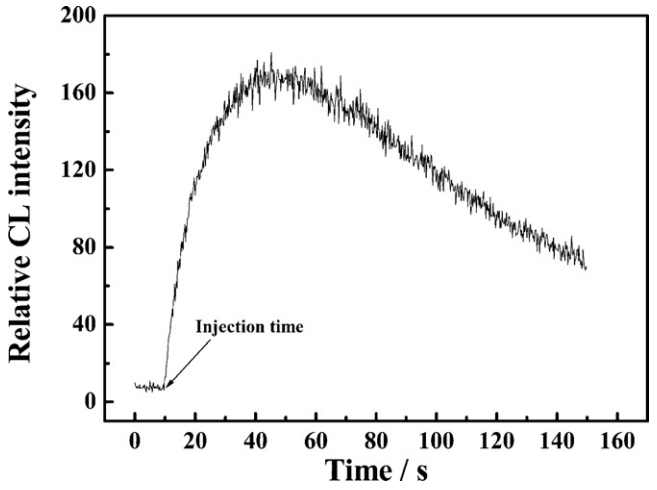

Fig. 8. The kinetics curves of DPC–EM. DPC 5×10^{-4} mol L^{-1}; KOH 0.1 mol L^{-1}; the present EM concentration was 4×10^{-7} g mL^{-1}.

also recorded to study the kinetic characteristic of the CL reaction of EM and DPC. Fig. 8 demonstrated the maximum CL intensity appeared within 30 s since the EM solution was injected into DPC solution (in KOH solution). As seen from Fig. 8, the CL reaction of DPC and EM is not a conventional quick reaction. Compared with Fig. 2, the ascorbic acid could enhance the CL intensity, which acted as a sensitizer. The CL system based on the reaction of EM and DPC was applied to the determination of EM with a detection limit of 3.1×10^{-9} g mL^{-1} (3σ).

Table 3
Results of recovery tests on serum samples.

Sample	EM content in sample (10^{-9} g mL^{-1})	Measurement concentration of EM (10^{-9} g mL^{-1})		Recovery (%)	R.S.D. (%, $n = 3$)
		Added[a]	Found[a]		
No. 1	5.41	1.00	6.45	104.0	0.9
		2.00	7.38	98.5	1.1
		5.00	10.52	102.2	0.8
No. 2	8.72	4.00	12.78	101.5	2.1
		8.00	16.64	99.0	0.7
		12.00	20.80	100.7	1.3

[a] Average value of the measure ($n = 3$).

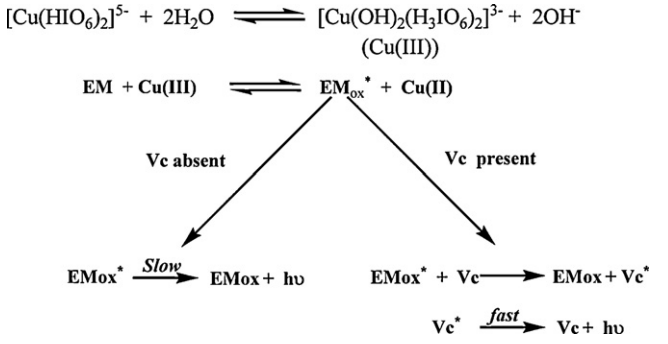

Fig. 9. A reaction mechanism suggested for the CL reaction.

3.7.3. Reaction mechanism

Based on the previous study of oxidation of threonine and isoniazid by Cu(III) [9,10], the formation of $[Cu(H_3IO_6)_2(OH)_2]^{3-}$ was suggested to be the reactive species of water-soluble Cu(III) periodate complex in alkaline medium. While the ascorbic acid is character of reducibility, DPC could directly oxidize the ascorbic acid. The kinetics curve of the CL reaction of DPC-Vc and DPC-Vc-EM was similar. The CL intensity could be enhanced in the presence of the EM. The CL reaction of DPC and Vc-EM is a quick reaction. The CL reaction mechanism of DPC-Vc-EM was proposed in Fig. 9.

4. Conclusion

In this work, a novel oxidant DPC has been used for its oxidant effect on the EM, DPC can be readily prepared and stabilised in alkaline media. The use of Cu(III) as a single oxidant in CL has been extensively explored. The novel CL emitted based on the CL reaction of Cu(III) and EM in the presence of ascorbic acid has been developed. The ascorbic acid was proved to be a sensitizer. It not only could improve the detection limit, but also remove the interference of ascorbic acid in the serum sample. The usage of ascorbic acid made it possible that the oxidation of EM by DPC was used for quantitative analysis of ergometrine maleate in real sample. The detection limit of EM was at nanogram level. The DPC–EM–Vc CL system has been successfully applied in the determination of EM in human serum without any pretreatment. It indicates that Cu(III) based CL system is a potential analytical method. Moreover, the Cu(III) can widen the application of CL method in the further study.

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