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Determination of melamine in dairy products by an electrochemiluminescent method combined with solid-phase extraction

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ABSTRACT

An electrochemiluminescence (ECL) enhancement method combined with solid-phase extraction has been developed for the determination of melamine in dairy products. It was found that melamine in a strong base solution is able to enhance the ECL of $Ru(bpy)_3^{2+}$ at glass carbon electrode. The optimum experimental conditions for the determination of trace melamine by ECL, such as scan mode and scan rate of the applied potential, the type of buffer solutions and their pH conditions, were investigated. Under optimized conditions, the enhanced ECL intensity was linearly proportional to the logarithm of melamine concentration in the range of 0.01–1.0 ppb, and the detection limit was 0.003 ppb. The method has been successfully demonstrated to determine melamine in dairy products including liquid milk, yogurt and milk powder samples. The relative standard deviations ranging from 5.3% to 11.2% and the recoveries from 95.2% to 102.4% were acquired by this method. A possible mechanism for the ECL enhancement effect was also proposed.

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1. Introduction

Melamine (1,3,5-triazine-2,4,6-triamine) (Fig. 1) is a triazine-based chemical intermediate commonly used in the manufacture of amino resins, plastics and flame retardants. It is not approved for use as a feed or food additive. However, it has been added illegally into feedstuffs or food to artificially distort their crude protein content. It received considerable public attention after the pet food contamination scandal in North America in 2007 and the melamine-tainted-milk powder incident in China in 2008. To protect public health and food safety, many countries and groups has established the criteria of Maximum Residue Limits (MRLs) for melamine in various everyday products [1]. For example, the European Union (EU) has set the MRL of melamine in dairy products/high protein foods as 2.5 ppm. The US Food and Drug Administration (FDA) have set the MRL of melamine in milk and dairy products/milk food as 0.25 ppm, and stressed that infant formula sold to US consumers must be completely free of melamine. On April 22nd 2010, the Ministry of Health of China published a new dairy safety standards and emphasized that food should not be tainted with melamine. In other words, any act of adding melamine into dairy products artificially is illegal, even if the amount is much lower than the MRL. Therefore, there is an urgent need to establish a simple, effective and highly sensitive method for the analysis of trace melamine in dairy products.

Many analytical techniques are available to determine melamine in dairy products, for example, chromatography [2–18], mass spectrometry (MS) [19,20], Enzyme-linked immunosorbent assay (ELISA) [21], colorimetric visualization [22–25], electrochemical method [26–31], near-infrared spectroscopy [32–35] and flow injection chemiluminescence [36]. The chromatographic and MS methods require expensive apparatus and professional operators. ELISA usually requires at least 10–12 samples for determination. Colorimetric visualization and electrochemical method are simple, but their precision and/or sensitivity are poor. Near-infrared spectroscopy method is a very good in situ determination one due to its simplicity and very short assay time, but the sensitivity is not high enough. Flow injection chemiluminescence based on a luminol–myoglobin system was also proposed for the determination of melamine, but myoglobin is very expensive and the method requires significant quantity of the sample. Detection and quantification of trace levels of melamine in raw milk and dairy products using HPLC, preceded by separation and pre-concentration on a Cleanert PCX SPE column was recently proposed [18,37,38]. Electrochemiluminescence (ECL) not only has the advantages of high sensitivity, low detection limit and high selectivity, but also it is easy [39–41]. To the best of our knowledge, there is no ECL method that has been developed for the determination of trace melamine in dairy products to date.

In this work, a strong ECL enhancement due to melamine at a glass carbon electrode (GCE) in a $Ru(bpy)_3^{2+}$ solution was demon-

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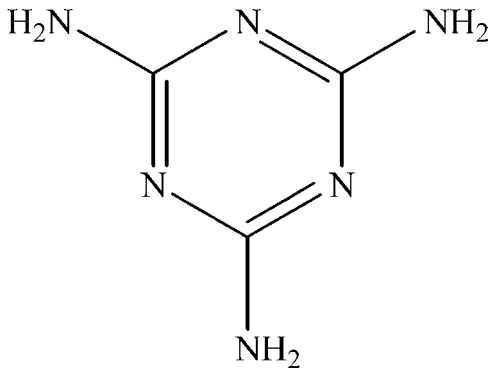

Fig. 1. Structure of melamine.

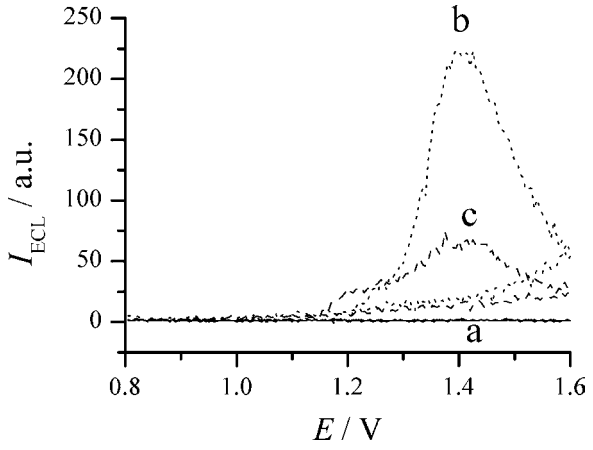

Fig. 2. I_{ECL}–E curves of 0.1 ppb melamine (a, solid line), 1.0×10^{-6} mol/L $Ru(bpy)_3^{2+}$ + 0.1 ppb melamine (b, dotted line) and 1.0×10^{-6} mol/L $Ru(bpy)_3^{2+}$ (c, dashed line) on the glass carbon electrode in NaOH–KCl buffer solution (pH 13.0). Scan rate: 0.05 V/s.

strated. Based on the enhancement behavior, a simple, highly sensitive ECL approach for the determination of melamine in dairy products was thus developed. The sensitivity achieved by this ECL method is the highest among the reported methodologies. The enhancement mechanism of melamine to the ECL of $Ru(bpy)_3^{2+}$ was also discussed.

2. Experimental

2.1. Chemicals and solutions

$Ru(bpy)_3Cl_2{\cdot}6H_2O$ was purchased from Sigma Chemical Company (St. Louis, MO, USA). Melamine was purchased from China National Medicines Co. Ltd. (Shanghai, China). All other chemicals were of analytical grade and used without further purification. Water obtained from a Milli-Q water purification system (Millipore, Bedford, MA, USA) was used throughout. Samples of dairy products were purchased from the local market and used directly. Cleanert PCX-SPE cartridges (3 mL/60 mg) were obtained from Beijing Agela Technologies Company (Beijing, China).

Stock solutions of $Ru(bpy)_3^{2+}$ at the concentration of 1.0×10^{-2} mol/L were stored in a refrigerator at 4 °C in dark, and were used to prepare working standard solutions by appropriate dilution with water. Working standard solutions of melamine at different concentrations were prepared in NaOH–KCl buffer solution (pH = 13) just prior to use.

2.2. Apparatus

ECL measurements were carried out using a home-made setup consisting of a BPCL Ultra-Weak Chemiluminescence Analyzer (Institute of Biophysics, Chinese Academy of Sciences, Beijing, China) and a CHI 660A electrochemical analyzer (Shanghai Chenhua Instrument Co., Shanghai, China), which was controlled by a personal computer. A conventional three-electrode configuration was employed, with a glass carbon electrode used as the working electrode, a platinum wire as the counter electrode and an Ag/AgCl (sat. KCl) electrode as the reference electrode. A tailor-made 5 mL cylindroid quartz cell was used as the ECL cell, and was directly placed in the front of the photomultiplier tube.

Fluorescence experiments were run using a Hitachi (Tokyo, Japan) F-4600 spectrofluorimeter. Excitation was at 452 nm, the lowest energy absorption for the $Ru(bpy)_3^{2+}$ luminophore, with detection between 480 nm and 900 nm. UV–Vis absorption spectra were recorded with a Persee General (Beijing, China) TU-1901 spectrophotometer.

2.3. ECL procedure

A 10 μL of 1×10^{-4} mol/L $Ru(bpy)_3^{2+}$ and 500 μL of the pretreated samples or melamine working standard solutions were added to the ECL cell subsequently, and then 490 μL of NaOH–KCl buffer solution (pH = 13) was added. The curves of ECL intensity versus applied potential (I_{ECL}–E) and the curves of current versus applied potential (i.e. CV) were recorded simultaneously. Applied potential was in the range of +0.8–1.6 V with the scan rate of 0.05 V/s. Determination was based on the enhanced ECL intensity ΔI, $\Delta I = I_s - I_0$, where I_0 was the background ECL intensity of $Ru(bpy)_3^{2+}$ in the absence of melamine, and I_s was the sample signal. All experiments were done at room temperature (25 ± 1 °C).

Prior to each measurement, the working electrode was polished consecutively with 1.0 μm, 0.3 μm and 0.05 μm α-Al_2O_3 aqueous slurries on a chamois leather to obtain a mirror surface, sonicated and thoroughly rinsed with water, and then dried using compressed air at room temperature.

2.4. Sample preparation

Procedure of the sample preparation was nearly identical with previous references [18,37,38]. A 2.0 g of dairy products was weighed accurately and was placed into a 50 mL centrifuge tube. Then, 20 mL of 1.0% trichloroacetic acid solution and 2 mL 2.2% lead acetate solution were added in order to eliminate proteins and extract the analytes. The mixture was ultrasonicated for 20 min, and then allowed to stand for 2 min. The solution was then centrifuged for 10 min at 10,000 rpm. A 10 mL volume of the supernatant obtained was added into a PCX-SPE cartridge, which was conditioned with 3 mL of methanol and 3 mL of water. After the SPE cartridge was washed in turn with 5 mL of water and 5 mL of methanol, melamine was eluted with 6 mL of 25% ammonia solution–methanol (1:20, v/v). The eluate was evaporated to dryness at 50 °C under a stream of nitrogen and the residue was re-dissolved in 1.0 mL of NaOH–KCl buffer solution (pH = 13).

3. Results and discussion

3.1. ECL behavior of melamine

The control experiments showed that melamine did not give ECL at GCE in the absence of $Ru(bpy)_3^{2+}$ (Fig. 2a). However, the ECL intensity of $Ru(bpy)_3^{2+}$ at GCE in the presence of melamine (Fig. 2b) was much higher than that using $Ru(bpy)_3^{2+}$ alone (Fig. 2c).

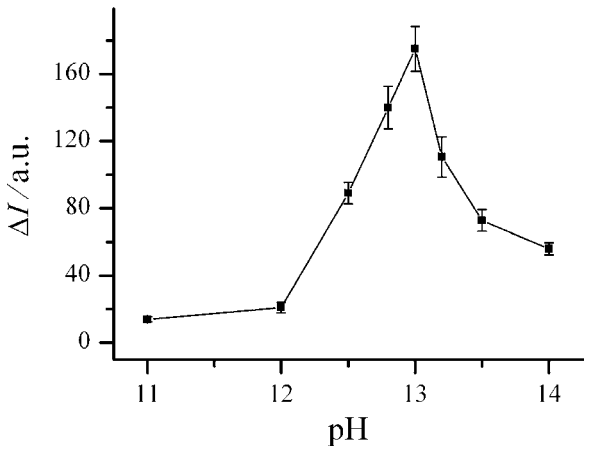

Fig. 3. Effect of pH on ΔI. Other experimental conditions were same as those in Fig. 2.

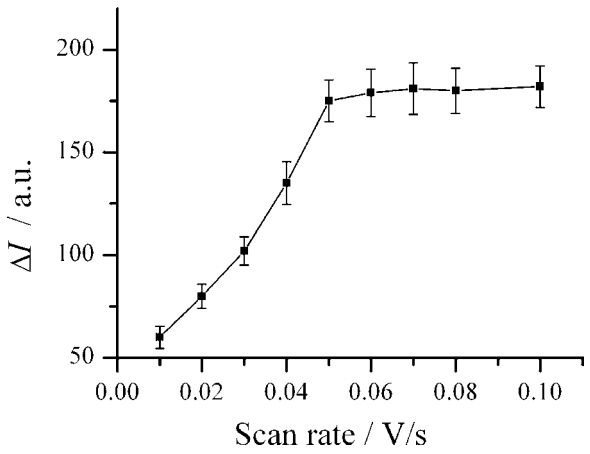

Fig. 4. Effect of scan rate on ΔI. Other experimental conditions were same as those in Fig. 2.

The significantly increased ECL signal indicates that melamine even under 0.1 ppb is able to enhance the ECL of $Ru(bpy)_3^{2+}$ effectively.

3.2. Optimization of experimental conditions

The enhanced ECL intensity ΔI was in correlation with many factors including the type of the buffer solution used and its pH conditions, the scan mode and scan rate of the applied potential. In order to obtain a higher sensitivity of ECL, all these factors were optimized.

3.2.1. Effect of buffer solution and its pH conditions

The ECL behavior of $Ru(bpy)_3^{2+}$/melamine system was investigated in different buffer media (pH = 13), such as Britton–Robinson buffer solution, NaOH–H_3BO_3 buffer solution, NaOH–Na_2SO_4 buffer solution and NaOH–KCl buffer solution. Results showed that the maximum ΔI was obtained in a NaOH–KCl buffer solution (prepared by mixing 66.0 mL of 0.2 mol/L NaOH solution and 25.0 mL of 0.2 mol/L KCl solution; the resulting solution was then diluted to 100 mL with water).

In addition, ΔI showed significant dependence on the pH of the NaOH–KCl buffer solution. As shown in Fig. 3, ΔI was not high when the pH value was less than 11.0. When the pH was adjusted in the range of 11.0–13.0, ΔI increased remarkably with the increase in pH of the solution. And the trend for ΔI was declined when pH > 13.0. Therefore, NaOH–KCl buffer solution at pH = 13.0 was selected as the optimal condition for ECL experiments.

3.2.2. Selection of the scan mode and scan rate of applied potential

Cyclic voltammetry, square wave voltammetry and symmetric double step pulse voltage scanning were used to examine the ECL behavior of $Ru(bpy)_3^{2+}$/melamine system. The results showed that the higher signal-noise-ratio was obtained under the cyclic voltammetry mode.

Using scanning mode of cyclic voltammetry, the effect of scan rate on the enhancement of ΔI of $Ru(bpy)_3^{2+}$ was examined and the results were shown in Fig. 4. It was found that ΔI increased when the scan rate was below 0.05 V/s, and reached a plateau afterwards. Therefore, 0.05 V/s was chosen as the optimal scan rate for subsequent studies.

3.3. Linear response range and detection limit

Under the optimized conditions (in a NaOH–KCl buffer solution at pH = 13.0, scan rate at 0.05 V/s), the linear response range

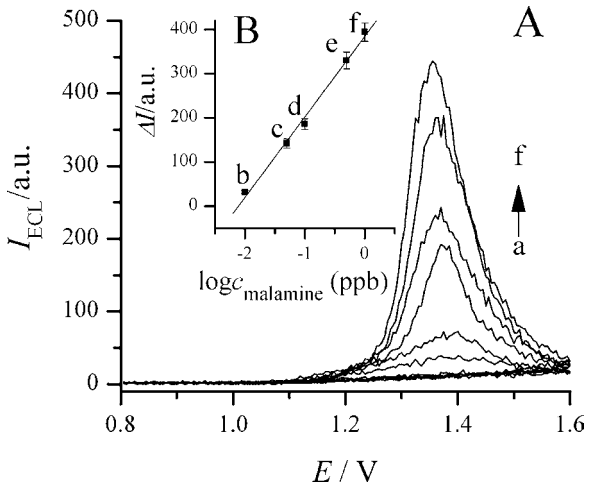

Fig. 5. I_{ECL}–E curves of 1.0×10^{-6} mol/L $Ru(bpy)_3^{2+}$ with different concentration of melamine (A), and calibration curves between ΔI and $\log c_{melamine}$ (ppb) (B). a → f: 0, 0.01 ppb, 0.05 ppb, 0.1 ppb, 0.5 ppb, 1.00 ppb; experimental conditions were same as those in Fig. 2.

and the detection limit for the melamine was established. As shown in Fig. 5, ΔI was linearly proportional to the logarithm of the melamine concentration within the range of 0.01–1.0 ppb. The detection limit was 0.003 ppb. The regression equation was $\Delta I = 384.22 + 181.98 \times \log c_{melamine}$ (ppb) with a correlation coefficient of 0.9969. The low detection limit indicated that this method is very sensitive for the determination of trace melamine. The sensitivity of this method for the determination of melamine in some dairy products was listed in Table 1. The detection limits (LOD) or quantitation limits (LOQ) were also compared with other reported methods. It can be found that this method exhibits advantages in the detection limit, which was same as the most sensitive method reported in literature.

3.4. Interference

In order to assess the proposed method for the analysis of melamine in dairy products, the interference effects of some substances which were expected to be present in the dairy samples were examined. The solutions used for this purpose contained 0.1 ppb melamine and interfering species. The upper limit of an interfering species was estimated under the conditions that the relative error for determination of a standard melamine solution was less than 5%. The tolerable concentration ratios were deter-

Table 1
Comparison of main methods reported for the determination of melamine in dairy products.

Analytical method	Product type	LOD/LOQ	References
This method	Dairy products	0.003 ppb (LOD)	
GC–MS	Milk sample	0.001 μg/mL (LOD)	[4]
	Dairy products	0.01 mg/kg (LOD)	[7]
GC–MS/MS	Milk products	0.004 mg/kg (LOD)	[5]
HPLC	Milk	0.02 mg/kg (LOQ)	[6]
	Liquid milk	18 μg/kg (LOD)	[18]
	Milk products	0.021 μg/mL (LOD)	[11]
LC–MS–MS	Milk	0.025 mg/kg (LOD)	[15]
	Milk-based products	0.01–0.1 mg/kg (LOD)	[13]
	Liquid milk	0.03 μg/g (LOD)	[10]
HILIC-UV	Milk powder	0.005 mg/mL (LOD)	[8]
HILIC-ESI-MS/MS	Raw milk	0.05 mg/kg (LOQ)	[3]
	Dairy products	0.05 mg/kg (LOQ)	[3]
CE	Liquid milk	0.5 mg/kg (LOQ)	[2]
	Milk samples	0.047 μg/mL (LOD)	[16]
	Dairy products	0.01 μg/mL (LOD)	[17]
MECC-AD	Milk products	2.1 μg/mL (LOD)	[12]
MS	Raw milk	500 ppb (LOD)	[19]
	Milk powder	170 μg/kg (LOD)	[20]
ELISA	Milk products	7.6–9.4 μg/L (LOD)	[21]
Colorimetric visualization	Raw milk	2.5 ppb (LOD)	[22]
	Milk products	25 ppb (LOD)	[23]
	Raw milk	0.4 mg/L (LOD)	[24]
	Milk products	0.15 ppm (LOD)	[25]
Electrochemical	Milk products	3.0×10^{-9} mol/L (LOD)	[26]
	Milk products	9.6×10^{-9} mol/L (LOD)	[27]
Near-infrared spectroscopy	Milk powder	1 ppm (LOD)	[32,34]
FI-CL	Milk products	3.0 pg/mL (LOD)	[36]

mined as followings: >1,000,000 for all interfering species studied including NO_3^-, Ac^-, Cl^-, CO_3^{2-}, SO_4^{2-}, PO_4^{3-}, Zn^{2+}, NH_4^+, Cu^{2+}, Fe^{3+}, Ba^{2+}, Ca^{2+}, Mg^{2+}, Ni^{2+}, Cr^{3+}, Fe^{2+}, fructose, borate, lactose, glucose, urea, lysozyme, uric acid, VB_1, VB_2, VB_6 and VC. The anti-interference ability of this method was much better than that of the flow injection chemiluminescence [36], due to the pretreatment of the sample. Defatting and protein precipitation were accomplished using trichloroacetic acid solution and lead acetate solution in the first step. All interfering species above-mentioned would be washed away by water and methanol in the washing procedure of the subsequent solid-phase extraction.

3.5. Application

The proposed method was applied to quantify melamine in the liquid milk, yogurt and milk powder samples. As shown in Table 2, good relative standard deviations (RSDs) of 5.3–11.2% and recoveries of 95.2–102.4% were obtained. Thus, this proposed method shows potential application for the determination of melamine in dairy products.

3.6. Possible mechanism of ECL enhancement

The preliminary study showed that the maximum emission wavelength of the ECL emission spectrum for $Ru(bpy)_3^{2+}$, in the absence and the presence of melamine, were both at 620 nm in a NaOH–KCl buffer solution at pH = 13. This indicated that the electrochemiluminescence is corresponding to the light emission of $[Ru(bpy)_3^{2+}]^*$ species. Considering that melamine itself gave no ECL signal, the ECL enhancement of $Ru(bpy)_3^{2+}$/melamine system was no doubt due to the effect of melamine on the ECL of $Ru(bpy)_3^{2+}$. UV–Vis absorption spectra strongly suggested that no new intermediate was produced as simply mixing $Ru(bpy)_3^{2+}$ and melamine in a solution. It is likely that the product from electrochemical reaction enhanced the ECL of $Ru(bpy)_3^{2+}$.

To probe the nature of the products formed during electrolysis, bulk electrolysis coupled with fluorescence detection was also used. The potential was stepped to +1.6 V to oxidize $Ru(bpy)_3^{2+}$ and

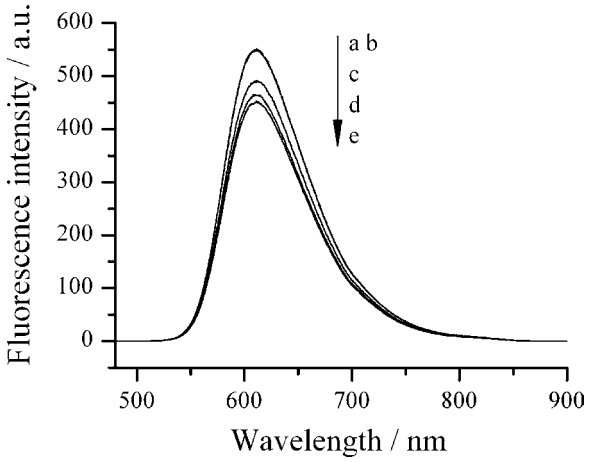

Fig. 6. A series of fluorescence spectra. Concentration of melamine was 0.6 ppb and other experimental conditions were same as those in Fig. 2c. (a) $Ru(bpy)_3^{2+}$, electrolyzed for 0 min; (b) $Ru(bpy)_3^{2+}$/melamine system, electrolyzed for 0 min; (c) $Ru(bpy)_3^{2+}$/melamine system, electrolyzed for 60 min; (d) $Ru(bpy)_3^{2+}$/melamine system, electrolyzed for 120 min; (e) $Ru(bpy)_3^{2+}$/melamine system, electrolyzed for 180 min.

melamine effectively. As shown in Fig. 6, the signal decreased with increasing time of electrolysis in $Ru(bpy)_3^{2+}$/melamine system. It indicated that a product of melamine is directly responsible for the luminescence signal and the ECL enhancement behavior again [42].

Based on our experimental results and ECL enhancement mechanisms previously reported [39–41,43], a mechanism for the ECL enhancement of $Ru(bpy)_3^{2+}$/melamine system was proposed as shown in Fig. 7. The two compounds were oxidized on the electrode to produce their corresponding products. Then, the active neutral free radical intermediate produced by the oxidation of melamine would react with the electrogenerated $Ru(bpy)_3^{3+}$, which would help make the production of the excited $Ru(bpy)_3^{2+*}$. Consequently, the ECL signal from $Ru(bpy)_3^{2+*}$ is thus strongly enhanced.

Table 2
Determination of melamine in liquid milk, yogurt and milk powder samples.

Sample	$c_{melamine}$ (ppb)					Recovery (%)
	Found before adding[a]	RSD (%)	Melamine added	Found after adding[a]	RSD (%)	
Liquid milk	0.687 ± 0.044	6.4	0.20	0.872 ± 0.056	6.4	98.3
	0.721 ± 0.051	7.1	0.25	0.980 ± 0.068	6.9	100.9
	0.654 ± 0.055	8.4	0.30	0.942 ± 0.078	8.3	98.7
Yogurt	0.165 ± 0.017	10.3	0.10	0.258 ± 0.020	7.8	97.4
	0.136 ± 0.012	8.8	0.15	0.275 ± 0.028	10.2	96.1
	0.142 ± 0.012	8.5	0.20	0.349 ± 0.025	7.2	102.0
Milk powder	0.031 ± 0.003	9.7	0.10	0.132 ± 0.010	7.6	100.8
	0.045 ± 0.004	8.9	0.20	0.251 ± 0.028	11.2	102.4
	0.038 ± 0.003	7.9	0.30	0.322 ± 0.017	5.3	95.2

[a] Mean ± SD, $n = 5$.

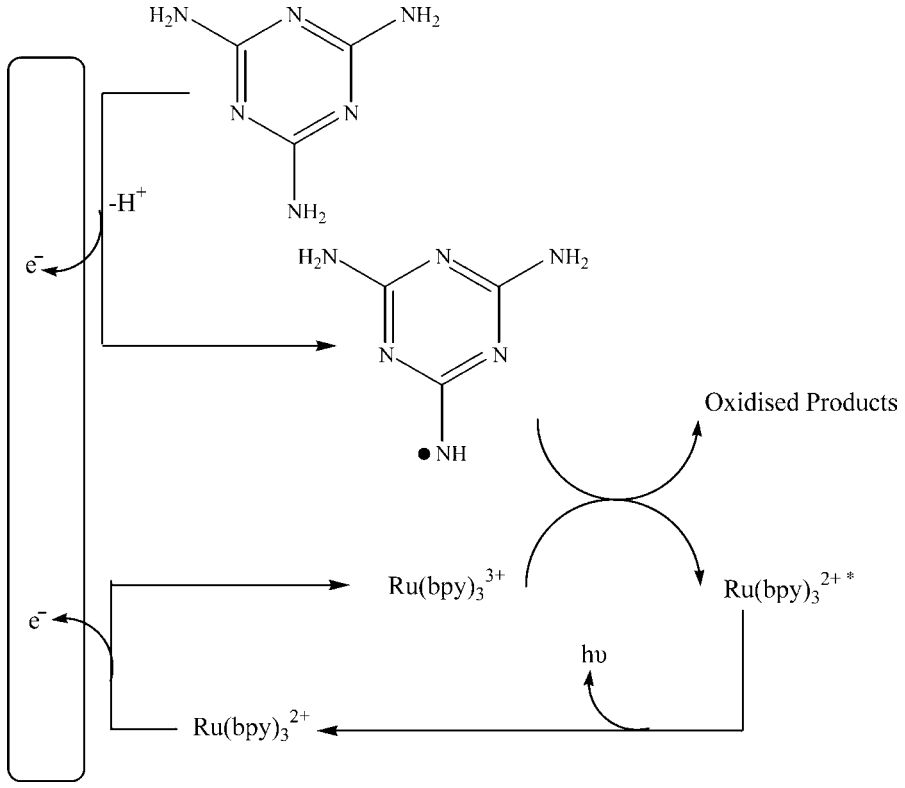

Fig. 7. Mechanism for the ECL enhancement of $Ru(bpy)_3^{2+}$/melamine system.

4. Conclusions

A simple, sensitive and accurate method was developed for the determination of melamine in dairy products based on the electrochemiluminescence combined with solid-phase extraction. It was found that melamine could be able to enhance the ECL of $Ru(bpy)_3^{2+}$ in strong base solution. The enhanced ECL intensity was linearly proportional to the logarithm of melamine concentration. The linear relationship was found over a wide concentration range of melamine. Under the optimized conditions, the detection limit for melamine was 0.003 ppb. In addition, a possible mechanism for the ECL enhancement of $Ru(bpy)_3^{2+}$ by melamine was proposed.

Acknowledgments

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