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Determination of hydralazine with flow injection chemiluminescence sensor using molecularly imprinted polymer as recognition element

Yan Xiong, Houjiang Zhou, Zhujun Zhang *, Deyong He, Chao He

Institute of Analytical Science, Department of Chemistry, Southwest China University, Beibei, Chongqing 400715, PR China

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Abstract

A novel flow injection chemiluminescence (CL) sensor for hydralazine determination using molecularly imprinted polymer (MIP) as recognition element is reported. Hydralazine–MIP was prepared through non-covalent copolymerization using methacrylic acid (MAA) monomer, hydralazine template and ethylene glycol dimethacrylate (EGDMA) cross-linker. Particles of the MIP were packed into a v-shape glass tube for on-line adsorption of the analyte of hydralazine. The adsorbed hydralazine could be sensed by its great enhancing effect on the CL reaction between luminol and periodate. The CL intensity is linear to hydralazine concentration in the range from 2×10^{-9} to 8×10^{-7} g/mL. The detection limit is 6×10^{-10} g/mL (3σ) and the relative standard deviation is 2.8% ($n = 7$) for 8×10^{-9} g/mL hydralazine. The selective experiment showed that the selectivity and sensitivity of the CL method could be greatly improved when MIP was used as recognition element in the flow-injection CL sensor. The sensor was reversible and reusable. It could be used for more than 100 times. It has been used directly to determine the hydralazine in human urine.

Keywords: Hydralazine; Flow injection chemiluminescence sensor; Molecularly imprinted polymer

1. Introduction

Hydralazine (Fig. 1) is used as an antihypertensive vasodilator drug. It was used to lower blood pressure in people with hypertension and the muscles that control the diameter of blood vessels were relaxed by hydralazine. This relaxation allowed the blood vessels to dilate (open wider), lowering blood pressure. Hydralazine also was used after heart valve replacement and in the treatment of congestive heart failure. But excessive or habitual uptake of hydralazine can cause toxic symptoms, such as headache, joint or muscle pain, swollen ankles and others [1]. To prevent overdose-induced toxication, the sensitive monitoring of hydralazine concentrations in a patient's blood or urine sample is necessary. Many analytical techniques for hydralazine detection, including mass spectrometry (MS) [2], gas chromatography (GC) [3], high-performance liquid chromatography (HPLC) [4,5], colorimetry [6] and spectrometry [7,8] have been developed. However, the instruments of these methods not only are expensive and complicated, but are also laborious and time consuming.

In recent years many extremely sensitive analytical techniques based on chemiluminescence (CL) reaction have received considerable attention. Simplicity of detection, low detection limit, wide linear ranges and short analysis times are some of the characteristics that make the method attractive. When coupled with flow-injection analysis (FIA), the CL-based FIA method provides cheap, rapid, simple and reproducible means of detection and, therefore, it has been successfully applied to many drug detection fields.

Though several CL methods have been reported for the determination of dihydralazine [9,10], there is only one report on the CL determination of hydralazine [11]. CL reaction of luminol and periodate can be enhanced by many compounds [12–17]. In our work, it is found that the CL intensity produced by the CL reaction between luminol and periodate can be greatly enhanced by hydralazine. According to this CL behavior, a simple, sensitive and rapid CL method for the determination of hydralazine is developed. However, CL method is limited in real sample analysis because of its weak selectivity.

* Corresponding author. Tel.: +86 23 68253863; fax: +86 23 68253863.
E-mail addresses: xiongyanliumingsheng@yahoo.com.cn (X. Yan), zzj18@hotmail.com (Z. Zhang).

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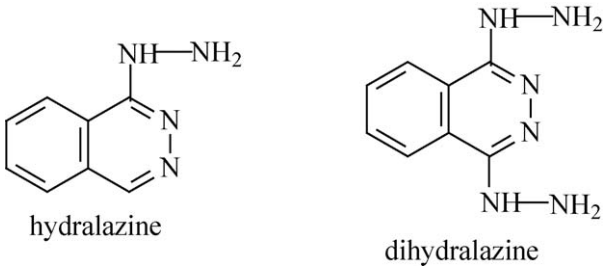

Fig. 1. The structure of hydralazine and dihydralazine.

Molecular imprinting is a process by which functional and cross-linking monomers are co-polymerized in the presence of the target analyte, which acts as a molecular template. Subsequent removal of the templates leaves behind binding sites that are complementary to the target analyte in the resultant molecularly imprinted polymer (MIP). MIPs possess advantages of physical robustness, rigidity, resistance to elevated temperatures and pressures, and inertness towards acids, bases, metal ions and organic solvents compared to biomolecules. MIPs have been extensively used in biosensors, to mimic enzyme catalysis, for solid-phase extraction (SPE) and HPLC stationary phase [18–25]. Thus, molecularly imprinted polymers (MIPs) are considered a more reliable, robust and simple strategy for specific recognition compared to other separation techniques.

The use of MIP as recognition element in flow injection CL sensors has been developed in recent years. The specific recognition function of MIP can improve the selectivity of CL analysis. Lin and Yamada have devised a MIP–CL flow through sensor for 1,10-phenanthroline [26]. Du et al. have developed the MIP–CL sensor for determination of epinephrine [27]. In our laboratory, we have already reported the MIP–CL sensors for determination of clenbuterol [28] and salbutamol [29].

In this paper, a novel flow injection CL sensor for hydralazine determination using MIP as recognition element is developed. Hydralazine–MIP can be prepared by a non-covalent imprinting approach, in which radical co-polymerization of methacrylic acid (MAA, functional monomer) and ethylene glycol dimethacrylate (EGDMA, cross-linker) is carried out in the presence of hydralazine template molecules. Particles of the resultant MIP were packed into a piece of v-shape glass tube, which served as a flow cell positioned in front of a photomultiplier. Hydralazine is on-line selectively adsorbed on the MIP, reacting with luminol and periodate to produce strong CL. After the CL reaction, the absorbed hydralazine was destroyed and removed by the flow solution with the cavities left on the MIP for the adsorption of the next sample. The sensor is reusable and reversible and reusable. It can be used for more than 100 times. It has been used successfully to determine hydralazine in human urine.

The CL sensor proposed in this work shows some distinguished advantages.

First, in order to achieve the specific recognition, chromatography [3–5] is commonly adopted for hydralazine determination, in which impurities can be gotten rid of. However, chromatography methods not only need expensive and complicated instruments, but also need laborious and time consuming manipulation. MIP is easily prepared and the cost of MIP is comparatively cheap. Usually MIP is prepared with three-dimensional network by copolymerization. Removal of the templates molecule results in a functional polymeric matrix with recognition sites complementary in size, shape and functionality to the template molecule. So MIP provides special recognition and selective function for template molecule. Using MIP as recognition element in the CL sensor, the selectivity of the CL method can be greatly improved and the interference of some species commonly present in urine sample can be gotten rid of. As a result, the MIP-based CL sensor is used directly to determine the analyte of hydralazine in real urine sample.

Second, when MIPs were used as recognition element in the CL sensor, the structure of the templates adsorbed on the MIPs can be destroyed through the CL reaction and the templates that have been reacted can be easily washed off from the MIPs using water as eluent. It avoids the organic reagents and buffer solutions from being used as the eluents, which can affect the CL reaction. As a result, the analysis procedure becomes simple.

At last, the MIPs can on-line enrich the analyte of hydralazine and the enrichment effect of the MIPs can greatly improve the sensitivity of the CL analysis.

2. Experimental

2.1. Reagents and materials

2,2′-Azobisisobutyronitrile (AIBN, initiator) was of chemical purity grade and the other reagents were of analytical grade. Doubly distilled water was used for the preparation of solutions and throughout the experiments. MAA and AIBN were obtained from Shanghai Chemical Reagent Company (Shanghai, China). EGDMA was obtained from Guangzhou Shuangjian Trade Company (Guangzhou, China). Acetonitrile, acetone, acetic acid, methanol and luminol were obtained from Chongqing Chemical Reagent Company (Chongqing, China). Hydralazine was kindly supplied by Dafeng Tiansheng Pharmaceutical Co. Ltd. (Tianjing, China). Hydralazine stock solution of 1 mg/mL was prepared in water. It was stable when stored at 4 °C for several months. Working solutions were prepared afresh for daily use. Stock solutions of KIO_4, NaOH and luminol were prepared by dissolving 1 g in 100 mL water, respectively. EGDMA and MAA were purified by distilling and AIBN was purified by recrystallizing before use.

2.2. Apparatus

The schematic diagram of flow system used in this work is shown in Fig. 2. Two peristaltic pumps (type HL-2, manufactured at Huxi Instrument and Meter Plant, Shanghai, China) were used to deliver the flow streams. PTFE tubing (0.8 mm i.d.) was used to connect all components in the flow system. One hundred and fifty microliters mixture solutions of potassium periodate and luminol were injected into the carrier stream (water) using an eight-way injection valve for sampling and the solutions then reached the flow cell to react with the adsorbed hydralazine.

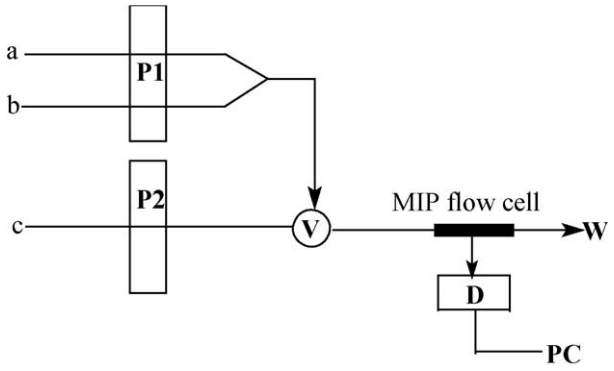

Fig. 2. Schematic diagram of the flow sensing system: (a) luminol solution, (b) potassium periodate solution, (c) hydralazine or doubly distilled water, (V) six way valve, (W) waste solution, (D) detector, (PC) personal computer and (P1, P2) peristaltic pumps.

The emitted CL signal was detected and recorded with a computerized IFFM–D flow-injection chemiluminescence data processing software (Xi'an Remex Electronic High-Tech Ltd.). The UV absorbance was detected by UV-2001 spectrophotometer (Hitachi Ltd., Japan).

2.3. Hydralazine–MIP preparation

The MIP was synthesized in a 100 mL pyrex flask vial containing 600 mg of hydralazine (3 mmol, print molecule), 1 mL of methacrylic acid (12 mmol, functional monomer) and 25 mL of methylbenzene (solvent). 12 mL of ethylene glycol dimethacrylate (EGDMA, 60 mmol, cross-linker) and 328 mg of 2,2′-azobis-(2-methylproprionitrile) (AIBN, 2 mmol, initiator) were added. The mixture was purged with nitrogen for 20 min and sonicated in a water bath for 5 min. Polymerization was then caught through in a 60 °C water bath for 24 h. A colorless translucent bulk of solid MIP was obtained. The MIP was broken and ground in a mortar with a pestle. The ground MIP particles were sieved to collect the particles of 50 μm. The sieved powder was washed with methanol/acetic acid (9/1, v/v) mixture until hydralazine was not detected in the filtrate solution. The washed solid powder was dried in vacuum. A control polymer was also prepared similarly without the use of hydralazine. The possible polymerization process is shown in Fig. 3.

2.4. MIP flow cell make-up

Fifty milligrams MIP was packed into a v-shape colorless glass tube (5 mm i.d. × 5 cm length), which served as the flow cell. The tube was positioned in front of the photomultiplier and both ends were stuffed with adequate glass wool to prevent the MIPs from being taken away by the flow solution. Before the first use of the flow cell, CL reagents of luminol and periodate were flowed through the cell to react with hydralazine on the MIP. Until the CL intensity became blank, distilled water was used to remove the residues of the CL reaction away and clean the polymer. The cavities complementary to hydralazine were left on the MIP for hydralazine adsorption.

2.5. Separation and preconcentration of hydralazine

With the flow injection system shown in Fig. 2, the procedure for separation and preconcentration determination of hydralazine is summarized in four steps.

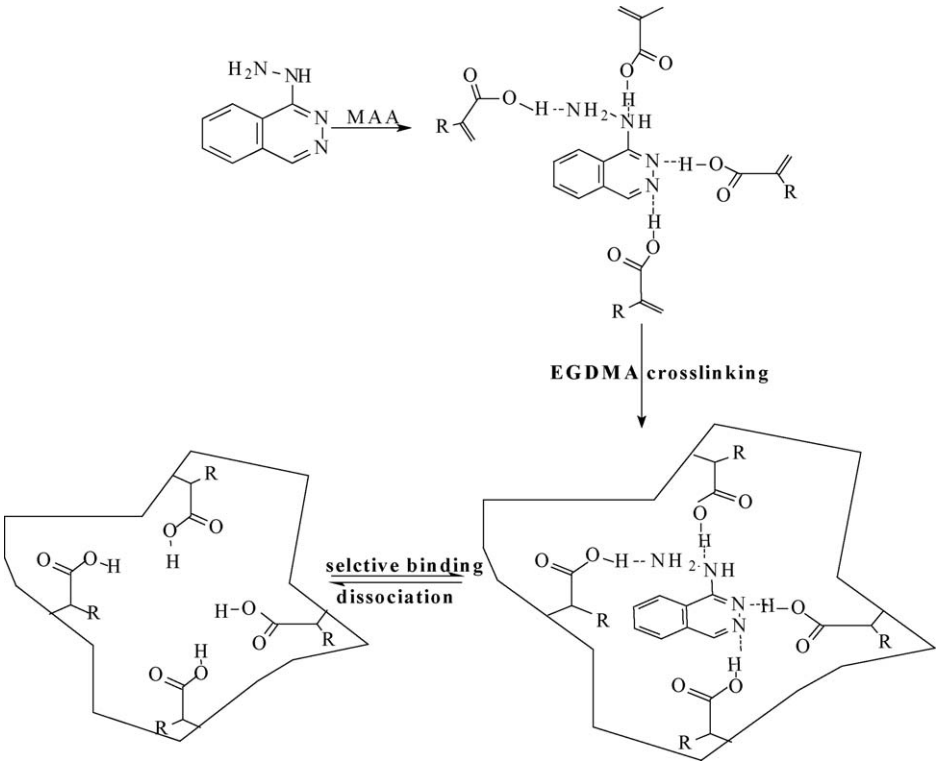

Fig. 3. The preparation of hydralazine imprinted polymer.

Step 1 (preconcentration of hydralazine): pump 1 is stopped and hydralazine solution is delivered to flow through the flow-cell by pump 2. As a result, hydralazine is selectively adsorbed on MIP.

Step 2 (washing polymer to remove other substances except hydralazine): pump 1 is also stopped but water carrier replacing the hydralazine solution is delivered by pump 2 to flow through the cell to take the purities except the adsorbed drug away.

Step 3 (chemiluminescence determination of hydralazine): after a proper washing time, using the eight-way injection valve, the emerging stream of potassium periodate and luminol delivered by pump1 is injected into the carrier water delivered by pump 2. When the CL reagents flow through the flow cell, they react with the hydralazine adsorbed on the polymer and produced strong CL.

Step 4 (cleaning the cavities on the polymer): when the CL intensity of the reaction comes to be blank, pump 1 is stopped immediately and water carrier delivered by pump 2 is flowed through the cell to clean the polymer cavities for the next determination.

2.6. Binding measurements

The binding characteristic of the hydralazine-imprinted polymer was estimated by dynamic method. The hydralazine solution was continuously flowed through the MIP-flow sensor with 50 mg MIP packed at the constant flow rate. Then the absorbance of free hydralazine in the efflux solution was detected.

3. Results and discussion

3.1. Optimum condition of luminol–KIO_4–hydralazine system

The reaction between luminol and periodate produced CL emission. To get a low background emission, luminol and periodate solutions were mixed together in a proper distance before they flowed through the flow cell. A strong CL emission was recorded when hydralazine was injected into the flow cell to react with the mixed solution. The CL intensity was proportional to the concentration of hydralazine.

To achieve the best effect of the CL reaction of luminol–KIO_4–hydralazine, the optimal conditions for the CL reaction system were investigated by measuring the ratio of the height of the CL signal (1×10^{-7} g/mL hydralazine) to noise influence (S/N) at different concentration level of luminol, KIO_4 and NaOH. The results showed that the CL emission intensity increased with higher concentration of luminol at fixed concentration of KIO_4 and NaOH. The background emission also increased. The maximum S/N ratio was found with 4×10^{-4} mol/L luminol solution and higher concentration gave lower S/N ratio (Fig. 4). The same phenomenon was found with KIO_4 solution and the results indicated that 8×10^{-5} mol/L KIO_4 solution gave the maximum S/N ratio (Fig. 5). With the selected concentration of lumiol solution and KIO_4 solution, the optimal concentration of NaOH in this reaction system was chosen and 4×10^{-2} mol/L NaOH concentration was selected for subsequent experiments. Higher concentration of NaOH caused increment of background emission. Thus, the chosen conditions for this CL system are 4×10^{-4} mol/L luminol, 8×10^{-5} mol/L KIO_4 and 4×10^{-2} mol/L NaOH.

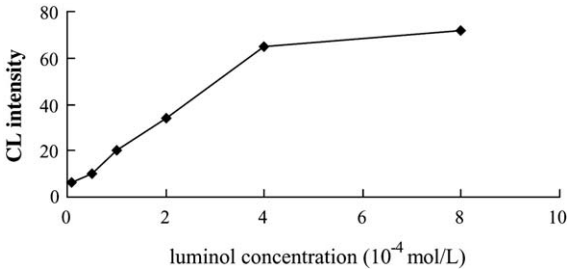

Fig. 4. Luminol concentration effect on CL intensity.

3.2. Discussion of the possible CL reaction mechanism and the possible sensing principle of the sensor

At the selected conditions, there was no CL emission from the reaction of hydralazine with KIO_4 in the absence of luminol. It was also difficult to observe CL emission when hydralazine was injected into the solution of luminol without KIO_4. However, CL emission occurred when hydralazine was injected into the mixed solution of luminol and KIO_4. Thus, the CL emission spectrum of this CL system was recorded accordingly to illustrate the emission property. The maximum wavelength was found at 425 nm, which suggested that the possible emission species was the oxidated 3-aminophthalate. It could be proposed that the dissolved O_2 can be reduced to H_2O_2 by hydralazine since it has a similar reducing group as NH_2NH_2 [30]. Then the active oxygen can be produced by the chemical reaction between KIO_4 and H_2O_2 [31]. Hence, strong CL emission produced when luminol reacted with active oxygen. The mechanism could be shown as follows:

$$\text{Hydralazine} + O_2 \rightarrow H_2O_2$$

$$IO_4^- + H_2O_2 \rightarrow [O]$$

$$\text{Luminol} + [O] \rightarrow h\nu$$

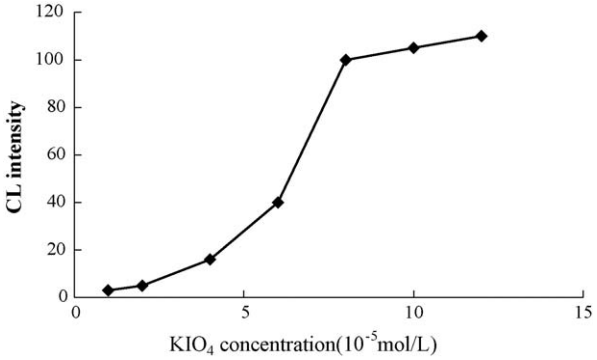

Fig. 5. KIO4 concentration effect on CL intensity.

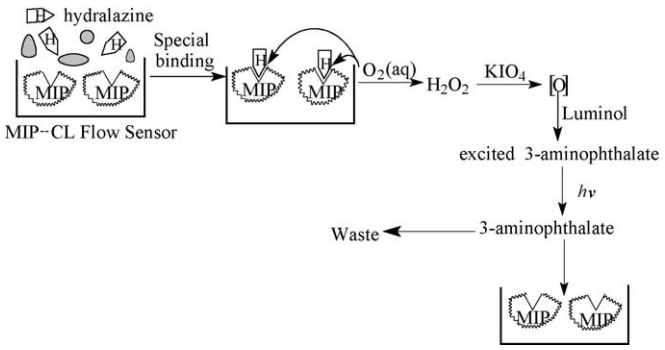

Fig. 6. The reaction principle of MIP–CL flow sensor and the possible mechanism of CL reaction.

The possible reaction principle of the sensor IS is shown in Fig. 6. Hydralazine was selectively adsorbed on the MIPs that were packed in the flow cell. Then CL reagents of luminol and KIO_4 flowed through the cell sensor and reacted with the adsorbed hydralazine to produce strong CL. After the CL reaction, the structure of hydralazine was destroyed and the cavities complementary to hydralazine were left on the MIP for the next determination of hydralazine.

3.3. Choice of conditions of the MIP–CL sensor

In this work, the experiments were conducted according to the schematic diagram shown in Fig. 2.

First, the preconcentration time of hydralazine was chosen. This aspect was examined under the condition that 50.0 mg polymer was filled into the flow cell sensor and the flow rate was fixed at 1.25 mL/min. It was noted that too long time would consume time and reagent but too short time would not have the hydralazine being absorbed. At last, 4 min was selected by examining 2×10^{-9} g/mL hydralazine from 2 to 8 min.

Second, the washing time with carrier water is the next critical aspect for the selective detection of hydralazine. Too long washing time would take hydralazine away together with other substances, resulting in decrease of sensitivity. The washing time was examined with the water carrier flow rate of 1.25 mL/min from 0 to 5 min. The experiments showed that 1.5 min could take other substances away other than hydralazine.

Third, the CL reagent volume of the emerging stream of KIO_4 and luminol solution is another key aspect. Too small volume could not react with adsorbed hydralazine at a time, while too large volume would need a long time to clean the excessive reagents. One hundred and fifty microliters sample volume was chosen from 50 to 400 mL.

At last, the cleaning time for the polymer was also very important for reproductivity. It was examined within 0–5 min with the flow rate of water carrier of 1.25 mL/min, and then 2 min was observed properly.

3.4. The flow CL sensor analytical performance for the determination of hydralazine

Under the selected conditions given above, CL intensity response to hydralazine concentration was linear in the range from 2×10^{-9} to 8×10^{-7} g/mL concentration with the detection limit of 6×10^{-10} g/mL (3σ). A typical response of CL intensity to 8×10^{-8} g/mL hydralazine with MIP-flow sensor is shown in Fig. 7. The regression equation is $I = 37.419C - 17.919$ (C being the hydralazine concentration (g/mL)) with a correlation coefficient of 0.9941 ($n = 7$). A complete analysis is performed with a relative standard deviation of 2.8% ($n = 7$). Without MIP as recognition material, the linear range, the detection limit and the relative standard deviation are from 4×10^{-8} to 8×10^{-7}, 1×10^{-8} g/mL (3σ) and 4.5%, respectively.

The results of the two analytical methods showed that using MIP as recognition material in the CL sensor, the sensitivity and selectivity of CL analysis could be greatly improved.

3.5. Hydralazine-imprinted polymer binding characteristics

In this work, the binding characteristic of hydralazine-imprinted polymer was estimated by the dynamic method.

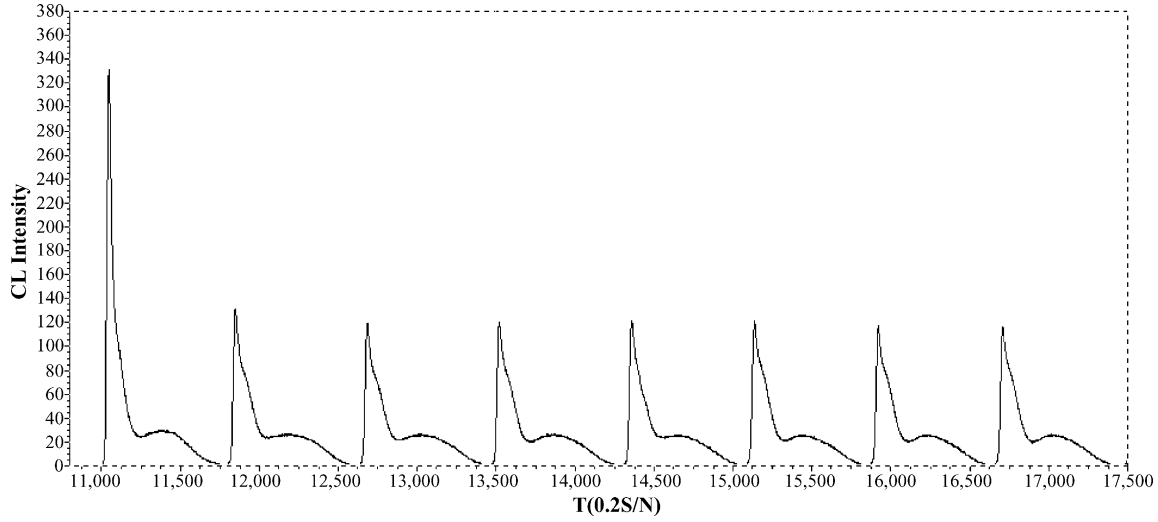

Fig. 7. The typical CL response by MIP–CL flow sensor for 8.0×10^{-8} g/mL hydralazine.

Table 1
Tolerable concentration ratios with respect to hydralazine for some interfering substances with and without MIP

Interfering substances	With MIP	Without MIP
Na^+, Cl^-, uric acid	1000	50
Urea	800	5
Starch, glucose, citric acid	500	3
Ascorbic acid	100	1
Dihydralazine	450	50

The hydralazine solution was continuously flowed through the MIP-flow sensor with 50 mg MIP packed at the flow rate of 1.25 mL/min. Then the absorbance of hydralazine in the efflux solution was detected every 3.5 mL. It was observed that the absorbance of hydralazine in the efflux solution was increasing with the increase of the hydralazine flowing time through the sensor. At last, the absorbance reached a constant value. The time for reaching constant value was different for different concentrations of hydralazine solution at the same flow rate. The lower concentration of hydralazine solution, the longer was needed. The equilibrium time for 1.0×10^{-5} mol/mL, 5.0×10^{-5} mol/mL hydralazine solution and the 1.0×10^{-4} mol/mL hydralazine solution at the flow rate of 1.25 mL/min were about 14, 8 and 5 min, respectively.

3.6. Selectivity

Under the chosen conditions and using the manifold depicted in Fig. 2, the interference of some species commonly present in urine was examined by analyzing a standard solution of 2×10^{-8} g/mL hydralazine. The tolerable limit of foreign species was considered with the relative error less than 5%. The results are listed in Table 1. Dihydralazine (Fig. 1), which has the similar structure with hydralazine, was chosen as an interfering substance to study the selectivity of the MIP. The result is listed in Table 1, too. These results showed that MIP could be used as recognition material in the CL analysis to improve selectivity and sensitivity.

3.7. Analytical application of MIP–CL flow sensor

The proposed method was used to detect hydralazine in urine sample obtained from a healthy person. A known amount of hydralazine standard solution was added to 2.5 mL of urine sample and the sample solution was then diluted to 25 mL with doubly distilled water. The recovery results are shown in Table 2.

Table 2
Results of recovery tests of hydralazine on urine samples

Sample	Amount added (ng/mL)	Amount found (ng/mL)	Recovery (%)	R.S.D. (%)
1	3.00	3.12	104.0	1.5
2	5.00	4.90	98.0	2.1
3	7.00	6.71	95.9	0.3
4	50.00	52.02	104.0	1.1
5	70.00	71.24	101.8	3.2

4. Conclusion

A novel CL sensor with hydralazine-imprinted polymers being used as recognition element was successfully developed in the present paper. The characteristic of the selective binding function of MIP to hydralazine enables the method to have the advantage of selectivity, which makes it possible to be applied to the analysis of hydralazine in urine sample directly. The MIP–CL flow sensor was successfully used for the analysis of hydralazine in human urine.

Acknowledgement

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