Talanta 85 (2011) 2614–2619

Contents lists available at ScienceDirect

Talanta

journal homepage: www.elsevier.com/locate/talanta

A novel microchip based on indium tin oxide coated glass for contactless conductivity detection

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ARTICLE INFO

Article history:
Received 10 June 2011
Received in revised form 11 August 2011
Accepted 13 August 2011
Available online 19 August 2011

Keywords:
Microchip capillary electrophoresis
Indium tin oxide
Contactless conductivity detection
Aminopyrine
Caffeine

ABSTRACT

A microfluidic chip manufactured from glass substrate and indium tin oxide (ITO) coated glass use for contactless conductivity detection was developed. The detecting electrodes were fabricated by screen-printing and chemical etching methods using an ITO-coated glass wafer. Then, the glass substrate containing separation channels was bonded with the bare side of the processed ITO-coated glass, thus producing an electrophoresis chip integrated with contactless conductivity detector. The prepared microchip displayed considerable stability and reproducibility. Sensitive response was obtained at optimal conditions (including the gap between electrodes, excitation frequency, and excitation voltage). The feasibility of this microfluidic device was examined by detection of inorganic ions, and further demonstrated by the quantification of aminopyrine and caffeine in a compound pharmaceutical. The two ingredients can be completely separated within 1 min. The detection limits were 8 $\mu g\,mL^{-1}$ and 3 $\mu g\,mL^{-1}$, respectively; with the correlation coefficient of 0.996–0.998 in the linear range from 10 $\mu g\,mL^{-1}$ to 800 $\mu g\,mL^{-1}$. The results have showed that the present method is sensitive, reliable and fast.

1. Introduction

Microchip capillary electrophoresis (MCE) technology has developed rapidly since it was first raised by Manz [1]. MCE has many advantages, for example, high efficiency, low sample consumption and frugal cost, and a wide field of applications. In recent years, a vast number of researches have focused on the development of detection system to offer improved analytical performance and ease of integration.

Electrochemical detection (ECD) includes three modes-amperometry, conductimetry and potentiometry [2]. Amperometric detection (AD) is the easiest implementation. It is accomplished by applying a constant potential to the working electrode and measuring the current as a function of time [3]. But the object detection and application field are limited. Potentiometric detection is conceptionally the most simple of the electroanalytical methods as it is not necessary to apply a voltage, but only necessary to measure a voltage between the measuring and reference electrodes. Thus, potentiometry can also be considered to be the most straightforward of all instrumental analytical techniques in terms of signal transduction [4]. However, slow response and relatively narrow application range are defective.

Conductivity detection is a universal tool to monitor conductance difference between analyte and background solution in the zone between two electrodes, and has been increasingly employed due to high sensitivity in small volume [5]. It is the favorite detection method to realize the concept of "lab on a chip" because of the development of microfabrication technology. It theoretically can be applied to all charged analytes, especially for small inorganic and organic ions, which are substantially low in electrochemical activity for amperometric detection or weak in optical absorbance for optical measurements [6]. Conductivity detection can be realized either by direct contact of the electrolyte and the electrodes [7], or by contactless fashion relying on capacitive coupling of an AC electrical field to solution [8,9]. In general, detecting electrodes are buried into the chip channel in the contact conductivity detection. The galvanic contact is problematic due to the incompatibility of high separation voltage on the one hand and sensitive detection circuit on the other. Chemical reactions and bubble formation may occur on electrodes, inducing additional noise on the read-out electronics and thus reducing detection sensitivity [10]. Contactless conductivity detection (CCD) is a particular form of conductivity-based detector, in which the electrodes are not in contact with the measured solution. The contactless mode presents two major advantages. First, the electronic circuit is simple and decoupled from high separation voltage. Second, the formation of bubbles and electrode fouling are avoided, thereby allowing a wide variety of electrode materials to be used.

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doi:10.1016/j.talanta.2011.08.029

Microchip electrophoresis with CCD was first described by Guijt et al. [11], who incorporated aluminum electrodes directly into the separation channel but covered them with a thin layer of a SiC insulating material to prevent their contact with the liquid in the channel. Lichtenberg [12] proposed a microchip where the detector was constructed with two opposite platinum electrodes placed close to the microchannel. The detection electrodes were isolated from the channel by a thin layer of glass. Pumera et al. [13] described a simple arrangement for MCE where two external electrodes consisting of strips of aluminum foil were affixed on the top of the poly(methyl methacrylate) (PMMA). Wang et al. [14,15] used a similar electrode fabrication procedure to create a movable CCD. The aluminum strip electrodes were attached to a cover slip, which was independent of the PMMA microchip. We have reported a microchip-based conductivity detector, where the external electrodes were microfabricated by printed circuit board [6]. It is convenient in operation because the chip is independent from electrodes, but there is an air layer between chip and electrodes which leads a bad analytical reproducibility.

In this work, we present a novel contactless conductivity detector that is made of common ITO-coated glass for microchip capillary electrophoresis. The detecting electrodes can be readily fabricated based on screen-printing technique. Furthermore, the low-cost and easily available ITO-coated glass offers alternative opportunity for mass-production of microchip-CCD devices in ordinary laboratories. The proposed method is fairly attractive because the fabrication of electrode is simple, and holds potential for a broad range of applications.

2. Experimental

2.1. Materials, reagents and samples

Tris (hydroxymethyl) aminomethane (Tris) was obtained from Shanghai Sangon Biological Engineering Technology & Services Co. Ltd. (Shanghai, China). 2-(N-Morpholino) ethanesulfonic acid (MES) was obtained from AMRES Co. Ltd. (Hongkong, China). Histidine (His) was purchased from Shanghai Bio Life Science & Technology Co. Ltd. (Shanghai, China). Other reagents were from Guangzhou Chemical Reagent Co. Ltd. (Guangzhou, China). Reference standards of aminopyrine and caffeine were purchased from National Institute for the Control of Pharmaceutical and Biological Products (Beijing, China). Aminopyrine and caffeine tablets were produced by Haiwai Medicine Co. Ltd. (Changchun, China). All chemicals were of analytical reagent grade and used without further purification.

Standard solutions of aminopyrine and caffeine were prepared in water at the concentration of 2.0 g L^{-1}. Twenty pills of aminopyrine and caffeine tablets were pounded in mortar, a suitable amount of powder, containing 150 mg aminopyrine and 40 mg caffeine, was dissolved in 30.0 mL redistilled water. It was then agitated for 15 min on an ultrasonic bath; the solution was diluted to 100.0 mL and then filtered. Subsequently, the filtrate was diluted with 100-fold for electrophoresis analysis. The processed solutions were kept from light. All of solutions were filtered through 0.22 μm nylon filters prior to use.

2.2. Apparatus

2.2.1. Device fabrication

The MCE system was composed of a high voltage power supply, a contactless conductivity detector and an ITO coated glass microchip. The high voltage supply was made of piezoelectric ceramics, providing a potential of constant DC of 0.1–0.5 kV for injection and a potential of constant DC or pulse of 0.5–5 kV for separation [16]. The contactless conductivity detector provided oscillation frequency of 0–200 kHz and oscillation voltage of 0–100 V (peak to peak, V_{pp}). The detector was connected to a personal computer with an A/D converter (model PCL-711B, EVOC, Taiwan, China).

2.2.2. The manufacture of the assembled microchip system

The construction of this system was shown in Fig. 1.

(i) Preparation of mask

The layout of the microchannel network was designed by using CorelDraw software and it was produced by photoreduction of a laser-printed drawing. As shown in Fig. 1B, the layout of the microchip was printed on a transparency film, using white for the regions where the microfluidic channels were formed.

The mask of electrodes for CCD in chip cover was prepared in the same manner (Fig. 1G).

(ii) Preparation of substrate plate

In this section, the procedures were referred to the previous study [17]. Injection channel and separation channel were photolithographed and chemically etched on a chrome wafer (type SG2506, Shaoguang Chrome Blank Co., Changsha, China) shown in Fig. 1D. The chrome wafer was first exposed to UV radiation (Fig. 1E), and then immersed in 0.5–0.7% NaOH solution. After dechromisation, the glass substrate was then placed into the etching solution ($HF:NH_4F:HNO_3$ = 1:0.5:0.5) for 40 min at 50 °C in water bath. As depicted in Fig. 1F, separation channel (g) and the double-T injection channels (e and f) were in the same width and depth [18]. Four access holes of 1.8-mm diameter use for the reservoirs were drilled to connect with the corresponding channels.

(iii) Preparation of cover plate

The ITO coated glass (1.3 mm thick) was obtained from Leybold Optics Corp. (Shenzhen, China). The glass was first sliced into small pieces (6.7 cm × 3.5 cm). Before use, these slices were cleaned with sonication in chemical solution ($HCl:FeCl_3:H_2O$ = 1:3:1) [19], deionized water and ethanol, each for 10 min, respectively. The planar electrodes mask for CCD was shown in Fig. 1G, in which the parts "–"of "T" acted as electrodes, while the parts "|"of "T" were used to connect the electrodes to circuit. The distance between the two electrodes was 2.0 mm. In the middle of two electrodes was a 0.2-mm-wide ground wire to decrease noise level. The electrodes were fabricated on a silk-screen (Fig. 1H). For screen-printing, oil ink (essential ingredient of acrylic resin) was transmitted through the silk-screen onto the ITO layer by brush (Fig. 1J). Once the planar electrodes were completed, a wet chemical etching procedure was carried out with chemical solution as mentioned above, which produced the desired ITO conductivity electrodes.

(iv) Thermal bonding

Prior to chip bonding, the structured glass and the processed ITO-coated glass were thoroughly cleaned with absolute ethanol, detergent, and water in sequence to remove contaminants on the surface. The two parts were to be thermal bonding according to a programmed temperature procedure (up to 570 °C) in a muffle furnace [17]. The electrodes of the assembled microchip (Fig. 1N) were coated with conducting silver resin to provide electric contact with the detector.

2.3. Electronic circuit

The fundamental principle of CCD is that two electrodes were placed in close proximity to the microchannel where electrophoresis was performed [20]. As shown in Fig. 2A an AC signal applied to one of the electrodes, was coupled capacitively through the

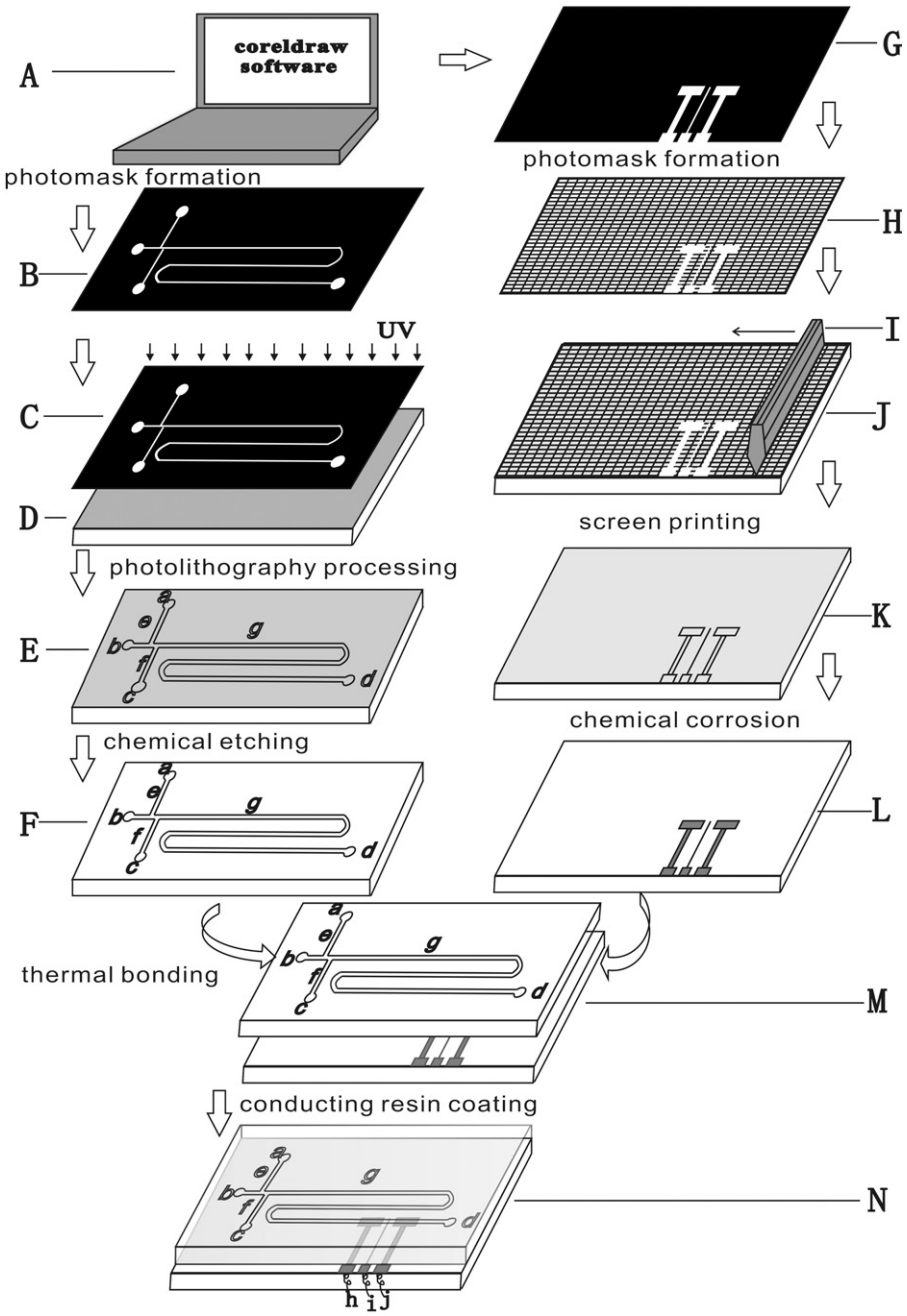

Fig. 1. Schematic illustration of the assembled microchip-contactless conductivity detector system: (A) computer; (B and C) photomask of chip; (D) chrome blank; (E) glass substrate with channels design; (F) substrate plate; (G) the mask of electrodes; (H) silk screen with planar electrodes pattern; (I) brush; (J) ITO coated glass; (K) ITO coated glass with shaped electrodes; (L) cover slip with conductivity electrodes; (M) the fully assembled microchip and (N) the microchip with conducting resin; (a) sample reservoir; (b) buffer reservoir; (c) sample waste reservoir; (d) buffer waste reservoir; (e and f) double-T injection channel; (g) separation channel; (h) input electrode terminal; (i) earth electrode terminal and (j) output electrode terminal.

microchannel to the second pickup electrode. The coupled signal of the same frequency was weaker at the pickup electrode with a negative phase shift. The pickup current was subsequently amplified and converted to a voltage and a DC signal by using a low-pass filter, and then biased to zero before it was sent to a Digital Acquisition (DAQ) system for further disposing. Fig. 2B is the equivalent circuit of this CCD system, its total impedance (Z) can be expressed as:

$$Z = \frac{1}{j\omega C_y} + \frac{1}{1/r + j\omega C_x} = \frac{r}{1+\omega^2 C_x^2 r^2} + j\frac{1+\omega^2 C_x(C_x+C_y)r^2}{\omega C_y(1+\omega^2 C_x^2 r^2)}$$

where r and C_x are resistance and capacitance of the electrolyte solution in the zone between two electrodes, respectively; C_y is the total capacitance between electrodes and solution.

When the ions separated in microchannel pass through the section of the channel within the detection electrodes, the total impedance of the system is altered and a change in the level of the pickup signal is produced for a short interval of time. This leads to a sudden change in the level of the resultant output signal and appears as a peak on the DAQ system.

2.4. Electrophoresis procedures

The channels of the new microfluidic chip were flushed with 0.1 mol L^{-1} NaOH, deionized water and electrolyte solution. Then,

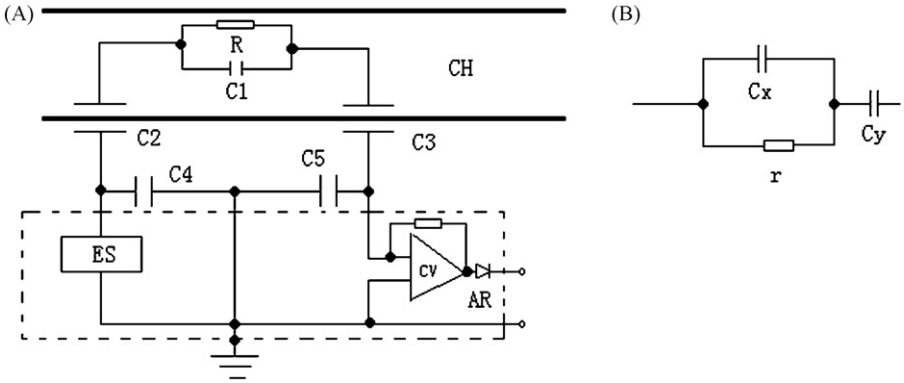

Fig. 2. (A) The circuit of the contactless conductivity system. CH: channel in the microchip; R and C_1: resistance and capacitance of the electrolyte solution in the zone between two electrodes; C_2 or C_3: capacitance between electrode and solution; C_4 or C_5: capacitance between electrode and earth; ES: excitation signal source; CV: current-to-voltage converter; AR: absolute rectifier. (B) The equivalent circuit of the CCD system. r and C_x: resistance and capacitance of the electrolyte solution in the zone between two electrodes, respectively; C_y: the total capacitance between electrodes and solution.

reservoirs (b)–(d) on the microchip (Fig. 1N) were filled with running buffer solution, while reservoir (a) was loaded with sample solution. After an initial 20 s electrokinetic sampling in the injection channel by applying a potential of +0.5 kV between the sample reservoir (a) and the reservoir (c), a 2.0 kV high voltage was applied to the running buffer reservoir (b) with the buffer waste reservoir (d) grounded and all other reservoirs floating. Experiments were performed at the room temperature (24 ± 1 °C).

3. Results and discussion

3.1. Optimization of the operation parameters

3.1.1. The gap between the electrodes

Several chips with various electrode gaps from 1.0 mm to 3.0 mm were investigated, with 0.5 mmol L^{-1} KCl as model analyte. The running buffer consisted of 15 mmol L^{-1} MES and 15 mmol L^{-1} His, other parameters such as excitation frequency and excitation voltage, were in their optimized values (to be discussed). In principle, the cell output voltage decreases when widening the gap between the electrodes [21]. Note, that when the two sensing electrodes were assembled at a short distance (1.0 mm), the detection signal was very weak. As the gap increased, the peak height gradually enhanced; however, the maximum response appeared at the distance of 2.0 mm. This may be explained that a smaller detection gap results in analytical signal directly passing from the excitation electrode to the pickup electrode, consequently, the response from the separation channel is, partly neglected. Therefore, the distance of 2.0 mm was chosen for the following experiments (Fig. 3).

3.1.2. Frequency

As expected, the response of the CCD highly depended on the frequency applied. The influences of the excitation frequency on peak height and the signal-to-noise ratio (S/N) at different electrode gaps (1.5 mm, 2.0 mm, 2.5 mm, and 3.0 mm) are shown in Fig. 4A. The frequencies were recorded from 30 kHz to 200 kHz under a constant excitation voltage of 30 V_{pp}. The results indicated that no obvious signal was detected at frequency lower than 30 kHz. When the frequencies increased from 30 kHz to 200 kHz, the peak height and the S/N progressively increased to maximum, and then dramatically decreased. The S/N ratio at 2.0 mm of electrode distance increased from 30 kHz to 150 kHz, which showed similar tendency as the peak height, and the maximum peak height value was obtained at 170 kHz. The optimum frequency of the 2.0 mm gap ranged from 100 kHz to 150 kHz.

3.1.3. Excitation voltage

Previously published results [22,23] have showed that excitation voltages beyond 20 V_{pp} (up to several hundred volts) applied to the CCD in conventional capillary electrophoresis offer better sensitivity. This condition was in accordance with that described in Fig. 4B. An optimal voltage of 50 V_{pp} was used in the following experiments.

3.2. Analytical performance

Fig. 5 compared the performance of the screen-printed contactless conductivity detector electrodes (B) with the copper electrodes (A) commonly used in our laboratory [18]. The two CCDs have identical dimensions of the chip channels but the Cu electrodes are independent from the microchip system. The performances of the two detectors were evaluated by separating and detecting glycine and lysine. As can be seen in Fig. 5, the sensitivity of the ITO electrode was slightly higher compare to the conventional Cu electrode. Such improvement of sensitivity at the ITO electrode may be attributed to nature of materials used for electrode fabrication.

The present method was further validated by determination of three inorganic ions. As shown in Fig. 6A, the novel ITO electrode had sensitive response to all of the ions, and the satisfactory reproducibility was obtained (Table 1). Further work was extended to

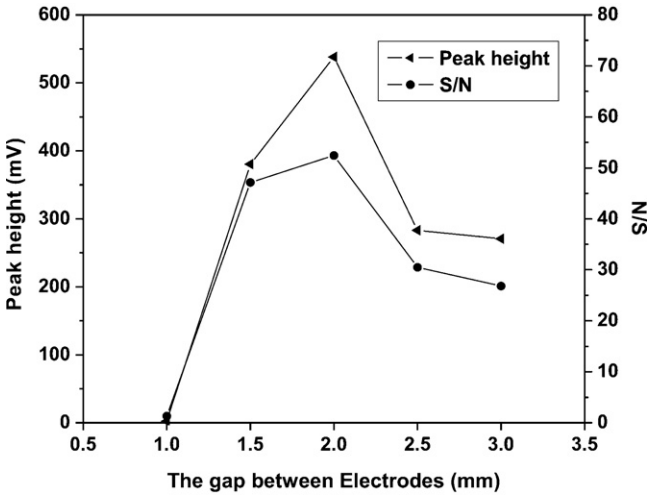

Fig. 3. Effect of the gap between sensing electrodes on peak height and S/N of 0.5 mmol L^{-1} of potassium ion.

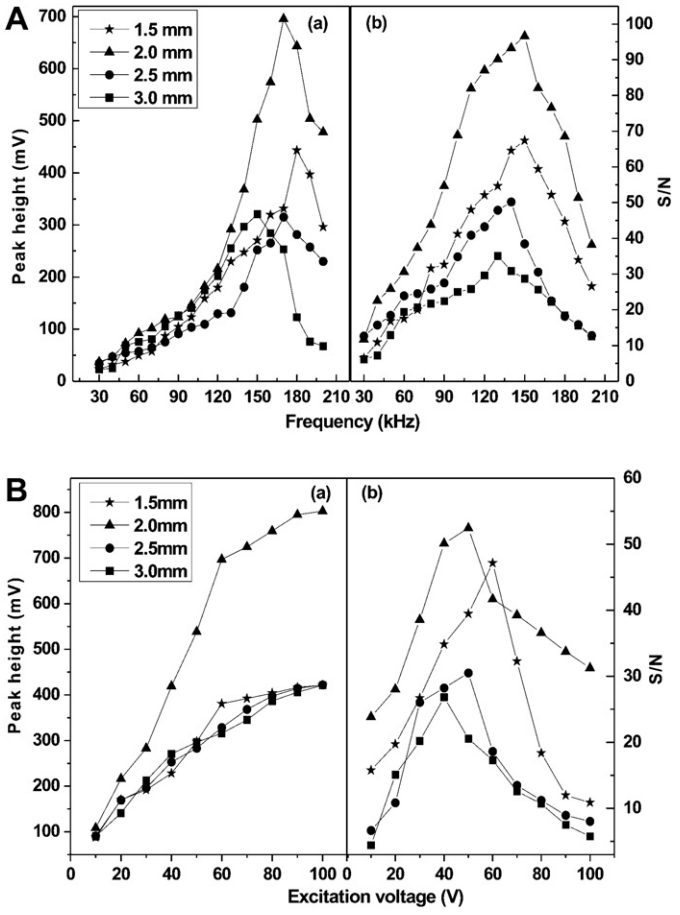

Fig. 4. (A) Effect of excitation frequency on peak height (a) and S/N (b) of 0.5 mmol L^{-1} of potassium ion at a constant excitation voltage of 30 V_{pp} for different distances between electrodes and (B) effect of the excitation voltage on peak height (a) and S/N (b) at optimal excitation frequency, other conditions as in Fig. 4A.

the analysis of pharmaceutical formulation. Aminopyrine [24] and caffeine [25] are normally used for cold remedy. Fig. 6B showed the separation of aminopyrine (peak 1) and caffeine (peak 2), followed by the contactless conductivity detection using ITO electrodes. The

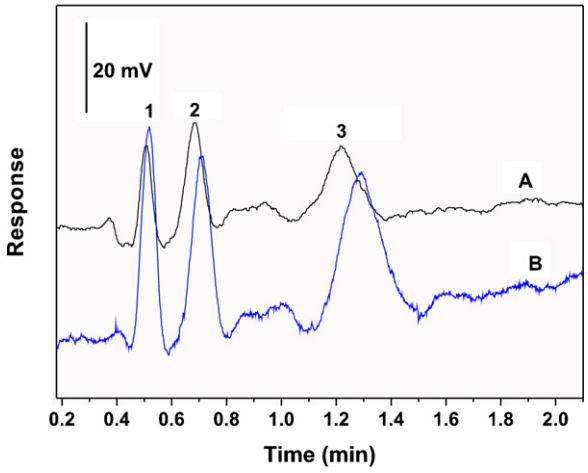

Fig. 5. Electrophorograms for a mixture containing 1 mmol L^{-1} glycine and lysine with Cu electrode (A) and screen-printed ITO electrode (B). Electrodes gap 2.0 mm, excitation voltage 50 V_{pp}, frequency 120 kHz. Electrophoretic conditions: background electrolyte, 15 mmol L^{-1} MES/His; separation voltage 2.0 kV, injection 0.5 kV for 10 s. Peak assignments: (1) glycine, (2) lysine and (3) H_2O.

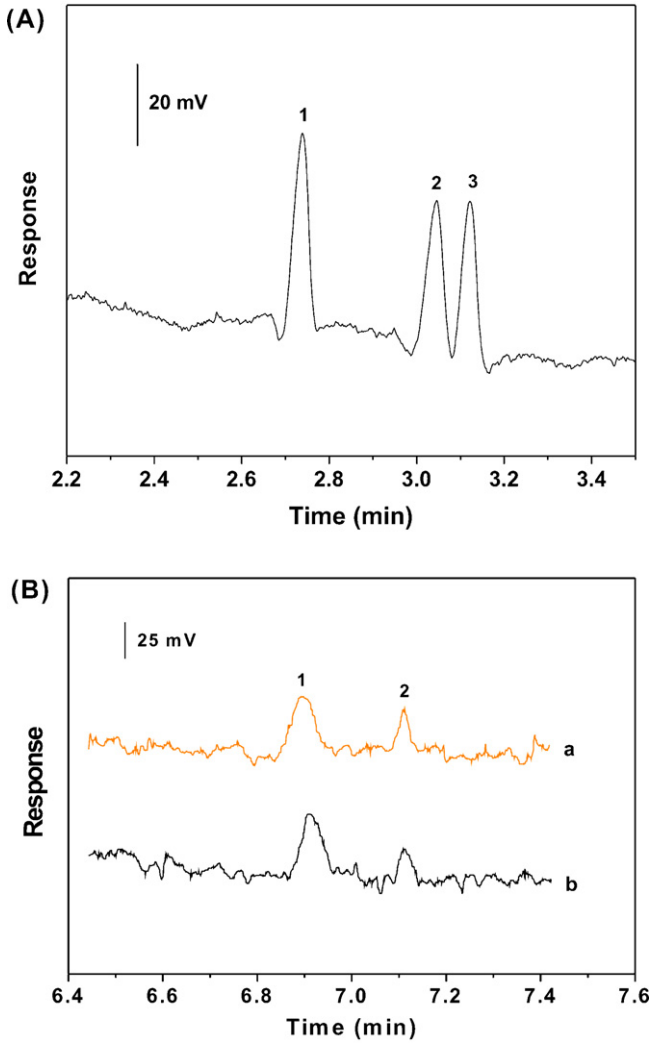

Fig. 6. (A) Applications of the optimized CCD cell design to the analysis of three inorganic ions. Three inorganic ions: K^+, Na^+, Mg^{2+} (0.1 mmol L^{-1} each). Conditions as in Fig. 5. Peak assignments: (1) K^+, (2) Na^+ and (3) Mg^{2+}. (B) Electrophorograms of microchip capillary electrophoresis using ITO electrode detection of (a) standard mixture solution of aminopyrine (peak 1, 15 μg mL^{-1}) and caffeine (peak 2, 4 μg mL^{-1}) and (b) sample solution of compound aminopyrine and caffeine tablets. Conditions: running buffer of 10 mmol L^{-1} Tris and 5 mmol L^{-1} H_3BO_3 (pH 8.8); other conditions as in Fig. 5.

background electrolyte consisted of 10 mmol L^{-1} Tris–5 mmol L^{-1} H_3BO_3 (pH 8.8) and the separation voltage was 2.0 kV. In this condition, aminopyrine and caffeine can be fully separated within 60 s with well-defined peaks.

Calibration curves were accomplished by employing a series of concentrations of aminopyrine and caffeine range from 10 μg mL^{-1} to 1000 μg mL^{-1}. The regression equations and the linear correlation coefficients were listed in Table 2. The LODs (S/N = 3) for aminopyrine and caffeine were 8 μg mL^{-1} and 3 μg mL^{-1},

Table 1
The reproducibility of the MCE–CCD device for analysis for potassium ion (0.1 mmol L^{-1}).

	t_m (min)	Peak height (mV)
Chip 1	0.735	50.928
Chip 2	0.698	52.016
Chip 3	0.713	51.628
Chip 4	0.724	52.927
Chip 5	0.708	51.762

Table 2
Statistical results for linearity, LOD, regression analysis, and sample determination.

Parameters	Aminopyrine	Caffeine
Calibration range ($\mu g\,mL^{-1}$)	10.0–800	10.0–800
Limit of detection (LOD) ($\mu g\,mL^{-1}$)	8.0	3.0
Regression equation[a]		
Slope (B)	0.0011	0.0009
Intercept (A)	0.226	0.344
Correlation coefficient, r	0.998	0.996
Sample		
Lable claim (mg)	150.0	40.0
Amount found (mg)	149.1 ± 2.3	39.2 ± 1.4

[a] The regression equation was $Y = A + BX$ (Y is the peak area; X is the concentration) ($\mu g\,mL^{-1}$).

Table 3
Recoveries of standard additions ($n = 6$).

Components	Found ($\mu g\,mL^{-1}$)	Added ($\mu g\,mL^{-1}$)	Total found ($\mu g\,mL^{-1}$)	Recovery (%)	RSD (%, $n = 6$)
Aminopyrine	149.6	120.0	269.3	99.8	1.5
		180.0	328.8	99.6	1.7
		300.0	448.5	99.6	1.0
Caffeine	39.5	32.0	71.1	98.8	1.9
		48.0	87.2	99.3	1.8
		80.0	118.4	98.6	2.0

respectively. The determination results of the sample showed that 99.3% and 97.5% of the labeled amount for aminopyrine and caffeine were found. The recovery test was made by adding three known concentrations of standard solutions of aminopyrine and caffeine into the sample solution, and the recoveries were in the range of 98.6–99.8% (Table 3).

4. Conclusions

The new assembled microchip integrated with ITO electrode for CCD has been demonstrated. The ITO-coated glass plays not only a cover of microchip but also the CCD electrodes. It can separate substances well as general microchips do. The well-established MCE–CCD system can be achieved in the optimal operation parameters, including 2.0 mm of electrode distance, 120 kHz of oscillation frequency and 50 V_{pp} of excitation voltage. The application for determination of aminopyrine and caffeine in pharmaceutical tablets was also performed, which manifested that this developed analytical system was feasible.

The constants of interval of two electrodes, as well as the distance between electrodes and the separation microchannel can guarantee the analytical reproducibility and industrializing producibility. In addition, the advantages of this system exist in the followings: (a) in contrast with the previously published literatures, ITO electrodes can be easily fabricated; (b) a thick cover layer (1.3 mm) can be used without obvious loss of signal response, perhaps owing to higher excitation frequency; (c) this novel microchip-CCD analytical platform is convenient to be incorporated into microchip without external electrode plate.

Acknowledgements

Financial supports from the National Natural Science Foundation of China (NSFC, Grant Nos. 20727006 and 21075139) and Guangdong Provincial Science and Technology Project (Grant No. 2008A030102009), technical support from engineer Hexian Situ are gratefully acknowledged.

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