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Speciation of inorganic and methylated arsenic compounds by capillary zone electrophoresis with indirect UV detection Application to the analysis of alkali extracts of As_2S_2 (realgar) and As_2S_3 (orpiment)

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Abstract

A capillary zone electrophoresis (CZE) method with indirect UV detection was developed to simultaneously separate inorganic and organic arsenic compounds including arsenite (iAs^{III}), arsenate (iAs^{V}), monomethylarsonate and dimethylarsenic acid (DMA^{V}). 2,6-Pyridinedicarboxylic acid (PDC) and *n*-hexadecyltrimethylammonium hydroxide (CTAOH) were selected to compose a background electrolyte (BGE), where PDC was used as chromophore and CTAOH functioned as electroosmotic flow (EOF) modifier to reduce/eliminate EOF. The choice of detection wavelength, the optimization of BGE pH, and effects of applied electric field strength and temperature on separation were further investigated. The limits of detection for the targeted analytes were between 0.19 and 0.23 ppm as molecule. Good linearity of more than three orders of magnitude was obtained. Repeatability of migration times and peaks areas were 0.8–1.7 and 3.4–6.9% R.S.D.; whereas reproducibility were 1.2–2.2 and 3.6–7.1% R.S.D., respectively. The established CZE method was then applied to analyze the alkali extracts of realgar (As_2S_2) and orpiment (As_2S_3). The main components in both alkali extracts were identified to be iAs^{III} and iAs^{V}.

Keywords: Realgar; Orpiment; Arsenic; Metal cations; Organometallic compounds; Sulfur compounds

1. Introduction

Arsenic compounds are ubiquitous in nature. The most commonly encountered arsenic forms are 3+ and 5+ valent states, including inorganic arsenite (iAs^{III}) and arsenate (iAs^{V}) as well as the methylated species consisting of the monomethylarsenic acid (MMA^{V}) and dimethylarsenic acid (DMA^{V}). Exposure to arsenic by the general population occurs mainly through ingestion of arsenic present in drinking water and food. In freshwater systems, iAs^{III} and iAs^{V} are major arsenic species, while minor amounts of MMA^{V} and DMA^{V} also exist [1]. There is little information on the nature of arsenic species in human diet, apart from seafood. Most dietary arsenic originates from fish, shellfish, and seaweed products, where the major arsenic species found is the nontoxic arsenobetaine (AB) [2]. Most of the inorganic arsenic species are metabolized in humans and many mammalians to methylated arsenic species including MMA^{V} and DMA^{V}, which are more readily excreted into the urine than the inorganic arsenic species [3]. Examples of the reported concentrations of arsenic in human urine from the general population are (mean ± standard deviation, μg/l) 9 ± 7 in America [4], 17 ± 11 in Europe [5], and 121 ± 101 in Japan [6]. Association between acute and long-term exposures of humans to the inorganic arsenic compounds and various forms of cancer and other health problems has been well documented [7]. The effectual actions of arsenic species as chemotherapeutic anticancer agents have also been reported from epidemiological or clinical studies [8,9]. For example, in the early 1970s a group of clinical researchers in China began to treat some types of cancer with arsenic trioxide (As_2O_3), and encouraging results have been observed in the treatment of esophageal carcinoma, malignant lymphoma, and leukemia, particularly chronic myelocytic

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leukemia (CML) and acute promyelocytic leukemia (APL) [8]. In a study, As_2O_3 was intravenously administered in the form of 1% solution at a dose of 0.16 mg/kg daily diluted with 5% glucose in normal saline for 2–3 h. Patients achieved complete remission (CR) after received As_2O_3 treatment for 28–44 days [9]. Conclusively, it is well established that arsenic compounds present a paradox because they could act as both potent carcinogens and beneficial chemotherapeutic agents depending on their valent states and chemical forms. Therefore, arsenic speciation is of extensive research focus. In many studies, high-performance liquid chromatography (HPLC) was used for separating arsenic compounds, and a variety of detection methods including hydride generation atomic absorption/fluorescence spectrometry (HG-AAS/AFS) [10,11], inductively coupled plasma atomic emission spectrometry (ICP-AES) [12], and the more sensitive ICP mass spectrometry (ICP-MS) [13,14] were used for their detection. However, the separation efficiency of HPLC is relatively low particularly for the analysis of inorganic arsenic compounds with low molecular weights. In contrast, capillary electrophoresis (CE) was found to be an efficient alternative to simultaneously separate and determine arsenic species owing to its very high efficiency, ease of operation, low cost, and universal availability compared to the HPLC method [15,16].

Realgar (As_2S_2) and orpiment (As_2S_3) are minerals abundantly distributed in the earth's crust. The medical use of realgar and orpiment has been traced back thousands of years. For example, in ancient China, Realgar (*Xiong-Huang*) was utilized in the treatment of carbuncles, boils, insect- and snake-bites, abdominal pain due to intestinal parasitosis, infantile convulsion, malaria, and psoriasis and skin diseases caused by plasmodium and schistosoma japonicum [17], while orpiment (*Ci-Huang*) was administered as an anthelmintic and as a remedy for skin problems [17]. Historically, Greeks, Arabs and Romans have also long realized the therapeutic benefits of arsenic compounds [17]. Recently, the anticancer effects of realgar and orpiment particularly in leukemia have become the focus of research [18–20]. Although their clinical effectiveness especially on CML and APL are evident, it is relatively unclear whether the effectiveness is due to the sulfides or the oxides formed from the parent compounds. The major difficulty limiting research in these areas is the lack of specific and sensitive analytical methods. Realgar and orpiment are both water insoluble. It is believed that when these arsenic powders are administered orally, these compounds are partially dissolved in the alkaline intestinal media (pH 7–8) before absorption. The alkali extracts of realgar and orpiment have been analyzed by using ion chromatography (IC) with HG-AAS detection [21]. The findings indicated that main components in the alkali extract of realgar were iAs^{III} and more iAs^{V}, and those in the alkali extract of orpiment were iAs^{III}, iAs^{V} and DMA^{V} with varying proportions [21]. However, the realgar and orpiment used in that study probably contained some impurity, such as different types of iAs^{III}, arsenic trioxide and other trace elements. Thus, to avoid interference of impurities to analytical results, in this study, samples of As_2S_2 and As_2S_3 with high purity of above 98% were used. We also established a highly efficient capillary zone electrophoresis (CZE) method to separate the inorganic and methylated arsenic compounds. Thereafter, this CZE method was used to analyze the alkali extracts of As_2S_2 and As_2S_3.

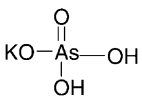
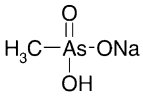
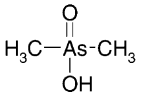
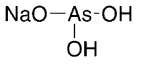
2. Experimental

2.1. Materials

2.1.1. Arsenic compounds of interest

Potassium arsenate (KH_2AsO_4, iAs^{V}), dimethylarsenic acid [$(CH_3)_2AsO(OH)$, DMA^{V}], arsenic trioxide (As_2O_3) were purchased from Sigma (St. Louis, MO, USA). Sodium monomethyl arsonate ($CH_4AsNaO_3{\cdot}(3/2)H_2O$, MMA^{V}) was purchased from Chem Service (West Chester, PA, USA). The aqueous stock solutions of iAs^{V}, MMA^{V} and DMA^{V} each with the concentration of 1000 ± 5 ppm as molecule were respectively prepared by dissolution in Milli-Q water. Sodium arsenite (iAs^{III}) stock solution (1000 ± 5 ppm as iAs^{III}) was made by first dissolving 5.82 mg As_2O_3 in 10 ml 0.1 M NaOH and then being neutralized with concentrated HCl to pH 7.0 (± 0.2). The accurate As concentrations of all stock solutions were further measured by inductively coupled plasma optical emission spectrometry (ICP-OES, Thermo Jarrell Ash, IRIS/AP, Germany). Before storage, all stock solutions were filtered through a 0.2 μm nylon filter membrane and degassed in ultrasonic bath for 15 min. The molecular structures and pK_a values of the arsenic analytes are shown in Table 1.

Table 1
Arsenic compounds of interest

Chemicals with symbolic name	Structure	pK_a
iAs^{V}		2.3 6.9 11.4
MMA		3.6 8.2
DMA		9.3
iAs^{III}		9.2 13.5

2.1.2. Preparation of the alkali extracts of realgar and orpiment

Realgar (As_2S_2, 98% in purity) and orpiment (As_2S_3, 99.9% in purity) were purchased from Sigma. The alkali extracts were prepared by ultrasonically extracting the respective samples in 0.1 M NaOH for 30 min at an amount of 2 mg sample per 1 ml solution, and then being filtered through a 0.2 μm nylon filter membrane. The As concentrations in the obtained alkali extracts were measured by ICP-OES, being 998 and 1338 ppm as As, respectively. All stock solutions were stored at 4 °C in dark.

2.2. CZE separation

2.2.1. Instruments

All CZE separations were performed on a CE-L1 capillary electrophoresis system (CE Resources Singapore, Singapore). This system was equipped with a reversible-polarity power supply (0 to (±30) kV) and an on-column VUV-H22 UV detector. System control as well as data acquisition and analysis were processed by a CSW software (CE Resources). The separations were carried out on an uncoated fused-silica capillary (70 cm × 50 μm i.d. × 360 μm o.d.) with a detection window located 10 cm from its extremity. Negative potential applied at the injection port was referred to as negative polarity. A built-in temperature control system was designed to maintain separation temperature and minimize Joule heat generated, thus enabling the CE system to use high electric field and to achieve very low band dispersion. The system had both pneumatic and electrokinetic injection modes. In order to avoid sampling bias and achieve better reproducibility, the pressure-based injection mode was chosen. Sample plug was consistently injected at a low pressure of 0.36 psi for 10 s, which was equal to about 2.9 mm in length (L_{inj}) according to the Poiseulle–Hagen equation [22]:

$$L_{inj} = \frac{(d_{cap})^2 pt}{32\eta L_{cap}}$$

where d_{cap} is the capillary inner diameter in μm, p the pressure in psi, t the time of injection in s, η (0.000891 N s/m^2) the viscosity of water at 298 K, and L_{cap} is the length of capillary in cm.

2.2.2. Standard separation

Pyridine 2,6-dicarboxylic acid (PDC) (≥98% in purity) was purchased from Fluka (Buchs, Switzerland). *n*-Hexadecyltrimethylammonium hydroxide (CTAOH) (25% in methanol) was purchased from Tokyo Kasei Kogyo (Tokyo, Japan). PDC/CTAOH background electrolyte (BGE) containing 5 mM PDC and 0.5 mM CTAOH was freshly prepared at the beginning of each study day. Before use, it was vortexed for 2 min, filtered through a 0.45 μm filter membrane and degassed by ultrasonic for 15 min. Between runs, the capillary was subjected to preparation cycles including pre-rinse and precondition steps to ensure a clean and equilibrated surface for the following separations. At the end of the day, the capillary was rinsed for 3 min with 1 M NaOH and Milli-Q water for 5 min, respectively, and finally dried with air for 3 min.

3. Results and discussion

3.1. Separation of inorganic and organic arsenic species

The choice of a BGE is very important in method development employing CZE with indirect UV detection. In general, in CZE, ion migration velocity, separation, column efficiency, and peak shape are sensitive to changes in BGE characteristics. Typically, in indirect UV detection mode for anions, crucial BGE composition consists of electrolyte co-ion providing the UV chromophore and suitable additives such as EOF modifiers.

The most widely used electrolyte co-ion for inorganic anions is chromate, which provides high mobility anions with a UV chromophore [23]. It has been reported that band broadening and loss of resolution in the separation of some arsenic compounds were observed in a CZE method using a BGE containing chromate as a co-ion chromophore, borate buffer and *n*-hexadecyltrimethylammonium bromide (CTAB) as an EOF modifier (chromate–borate–CTAB) [24]. In our preliminary study, although separation efficiency was improved and a baseline separation was obtained as using chromate–borate–CTAB BGE to separate arsenic compounds in a CZE method as shown in Fig. 1, positive and negative signal peaks simultaneously appeared. The presence of the positive signal peaks disagreed with the separation principle of CZE with indirect detection and could not be clearly elucidated. In addition, the relative high limit of detection (LOD), e.g. around 10 ppm as molecule for iAs^V, limited the usage of such BGE in arsenic speciation analysis. Recently, the UV sensitive PDC with a medium mobility capacity was proven well suitable for the simultaneous analysis of both high and low mobility anions [25,26]. Moreover, PDC has strong UV absorbance in a broad wavelength range at 200–400 nm.

The electroosmotic flow (EOF), which directs to cathode end, is especially adverse for anionic analyte detection at anode. Reducing/eliminating the EOF is required for detecting anions successfully. Among approaches currently applied to control the EOF, cationic surfactant CTAOH was reportedly effective to change capillary inner surface charge by adsorption and consequently the EOF direction [25,26]. Therefore, PDC/CTAOH was selected as a BGE in this study, and no additional buffer component was used.

To select an optimum detection wavelength, a wavelength range of 195–225 nm was firstly tested. Fig. 2 displays the electrophoregrams of iAs^{III} obtained by using 5 mM PDC/0.5 mM CTAOH BGE (pH 10.5) at various detection wavelengths. With an increase in the detection wavelength from 195 to 210 nm, the sensitivity of detection represented

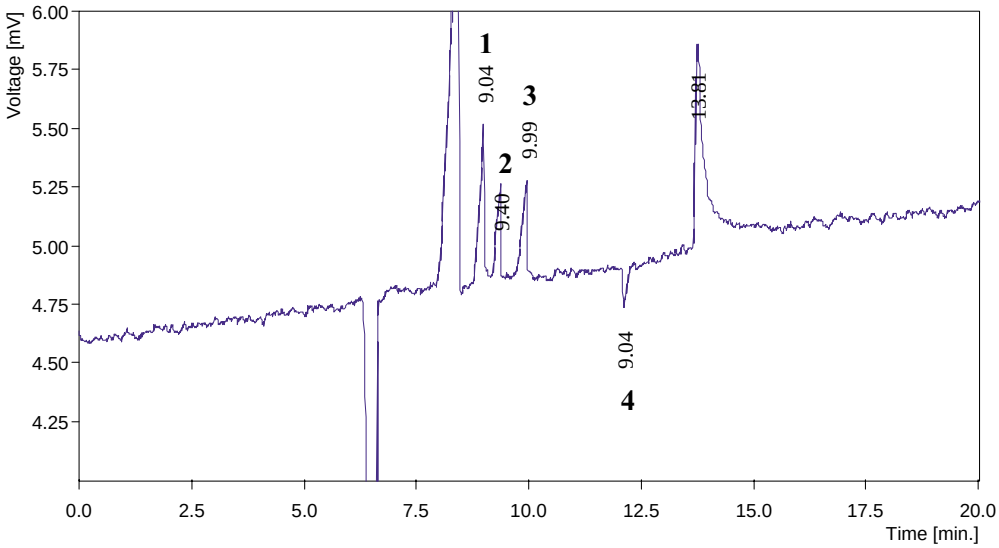


Fig. 1. The electrophoretic separation of arsenic compounds each with concentration of 100 ppm as molecule. BGE composing of 10 mM chromate, 12.5 mM borate and 0.5 mM CTAB with pH 9.4; $U_{setting} = -25$ kV and $I_{setting} = 15\,\mu$A; detection wavelength at 216 nm; at temperature of 20 °C. Peaks: (1) iAs^{V2-}; (2) iAs^{III2-}; (3) MMA^{V2-}; and (4) DMA^{V-}.

by the signal peak area increased, while the baseline noise decreased. Further increasing the detection wavelength to 225 nm, however, led to reduced detection sensitivity, with only a slight improvement in baseline. After considering the detection sensitivity and baseline noise, 215 nm was thus selected as the detection wavelength for all experiments unless specified otherwise.

In the CZE, ionic species are separated based on their charges and sizes, therefore, BGE pH has a crucial impact on the separation. According to the pK_a values of the analytes (Table 1), at pH 9.3 and above, theoretically, iAs^V, MMA^V and iAs^{III} have two negative charges, while DMA^V is negatively single-charged. Therefore, assuming the EOF has been successfully suppressed, iAs^V, MMA^V and iAs^{III} would first migrate towards the detector at the anode, while DMA migrates more slowly according to the electrophoretic separation principle. In other CZE studies [25,26], a high pH up to 12.1 was used in the BGE containing PDC, implying that PDC was chemically stable in a strong basic environment. However, an extremely high BGE pH is not encouraged because during an electrophoretic process, generally, an increase of the electrolyte pH will result in increasing the dissociation of silanol group of the capillary inner surface to silonate group, thus enhancing the capillary inner surface charges and the EOF consequently. Furthermore, iAs^{III} tends to be oxidized to iAs^V above pH 10 [27]. Therefore, a BGE pH range of 10.0 to 11.5 with a 0.5-step was selected and carefully adjusted.

Although the chosen PDC/CTAOH BGE was not further adjusted with other buffer components, stable baseline and reproducible migrations were obtained within the pH range, indicating their buffer capacity was high enough so that local pH and conductivity did not change as a result of sample injection and following separation. Fig. 3 shows the effects of different BGE pH values from 10.0 to 11.5 with a 0.5 (±0.02)-step on the electrophoretic separations of arsenic species. Although in the above pH range, theoretically, each arsenic analyte was at the same ionization degree: iAs^{III2-}, iAs^{V2-}, MMA^{2-} and DMA^-, migration profiles were different. At low BGE pH of 10.0, the first two anions, iAs^{III2-} and iAs^{V2-}, partially overlapped. Increasing the BGE pH to 10.5, baseline separation was achieved, and the migration order was iAs^{III2-}, iAs^{V2-}, MMA^{2-} and DMA^-, and was confirmed by comparing with the electrophoregrams of the individual analytes. At higher BGE pH at 11.0, the whole migration suddenly became much slower in comparison with that at pH 10.5; however, with a further increase to pH 11.5, the migration pattern remained similar to that at pH 11.0. It was well established that the EOF would increase with increase in the BGE pH. However, this situation can be changed with the addition of the EOF modifier CTAOH: the more negative capillary inner silica surface induced by higher BGE pH leads to attracting a greater amount of CTAOH, which will bind to it till saturation. The resultant net charges of the capillary inner surface thus determine the actual EOF, which, in turn, affects the anionic analyte mobility. This could be the cause for the observed changes in the extent of the analyte migration time with the increasing BGE pH. The effects of pH on BGE resistance are shown in Table 2 and summarized as follow: resistance decreased with an increase in the amounts of hydroxide anions added, which was probably due to the high mobility of the small hydroxide anions. As a result, electric field strength that was represented by dividing the applied voltage by the total capillary length (V/cm) decreased with the decrease of the BGE resistance. Obviously, there was a close relationship between the actual migration times of the analytes and the applied electric field strength that could be affected by

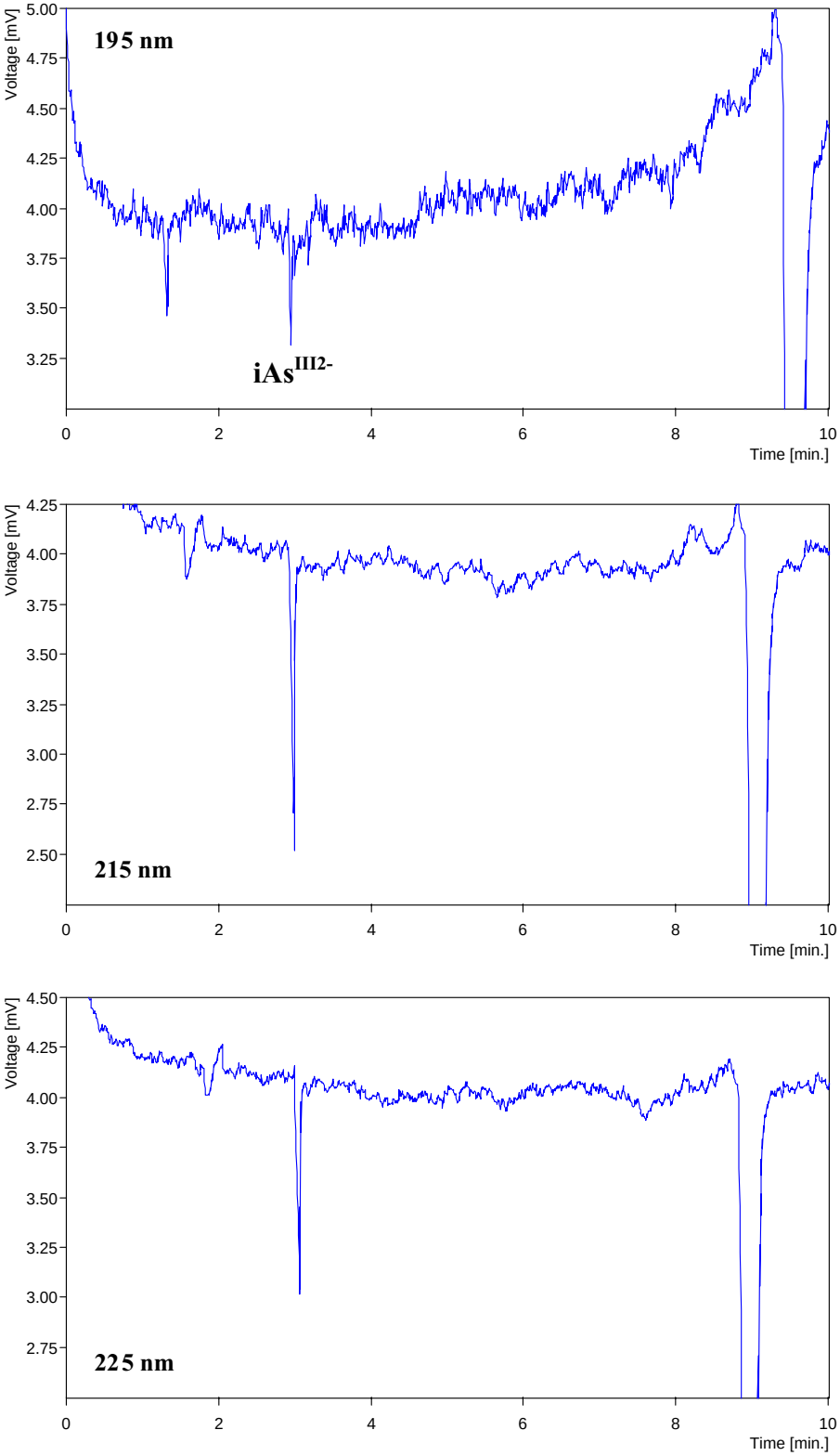
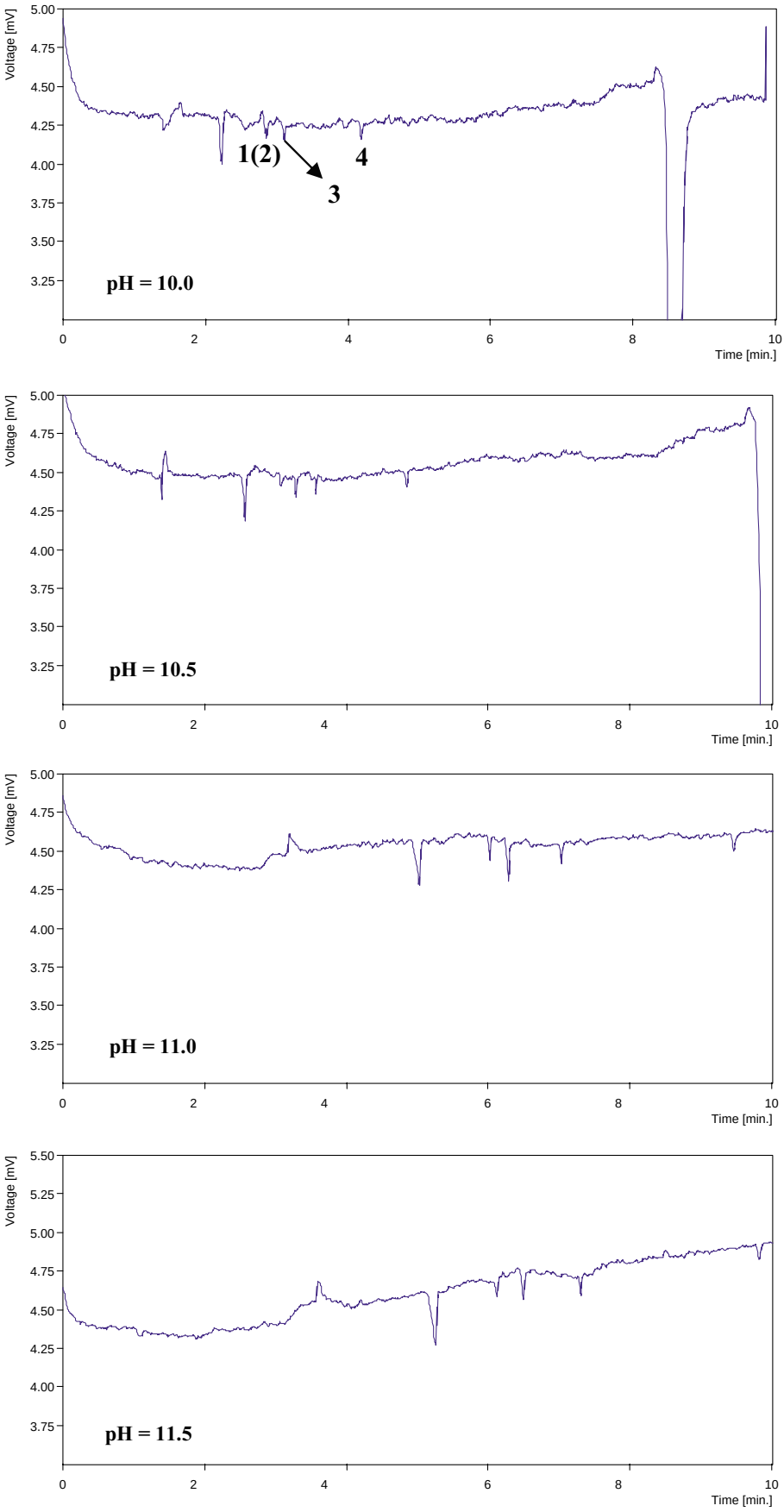


Fig. 2. The electrophoregrams of iAs^{III} with concentration of 10 ppm as molecule obtained at different detection wavelengths. BGE with pH 10.5 containing 5 mM PDC and 0.5 mM CTAOH; $U_{setting} = -30$ kV and $I_{setting} = 8$ μA; at temperature of 15 °C.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4.00
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0
2
4
6
8
10
Time [min.]
Voltage [mV]
pH = 10.5
Time [min.]
Voltage [mV]
pH = 11.0
Time [min.]
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3.75
pH = 11.5
Time [min.]

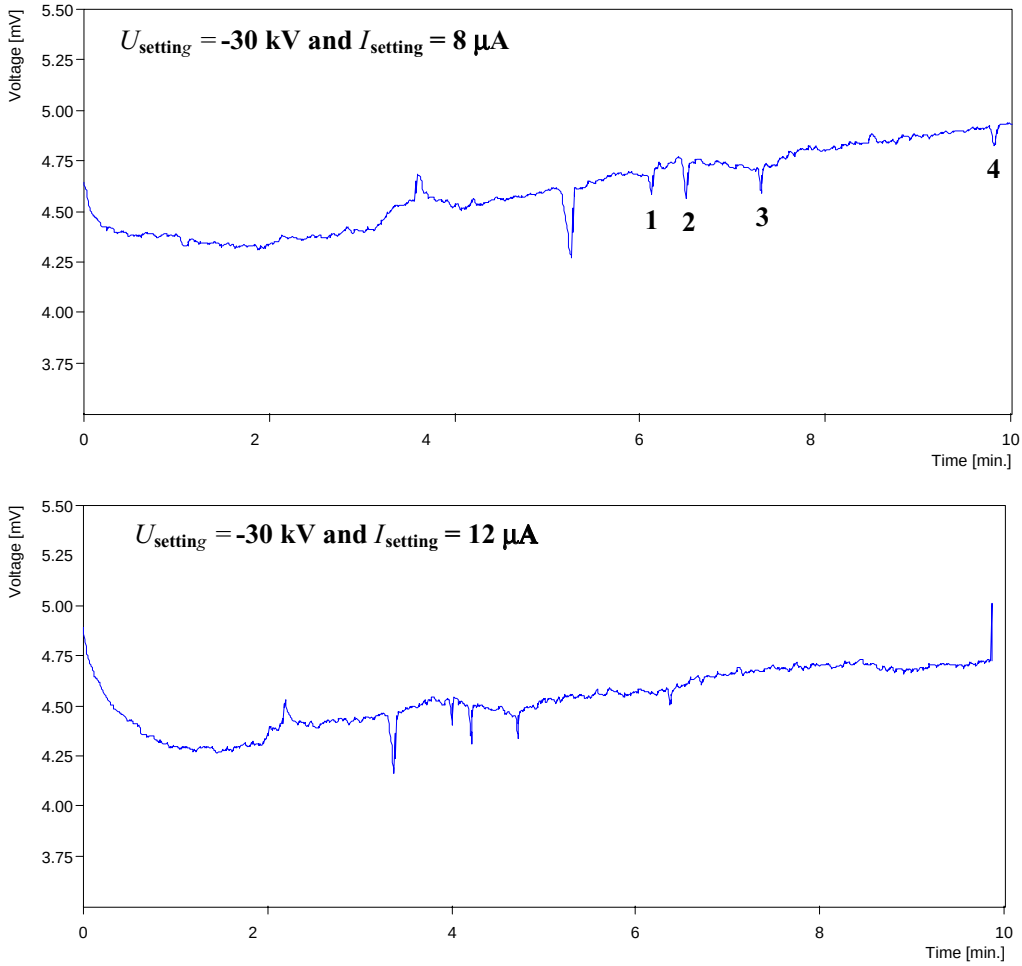

Fig. 4. The electrophoretic separation of arsenic compounds each with concentration of 1 ppm as molecule under different applied voltage and current: 5 mM PDC/0.5 mM CTAOH BGE at pH 11.5; at temperature of 15 °C. Peaks: (1), iAs^{III2-}; (2) iAs^{V2-}; (3) MMA^{V2-}; (4) DMA^{V-}.

Table 2
The influences of BGE pH on BGE resistance and electric field strength

BGE pH	BGE resistance (10^9 Ω)	Electric field strength (V cm^{-1})
10.0	3.65	397.14
10.5	3.21	348.57
11.0	1.70	180.00
11.5	1.68	178.32

the BGE pH. It was reported that decreasing electric field strength would reduce the Joule heat and the convection currents in the electrophoretic medium, thus the peak would appear sharper and higher in the electrophoregram. Also from Fig. 3, it was found that the detection sensitivity seemingly was improved at higher BGE pH such as at 11.0 and above. In the subsequent experiments, a BGE of pH 11.5 was thus selected.

The driving force behind the migration of ions in CZE is the electric field strength applied across the capillary. Effects of the applied electric field strength on the separation were further demonstrated by directly changing the current settings while fixing the voltage settings as shown in Fig. 4. The findings indicated that in the experimental range, increasing the applied electric field strength did not increase the detection sensitivity but did reduce the migration times, while it slightly increased the baseline noise probably due to Joule heat generated.

Temperature plays an important role in many separations, because both analyte mobility and the level of EOF are temperature-related. In general, electrophoretic mobility increases with increasing the temperature by about 2% per 1 K [28]. A temperature range of 20–35 °C was chosen and optimized. Fig. 5 gives the electrophoretic separations of four arsenic species in the temperature range. In the temperature

Fig. 3. The effects of BGE pH on the electrophoretic separation of arsenic compounds each with concentration of 1 ppm as molecule. BGE composing of 5 mM PDC and 0.5 mM CTAOH; $U_{setting} = -30$ kV and $I_{setting} = 8$ μA; at temperature of 15 °C. Peaks: (1) iAs^{III2-}; (2) iAs^{V2-}; (3) MMA^{V2-}; (4) DMA^{V-}.

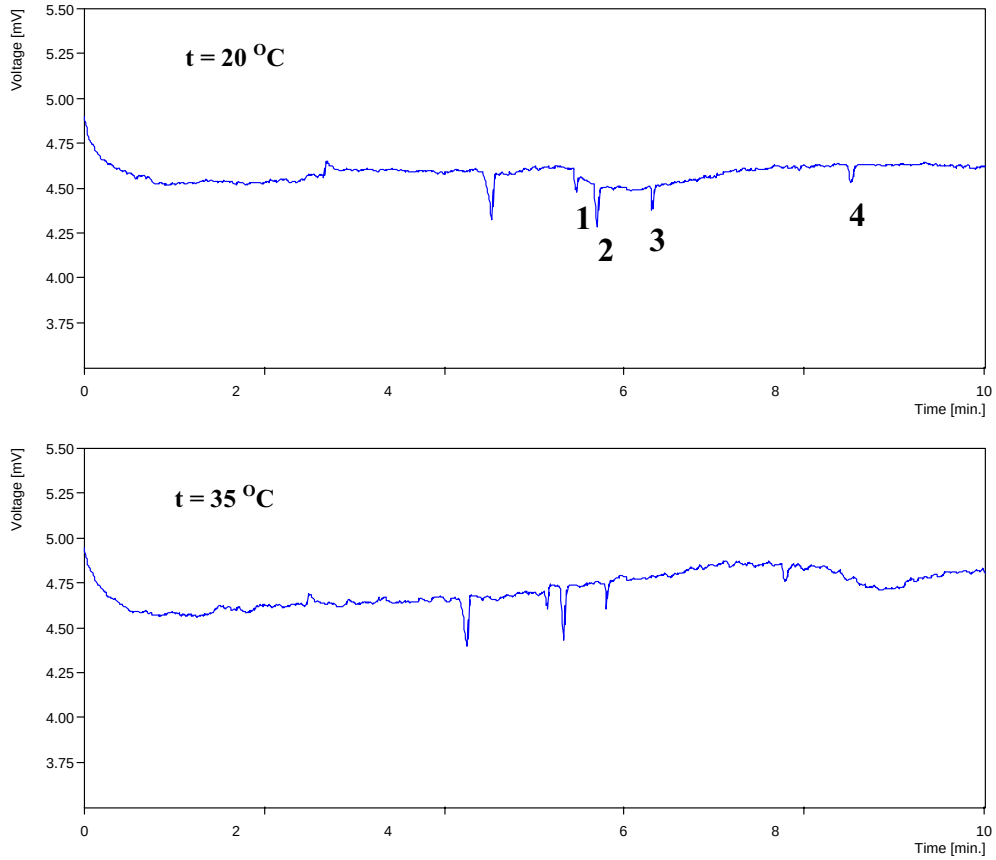

Fig. 5. The electrophoretic separation of arsenic compounds each with concentration of 1 ppm as molecule under different operation temperature: 5 mM PDC/0.5 mM CTAOH BGE at pH 11.5; $U_{setting} = -30$ kV and $I_{setting} = 8\,\mu$A. Peaks: (1) iAs^{III2-}; (2) iAs^{V2-}; (3) MMA^{V2-}; (4) DMA^{V-}.

range selected, separation was almost not affected by the change of temperature, except the slightly faster migration and noisier baseline was observed with an increase in temperature.

3.2. Calibration parameters

With 5 mM PDC/0.5 mM CTAOH BGE at pH 11.5, calibration curves were obtained for the respective arsenic species within the concentration range of 0.5–500 ppm. The sensitivity, linearity, repeatability and reproducibility of the method were determined as shown in Table 3. The mean correlation coefficient (r^2) of each calibration curve with dynamic range of more than three orders of magnitude of concentration exceeded 0.99, indicating good linearity. Repeatability expressed as relative standard deviations (R.S.D.%) with respect to migration times and peak areas from six successive injections at the analyte concentration of 10 ppm, was found to be 0.8–1.7% for migration times and 3.4–6.9% for peak areas, respectively. The reproducibility for the same parameters was obtained by analyzing analyte species (10 ppm) on three successive days. R.S.D. of 1.2–2.2% for migration times and around 3.6–7.1% for peak areas were obtained for all analyte species.

3.3. Identification of arsenic species in the alkali extracts of realgar and orpiment

It was reported that the main components in the alkali extracts of realgar were iAs^{III} and iAs^{V}, and in the alkali extract of orpiment were iAs^{III}, iAs^{V} and DMA^{V} as identified by IC-HG-AAS method [21]. By using the established CZE method the alkali extracts of realgar and orpiment were analyzed. By comparing with the migration times with the respective standards, the corresponding electrophoregrams indicated that there were two main components, iAs^{III} and iAs^{V} in the extracts. The alkali extracts were then spiked with standards. Fig. 6(a) and (b) showed that no additional signal peak appeared after the samples were spiked separately with iAs^{III} and iAs^{V}, while the peak intensities of iAs^{III} and iAs^{V}, respectively, increased, supporting the above assumptions. These results were partially consistent with the other study [21]. Although an amount of DMA^{V} was found in the alkali extract of orpiment, it was reasonable to suspect that the DMA^{V} might be present as impurity in their samples. With the established calibration curves, the concentrations of iAs^{III} and iAs^{V} in both extracts with 1.5 ppm as As were determined. In the alkali extract of realgar, there were 1.90 ppm (84.61%) of iAs^{III} and 0.39 ppm (15.39%) of iAs^{V}; whereas in the alkali extract

Table 3
Parameters of the calibration curves[a]

Analyte	Detection limit[b] (ppm)	Linear dynamic range (order of magnitude)	r^2	Repeatability, R.S.D. (%)		Reproducibility, R.S.D. (%)	
				Migration time	Peak area	Migration time	Peak area
iAs^{III}	0.23	>3	0.9984	1.1	3.9	1.7	4.3
iAs^{V}	0.19	>3	0.9993	0.8	3.4	1.2	3.6
MMA^{V}	0.19	>3	0.9991	0.9	4.1	1.5	4.8
DMA^{V}	0.22	>3	0.9987	1.7	6.9	2.2	7.1

[a] Conditions: injection 10 s at low pressure; BGE, 5 mM PDC/0.5 mM CTAOH with pH 11.5; detection wavelength 215 nm; voltage setting of −30 kV and current setting of 8 μA.
[b] Relative detection limits were calculated as concentration that will give signals equivalent to three times the peak-to-peak noise of the baseline.

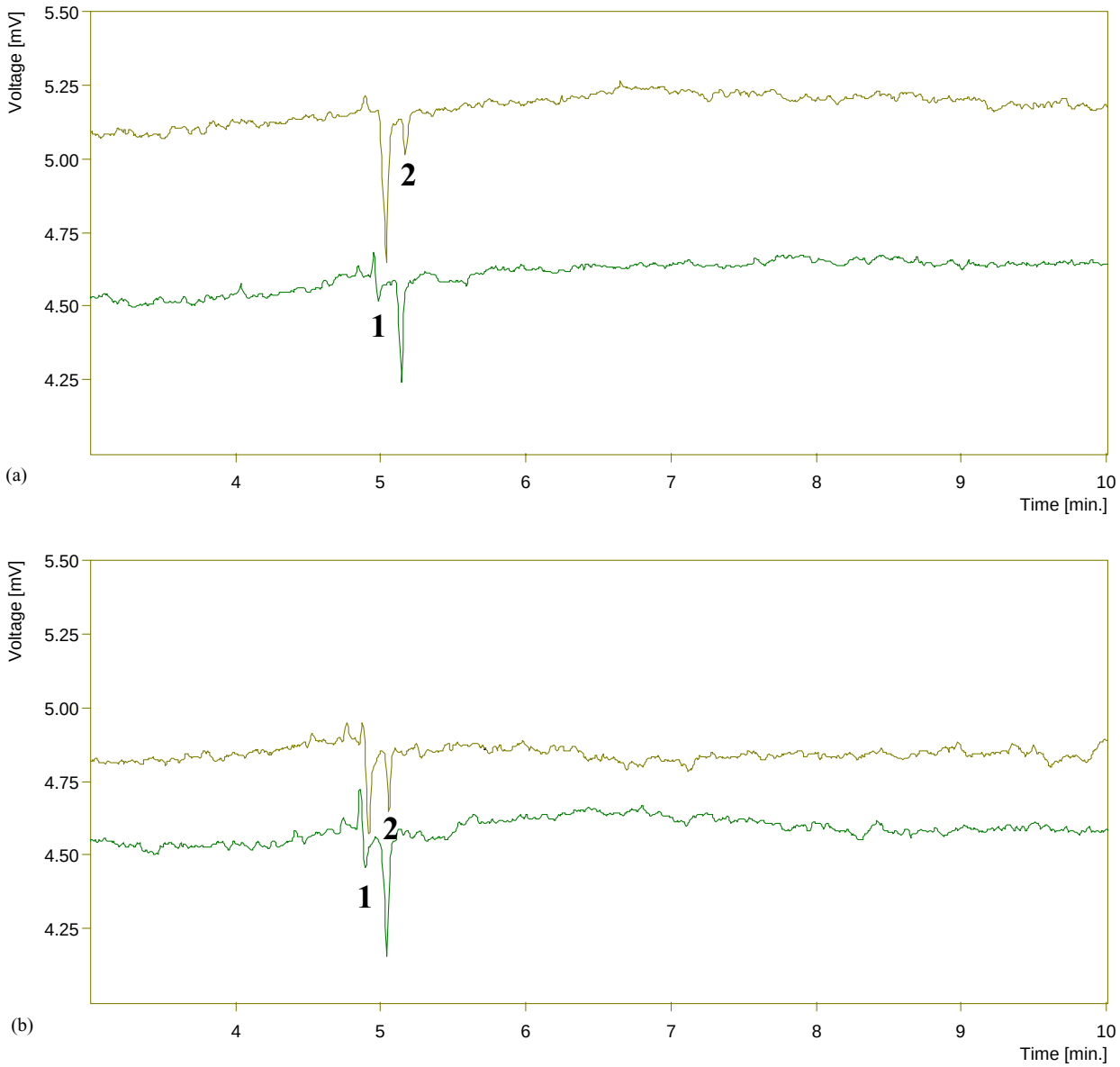

Fig. 6. The electrophoregrams of the alkali extracts of realgar (1.5 ppm as As) (a) and orpiment (1.5 ppm as As) (b) respectively spiked with 1 ppm iAs^{III} (upper line) and 1 ppm iAs^{V} (lower line): 5 mM PDC/0.5 mM CTAOH BGE at pH 11.5; $U_{setting} = -30$ kV and $I_{setting} = 8$ μA; at temperature 20 °C. Peaks: (1) iAs^{III2-}; (2) iAs^{V2-}.

of orpiment, there were 1.61 ppm (75.21%) of iAs^{III} and 0.60 ppm (24.79%) of iAs^{V}, respectively. This study is the first report on using CZE method to analyze the alkali extracts of realgar and orpiment, adding valuable information on the components constituting these compounds. Our finding indicated that the alkali extracting procedure could produce similar redox reactions between the respective As_2S_2 and As_2S_3 molecules and hydroxide to give arsenite (iAs^{III}) and arsenate (iAs^{V}).

4. Conclusion

The proposed CZE method with indirect UV detection showed the excellent suitability for the simultaneous separation and determination of the inorganic and organic arsenic compounds by using the PDC/CTAOH BGE. It provided the excellent linearity, repeatability, reproducibility, and sensitivity to identify the components in the alkali extracts of realgar and orpiment. The main components in both extracts were found to be iAs^{III} and iAs^{V}. The established CZE method could be applied to standardize pharmaceutical arsenic formulations. The LOD of this method could also be improved by coupling the CZE with ICP-MS for the detection of these arsenic compounds in biological samples.

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