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Simultaneous determination of nitrate and nitrite ions in seawater by capillary zone electrophoresis using artificial seawater as the carrier solution

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Abstract

We are proposing a novel capillary zone electrophoresis for the simultaneous determination of nitrate and nitrite ions in seawater. An artificial seawater was adopted as the carrier solution to eliminate the interference of high concentrations of chloride ion etc., in seawater. For the purpose of reversing the electroosmotic flow, 3 mM cetyltrimethylammonium chloride was added to the carrier solution. A 100 μm I.D. capillary was used to extend the optical path length. Regardless of the salinity of sample solutions, the peak areas and migration times for nitrate and nitrite ions were within $\pm 3\%$, while the peak heights for these ions decreased linearly with increasing salinity of sample solutions. The limits of detection for nitrate and nitrite ions were 0.04 and 0.07 mg/l ($S/N=3$, based on three-times the baseline noise), respectively. The values of the relative standard deviation (RSD) of peak areas for these ions were 1.5 and 2.4%. The RSDs of migration time for these ions were 0.85 and 0.78%. The proposed method was applied to the determination of nitrate and nitrite ions in seawater samples taken from the surface and the seabed. The recovery of nitrate and nitrite ions was 97–114%. © 1999 Elsevier Science B.V. All rights reserved.

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1. Introduction

The annual incidence of Red Tide observed in a sea area around Japan by the Maritime Safety Agency has been in the range of 40 to 90 during 1987–1997 [1]. It has no tendency to decrease. This suggests that eutrophication is currently in progress in the area. It is, therefore, important to determine

nutrients such as nitrate, nitrite, ammonium and phosphate ions in seawater.

Nitrate and nitrite ions in seawater have been determined by extractive spectrophotometric methods [2] and flow-injection analysis (FIA) using the conventional spectrophotometric detection of azo dye [3,4] or the nitrite–proflavin compound [5] for nitrite ions. In these procedures, the nitrate ion was reduced to nitrite ion in a copperized-cadmium column and the absorbance for the nitrate and nitrite ions was measured; the concentration of nitrate ions was determined from the difference between the total

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concentration of nitrate and nitrite ions and that of nitrite ions. The following methods have been also used: ion interaction reversed-phase liquid chromatography (RPLC) [6], ion chromatography (IC) with a high-capacity anion-exchange column and a UV detector [7], and chemiluminescence using the reaction of nitric oxide with ozone [8]. Namiki et al. [9] used the absorbance based on nitrate ion after eliminating the interference from bromide ion with a solid-phase adsorption column for the determination of total nitrogen in seawater. Namiki et al. [10] also proposed the method for the total nitrogen in seawater: nitrate and nitrite ions were reduced to ammonia using a copperized-zinc column and the ammonia was measured by indophenol colorimetry. Nitrate ions in seawater were determined by IC with a pre-column and two kinds of eluent [11] and nitrite ions were determined by the spectrophotometric method using diazotization-coupling with column preconcentration [12].

Recently capillary zone electrophoresis (CZE) has been applied to the analysis of environmental samples. However, there are a few reports concerning the determination of nitrate and nitrite ions in seawater by CZE. In general, seawater contains high concentrations of chloride and sulfate ions etc., compared to nitrate and nitrite ions. High concentrations of matrix ions in sample solutions such as seawater interfere with the CZE determination of low concentrations of analyte ions [13,14]. Matrix matching of electrolyte with sample has been adopted to eliminate the interference [15,16]. Sakai et al. [17] used 750 mM chloride as the carrier solution to eliminate the interference caused by chloride ions for the determination of nitrate and nitrite ions in seawater; the peak areas for these ions (with constant concentrations) varied with the concentration of chloride ions in seawater. Several anions, including nitrate and nitrite ions, in subterranean waters that contain high concentrations of salts have been determined using 100 mM sodium chloride as the carrier solution by Song et al. [18]. They found that the peak area for a constant concentration of an anion remained constant regardless of the concentration of salt in the sample. However, the relative standard deviations (RSDs) of the peak areas and the limits of detection (LODs) for nitrate and nitrite ions were relatively high. Nitrate and nitrite ions in

biological samples such as blood plasma, serum and urine etc., have been also determined by micellar electrokinetic capillary chromatography (MECC) [19] and CZE [20,21].

In our previous paper [22], we used diluted artificial seawater as the carrier solution for the determination of bromide ions in seawater. It was shown that the method might be applicable to the determination of nitrate and nitrite ions in seawater, by improving the detection power and the resolution between nitrate and nitrite ions. In this paper, above method was improved for the determination of nitrate and nitrite ions in seawater. Optimum analytical conditions, such as wavelength, applied voltage, injection volume, and concentration of cetyltrimethylammonium chloride (CTAC) were established. The effects of the concentration of artificial seawater in sample solutions on the peak area, peak height and migration time were also investigated. Finally, the proposed method was applied to the determination of nitrate and nitrite ions in seawater samples taken from the surface and the seabed, collected around the coastal area of Osaka Bay in September 1998. The recovery of nitrate and nitrite ions was also examined.

2. Experimental

2.1. Apparatus

A Perkin-Elmer (Foster City, CA, USA) Model 270A capillary electrophoretic analyzer was used with a UV-Vis absorbance detector. The rise-time for the detector was set at 0.50 s. A polyimide-coated fused-silica capillary (GL Sciences, Tokyo, Japan), 100 μm I.D. \times 375 μm O.D., served as the capillary electrophoresis column. The total length of the column was 72 cm and the effective length was 50 cm. Peak area and peak height were measured using a Hitachi (Tokyo, Japan) Model D-2500 Chromato-Integrator. A Unicam Analytical Systems (Cambridge, UK) PW9421 pH meter was used. A Denki Kagaku Keiki (Tokyo, Japan) Model HPH-110 pH meter and a Yellow Springs Instrument (Yellow Springs, OH, USA) Model 30 S-C-T meter, and a Yellow Springs Instrument Model 58 DO meter were

used for measuring the pH, salinity and dissolved oxygen (DO) of seawater samples in situ, respectively. A glass reagent-bottle with a thin rope and a Rigosha (Tokyo, Japan) Vandorn water sampler (2 l) were used for sampling seawater samples taken from the surface and the seabed. Seawater samples were filtered through a 0.45- μm pore size membrane filter (Toyo Roshi, Tokyo, Japan) and analyzed within 2–6 h after collection. Then the seawater samples were stored in 500-ml polypropylene bottles (Sanplatec, Osaka, Japan) in a refrigerator for standard addition experiments.

2.2. Reagents

All reagents were of analytical-reagent grade and were used as received. CTAC was obtained from Tokyo Chemical Industry (Tokyo, Japan). Standard solutions of nitrate and nitrite ions (0.25–2.0 mg/l) were prepared from 1000 mg/l potassium nitrate (Wako, Osaka, Japan) and sodium nitrite (Nacalai Tesque, Kyoto, Japan) solutions. Distilled, demineralized water, obtained from a Yamato Kagaku (Tokyo, Japan) Model WG220 automatic still and a Nihon Millipore (Tokyo, Japan) Milli-QII system, was used throughout. The preparation of artificial seawater was based on a Japanese Standard [23]. The preparation procedure and the composition of the seawater are described in our previous paper [19]. All solutions used in this study were filtered through a 0.45- μm membrane filter before use.

2.3. Procedure

The detection wavelength was set at 210 nm. The thermostat was maintained at 30°C. A new capillary was washed with 1 M sodium hydroxide for 40 min and then with water for 10 min. Then the capillary was filled with a carrier solution (artificial seawater containing 3 mM CTAC, pH 7.9) by vacuum for 3 min. A small amount of the sample (65 nl) was vacuum injected into the capillary electrophoresis apparatus for 1 s. A voltage of 6 kV was applied, with the sample inlet side as the cathode. Each step was run automatically. Calibration graphs were prepared by use of synthetic standards.

3. Results and discussion

3.1. Wavelength

Over the wavelength range of 200–220 nm, artificial seawater, containing 1.0 mg/l nitrate and nitrite ions, was analyzed to select the optimum wavelength for the determination of nitrate and nitrite ions. Maximum peak areas for nitrate and nitrite ions were observed at 205 and 210 nm, respectively, as illustrated in Fig. 1. Similar results were obtained for the peak heights. The peak area for nitrite ion was 1.7-times smaller than that for nitrate ion at 210 nm. This is explained by the difference of the molar absorptivity of nitrate and that of nitrite ions. The molar absorptivities of nitrate and nitrite ions are $8.3 \cdot 10^3$ and $4.9 \cdot 10^3$, respectively [6]. That is to say, the ratio of these values is 1.7. Therefore, 210 nm was adopted as the optimum wavelength for the determination of nitrate and nitrite ions.

3.2. Applied voltage

The effects of applied voltage on the reproducibilities of the peak areas, peak heights and migration

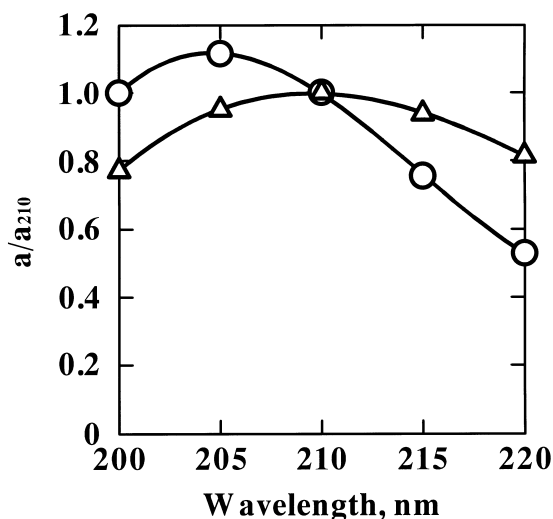


Fig. 1. Effects of wavelength on the peak areas for nitrate and nitrite ions. a = Peak area (arbitrary units); a_{210} = peak area at 210 nm; \circ = NO_3^- ; \triangle = NO_2^- . Carrier solution, artificial seawater containing 3 mM CTAC, pH 7.9; sample, artificial seawater containing 1.0 mg/l NO_3^- and 1.0 mg/l NO_2^- .

times (CZE parameters) for nitrate and nitrite ions were studied over the range 4–8 kV. The RSD values for peak areas were minimized at 6 kV, as evident in Table 1, whereas there were only small differences in those for peak heights. The RSD values for migration times at 4 kV were larger than those at 6 and 8 kV. In addition, the migration times increased with a decrease in the applied voltage. The migration times at 4 kV were 1.7-times longer than those at 6 kV. Therefore, 6 kV was adopted as the optimum voltage for the determination of nitrate and nitrite ions. Although the current was higher than that normally encountered in CZE, sharp peaks and stable baselines were obtained by operating at the relatively low applied voltage.

3.3. Injection volume

The effects of injection volume on the separation of nitrate and nitrite ions were studied. The injection volume was varied by changing the period for vacuum injection from 1 to 3 s. The injection periods, 1, 2 and 3 s, correspond to the injection volumes, 65, 130 and 195 nl, respectively. The volume of material injected per unit time (V_t , nl/s) is the function of the pressure drop (ΔP), the internal diameter of the capillary (D), the viscosity (η), and the length of the capillary (L) [24]. When the period for vacuum injection was 1 s, nitrate ion was resolved from nitrite ion, as shown in Fig. 2. However, the separation of nitrate and nitrite ions deteriorated at 2 s. Finally, only one peak was observed at 3 s. Therefore, sample solutions were vacuum injected for 1 s in all subsequent experiments. The number of theoretical plates (N) for nitrate and nitrite ions at 1 s were 13 000 and

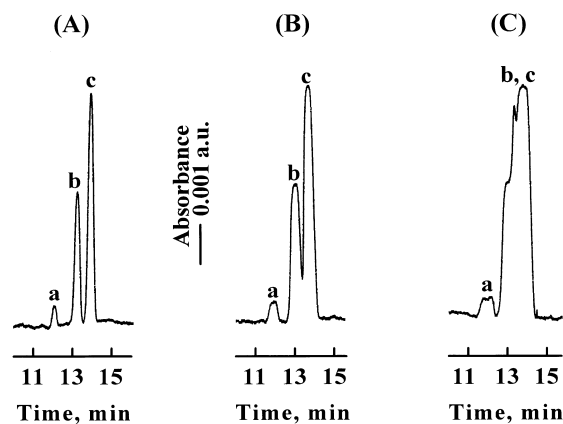


Fig. 2. Effect of injection volume on the separation of nitrate and nitrite ions. The period for vacuum injection, (A)=1 s, (B)=2 s, (C)=3 s. (a) Br^- ; (b) NO_2^- ; (c) NO_3^- . Carrier solution and sample are as in Fig. 1.

14 000, respectively. These values were calculated by the equation proposed by Tsuda [25] using the peak areas, the peak heights and the migration times. A small peak, appeared before the nitrite ion in Fig. 2. Considering from the absorptivity of bromide ion ($2.3 \cdot 10^3$ at 210 nm [6]) and the results of standard addition experiments, the peak corresponds to bromide ions in the sample solution.

3.4. Concentration of CTAC in carrier solution

The concentration of CTAC in the carrier solution was varied in the range 0–3 mM. The resolution increased with an increase in the concentration of CTAC, as evident in Fig. 3. Jimidar et al. [26] examined the influence of the concentration of cetyltrimethylammonium bromide (CTAB) on the effective mobility of some inorganic anions using

Table 1

Effect of applied voltage on the reproducibility of the peak area, peak height and migration time for nitrate and nitrite ions^a

Voltage (kV)	RSD (%)							
	Area		Height		Time		Migration time (min)	
	NO_3^-	NO_2^-	NO_3^-	NO_2^-	NO_3^-	NO_2^-	NO_3^-	NO_2^-
4	1.7	4.2	1.5	1.2	3.9	3.6	23.6	22.4
6	1.5	2.4	0.42	0.71	0.85	0.78	13.9	13.2
8	4.5	6.3	0.32	1.6	0.39	0.56	10.8	10.2

^a Sample: Artificial seawater containing 1.0 mg/l NO_3^- and 1.0 mg/l NO_2^- . Number of determinations: eight.

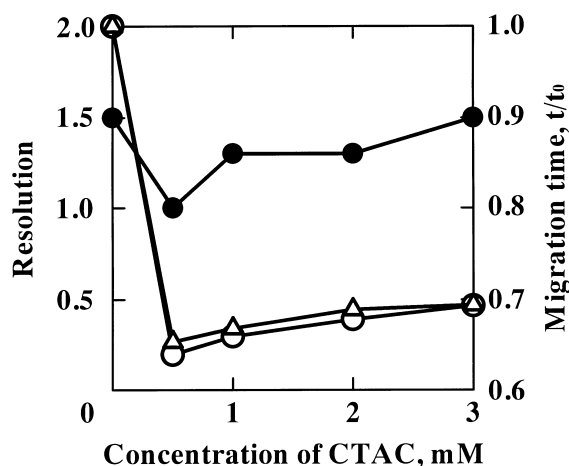


Fig. 3. Effects of the concentration of CTAC in carrier solution on the resolution and migration times for nitrate and nitrite ions. ●=Resolution; ○=migration time for NO_3^- ; △=migration time for NO_2^- ; t =migration time; t_0 =migration time with the carrier solution containing no CTAC. Carrier solution, artificial seawater containing 0–3 mM CTAC, pH 7.9; sample is as in Fig. 1.

chromate electrolyte as the carrier solution. They showed that the effective mobility of nitrate and nitrite ions decreased with an increase in the concentration of CTAB up to 2.5 mM and the tendency for nitrate ion was larger than that for nitrite ion. Kaneta et al. [27] also showed that the effective mobility of nitrate ion decreased with increasing concentration of CTAC up to 2.5 mM using phosphate carrier solution. Our result is consistent with these results. On the other hand, the migration times also increased as the concentration of CTAC increased. The values on the right ordinate show the ratio of the migration time to that without CTAC. Therefore, 3 mM was adopted as the optimum concentration of CTAC.

3.5. Concentration of artificial seawater in sample solutions

Sample solutions containing 1.0 mg/l nitrate and nitrite ions in 0–100% artificial seawater were prepared and analyzed by the method to examine the effects of the concentration of artificial seawater on the CZE parameters for nitrate and nitrite ions. The results are illustrated in Figs. 4 and 5. The values on the ordinate show the ratios of the peak area, peak

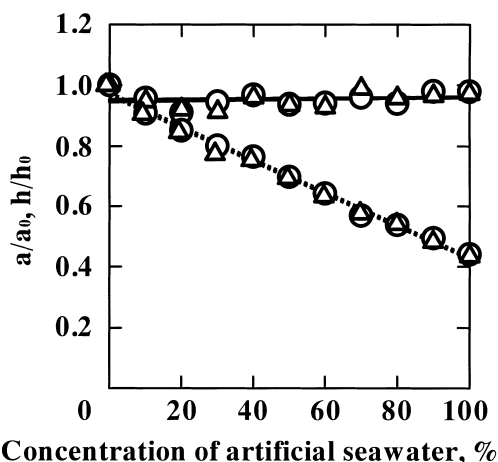


Fig. 4. Effects of the concentration of artificial seawater in sample solutions on the peak area and peak height for nitrate and nitrite ions. a , Solid line=peak area; a_0 =peak area for the sample solution containing no artificial seawater; h , dotted line=peak height; h_0 =peak height for the sample solution containing no artificial seawater; ○= NO_3^- ; △= NO_2^- . Carrier solution and the concentrations of NO_3^- and NO_2^- are as in Fig. 1.

height and migration time to those for the sample solution which contains no artificial seawater. The peak area and migration time were approximately constant over the range of 0 to 100% artificial seawater. The RSD of the peak area for nitrate ion

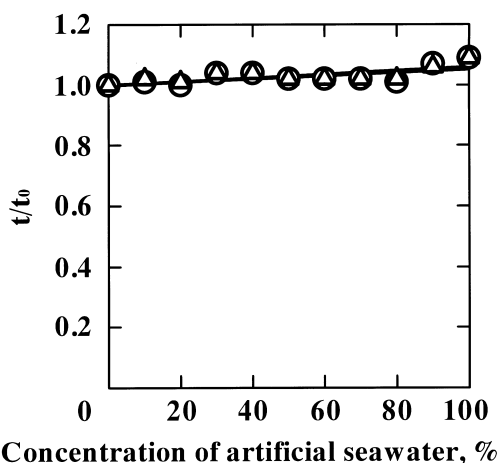


Fig. 5. Effects of the concentration of artificial seawater in sample solutions on the migration time for nitrate and nitrite ions. t =Migration time; t_0 =migration time for the sample solution containing no artificial seawater; ○= NO_3^- ; △= NO_2^- . Carrier solution and the concentrations of NO_3^- and NO_2^- are as in Fig. 1.

was 2.6%, which was calculated using the 11 values of the peak areas given in Fig. 4. The RSD of the peak area for nitrite ion was 2.9%. The RSDs obtained by our experiment were much lower than those obtained by Song et al. [18], 7.2–20.3% for nitrate ion and 5.1–18.0% for nitrite ion; the concentrations of nitrate and nitrite ions in our experiment, 1.0 mg/l ($7.1 \cdot 10^{-2}$ mM), were lower than the concentrations (1–40 mM) used in their experiment. The RSDs of the migration times for nitrate and nitrite ions were 2.8 and 2.5%, respectively. The peak height increased linearly with decreasing concentrations of artificial seawater. This is due to the stacking effect caused by the difference between the field strength of the sample zone and that of the electrolyte zone. Therefore, the concentrations of nitrate and nitrite ions in seawater can be determined by the working curve method using the peak area in spite of differences in the salinity of the sample solutions.

3.6. Analysis of seawater samples

Calibration graphs for nitrate and nitrite ions were linear using both peak area and peak height, up to at least 2.0 mg/l. The regression equations relating area response (y , arbitrary units) to concentration (x , 0–2.0 mg/l) for nitrate and nitrite ions were $y=1.06 \cdot 10^5 x - 2230$ (correlation coefficient 0.9998) and $y=5.93 \cdot 10^4 x + 264$ (0.9999), respectively. The regression equations relating height response to concentration for nitrate and nitrite ions were $y=5.55 \cdot 10^3 x - 96$ (correlation coefficient 0.9999) and $y=3.27 \cdot 10^3 x + 23$ (1.000). The magnitude of the slopes of the regression lines for the area response was 18- or 19-times larger than that for the height response. In the previous section, we demonstrated that the concentrations of nitrate and nitrite ions in seawater could be determined by the working curve method using the peak area in spite of differences in the salinity of the sample solutions. Therefore, the peak area was used for calculation of the concentrations of nitrate and nitrite ions in seawater. The LODs for nitrate and nitrite ions were 0.04 and 0.07 mg/l ($S/N=3$), respectively. Song et al. [18] reported that the LODs for nitrate and nitrite ions increased with increasing concentration of sodium chloride in the sample from 12.5 to 100 mM; the LODs were 0.332

and 0.368 mg/l ($S/N=2$) when the concentration of sodium chloride was 100 mM. That is to say, the LODs for nitrate and nitrite ions in our experiment were one-ninth and one-fifth of the LODs obtained by Song et al. The comparison of the LODs and RSDs for nitrate and nitrite ions obtained by the proposed method with those obtained by CE for the biological samples is as follows. The LODs for nitrate and nitrite ions were 1/18th and one-third of the LODs obtained by Bjerregaard et al. [19] using a high sensitivity optical cell assembly (Z-cell), 0.70 and 0.21 mg/l, respectively. The RSDs of peak areas and migration times for nitrate and nitrite ions were almost equal to those obtained by them, 1.75, 2.37, 0.29 and 0.24%, respectively. The LOD for nitrate ion was one-third of the LOD obtained by Ueda et al. [20] using 750 mM sodium chloride containing an electroosmotic flow modifier as the buffer, 0.1 mg/l. The RSDs of peak areas and migration times for nitrate and nitrite ions were almost equal to those obtained by them, 1.1, 1.8, 0.4 and 0.4%, respectively. The LODs for nitrate and nitrite ions were 1/8th and 1/4th of the LODs obtained by Friedberg et al. [21] using sample stacking caused by acetonitrile, 0.3 mg/l, respectively. The RSDs of peak heights for nitrate and nitrite ions were much smaller than those obtained by them, 5.7 and 3.7%, respectively.

The proposed method was applied to the determination of nitrate and nitrite ions in seawater taken from the surface and seabed and collected from around the coastal area of Osaka Bay, between Port of Amagasaki and Port of Kobe, on 14 and 16 September 1998. The seawater samples from the Port of Amagasaki and mouth of Muko river contained relatively high concentrations of nitrate and/or nitrite ions, as presented in Table 2. The concentrations of nitrate and nitrite ions in the seabed seawater sample from the Port of Amagasaki were lower than those in the surface seawater sample. Nitrate ion was detected in surface seawater sample from the mouth of Muko river, but neither nitrate nor nitrite ion was detected from the seabed seawater sample. Considering from the low DO around the seabed in these waters, nitrate and nitrite ions might be reduced to ammonium ions. We mentioned that 3.0 and 0.68 mg/l nitrate ions were detected in the surface and seabed seawater samples from the Port of Amagasaki in the previous paper [19]. Above results suggest that

Table 2
Analytical results for nitrate and nitrite ions in seawater^a

Sampling site	Depth (m)	Temperature (°C)	pH	S (‰)	DO (mg/l)	NO ₃ ⁻ (mg/l)	NO ₂ ⁻ (mg/l)
Port of Amagasaki	0	27.8	7.19	23.4	9.36	1.16	0.13
Port of Amagasaki	3.0	27.8	6.90	27.5	3.41	0.49	0.09
Mouth of Muko river	0	27.4	6.98	29.9	6.29	0.13	ND
Mouth of Muko river	3.0	27.0	6.70	31.3	0.56	ND	ND
Rokko Island	0	27.0	7.20	30.7	8.58	ND	ND
Rokko Island	10.0	26.5	6.83	31.6	3.50	ND	ND
Pond at KUMM	0	26.5	7.17	30.3	7.05	ND	ND
Pond at KUMM	4.0	26.3	7.18	30.7	4.41	ND	ND

^a KUMM: Kobe University of Mercantile Marine. Sampling dates: 14 and 16 September 1998.

eutrophication is still in progress in these areas. The concentrations of nitrate and nitrite ions shown in Table 2 were the same magnitude as those obtained by Nakamura et al. [7], 2.61–3.01 mg/l for nitrate ion and 0.15–0.25 mg/l for nitrite ion; nitrate and nitrite ions were not detected in the samples taken from other waters. Fig. 6A represents an electropherogram of surface seawater from the Port of

Amagasaki. Fig. 6B was obtained for the analysis of the surface seawater, diluted 10/9th-fold, with 1.0 mg/l nitrate and nitrite ions added. It was also confirmed that the small peak before the nitrite peak corresponded to bromide ion by standard addition experiments.

The seawater samples, diluted 10/9th-fold, with adding 0–2.0 mg/l nitrate and nitrite ions, were also

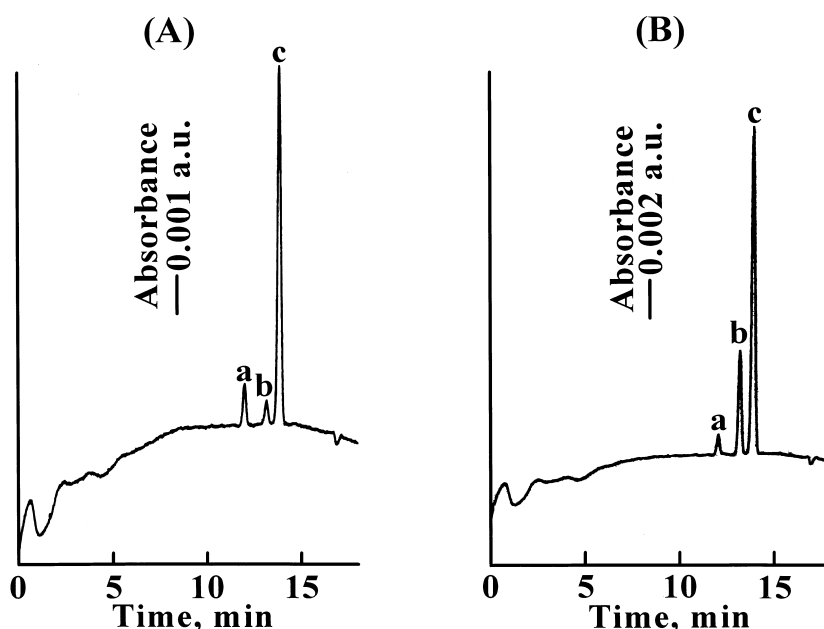


Fig. 6. Electropherograms for the separation of nitrate and nitrite ions in seawater samples. (A) Sample, surface seawater from the Port of Amagasaki; (B) sample, the surface seawater, diluted 10/9th-fold, with addition of 1.0 mg/l NO₃⁻ and 1.0 mg/l NO₂⁻. (a) Br⁻; (b) NO₂⁻; (c) NO₃⁻. Carrier solution is as in Fig. 1.

Table 3
Recovery of nitrate and nitrite ions in seawater^a

Sampling site	Depth (m)	Added (mg/l)		Found (mg/l)		Recovery (%)	
		NO ₃ ⁻	NO ₂ ⁻	NO ₃ ⁻	NO ₂ ⁻	NO ₃ ⁻	NO ₂ ⁻
Port of Amagasaki	0	0	0	1.06	0.11	–	–
Port of Amagasaki	0	0.5	0.5	1.55	0.64	98	106
Port of Amagasaki	0	1.0	1.0	2.04	1.13	98	102
Port of Amagasaki	3.0	0	0	0.44	0.07	–	–
Port of Amagasaki	3.0	0.5	0.5	0.95	0.56	102	98
Port of Amagasaki	3.0	1.0	1.0	1.44	1.09	100	102
Port of Amagasaki	3.0	1.5	1.5	1.89	1.74	97	111
Rokko Island	0	0.5	0.5	0.53	0.57	106	114
Rokko Island	0	1.0	1.0	1.03	1.07	103	107
Rokko Island	0	1.5	1.5	1.52	1.49	101	99
Rokko Island	0	2.0	2.0	1.98	2.09	99	105
Rokko Island	10.0	0.5	0.5	0.53	0.51	106	102
Rokko Island	10.0	1.0	1.0	1.03	0.99	103	99
Rokko Island	10.0	1.5	1.5	1.50	1.48	100	99
Rokko Island	10.0	2.0	2.0	1.97	1.98	99	99

^a Samples: 10/9th-fold diluted seawater with addition of 0–2.0 mg/l nitrate and nitrite ions.

analyzed by the method. The recovery of nitrate and nitrite ions was 97–114%, as shown in Table 3.

4. Conclusions

The CZE method using artificial seawater as the carrier solution is simple, rapid and possesses sufficient precision and freedom from the interference of chloride ion etc. and from differences in salinity to be useful for the determination of nitrate and nitrite ions in seawater. It does not require any pretreatment, although the further improvement of the LODs is necessary for lower concentrations of nitrate and nitrite ions as evident in Table 2. The use of a high sensitivity detector, which has longer optical path length, is worthy of a further study to improve the LODs for nitrate and nitrite ions.

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