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# Determination of Nitrate and Nitrite in Hanford Defense Waste (HDW by Reverse Polarity Capillary Zone Electrophoresis (RPCE)

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Numatec Hanford Corporation

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**DETERMINATION OF NITRATE AND NITRITE IN HANFORD DEFENSE WASTE  
(HDW) BY REVERSE POLARITY CAPILLARY ZONE ELECTROPHORESIS (RPCE)  
METHOD.**

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## Abstract

This paper describes the first application of reverse polarity capillary zone electrophoresis (RPCE) for rapid and accurate determination of nitrate and nitrite in Hanford Defense Waste (HDW). The method development was carried out by using Synthetic Hanford Waste (SHW), followed by the analysis of 4 real HDW samples. Hexamethonium bromide (HMB) was used as electroosmotic flow modifier in borate buffer at pH 9.2 to decrease the electroosmotic flow (EOF) in order to enhance the speed of analysis and the resolution of nitrate and nitrite in high ionic strength HDW samples. The application of this capillary zone electrophoresis method, when compared with ion chromatography for two major components of HDW, nitrate and nitrite slightly reduced analysis time, eliminated most pre-analysis handling of the highly radioactive sample, and cut analysis wastes by more than 2 orders of magnitude. The analysis of real HDW samples that were validated by using sample spikes showed a concentration range of 1.03 to 1.42 M for both nitrate. The migration times of the real HDW and the spiked HDW samples were within a precision of less than 3% relative standard deviation. The selectivity ratio test used for peak confirmation of the spiked samples was within 96% of the real sample. Method reliability was tested by spiking the matrix with 72.4 mM nitrate and nitrite. Recoveries for these spiked samples were 93-103%.

**Key Words:** Reverse polarity capillary zone electrophoresis; Electroosmotic flow modifier; Buffer/sample ionic strength ratio; Nitrate and Nitrite.; Hanford Defense Waste.

## 1. Introduction:

The characterization of more than 60 million gallons of radioactive and chemical wastes stored in 177 underground tanks at the US-Department of Energy Hanford Site near Richland, Washington, has been difficult due to the complex composition of the wastes [1, 2] and high analytical costs [3] involved in the process. These wastes, known as Hanford Defense Waste (HDW), were accumulated during World War II and the Cold War when the mass production of plutonium for making nuclear weapons was a top priority of the United States. The tanks where the wastes are stored, vary greatly in chemical and radiochemical compositions as a result of many different waste processing and treatment procedures that have occurred over the years. Tank composition has been further complicated by the mixing of one tank with another. The HDW has a very high pH and a very high ionic strength. Part or all of the liquid contents of some of the tanks were pumped out resulting in a multiphase mixture of solid sludge, saltcake, slurry, and supernatant liquor which are largely made up of various metals ions including radionuclides in addition to nitrate, nitrite, hydroxide, carbonate, and a number of organic complexants. The complexants were used in suppressing the formation of heavy metal hydroxides and facilitating the separation of some of the radionuclides. The tanks have high nitrate content because metal nitrates, nitric acid, and sodium nitrite were major constituents used in the processing and recovery of plutonium [4-6].

Methods for determining nitrate and nitrite such as spectrophotometry [7] and the flow injection analysis cadmium reduction method [8] cannot be used for the analysis of HDW because dissolved organic matter and metal ions present in HDW are known to interfere. Ion

chromatography (IC) is the method commonly used for the analysis of nitrate and nitrite in HDW [9, 10]. Some ion interferences have been reported in IC method for anions [9]. Moreover, the IC method generates high volumes of additional mixed wastes that are expensive to dispose, resulting in increased cost of analysis per sample.

One analytical method which has not been investigated for the determination of nitrate and nitrite in HDW is capillary zone electrophoresis (CZE). Several CZE analyses of nitrate and nitrite in aqueous and other samples have been reported [11-16]. These CZE methods involved the use of acidic buffers, and indirect detection based on organic acids and chromate buffers at pH 6-8 range. In the CZE of a very complex matrix like the Hanford Waste, the buffer/electrolyte and pH must be selected to insure that precipitation of the sample does not occur during the runs. Using acidic buffers require undesirable sample preparation steps to acidify the highly basic HDW. The high ionic strength and high absorbing background of HDW also help defeat detection by indirect methods.

The overall objective of this project was to develop a simple, reliable, and rapid method based on reverse polarity capillary zone electrophoresis (RPCE) for direct UV detection of nitrate and nitrite anions in HDW under basic sample conditions. Reverse polarity capillary zone electrophoresis is a technique that will lead to waste minimization and waste avoidance, and will result in significantly reduced overall costs of the Hanford Site characterization. RPCE involves the application of negative voltage at the injection position of the CE instrument. In this study, the electroosmotic flow (EOF) was reduced by adding hexamethonium bromide (HMB) to the buffer. This method was anion specific because cations and neutral compounds migrate away

from the detector and were not detected. Because analyzing HDW samples under high pH involves minimal sample preparation and sample handling, the RPCE method will be used for the analysis of real HDW without adjusting the pH of the sample. The specific objectives of this study were to determine if RPCE could be used for the separation of nitrate and nitrite in a high ionic strength sample, examine anions and complexants commonly found in HDW which may interfere with the analysis and demonstrate the method with both Synthetic Hanford Waste (SHW) sample and real HDW samples. Micro-sample sizes were used for the analyses because of the advantage of minimizing the exposure of workers and equipment to radiation.

## **2. Materials and Methods:**

### **2.1 Instrumentation:**

Two capillary electrophoresis instruments were used in this investigation. First, a Dionex Capillary Electrophoresis System (CES) I (Dionex Corporation, Sunnyvale, California) was used for the study involving Synthetic Hanford Waste (SHW) samples. The CES I was equipped with a tunable UV/visible detector with a wavelength 190-800 nm range, a positive and a negative polarity high voltage power supplies with 0-30 kV range. It allowed three methods of sample introduction: gravity, electromigration and pressure. A Dionex Advanced Computer Interface (ACI) was used to interface the CES I to a Dionex AI-450 Laboratory Automation System for data acquisition, storage, and analysis.

Second, a Waters model Quanta 4000E capillary ion analyzer (CIA) (Waters Millipore Corporation, MA) connected to a 486 PC for instrument control, was used for the preliminary



analysis of the real HDW samples. The high voltage power supply for the Quanta 4000E was 0-30 kV suitable for both positive and negative polarities. It had the capability of operating under constant electric current. The UV detector was configured for a limited fixed-wavelength filters of which the 215 nm filter was used in this study. It had both hydrostatic and electromigration methods of sample introduction.

## 2.2 Reagents and Chemicals.

Sodium nitrate Analytical Reagent, ACS Specifications (Mallinckrodt Chemical Works, St. Louis MO), and sodium nitrite Baker Analyzed Reagent, ACS Specifications, (J.T. Baker, Inc., Phillipsburge, NJ) were dried at 110 °C overnight, cooled and stored in a dessicator. Standard stock solutions containing 71.42 mM nitrate/nitrite were made following EPA Method 300.0 specifications [17]. Sodium borate SX365 CB857 (Matheson Coleman & Bell, Norwood, OH), and hexamethonium bromide (ICN Biochemicals, Inc. Cleveland, OH) were used for buffer preparations. The aqueous solution of hexamethonium bromide is acidic [\*\*\*]. The pH of 100mM solution in water was 4.62. HMB partially converts to the hydroxide form on mixing with borate to maintain the pH of the buffer at pH 9.3. Reagent Water from 18MΩ nanopure UV ultrapure water system (Barnstead/Thermolyne, Dubuque, Iowa) was used for the preparation of buffer and all standard solutions.

## **2.3 Experimental:**

### **2.3.1 Optimization of RPCE applied voltage for the separation of high concentration levels of nitrate and nitrite:**

The optimum negative voltage for the separation of nitrate and nitrite was examined for up to -22 kV across a 50  $\mu\text{m}$  i.d., x 50 cm long bare silica column for 25 mM borate/25 mM HMB buffer. Gravity injection of 1.0 mM nitrate and 1.0 mM nitrite standards were carried out for 60 seconds at 100 mm head height of the injector. Absorption maxima wavelength for nitrate and nitrite at 220 nm was used for direct UV detection.

### **2.3.2 Interference study of anions and complexants commonly found in Hanford Defense Waste tanks:**

The separation of nitrate and nitrite in the presence of chloride, sulfate, carbonate, and phosphate anions was carried out under the electrophoretic conditions of section 2.3.1. For detection of nitrate and nitrite in the presence of the complexants, the detector wavelength was set at 240 nm, the absorption maxima wavelength for the complexants. The concentration of the complexants standards added were 10 mM oxalate, 1.8 mM nitrilotriacetic acid (NTA), 0.90 mM ethylenediaminetetraacetic acid (EDTA), and 0.90 mM N-hydroxyethylethylenediaminetriacetic acid (HEDTA) at sample pH of 12. The mixture was made up in reagent water.

### 2.3.3 Separation of nitrate/nitrite in the Synthetic Hanford Waste<sup>1</sup> (SHW):

The Synthetic Hanford waste (SHW) was a simulated HDW sample matrix, containing 1.02 M  $\text{Na}_2\text{Al}_2\text{O}_4$ , 1.61 M NaOH, 2.59 M  $\text{NaNO}_3$ , 2.24 M  $\text{NaNO}_2$ , and 0.42 M  $\text{Na}_2\text{CO}_3$ , estimated to be nearest in composition and behavior of the major components of HDW. This mixture is very high in ionic strength, highly viscous and has  $\text{pH} > 13$ . The sample preparation of SHW, as was true of HDW, was often formidable, with massive formation of hydroxide precipitates, and excessive foaming as the sample pH was lowered from pH 11 to pH 4. Therefore, sample pH was kept above pH 11. The sample preparation of SHW was a simple 4-fold serial dilution of 1:10 in each step to a final sample concentration of 0.016 vol.%. Standard addition calibration was used for quantification by adding 0.000, 0.0235 mM, 0.118 mM, 0.589 mM, and 1.18 mM nitrite standards and 0.000, 0.0871 mM, 0.436 mM, 2.18 mM, and 4.36 mM nitrate standards to the sample. Replicate RPCE runs were carried out under the electrophoretic conditions of -20 kV applied across a 50  $\mu\text{m}$  i.d., x 50 cm long bare silica column in a 25 mM borate/25 mM HMB buffer. Gravity injection for 60 seconds at 100 mm injector head height was used.

### 2.3.4 Analysis of real HDW samples:

For the purposes of sample identification, the real HDW samples analyzed were assigned the numbers S96R000308, S96R000309, S96R000311, and S96R000312. For simplicity, only the last 3 digits were reported in the results and discussion section. The procedure used for the preliminary analysis of the real HDW samples involved a serial dilution of HDW sample as follows: 100  $\mu\text{L}$  of HDW sample was added to 1.0 mL of reagent water, and further diluted a second time resulted in an overall 1:121 dilution. A 1:6 further dilution of the sample was

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<sup>1</sup> SHW supplied by Westinghouse Hanford Company, Richland, Washington.

carried out to give a final mixing ratio of 0.138 vol.%. Spiking of the real HDW samples was carried out by adding 25  $\mu\text{L}$  of standard that gave a final concentration of 1.0 mM of nitrate and nitrite in the sample. The percent recovery of the matrix spike was used as a method of reliability test in addition to using standard addition calibration method to check accuracy of the data obtained by using external calibration. The RPCE replicate runs were carried out in a 50  $\mu\text{m}$  i.d. x 50 cm long capillary column. A constant current maintained at 75  $\mu\text{A}$ , rather than a constant voltage was therefore applied across the capillary because constant current gave a more reproducible migration times than constant voltage. In addition, a 30 seconds 0.5 M hydroxide rinse was carried out between runs so as to maintain a more reproducible migration time during the analysis. Gravity injection was used for sample introduction by raising the injector to a height of 9.8 cm for 30 seconds sampling time.

### **3 Results and Discussion:**

#### **3.1 Optimization of RPCE applied voltage for the analysis of SHW:**

The applied voltage for the separation of nitrate and nitrite in SHW was investigated in order to obtain the optimum voltage that achieved a good detection limit, high separation efficiency, and rapid analysis. The electropherogram of nitrate and nitrite standards containing 1.0 mM of each analyte under the RPCE conditions of -20 kV applied voltage is shown in Figure 1. The separation gave very good peak shape and high resolution, but slightly noisy baseline. The highest achievable voltage in the 25 mM borate/25 mM HMB buffer was -22 kV. When higher than -22 kV was applied, high electric current was generated which produced power output that exceeded the power rating of the instrument and resulted in an electrical short. Thus, optimum

voltage for the separation of nitrate and nitrite in SHW was therefore -20 kV across a 50  $\mu\text{m}$  i.d., and 50 cm long bare silica capillary column. When greater than -20kV voltage was used for the RPCE runs, a noisy background from Joule heating occurred [18]. This noise limited the lowest detectable concentrations of nitrate and nitrite to 0.05 mM. The separation of lower sample concentrations required the application of low voltage across the column in order to reduce the temperature effect discussed above. Alternatively, low ionic strength buffer can be used for the analysis of samples containing less than 0.05 mM nitrate and nitrite [11]. Although more than -20 kV voltage could be used if the buffer concentration was lower, the high ionic strength of the HDW samples degraded resolution in lower buffer concentration. The resolution between nitrate and nitrite at -20 kV in the buffer used in this study was very high ( $> 4$ ), and migration times for nitrite and nitrate were within 3 minutes. It has been shown that buffer/sample ionic strength ratio affects the resolution of anions in RPCE [19]. Resolution increases with an increase in buffer/sample ionic strength ratio, hence, a high ionic strength buffer was recommended for the analysis of high ionic strength samples.

### **3.2 Study of potential interference anions and complexants commonly found in Hanford Defense Waste tanks:**

Figure 2 shows an electropherogram of a mixture of nitrate and nitrite and some complexants commonly found in HDW. This Figure demonstrated that the complexants did not interfere with the separation of nitrite and nitrate in the waste. The migration times of nitrate, nitrite, and oxalate were under 3 minutes, while the migration time of NTA, EDTA, and HEDTA were 4.61 minutes, 4.74 minutes, and 5.76 minutes respectively. Nitrate and nitrite displayed narrow peak

shapes, while the complexants especially oxalate and NTA showed tailing and leading edges respectively as a result of their conductivity mismatch with the conductivity of the buffer [19].

The separation of nitrate from oxalate can be difficult because their equivalence conductance [20] and electrophoretic mobilities are very close [21]. In fact, an investigation reported by Wildman, *et al.* [22], showed that nitrate co-migrated with oxalate in a separation with NICE-Pak OFM Anion-BT (Waters Corporation) buffer/electroosmotic flow modifier. Figure 2 on the other hand, shows that RPCE produces a good separation of nitrate, nitrite, oxalate and other complexants. The better resolution between oxalate and nitrate in RPCE compared to the NICE-Pak OFM Anion-BT method can be explained as a function of the electroosmotic flow (EOF). The electroosmotic modifier used in the RPCE method reduced the EOF but did not reverse its normal direction i.e., the EOF still was flowing in a direction counter to the anions. The analysis using the NICE-Pak OFM Anion-BT on the other hand, had the EOF reversed and hence, EOF was in the same direction as anions. The contribution of the reduced electroosmotic flow velocity to the separation efficiency accounted for the good resolution observed [11]. Because of the low absorptivity of oxalate at 240 nm wavelength, the oxalate concentration used in the study was about 7 times greater than that of nitrate. Figure 2 demonstrated that at high oxalate concentrations, better than base line resolution was achieved. Sulfate, chloride, carbonate and phosphate anions were tested during the analysis, and none of these interfered. Another major advantage of the RPCE over conventional CZE was that cations and neutral compounds migrate in the opposite direction of the anions away from the detector. Thus cations and neutrals were never observed and hence did not interfere with the method [19].

### 3.3 Separation of nitrate and nitrite in the Synthetic Hanford Waste:

The separation of nitrate and nitrite in 0.016 vol.% SHW is shown in Figure 3. The electropherogram showed a rapid separation of nitrite and nitrate within 3 minutes and a high resolution ( $> 4$ ). Standard addition method was used for quantification, and the results obtained were within 95% agreement with the amount used in the preparation of the SHW. The sensitivity of the standard addition curves (62.5 area counts/mM and 70.8 area counts/mM for nitrite and nitrate respectively) were in good agreement with those obtained by method of extrapolation. One of the advantages of the successful analysis of SHW is that the RPCE method can now be used for the analysis of real HDW containing very high radioactivity in which a simple dilution will be significant in reducing the level of sample radioactivity and lower radiation dose exposure to the analyst during analysis. Sample radioactivity can be lowered by more than 3 orders of magnitude. Moreover, no further sample preparation nor pH adjustments was necessary before the RPCE run.

### 3.4 Analysis of real HDW samples:

Because of the high salt concentrations of the real HDW samples, constant voltage applications during the RPCE analysis produced electric current in the sample zone that was usually higher than the electric current in the buffer zone. This caused a fluctuation in the migration times of the analytes. For this reason, a constant current of 75  $\mu\text{A}$  rather than a constant voltage was used for the analysis. Table I is the summary of results obtained for the analysis of 4 real HDW samples showing a concentration range of 1.03 to 1.42 M of nitrate and nitrite in the samples. The standard deviations for 3 replicate runs carried out for the spiked samples were all within 2% RSD. The precision obtained for 3 replicate runs for each sample based on external calibration curve were slightly higher than those of the spiked samples, but within 6% RSD.

Sample spikes were used to positively confirm the identification of nitrate and nitrite in HDW and as a method of accuracy check. Peak confirmation by spiking with the standard in high ionic strength sample may result in peak width broadening due to ionic strength dispersion effects in CZE [19].

The electropherograms of sample S96R000308 and spiked sample S96R000308 are shown in Figures 4A and 4B. Figure 4A showed that there were no interfering peaks within the migration times of nitrate and nitrite. Figure 4B is the electropherogram of the spiked sample. This Figure demonstrated that increase in peak area rather than peak height was proportional to the amount of the analyte added, in agreement with the results shown in Table I. In order to positively confirm the identification of the peaks after spiking, Okemgbo and others proposed the use of selectivity ratio,  $S$ , ( $S = \Delta\mu/\mu_{av}$ ), where  $\Delta\mu$  is the difference between electrophoretic mobilities of adjacent ions and  $\mu_{av}$  is their average electrophoretic mobilities) rather than proportional peak height due to ionic strength based peak broadening in spiked samples [19]. This principle was applied to the nitrate and nitrite peaks as shown in Table II. The migration times of both nitrate (3.621 and 3.601 minutes) and nitrite (3.393 and 3.392 minutes) in the real and spiked HDW samples were within 99% match for samples SR96000308. Similarly, the selectivity ratio (0.065 and 0.062 for the real and spiked HDW samples respectively) showed greater than 95% agreement. Furthermore, results obtained by standard addition calibration method were used for accuracy check of the data obtained by using external calibration. The % recoveries of spiked nitrate and nitrite standards were 95% or better except for sample S96R000312 which was 93%. This high % recoveries and high precision indicated good reliability of the method.



The SHW sample that was used in the RPCE method development contained higher concentrations of nitrate and nitrite (2.59 M and 2.24 M respectively) than the result obtained for real HDW sample analyzed (1.07 - 1.42 M range for nitrate and nitrite). The difference was due to the fact that the SHW matrix was made for the worse case situation of HDW. In addition, these results suggest that the HDW samples may come from tanks that have not gone through any waste concentration/evaporation treatment. The electropherograms were obtained under constant current of 75  $\mu$ A (corresponding to about 18 kV voltage), across a 50  $\mu$ m i.d. x 50 cm long fused silica capillary column. Although the application of higher current gave a faster analysis, pronounced noisy baseline also occurred, making peak area precision wider. Although a direct UV detection, at 215 nm was used during the RPCE, the peaks were inverted because the CE instrument was previously installed for indirect UV detection. Figures 5A and 5B are the standard addition calibration for nitrate and nitrite in HDW sample S96R000311. The linearity was better than  $r^2 = 0.970$  and the extrapolated results for nitrite and nitrate were 1.28 M and 1.05 M respectively. These results are within 10% agreement of 1.15 M and 1.03 M for nitrite and nitrate respectively obtained by external standard calibration shown in Table I.

#### 4. Conclusion:

The reverse polarity capillary zone electrophoretic analysis of Hanford Defense Waste for nitrate and nitrite was rapid, simple, precise, accurate, involved micro-sample size, and generated very low laboratory waste. Minimum sample handling with only simple dilution of the sample by a

factor of 3 orders of magnitude without any further sample preparation or pH adjustments was involved. This cut down the overall time of analysis per sample. The high dilution factor of the sample also reduced the level of radiation by the same factor and therefore reduced the level of radiation exposure to the analyst. The method will give high sample throughput (> 10 samples/hr.) in the Hanford Defense Waste characterization because RPCE can be run with minimum attention (it is automated). Although the SHW samples used in RPCE method development contained higher concentrations of nitrate and nitrite than the analyzed real HDW samples, SHW samples were a good reference point of high ionic strength and high pH for method development. RPCE is therefore recommended for the analysis of nitrate and nitrite not only in the HDW wastes stored in the tanks, but also for screening other high ionic strength samples.

**Acknowledgment:**

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**TABLE I**  
**RESULTS OF TEST ANALYSIS OF REAL HANFORD DEFENSE WASTE**

SAMPLE	[NO <sub>2</sub> <sup>-</sup> ] (M)	% Recovery	[NO <sub>3</sub> <sup>-</sup> ] (M)	% Recovery
S96R000308	1.42 ± 0.03	-	1.41 ± 0.03	-
S96R000308spiked	2.96 ± 0.06	101	2.75 ± 0.05	95
S96R000309	1.23 ± 0.03	-	1.07 ± 0.01	-
S96R000309 spiked	2.67 ± 0.01	100	2.56 ± 0.02	103
S96R000311	1.15 ± 0.02	-	1.03 ± 0.02	-
S96R000311 spiked	2.59 ± 0.05	99	2.46 ± 0.05	99
S96R000312	1.29 ± 0.03	-	1.26 ± 0.07	-
S96R000312 spiked	2.63 ± 0.06	93	2.55 ± 0.02	95

TABLE II  
COMPARISON BETWEEN REAL HDW AND SPIKED HDW SAMPLES FOR  
MIGRATION TIMES AND SELECTIVITY RATIO,  $S$ .

SAMPLE	$t_r(\text{NO}_2^-)$ (min.)	$t_r(\text{NO}_3^-)$ (min.)	$S = \Delta t / \mu_{av}$
308	3.393±0.007	3.621±0.007	0.065±0.001
308spike	3.392±0.009	3.608±0.009	0.062±0.002
311	3.403±0.005	3.630±0.005	0.065±0.001
311spike	3.402±0.004	3.626±0.008	0.064±0.001

**FIGURE CAPTIONS**

**Figure 1** The electropherogram of 1.0 mM nitrate and 1.0 mM nitrite standard solution. Buffer: 25 mM Borate, 25 mM Hexamethonium bromide, pH 9.2, direct UV detection at 220 nm wavelength. Applied voltage: - 20kV across a bare silica capillary column of 50 $\mu$ m i.d. by 50cm long. Gravity injection time was 60s.

**Figure 2** The electropherogram of a mixture of 1.5 mM nitrate, 0.70 mM nitrite, 10 mM oxalate, 1.8 mM nitrilotriacetic acid, 0.90 mM ethylenediaminetetraacetic acid, and 0.90 mM N-hydroxyethylethylenediaminetriacetic acid. Sample pH = 12, UV wavelength = 240 nm. Electrophoretic conditions: Buffer: 25 mM borate/25 mM hexamethonium bromide, pH 9.2. Applied voltage: - 20kV was 60s.

**Figure 3.** The electropherogram of Synthetic Hanford Waste showing the separation and detection of 1 mM nitrate and 1 mM nitrite in this complex sample. Electrophoretic conditions of Figure 1 were used during the separation.

**Figure 4A** The electropherogram of sample S96R000308, demonstrating the rapid separation and detection of nitrate and nitrite in the real sample. Electrophoretic conditions of 75  $\mu$ A constant current (negative polarity) across a bare silica capillary column of 50 $\mu$ m i.d. by 50cm long. Gravity injection time was 30s. **4B** is the electropherogram of the same sample in Figure 4A which has been spiked with 1 mM of nitrate and 1 mM of nitrate. The response showed a



proportional increase in peak area with the amount of standard added. Electrophoretic conditions as in Figure 4A.

**Figure 5A** The standard addition calibration curve for nitrate in HDW sample S96R000311. The linearity was better than  $r^2 = 0.97$  and the extrapolated results were in good agreement with the results in Table II. Electrophoretic conditions as in Figure 4. **5B** The standard addition calibration curve for nitrite in HDW sample S96R000311. The linearity was better than  $r^2 = 0.97$  and the extrapolated results were in good agreement with the results in Table II. Electrophoretic conditions as in Figure 4.

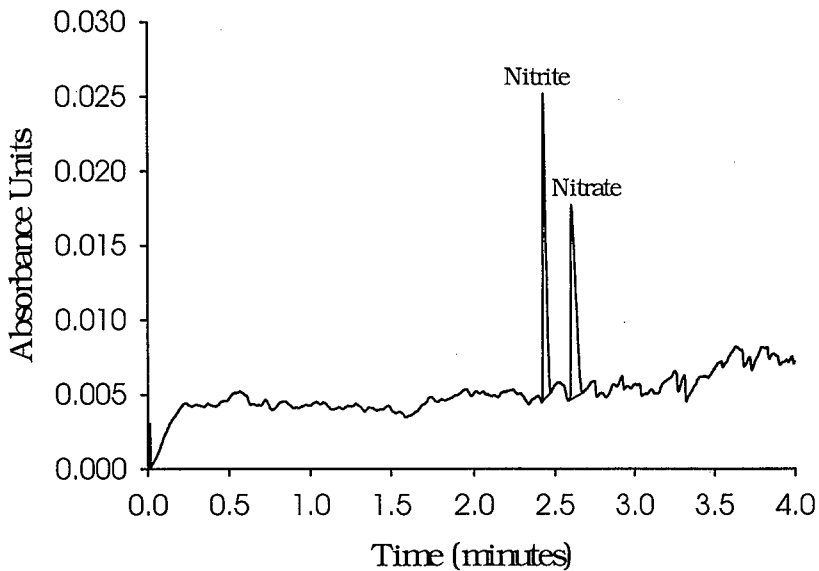


Figure 1. Okemgbo et al. Determination of Nitrate and Nitrite in Harford Defense Waste....

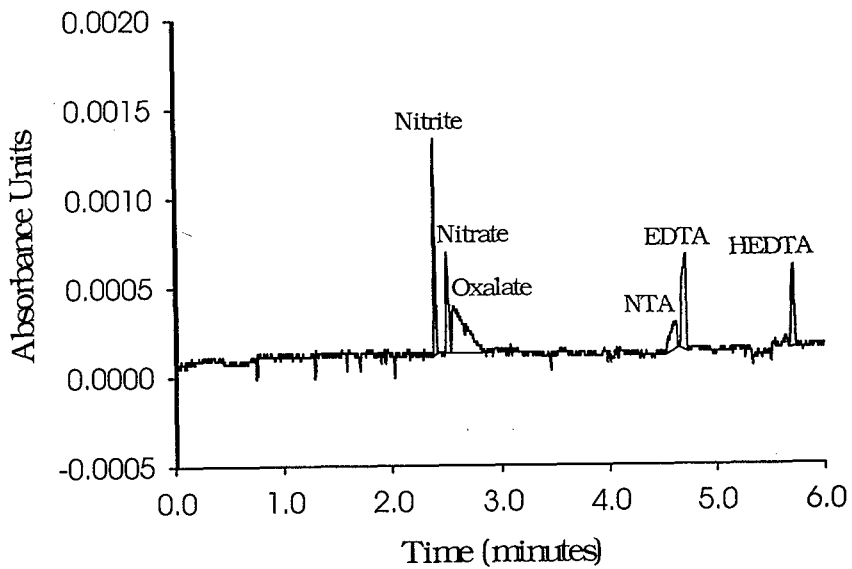


Figure 2. Okengbo et al. Determination of Nitrate and Nitrite in Harford Defense Waste....

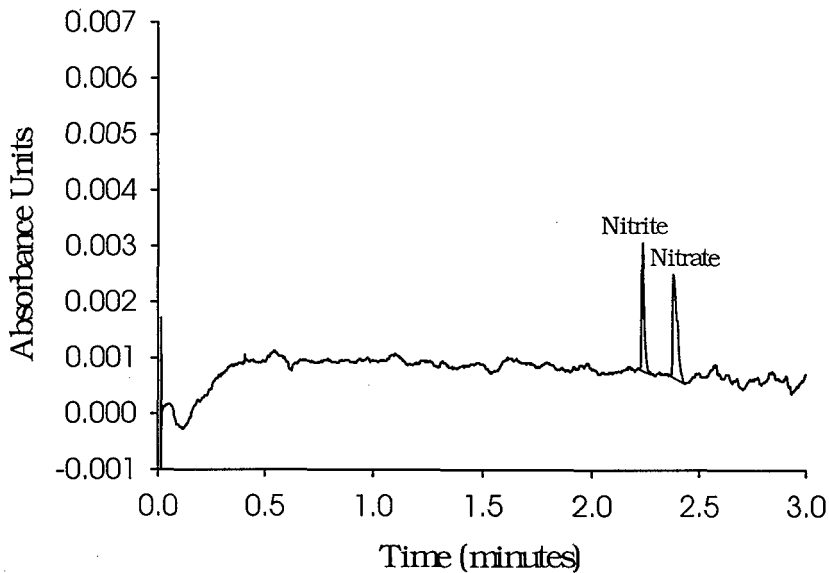


Figure 3. Okemgbo et al. Determination of Nitrate and Nitrite in Harford Defense Waste.....

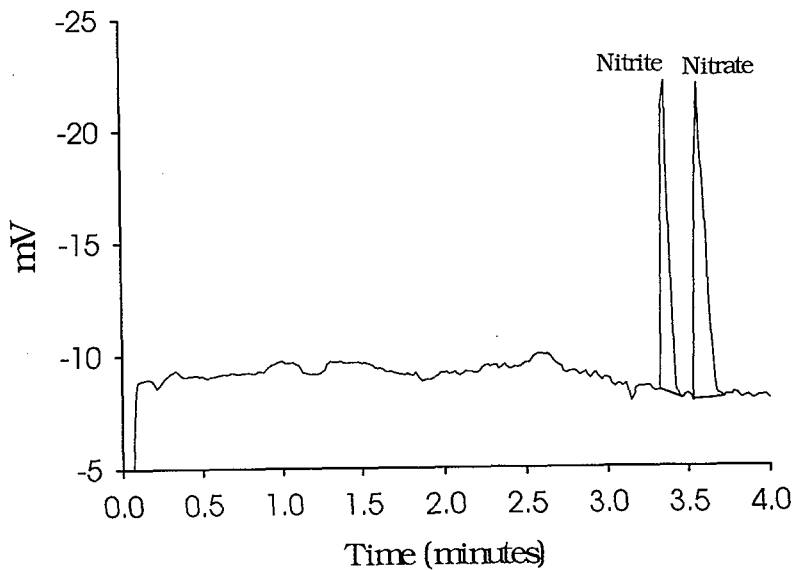
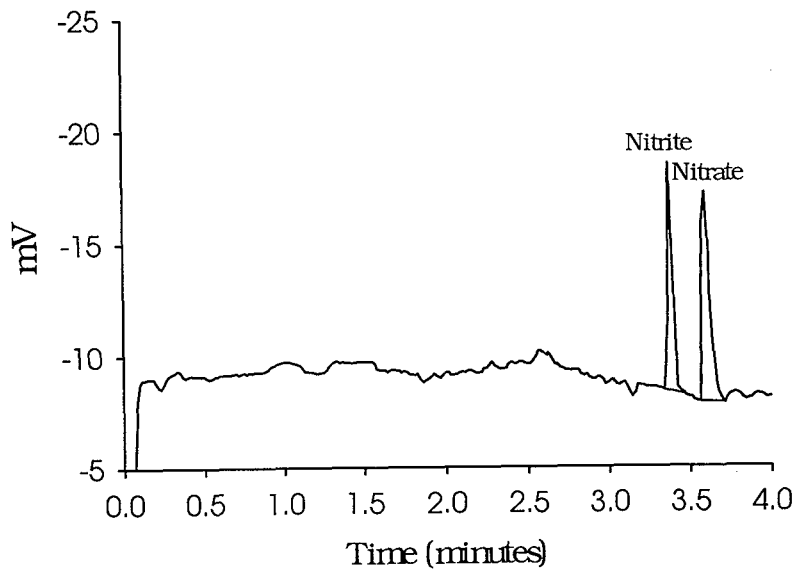


Figure 4. Okemgbo et al. Determination of Nitrate and Nitrite in Hanford Defense Waste....

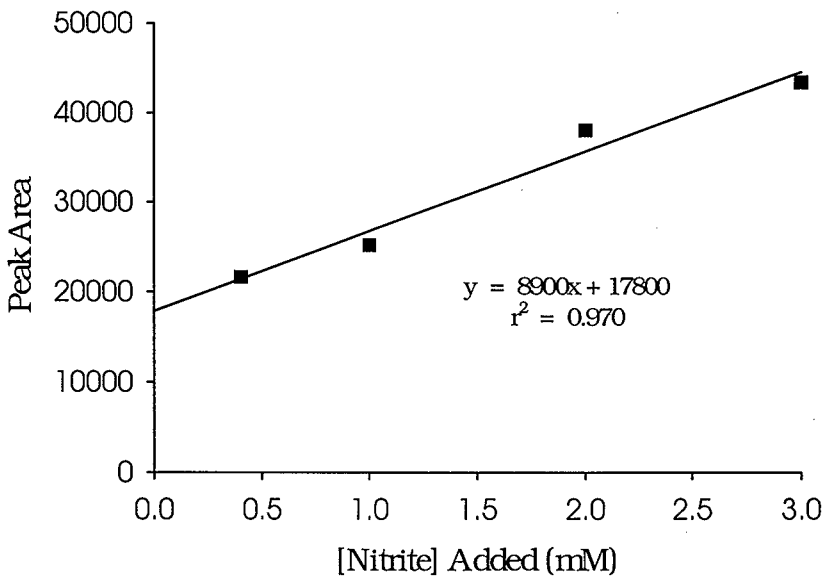
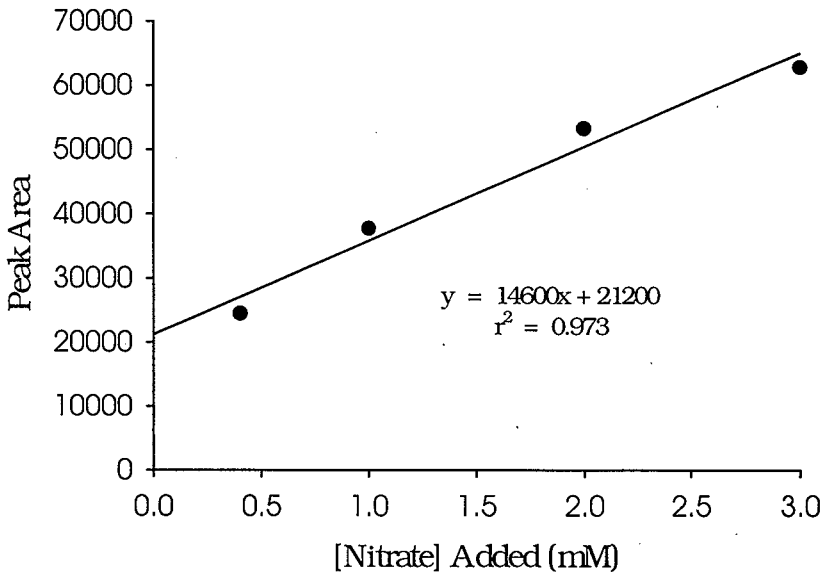


Figure 5. Okemgbo et al.. Determination of Nitrate and Nitrite in Hanford Defense Waste...