Separation Science and Technology
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713708471

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To cite this Article Kabay, N. , Yüksel, M. , Samatya, S. , Arar, Ö. and Yüksel, Ü.(2006) 'Effect of Process Parameters on Separation Performance of Nitrate by Electrodialysis', Separation Science and Technology, 41: 14, 3201 — 3211
To link to this Article: DOI: 10.1080/01496390600851681
URL: http://dx.doi.org/10.1080/01496390600851681

Separation Science and Technology, 41: 3201–3211, 2006

ISSN 0149-6395 print/1520-5754 online
DOI: 10.1080/01496390600851681

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Abstract: The electrodialytic separation of nitrate from water was investigated as a function of feed characteristics such as applied voltage, feed flow rate, and nitrate concentration in the solution. The separation performance was evaluated in terms of percent removal of nitrate, process time, and energy consumptions. TS-1-10 electrodialysis equipment (Tokuyama) was used in experimental studies. During the unsteady state run of the ED system, both inlet and outlet concentrations of nitrate and conductivities of streams were measured at certain time intervals until the current drops to 0.01 A. It was obtained that the percent removal of nitrate from the solution increased when the concentration of nitrate in the feed solution increased. The operation time became shorter when a high potential was applied.

Keywords: Electrodialysis, nitrate, water, separation, ion exchange membranes

INTRODUCTION

Of the many membrane processes available for the separation of ions from solutions, only two, reverse osmosis (RO) and electrodialysis (ED), have reached the practical application stage for the removal of inorganic contaminants from drinking water and wastewater. Both processes remove salts from

Received 18 January 2006, Accepted 2 May 2006

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seawater and brackish water and thus are commonly classified as desalination processes (1).

In recent years, nitrate concentration in ground water has been increasing due to the excess use of fertilizers in agriculture, animal and human wastes, and municipal and industrial wastes (2). Contamination of drinking water by nitrate may result in important health problems. Nitrate causes methemoglobinemia when ingested by infants. Furthermore, nitrosamines are carcinogenic compounds that may be formed from nitrate (2). To protect the public from the adverse effects due to the high nitrate intake, nitrate consumption should be limited and thus some standards have been established (3). According to TSE (Turkish Standards Institute)-266 and WHO, drinking water must contain no more than 50 mg NO_3^-/L and EPA (Environmental Protection Agency) established a maximum contaminant level of 45 mg-NO_3^-/L. The European Community recommends levels of 25 mg NO_3^-/L in drinking water (4, 5).

Several processes are available for the treatment of drinking water with high nitrate concentration. The processes include ion exchange, reverse osmosis, electrodialysis, biological denitrification, catalytic reduction (6–11). A biological denitrification method using degradation of the microorganism offers the possibility of a very specific and selective reduction of nitrate to nitrogen. However, there are some limitations due to the contamination of drinking water with germs and metabolic substances. Because of this, an extensive reconditioning of the drinking water by filtration and germicidal treatment is necessary (10). Reverse osmosis (RO) and electrodialysis (ED) have larger automation possibilities. Lower nitrate level in feed, ease of controls for process parameters, and not needing for extensive post treatment are advantages of RO and ED (10–12). The advantage of a catalytic reduction process is the rapid removal of nitrate from water (13, 14). The electrodialysis combines the advantages of selectivity and low chemical demands (15, 16).

Elmidaoui et al. (2001) performed an electrodialysis operation to remove nitrate from ground water. A preselection of several commercial membranes (AFN, ACS, AMX, ADP, and ADS) was carried out to determine the best anionic exchange membrane for nitrate removal. The selected membrane transports anions in the following order: nitrate > chloride > bicarbonate > sulfate. The results show that a desired product water quality can easily be obtained by electrodialysis.

The disadvantage of electrodialysis is the destiny of the removed concentrated nitrate. Due to increasing environmental concern and constraints, the electrodialysis waste effluent being nitrate rich may not receive consent for discharge into water-courses without further treatment being carried out. To avoid the discharge of the concentrated nitrate in nature, a bioreactor was coupled to the concentrate stream to reduce the nitrate content by biological denitrification. Biological denitrification permanently removes nitrate by reducing it to nitrogen. Studies have been conducted on groundwater containing 850–1100 mg/L of total dissolved solids (TDS) and 80 mg/L of nitrate to remove nitrate by coupling electrodialysis and a bioreactor. By coupling a bioreactor to the brine stream, a significant reduction of nitrate in the brine was observed (4).

The electrodialysis is a widely used electro-membrane process especially for desalination of brackish water and sodium chloride recovery from seawater. The removal of ionic components from an aqueous solution through ion exchange membranes is carried out under the driving force of an electrical field (16). When a direct current potential is applied between two electrodes, the positively charged cations move to the cathode, passing through the negatively charged cation exchange membrane and retained by the positively charged anion exchange membrane. While the negatively charged anions move to the anode, passing through the anion exchange membrane and retained by the cation exchange membrane. At the end, ion concentrations increase in alternate compartments with a simultaneous decrease of ions in other compartments.

In our previous studies, we investigated nitrate removal from aqueous solution and ground water by nitrate selective ion exchange resins (17, 18). It will be useful to compare the results of the ion exchange studies with that of the electrodialysis method for selecting the best process for nitrate removal from ground water. The aim of this work was to investigate the effect of process parameters such as applied voltage, flow rate, and initial feed concentration on nitrate removal from water that dosed with nitrate ions using the electrodialysis method.

EXPERIMENTAL

TS-l-l0 electrodialysis equipment (Tokuyama) was employed for experimental studies. A schematic view of the experimental set-up is shown in Fig. 1. This equipment contains a stack with ten pairs of Neosepta® CMX (cation exchange) and AMX (anion exchange) membranes with the area of 1 dm^2 (Table 1). Three pumps have capacities of max. 1.8 L/min. A rectifier is provided to supply a DC power at constant voltage (max. 18 V) or constant current (max. 3 A). Three solution tanks (each 1 L) are used for holding the diluted (D), the concentrated (C), and the electrode rinse solutions. The electrodes are platinum plated titanium (anode) and stainless steel (cathode) (19). When a direct current potential is applied, the nitrate ions migrate towards the anode. They leave the dilute compartment and move through the anion exchange membrane and are retained by the cation exchange membrane in the concentrate compartment. Samples were taken periodically from dilute and concentrate compartments. The batch tests were carried out with $NaNO_3$ solutions prepared in deionized water at different concentrations (100; 150; 250; 350; 450; 600 mg-NO_3^-/L) and using various potentials (5 and 10 V) and flow rates (0.8, 1.2 and 1.6 L/min). The

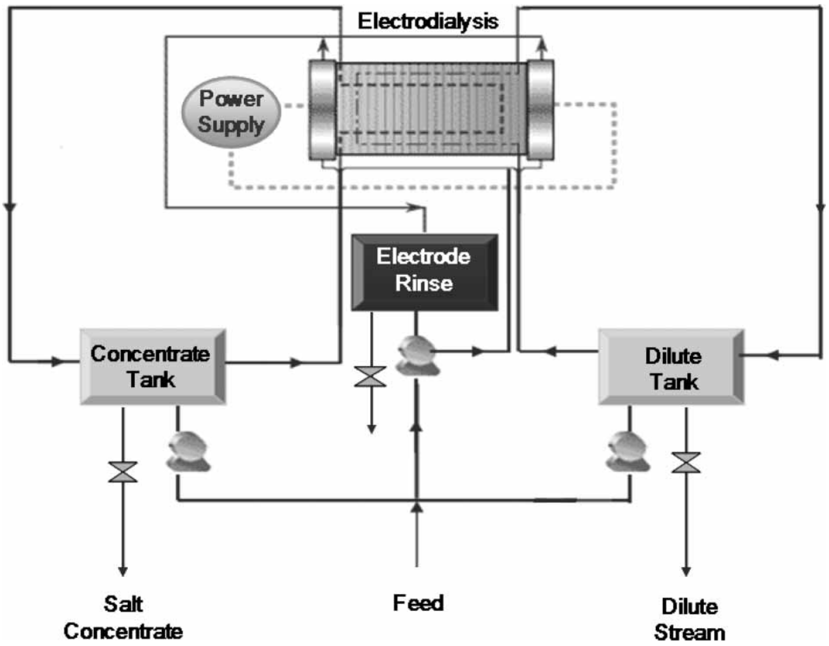

Figure 1. Batch-mode operation of electrodialysis system.

concentration of nitrate was determined by ion chromatography (Model Shimadzu LC 10 A*i*).

RESULTS AND DISCUSSION

Effect of Initial Feed Concentration on Separation Performance

In order to investigate the effect of initial feed concentration on the separation performance of nitrate from water, the experiments were performed using

Table 1. Properties of Neosepta® membranes

	CMX	AMX
Type	Strongly acidic cation permeable	Strongly basic anion permeable
Characteristics	High mechanical strength (Na-form)	High mechanical strength (Cl-form)
Electric resistance (Ω-cm^2)	2.0–3.5	2.0–3.5
Burst strength (kgf/cm^2)	3.5–6.0	4.5–5.5
Thickness (mm)	0.16–0.20	0.14–0.18

various concentrations of nitrate. For each concentration, limiting current measurements were carried out before performance tests. The applied currents were changed keeping the solution concentration constant. The feed and permeate solutions pumped from the same reservoir were circulated through the feed and permeate compartments, respectively, and then recycled to the reservoir. By doing it in this manner, the solution concentration in the feed could be maintained constant at a desired level during the experiments. If the applied current changes, obtained potential values will change linearly until limiting current point. After this point, there is a sharp change in slope. This switching point is determined as limiting current point. The concentrated (C) and dilute (D) streams were monitored for nitrate ion concentration, current, and conductivity values by time. The nitrate ion concentrations of the dilute and the concentrated stream were measured in certain time intervals and presented on charts, in dimensionless concentrations, defined as the ratio of concentration at any time to initial concentration (C/C_o). The histories of nitrate ion concentrations at each of the experiments performed using different initial concentration of nitrate are given in Fig. 2. From Figs. 2 and 3, it is seen that the operation time needed for reaching 0.01A which is the lowest current and lowest concentration of nitrate reached becomes longer as the initial concentration of nitrate in the feed solution increases.

The effect of initial nitrate concentration of the feed on the percent removal of nitrate is shown in Fig. 3. It was obtained that the separation performance increased when the initial concentration of nitrate in the feed solution increased. This is due to the increased mobilities of nitrate ions in a solution having a high ionic strength. On the other hand, the total process time increased with an increasing initial nitrate concentration of the feed.

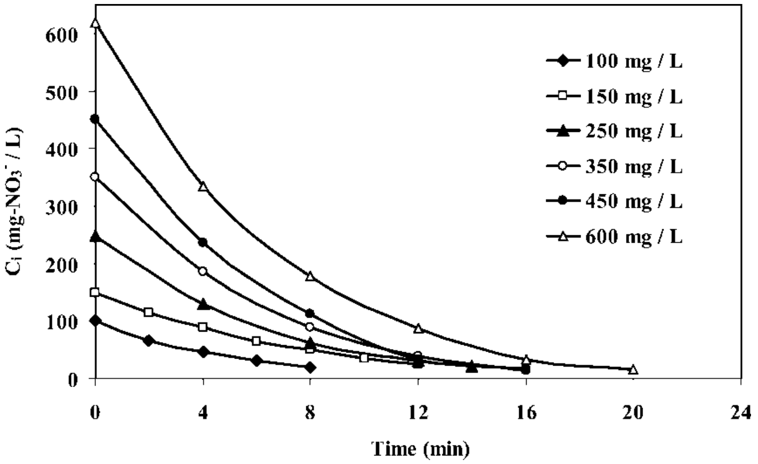

Figure 2. Influence of the initial concentration of nitrate on the removal period during electrodialysis.

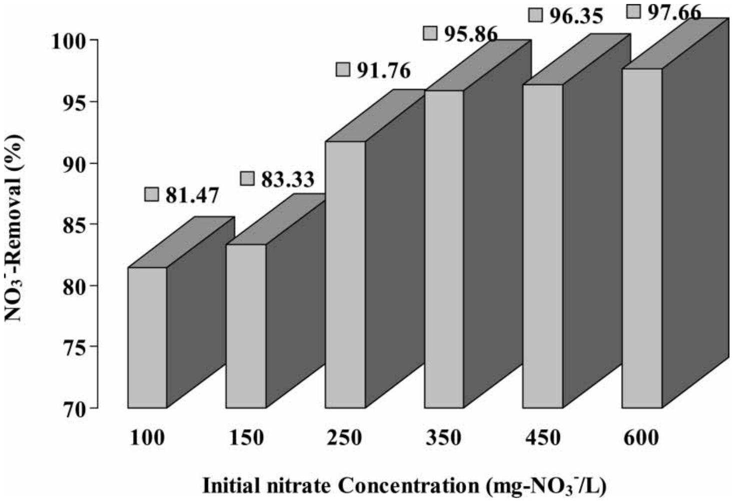

Figure 3. Influence of the initial concentration of nitrate on the percent removal during electrodialysis.

Effect of Voltage on Separation Performance

The effect of voltage on nitrate removal was investigated at 1.6 L/min as constant flow rate and room temperature. The initial feed concentration was 450 mg-NO_3^-/L. The dimensionless form of concentration changes in dilute and concentrate compartments versus time graphs are shown in Fig. 4. As seen from Figs. 4 and 5, the operation time needed for reaching 0.01A, which is the lowest current, becomes much shorter with a high percent removal of nitrate as the potential increases.

Effect of Flow Rate on Separation Performance

The operation was carried out using different feed flow rates (0.8, 1.2, 1.6 L/min) and nitrate solution containing 450 mg-NO_3^-/L and using a voltage of 10.0 V at room temperature. The results are given in Figs. 6 and 7. It was concluded that the that flow rate did not have a meaningful effect on nitrate removal by ED. For the different feed flow rates the operation time is 16 min and the percent of nitrate removal is about 96%.

Specific Power Consumptions

Specific power consumption can be described as the energy needed to treat the unit volume of the solution. The effects of operating parameters (applied

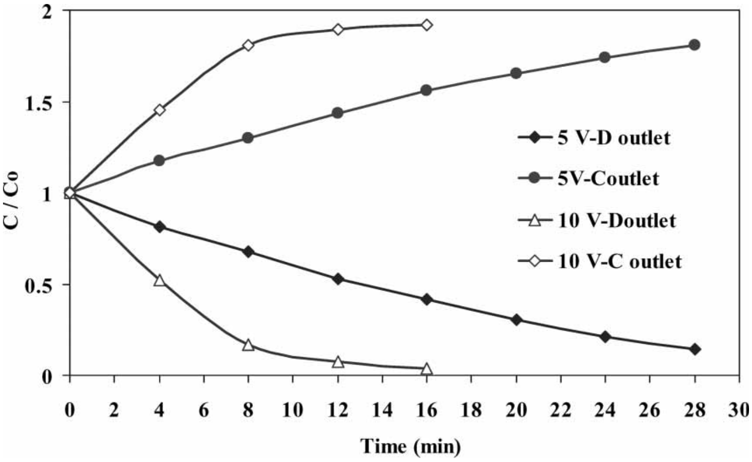

Figure 4. Effect of applied voltage on the separation performance of nitrate.

voltage and feed flow rate) on specific power consumption (SPC) were studied. SPC was calculated using Eq. (1).

$$\mathrm{SPC} = \frac{E\int_0^t I(t)dt}{V_D} \tag{1}$$

where E is the applied potential, I the current, V_D the dilute stream volume, and t the time. The calculated SPC values for each potential and flow rate for

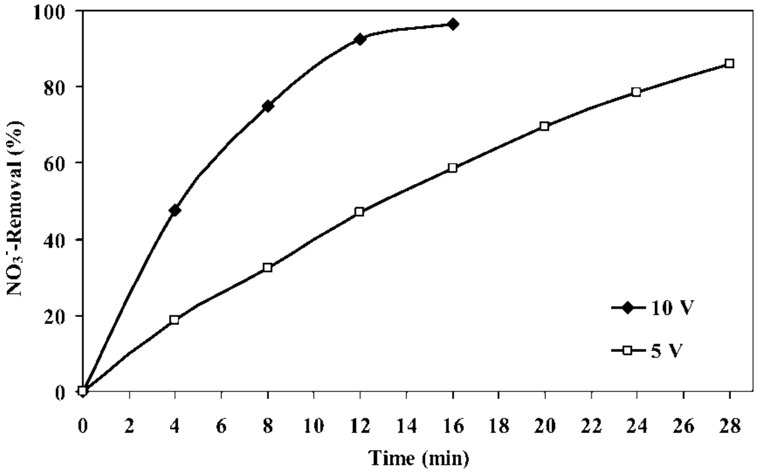

Figure 5. Effect of applied voltage on the separation performance of nitrate.

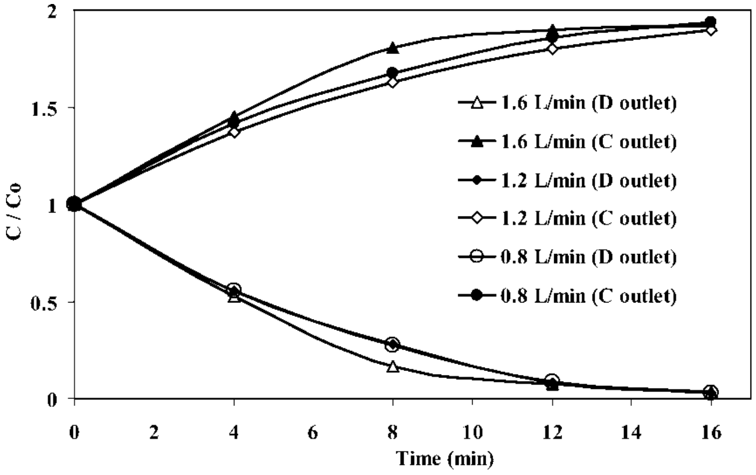

Figure 6. Effect of flow rate on the separation performance of nitrate.

450 mg-NO_3^-/L are given in Table 2 and Fig. 8, respectively. SPC is remarkably affected by the change in applied potential. It was obtained that the specific power consumption increases when a higher voltage is applied for 450 mg-NO_3^-/L. SPC value increased from 0.08 to 0.21 Wh/L when the applied potential increased from 5 to 10 V. On the other hand there is not any remarkable effect of chosen flow rate on the SPC since the operation time and percent removal of nitrate did not change as a function of flow rate. In our previous studies, it was also found that SPC was influenced by the

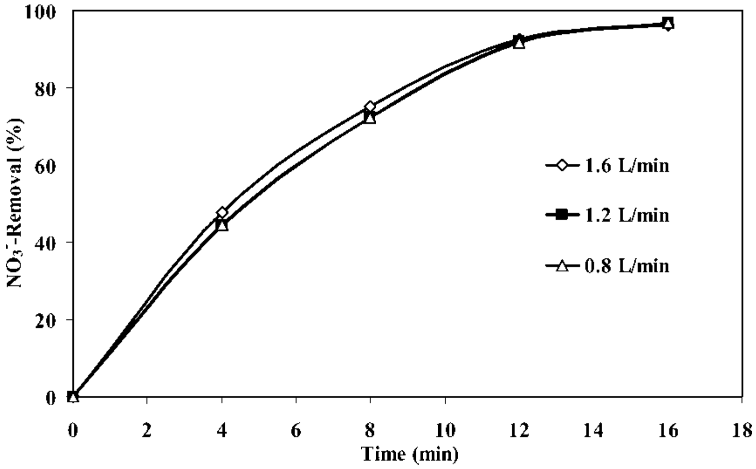

Figure 7. Effect of flow rate on the separation performance of nitrate.

Table 2. Effect of electrical potential on specific power consumption

Applied potential (V)	SPC (Wh/L)
5	0.08
10	0.21

(Solution: 450 mg-NO_3^-/L-$NaNO_3$).

potential applied and SPC was not so sensitive to the change in the flow rate (20, 21).

CONCLUSIONS

Electrodialysis is an effective method for removal of nitrate from water. The total process period was shorter than that of the ion exchange method. In addition, the elution stage of the ion exchange process is eliminated in the electrodialysis method. But percent removal of nitrate with the electrodialysis method decreased when the initial concentration of nitrate is low. This is most probably because of the increased mobilities of ions in the solution having a high ionic strength. But the total process time increased with increasing initial nitrate concentration of the feed. When the applied potential increased the operation time for nitrate removal decreased but the high values of specific power consumption were obtained. The tests were also performed

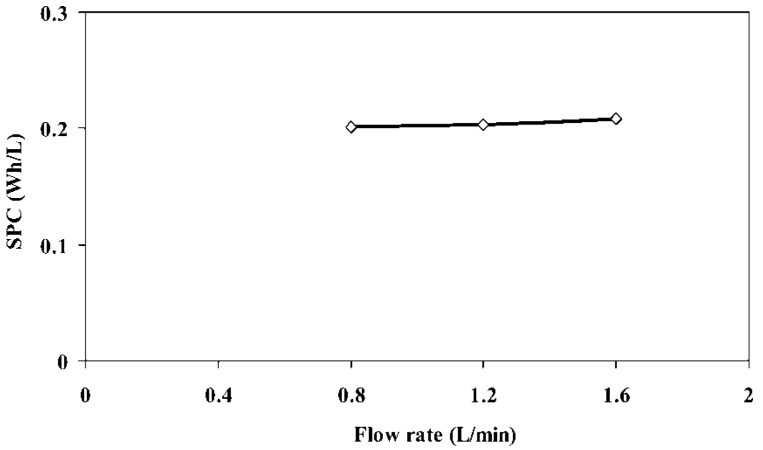

Figure 8. Effect of flow rate (0.8, 1.2, 1.6 L/min) on the specific power consumption of 450 mg-NO_3^-/L-$NaNO_3$.

in the presence of other anionic species such as chloride and sulfate ions along with nitrate ions (22). In addition, the separation of nitrate from ground water samples with high nitrate content was investigated using a hybrid process combining electrodialysis and ion exchange methods. The obtained results have already been submitted for publication elsewhere (22).

ACKNOWLEDGEMENTS

This study was supported by the Ege University Scientific Research Projects (2004 FEN 070 and 2005 FEN 016). We thank Profs. E. Henden and A. Celik for discussions. The kind help of H. Kahveci and O. Ipek during laboratory studies is gratefully acknowledged.

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