

Nitrate Removal from Ground Water: A Review

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Abstract: Nitrate contamination of ground water resources has increased in Asia, Europe, United States, and various other parts of the world. This trend has raised concern as nitrates cause methemoglobinemia and cancer. Several treatment processes can remove nitrates from water with varying degrees of efficiency, cost, and ease of operation. Available technical data, experience, and economics indicate that biological denitrification is more acceptable for nitrate removal than reverse osmosis and ion exchange. This paper reviews the developments in the field of nitrate removal processes which can be effectively used for denitrifying ground water as well as industrial water.

Keywords: Biological denitrification, IEMB - Ion exchange membrane bioreactor, MBR- Membrane bioreactor, ED - Electrodialysis.

Introduction

Nitrate contamination of ground water in various parts of the world has increased alarmingly. Nitrate levels have been increasing in drinking water supplies in most countries¹⁻⁵. Environmental protection agency (EPA) in 1990 indicated that 250,000 water supply sources had maximum contaminant levels (MCL) for nitrate⁶. The World Health Organization (WHO), 2004 found that 30% of the 2,000 sources surveyed in world had more than 24 mg /L.

Nitrate levels have increased due to increased usage of nitrogenous fertilizers, changes in land-use patterns and increased recycling of domestic wastewater⁷⁻⁸. While nitrate is considered to be relatively non-toxic to adults, in infants, NO_3^- is reduced to NO_2^- ; which combines with hemoglobin in the blood to form methamoglobin and leads to a condition commonly known as "blue baby syndrome." Health and Welfare World Guidelines⁹ had established a limit of 45 mgNO_3^-/L .

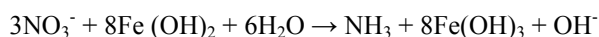
Many technologies for the removal of nitrate from water have been adopted based on scientific developments. A brief overview of all techniques is presented in this paper.

Treatment Techniques

Nitrate is a stable and highly soluble ion with low potential for coprecipitation or adsorption. Thus conventional treatment technologies cannot be used. This paper reviews various techniques in terms of their effectiveness, ease of operation and cost.

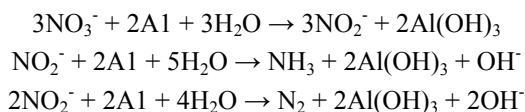
Chemical denitrification

Nitrate reduction can be induced under basic pH according to the following reaction¹⁰:

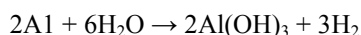


Experimental results showed that a Fe: NO_3^- ratio of about 15: 1 was required in the presence of copper catalyst for the reaction to proceed. This process generated a large quantity of iron sludge and formed ammonia that requires removal by air stripping. The process was associated with high costs.

In chemical denitrification by powdered aluminum ammonia was found to be the principal reaction product (60-95%) at pH of 10.25, which was removed by air stripping¹¹⁻¹². The denitrification was explained on the basis of the following reactions:



Aluminum reacted with water as per the following equation:



It was shown that 1.16 g of aluminum was required for the reduction of 1 g of nitrate. Catalytic reduction of nitrate with Pd and/Cu catalysts was another removal technique¹³. It was found that Pd-Cu combined catalysts at a ratio of 4 can maximize the nitrate reduction into nitrogen; above 80% total nitrate removal efficiency was realized. 84% denitrification efficiency was achieved at ambient temperature and pressure using zero-valent magnesium ($\text{Mg}(0)$)¹⁴ for $\text{Mg}(0):\text{NO}_3^-$ -N molar ratio of 5.8 and pH of 2 .

Reverse osmosis for denitrification

Nitrates could be removed by reverse osmosis cells under pressures ranging from 300 to 1,500 psi to reverse the normal osmotic flow of water. Membranes used were made of cellulose acetate, polyamides and composite materials. Problems associated with reverse osmosis membranes included fouling, compaction and deterioration with time. These problems resulted from deposition of soluble materials, organic matter, suspended and colloidal particles, and other contaminants, pH variations and chlorine exposure; thus the reverse osmosis process required pretreatment.

A 15-gpm spiral wound cellulose acetate reverse osmosis system was tested for 1,000 h and up to 65% nitrate separation was observed for influent NO_3^- concentrations ranging from 18 to 25 mg/L¹⁵.

Reverse osmosis using both polyamide and cellulose triacetate membranes were tested and sulfuric acid and sodium hexametaphosphate were added to feed water to prevent scaling. Polyamide membranes were more effective than cellulose triacetate membranes¹⁶.

A pilot plant using spiral wound modules with composite membranes was operated with a 2 m³/h capacity, at an operating pressure of 14 bar. Influent pretreatment consisted of passing the water through 5 µm cartridge filters and acid dosing to avoid scaling. The results showed high levels of denitrification¹⁷.

Electrodialysis (ED) for denitrification

In ED ions are transferred through membranes from a less concentrated to a concentrated solution by application of direct electric current. ED treated the water by selective removal of undesirable ions through a semi permeable membrane. An electrodialysis system required a supply of pressurized water [50-75 psi (345-578 kPa)] with pretreatment.

In the electrodialysis reversal (EDR) process, the polarity of the electrodes was reversed two to four times an hour to alter the direction of ion movement. The EDR process reduced scaling and chemical usage compared with conventional ED and was used for the production of drinking water from nitrate rich water. The nitrate removal efficiency of ED and RO processes was almost the same¹⁸.

A selective nitrate removal process was developed, NitRem, based on ED¹⁹. The process was effective in reducing nitrate concentrations from 50 mg/L or more to less than 25 mg/L. An attractive feature of the process was that it included the removal of nitrate without the addition of any chemicals.

It was observed that for nitrate reduction from 100 to 50 mg NO₃⁻/L the costs of EDR and RO were about the same¹⁸. It was expected capital and maintenance cost of membrane filtration to reduce with time²⁰.

Catalytic denitrification

A catalytic process was developed for the removal of nitrite and nitrate from water²¹. Palladium - alumina catalysts were effective in reducing nitrite to nitrogen (98%) and ammonia in the presence of hydrogen. The lead (5%), copper (1.25%), Al₂O₃, catalyst were found to completely remove nitrate from water having an initial nitrate concentration of 100 mg/L. The reaction was completed in 50 min. The process operated effectively at a temperature of 10°C and pH 6-8.

Electrocatalytic reduction for denitrification

An electrocatalytic reduction process was used to remove NO₃⁻ from groundwater²². A commercially available carbon cloth with a 30% surface coated Rh (rhodium) (1 µg cm⁻¹) was tested at an applied potential of -1.5 V versus standard calomel electrode (SCE) with a Pt auxiliary electrode. The results suggested that electrocatalysis reduced NO₃⁻ concentrations in groundwater from 73 to 39 mg/L on a timescale range of 40-60 min.

Ion exchange process

The ion exchange process involved passage of nitrate water through a resin bed containing strong base anion (SBA) exchange resins on which nitrate ions were exchanged for chloride or bicarbonate ions until the resin exhausted. The exhausted resin was regenerated using a concentrated solution of sodium chloride or sodium bicarbonate²³.

Fifteen ion exchange plants used in the united states²⁴ reduced nitrate from 18 to 6.8 mg/L²⁵. Addition of bentonite clay to the backwash water helped restore the resin, which was regenerated with 1 N NaOH and HCl. Nitrate removal capacity of the resins was reduced by

Silica and iron precipitates²⁶. An ion exchange process was tested for the removal of nitrate from ground water containing 16-23 mg NO₃⁻/L at flow rates of up to 31.5 L/m². s²⁷.

Treatment of sulfate water with resins is difficult as the nitrate removal capacity of the resin is reduced by the sulfate ions. It was observed that sulfate selectivity was reduced by increasing the distance between ion-exchange sites and nitrate selectivity can be increased by increasing the matrix and functional group hydrophobicity²⁸. Triethyl amine resins showed an increase in the bed life by 62% when treating water containing 1.5 meq/L nitrate and 6.5 meq/L sulfate²⁷. Regenerant usage decreased by 25-50%, thus the operating cost of the ion exchange process reduced.

The 0.043 m³/s ion exchange plant in California (USA) used a non-nitrate-selective resin, which was regenerated using a saturated brine solution²⁹. The total treatment cost was reported to be 24.2 cents/1,000 gal. The energy requirements of the plant were 0.244 kWh per 1,000 gal²⁷.

A pilot-scale study was conducted to evaluate nitrate removal from drinking water by ion exchange, reverse osmosis (RO) and electrodialysis (ED)¹⁶. The raw water contained 18-25 mg/L, 43 mg/L sulfate and 530 mg/L total dissolved solids (TDS). All processes were able to reduce nitrate concentration below 10 mg/L.

Ion exchange process was found to be five times more economical in comparison to RO process²⁹. A process was developed in which regeneration and exhaustion were performed in the same direction and reduced nitrate concentrations from 15.8 to 5.7 mg/L³⁰. The Carbon dioxide regenerated ion exchange resins (CARIX) process for removing nitrate, sulfate, and hardness from water was based on ion exchange principles³¹⁻³³. The exhausted exchange resins were regenerated through contact with a concentrated carbon dioxide solution. A CARIX pilot plant (0.047 m³/s) was constructed in Germany which was effective in reducing nitrate concentrations from 90 mg/L to less than 5.7 mg/L. The consumption of carbon dioxide amounted to 0.35 kg/m³ of treated water.

Denitrification using a membrane bioreactor

Immersed heterotrophic membrane bioreactor (MBR) produced high quality product water³⁴ when NO₃⁻ contaminated water was made to flow through the lumen of tubular microporous membranes. NO₃⁻ diffused through the membrane pores. Denitrification took place on the shell side of the membranes³⁵. The MBR achieved over 99% NO₃⁻ removal at an influent concentration of 200 mg NO₃⁻/L.

Combined ion exchange & membrane bioreactor for denitrification

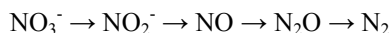
The IEMB concept combines dialysis and simultaneous biological degradation of nitrate in small concentrations³⁶⁻³⁷. The IEMB process operated with hydraulic retention times ranging from 1.4 to 8.3 h in the water compartment, proved to remove nitrates effectively, while preserving the water composition with respect to other ions, thus avoiding secondary contamination of the treated water.

Denitrification using nanofiltration

During the last decade, nano filtration (NF) made a breakthrough in drinking water production for the removal of nitrate³⁸. For the removal of nitrates the membranes NF70, NF45, UTC-20, and UTC-60 have been experimentally studied. The results showed that only a small fraction of nitrate was removed for most membranes, except for NF70 where a 76% nitrate removal was obtained.

Biological denitrification

Many bacteria belonging to different genera can grow anaerobically by reducing ionic nitrogenous oxides to gaseous products. Nitrates or nitrites served as the terminal electron acceptors instead of oxygen and resulted in generation of ATP³⁹. Such denitrification was dissimilatory nitrate reduction⁴⁰. When electrons are transferred from the donor to the acceptor, the organism gains energy which was applied for the synthesis of a new cell mass and the maintenance of the existing cellmass. The enzymes associated with denitrification are synthesized under anaerobic or partially aerobic conditions⁴¹. Nitrate reduction to nitrogen gas occurred as:



Each step was catalyzed by an enzyme system. Dissimilatory reduction of nitrate to nitrite was important for most bacteria, since the process involved energy conservation by increased substrate level phosphorylation reaction⁴².

Since denitrification was a respiratory process, an oxidisable substrate was needed as an energy source. Limitation of biological denitrification was possible bacterial contamination and presence of residual organics⁴³⁻⁴⁷.

Heterotrophic denitrification

Most denitrifying bacteria are heterotrophic and utilize complex organic substances as oxidisable substrates such as methanol, ethanol, methane, carbon monoxide, and acetic acid⁴⁸⁻⁵² for the conversion of nitrate to nitrogen. The cell formula $\text{C}_5\text{H}_7\text{NO}_2$ suggested by Hoover was used and the stoichiometric of various organic carbon substrates required for nitrate dissimilation are as listed in Table 1⁵³.

Table 1. Stoichiometric relationships of heterotrophic denitrification with various carbonaceous substrates⁵⁴⁻⁶².

S.N.	Substrate	Stoichiometric equation
1.	Ethanol	$* 5 \text{ C}_2\text{H}_5\text{OH} + 12 \text{ NO}_3^- \rightarrow 10 \text{ HCO}_3^- + 2 \text{ OH}^- + 9 \text{ H}_2\text{O} + 6 \text{ N}_2$ $* 0.613 \text{ C}_2\text{H}_5\text{OH} + \text{NO}_2^- \rightarrow 0.102 \text{ C}_5\text{H}_7\text{NO}_2 + 0.714 \text{ CO}_2 + 0.286 \text{ OH}^- + 0.980 \text{ H}_2\text{O} + 0.449 \text{ N}_2$
2.	Acetic acid	$* 5 \text{ CH}_3\text{COOH} + 8 \text{ NO}_3^- \rightarrow 8 \text{ HCO}_3^- + 2 \text{ CO}_2 + 6 \text{ H}_2\text{O} + \text{N}_2$ $* 0.819 \text{ CH}_3\text{COOH} + \text{NO}_3^- \rightarrow 0.068 \text{ C}_5\text{H}_7\text{NO}_2 + \text{HCO}_3^- + 0.301 \text{ CO}_2 + 0.902 \text{ H}_2\text{O} + 0.466 \text{ N}_2$
3.	Propanol	$0.278 \text{ C}_3\text{H}_7\text{OH} + \text{NO}_3^- \rightarrow 0.5 \text{ N}_2 + 0.833 \text{ CO}_2 + 0.611 \text{ H}_2\text{O} + \text{N}_2$
4.	Cellulose	$5(\text{C}_6\text{H}_{10}\text{O}_5)_n + 24n \text{ NO}_3^- \rightarrow 6n \text{ CO}_2 + 13n \text{ H}_2\text{O} + 12n \text{ N}_2 + 24n \text{ HCO}_3^-$
5.	Butanol	$0.208 \text{ C}_4\text{H}_9\text{OH} + \text{NO}_3^- \rightarrow 0.5 \text{ N}_2 + 0.833 \text{ CO}_2 + 0.542 \text{ H}_2\text{O} + \text{OH}^-$
6.	Aromatic hydrocarbon	$\text{C}_{61}\text{H}_{67} + 62.2 \text{ H}^+ + 62.2 \text{ NO}_3^- \rightarrow 31.1 \text{ N}_2 + 61 \text{ CO}_2 + 64.6 \text{ H}_2\text{O}$
7.	Organic matter	$\text{C}_5\text{H}_9\text{NO} + 3.36 \text{ NO}_3^- + 3.92 \text{ H}^+ \rightarrow 1.68 \text{ N}_2 + 0.36 \text{ C}_5\text{H}_7\text{NO}_2 + 3.2 \text{ CO}_2 + 3.92 \text{ H}_2\text{O} + 0.64 \text{ NH}_4^+$
8.	Methane	$5\text{CH}_4 + 8\text{NO}_3^- + 8\text{H}^+ \rightarrow 4\text{N}_2 + 5\text{CO}_2 + 14\text{H}_2\text{O}$
9.	Glucose	$\text{C}_6\text{H}_{12}\text{O}_6 + 2.8 \text{ NO}_3^- + 0.5 \text{ NH}_4^+ + 2.3 \text{ H}^+ \rightarrow 0.5 \text{ C}_5\text{H}_7\text{NO}_2 + 1.4 \text{ N}_2 + 3.5 \text{ CO}_2 + 6.4 \text{ H}_2\text{O}$

Pilot scale studies using heterotrophic denitrification were conducted using packed and fluidized columns⁶³. The reactors required a start-up period of two weeks to establish sufficient bacterial populations. The highest denitrification rate per unit reactor volume was observed for the fluidized sand bed (160 g N/m³.h at 10°C) and lowest for the packed bed reactors (12 g N/m³.h at 10°C). Nitrate concentrations were reduced to approximately 45 mg/L⁶⁴⁻⁶⁵.

Denitrification by immobilized *Pseudomonas denitrificans* cells was studied using a sodium alginate polymer and ethanol as the carbon source⁶⁶. The nitrate concentration reduced from 104 to 0.1 mg/L. The limitations of the process being that the low rate of diffusion of substrate and reaction products through the alginate matrix and the short life span of the alginate matrix. To overcome these problems⁶⁷⁻⁶⁸ a membrane-immobilized biofilm reactor was developed in which denitrifying bacteria and carbon energy source were segregated from the water to be treated.

Autotrophic denitrification

Some bacteria from the genera *Paracoccus*, *Thiobacillus*, *Thiosphaera* and others can accomplish denitrification autotrophically using hydrogen or various reduced sulfur compounds such as S⁰, S²⁻, S₂O₃²⁻, S₄O₆²⁻, or SO₃²⁻ as energy sources. Bacteria from the genera *Ferrobacillus*, *Gallionella*, *Leptothrix* and *Sphaerotilus* can utilize ferrous iron as an energy source for autotrophic denitrification. Under autotrophic growth conditions, carbon dioxide or bicarbonate was used as carbon source for microbial cell synthesis. Stoichiometric equations of autotrophic denitrification with various energy sources are listed in Table 2.

Table 2. Stoichiometric relationships of autotrophic denitrification with various electron donors.

S.N.	Electron donor	Stoichiometric equation
1.	Hydrogen	$2\text{NO}_3^- + 5\text{H}_2 \rightarrow \text{N}_2 + 4\text{H}_2\text{O} + 2\text{OH}^-$
2.	Ferrous iron	$\text{NO}_3^- + 5\text{Fe}^{2+} \rightarrow 0.5\text{N}_2 + 5\text{Fe}(\text{OH})_2 + 9\text{H}^+$
3.	Sulfide	$14\text{NO}_3^- + 5\text{FeS}_2 + 4\text{H}^+ \rightarrow 7\text{N}_2 + 10\text{SO}_4^{2-} + 5\text{Fe}^{2+} + 2\text{H}_2\text{O}$
4.	Thiosulfate	$* 8\text{NO}_3^- + 5\text{S}_2\text{O}_3^{2-} + \text{H}_2\text{O} \rightarrow 4\text{N}_2 + 10\text{SO}_4^{2-} + 2\text{H}^+$ $* 0.141\text{NO}_3^- + 0.125\text{S}_2\text{O}_3^{2-} + 0.0643\text{CO}_2 + 0.1\text{H}_2\text{O} \rightarrow 0.0129\text{C}_5\text{H}_7\text{NO}_2 + 0.064\text{N}_2 + 0.25\text{SO}_4^{2-} + 0.109\text{H}^+$
5.	Elemental sulfur	$10\text{NO}_3^- + 11\text{S}^0 + 4.1\text{HCO}_3^- + 0.5\text{CO}_2 + 1.71\text{NH}_4^+ + 2.54\text{H}_2\text{O} \rightarrow 0.92\text{C}_5\text{H}_7\text{NO}_2 + 11\text{SO}_4^{2-} + 5.4\text{N}_2 + 9.62\text{H}^+$

Thiobacillus denitrificans was used to reduce nitrate concentrations from 24 to 1 mg/L in packed bed reactors using elemental sulfur as an electron source⁶⁹. Schippers described denitrification using sulfur limestone filtration⁷⁰. Lewandowski encapsulated autotrophic denitrifiers in calcium alginate beads containing sulfur and calcium carbonate for autotrophic denitrification⁷¹. Nitrate concentrations were reduced from 27 to 6 mg/L in seven hours.

A process known as DENITROPUR was developed using hydrogenotrophic denitrification⁷². The process does not require the post treatment that heterotrophic denitrification requires. The reproduction rate of autotrophic bacteria is low, therefore, less biomass sludge was generated.

Conclusion

The three treatment processes that have been applied full-scale for nitrate removal include ion exchange, biological de-nitrification and reverse osmosis. The other methods discussed have limited potential for full-scale application. The literature indicates wider application of heterotrophic biological denitrification in comparison with autotrophic denitrification. The

technical and economic feasibility of heterotrophic denitrification has been confirmed at full scale in some European countries. The autotrophic reaction rate is low, therefore reactors with large volumes are required to achieve sufficient residence time for denitrification, thus increasing capital costs.

Removal of nitrates from drinking water is an important and developing area of research. Technology development has occurred in this area, but further optimization of current technologies is required. Biological denitrification reactor operation in regard to microbiological characteristics of biologically denitrified water demands attention. RO and ED performance data for nitrate removal is limited and the impact of rapid advances in these technologies should be examined.

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