REVIEW

Removal Techniques of Nitrate from Water

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Nitrate pollution of water is a major environmental problem all over the world. Consequently, numerous techniques have been developed to remove or reduction of nitrate in water. A review of removal techniques of nitrate from water has been carried out in this paper. Recent literatures related to various techniques including reverse osmosis, ion exchange, electrodialysis, biological denitrification, chemical denitrification, adsorption methods using different adsorbents like carbon base, agricultural waste, natural materials are systematically review to assess their performance. The paper reveals that adsorption could be the most promising technique of removal of nitrate from water in near future.

Keywords: Nitrate reduction, Reverse osmosis, Ion exchange, Electrodialysis, Denitrification, Adsorption.

INTRODUCTION

More than 70 % of the area of earth is covered by water. Therefore, apparently it seems that earth is abundant with water. However, availability of fresh water is a major problem across the world. Only 3 % of total water on the earth is useable. Out of this 3 %, nearly 70 % of fresh water is frozen, 29 % is present as soil moisture or lies in deep underground aquifers as groundwater and less than 1 % of the world's fresh water is in the lakes and rivers¹. However, it is regularly renewed by rain and snowfall and therefore, available on a sustainable basis. Safe drinking water is essential to humans and other life. Ever increasing demand increasing due to rapid population growth and dwindling supply of fresh water due to gradual destruct of fresh water resources have caused that water supply-demand balance in critical in many regions of the world. According to European Union³ water framework directives, a “pressure” is explained as an action that caused by human activities that have negative influence on water. The negative activities that cause the degradation of fresh water can be categorized into five types, such as point contamination sources, diffuse contamination sources, groundwater over exploitation, artificial recharge and seawater intrusion³. Among them, diffuse contamination due to human activities is a major cause of water pollution. Nitrate pollution of water due to intensive agricultural activities has become a major environmental problem since 1970s⁴. Nitrate is highly soluble in water³ and does not

readily bind to the soil causing it to be highly susceptible to leaching⁶. There are several potential sources of nitrate, including animal wastes, septic tanks, municipal wastewater treatment systems and decaying plant debris. However, nitrogen-enriched fertilizers for farming are considered as the main source of nitrate pollution in the environment⁷.

To reduce the health hazard, a nitrate standard of 50 mg/L nitrate- NO_3^- or 10 mg/L nitrate-N in drinking water has been set by WHO⁸ and US EPA⁹. However, it has been reported that nitrate concentration in drinking water has exceeded from the maximum acceptable concentration in many parts of the globe¹⁰⁻¹². Therefore, controlling nitrate level in potable water below the standard level has become a major concern. For instance, Defra¹³ estimated that UK government has been paid around £ G58 million per year by 2010 to keep the nitrate below permissible level in drinking water.

Numerous methods have been developed to reduce the nitrate concentration in water like reverse osmosis, ion exchange, electrodialysis, biological denitrification, chemical denitrification and adsorption¹⁴⁻²¹. All the methods have their inherent advantages and disadvantages. In the present study all the available methods are reviewed to assess their efficiency and economic suitability in removing nitrate from water. For this purpose, the recent literatures related to various nitrate removal techniques are systematically reviewed. It is expected that the study will help to enrich the knowledge of ongoing research on removal of nitrate from water.

Popular methods of removal of nitrate: There are number of popular and conventional treatment technologies available for reduction or removal of nitrate in water such as reverse osmosis, ion exchange and electrodialysis, biological denitrification²²⁻²⁷. These conventional methods are discussed below.

Reverse osmosis: Reverse osmosis (RO) was considered as *ex-* and *in situ* applications for the reduction of nitrate from drinking water^{14,28,29}. Reverse osmosis can be used to reduce a wide range of contaminants simultaneously²⁰. The governing basic in reverse osmosis method is selectional passing. In other words, just water molecules are permitted to pass and leaving other ions by a semi-permeable membrane³⁰. A single membrane of reverse osmosis can be uses to remove various contaminants *viz.* chloride, fluoride, sodium, *etc.*³¹. Contaminants are impeded by using pressure forces the water through membrane; therefore, solution concentration has direct relation to required pressure for feed water²⁰. The solute removal efficiency in reverse osmosis method depends on initial solute concentration³², however, the optimum nitrate reduction efficiency is 93 %¹⁵. Some different variables *viz.* initial nitrate concentration³² and the material of membrane³⁰ have negative effect on nitrate removal efficiency. The increasing of initial nitrate concentration from 25 to 200 mg/L causes decrease of removal efficiency³² from 93.5 to 82.5 %.

Ion exchange: The governing basic in ion exchange (IX) method is generally substitute chloride anions with not desirable compound such as contaminants, total dissolve solids and so on. The process of nitrate reduction with ion exchange is similar to water softening, but the difference between them is polymer resin²⁰. Nitrate pollutant from water is replaced with chloride by resin¹⁵. Eqn. 1 shows the reaction between nitrate pollutant and chloride on resin surface²⁰.

Exhausted resin of chloride is regenerated with a salt (NaCl) rich brine solution which washes out the nitrate from resin surface and replaces with chloride again. Eqn. 2 shows the reaction between nitrate on resin surface and chloride from brine²⁰.

Various factors such as initial nitrate concentration, total dissolve solids, regenerator³², type of resin and other ions like sulfate³⁰ are reported that nitrate removal efficiency for initial nitrate concentration 25 mg/L NO₃-N and TDS 400 mg/L was 99.8 %³² and nitrate removal efficiency for initial nitrate concentration 18-25 mg/L NO₃-N and TDS 530 mg/L and sulfate concentration 43 mg/L were between 45-60 %³³.

Electro dialysis: Electric current is the governing basic in electro-dialysis method (ED) to remove or to reduce pollutants from water. The system consists of cathodic and anionic semi-permeable membrane and two electrodes. Electric current causes anions and cations in solution to move toward anode and cathode, respectively. During the experiment, ions trap in waste flow and leaving de-mineralized water behind²⁰.

Reduction of inorganic pollutants like radium³⁴, perchlorate³⁵, bromide³⁶, fluoride¹⁹, iron and manganese³⁷ and nitrate³⁸ in drinking water were some applications of electro-dialysis. Pollutants removal efficiency of this method varies from 50 to 99 % depending on pollution level³⁹. For instance, electro-dialysis method reduced nitrate pollution from 42 mg/L NO₃-N to less than 10 mg/L NO₃-N, in other words its efficiency is 76 %⁴⁰.

Chemical dinitrification: Various chemical compounds have been used to reduce nitrate form contaminated water. Among those, the chemical compounds like zero-valent iron^{21,41,42}, nano-alumina¹⁷ and magnesium^{16,43} are most common. Description of different chemical dinitrification is given below.

Zero-valent iron: Since 1990s, contaminated ground-water with nitrate is treated using zero-valent iron as permeable reactive barrier⁴⁴. Zero valent iron and nitrate have reaction together and redox is their reaction. The outputs of reactions are nitrite, nitrogen and ammonium which cause nitrate reduction⁴⁵. A number of researchers including Flis⁴⁶, Huang & Zhang⁴⁷ and Su & Puls⁴⁸ mentioned different equations for nitrate reduction. For example, the eqns. 3 and 4 used to show the reaction between zero-valent iron and nitrate⁴⁹.

The range of nitrate removal efficiency in the solution depends on number of factors⁴¹ such as, pH⁵⁰, nanoparticles size⁴⁴, method used of produce of nanoparticles⁴⁵, the dose of nanoparticles to nitrate⁵¹ and contact time between particles and pollution²¹. For instance, the nitrate removal efficiency is 95 % at pH 4, contact time 60 min, initial nitrate concentration 50 mg/L and nanoparticles concentration 15 g/L²¹.

Zero-valent magnesium: Magnesium is one of metals which have been used to nitrate removal in solution for last many years.

It transforms the nitrate to nitrite and ammonia⁵². Eqns. 5-7 show the nitrate transform to nitrite and ammonium⁴³.

Various factors affect the efficiency of nitrate reduction in aqueous phase using zero-valent magnesium¹⁶. Equilibrium condition happen in short time and the rate of denitrification depend on contact time, initial nitrate concentration, the ratio of magnesium to nitrate and magnesium dose⁴³. For example, the nitrate removal efficiency is 93 % at pH 2, contact time 20 min, initial nitrate concentration 50 mg/L and nanoparticles concentration 2 g/L¹⁶.

Zero-valent alumina: Contaminated water with nitrate was treated using alumina powder by Murphy⁵³ and, Bhatnagar & Sillanpaa¹⁷. Principal product of nitrate reduction by this method is ammonia with some nitrite and nitrogen⁵². Eqns. 8-10 show nitrate transformation by zero-valent alumina.

In contrast to magnesium and iron nitrate reduction, this method reaches a maximum at pH 10.5^{52,53}. Nitrate removal efficiency by this method is also depends on various factors which effect nitrate reduction between 60-95 %⁵³.

Biological denitrification: Nitrate reduction to harmless nitrogen in special absence oxygen condition is the governing principle of biological denitrification (BD)²⁰. Nitrate is used as terminal electron acceptor in the respiratory process by bacteria when oxygen is absent²⁶. The reduction of nitrate to nitrogen are done in four steps as shown in eqn. 11⁵⁴.

Facultative anaerobes carry out biological denitrification which can be organic or inorganic^{55,56}. Heterotrophs and autotrophs are two major groups of denitrifiers. The difference between them is the way to get substance as energy source which heterotrophs need to organic and autotrophs need to inorganic substance⁵⁷. Denitrification rate depends on the type and concentration of carbon^{58,59} as well as carbon to nitrogen (C/N) ratio⁵⁶. Nitrate removal reported the efficiency of biological denitrification method with nitrate-degrading bacteria *Pseudomonas* and carbon source starch 1 % for initial nitrate concentration 500 and 460 mg/L are 86 and 89 %, respectively⁶⁰.

Summary of popular methods: According to previous reports^{22,25,26} ion exchange, electrodialysis and reverse osmosis are the most valuable methods to reduce nitrate in water. However, they have number of disadvantages and limitations which are discussed below.

(1) Nitrate removal using reverse osmosis suffers from fouling, compaction and deterioration with time³⁰, energy demand, waste disposal^{20,61}, concentrated waste brine and high cost^{23,24}. Furthermore, its efficiency is not satisfactory if the feed water quality is very low²⁰.

(2) Nitrate removal with the ion-exchange method is not a suitable for the water with high TDS⁴⁰. Ion-exchange method may release nitrate in environment during resin regeneration²⁴.

(3) Electrodialysis method has number of limitations including high system complexity, energy demand, waste disposal^{20,61}, difficult operation and maintain in rural areas⁴⁰.

(4) Biological denitrifications are potentially more complex. This type of method suffers from number of disadvantages such as high monitoring needs, sensitivity to environmental conditions and risk of nitrite formation^{20,61}.

(5) Ammonia and nitrogen gas are undesired products of using metallic zero valent methods⁶².

Comparison of various properties *viz.*, reliability, maintenance, cost, operation, *etc.* of different methods is shown in Table-1.

Limitations of different conventional methods led to more research on removal techniques of nitrate. New approach based on adsorption technique is proposed to reduce or omit those limitations in conventional treatment methods.

Adsorption: Convenience, no difficulty of process, low cost and ease of plan are the main advantages of adsorption method for water treatment¹⁷. Various inorganic contaminants *viz.*, fluoride⁶³, nitrate⁶⁴, bromate⁶⁵, perchlorate⁶⁶ *etc.* are removed or reduced using different type of adsorbents. Adsorbents can be categorized in to six groups namely, carbon base, natural, agricultural wastes, industrial wastes, biosorbent and miscellaneous⁶.

Carbon base adsorbents: Carbon base group adsorbents includes powdered activated carbon, carbon cloth, carbon nanotubes, commercial activated carbon, zinc chloride treated granular activated carbon and iron oxide-dispersed activated carbon fiber. Although different types of organic contaminants are removed sufficiently by using activated carbon, efficiency of activated carbon in ionic adsorption is poor. Modified carbon cloth used for nitrate removal and reported that adsorption of modified carbon cloth with sulfuric acid for nitrate was 2.03 mmol/g which was too high compared to non treated carbon^{6,67}. Khani and Mirzaei⁶⁸ used powdered activated carbon and carbon nanotubes for pollution reduction of nitrate and reported the adsorptions were 10 and 25 mmol/g, respectively. Non-activated granular carbon, produce by coconut shells and activated with zinc chloride showed reduction of nitrate with adsorption capacity 1.7 and 10.2 mg/L, respectively⁶⁹. The adsorption of nitrate has been studied using bamboo powder charcoal by⁷⁰ and reported upto 1.25 mg/g of nitrate removal.

Natural adsorbents: Natural adsorbents include clay, zeolite and sepiolite⁶ and leonardite⁷¹. Contaminants are reduced through ion exchange or adsorption by clays as natural scavengers. A comprehensive study investigated on the effect of different types of row clay, montmorillonite, kaolinite and illite on nitrate mobility in unsaturated zone^{72,73}. The results showed that anion adsorption is domain in kaolinite-sand mixture. Calcium montmorillonite modified by hydrochloric acid have more removal efficiency of nitrate, up to 22.28 %, than that modified by sulfuric acid although untreated bentonite, kaolinite and halloysite have poor nitrate adsorption, their nitrate removal capability increase if modified^{66,74}. Nitrate adsorption also have direct relation to the concentration of modifier⁶⁶. Modified and unmodified sepiolite are used to nitrate removal from water. Dodecyl ethyl dimethyl ammonium

TABLE-1
GENERAL COMPARISON BETWEEN DIFFERENT METHODS [AFTER SEIDEL *et al.*²⁰]

Priorities	IX	RO	EDR	BD	CD
High hardness not a major concern					
Reliability					
Training / Ease of operation					
Minimize capital cost					
Minimize ongoing O and M cost					
Minimize foot print					
Industry experience					
Ease of waste management					
Legend	Good	Medium	Proof	Unknown	

(DEDMA) bromide was used by Ozcan *et al.*⁷⁵ to sepiolite modification. Results showed nitrate reduction efficiency in both of adsorbents and modified sepiolite are significantly more than unmodified sepiolite⁷⁶. Investigated nitrate reduction using natural and modified zeolite. Various variables such as pH, temperature, adsorbent dosage, other ions and the amount of surfactant were tested on nitrate removal. Except pH and temperature other variable are found to have effect on removal of nitrate. The removal efficiencies of nitrate in groundwater using unmodified and modified zeolite are reported as 5 and 77 %, respectively. The result of nitrate reduction using raw zeolite showed that it is not suitable for water treatment. A comprehensive study is conducted on removal of nitrate from groundwater by using modified zeolite⁷⁷⁻⁸⁰. Hexadecyltrimethyl ammonium bromide was used to modify natural zeolite. The effect of different variables such as the amount of adsorbent, temperature and the amount of hexadecyltrimethyl ammonium bromide for modified natural zeolite are investigated. The results showed nitrate adsorption is significant by using modified zeolite. Nitrate removal evaluated from aqueous solution using sulfuric acid modified by leonadite⁷¹. The maximum nitrate removal of 77 % is found with pH equals to 5, contact time 1 h, initial nitrate concentration 50 mg/L and the dosage of adsorbent 1 g.

Agricultural wastes adsorbents: Agricultural wastes are included in different varieties of plants such as wheat straw, sugarcane bagasse, rice hull, coconut shells, almond shell and sugar beet pulp. The removal of nitrate has been conducted using modified and unmodified agricultural waste by some researchers. A series of experimental tests conducted on modified and unmodified almond shell. Zn⁰ and ZnSO₄ are used for modification. Results showed the removal efficiency of nitrate for modified almond shell are better than unmodified almond shell. Evaluation of nitrate removal showed 64-80 and 5-42 % for modified almond with Zn⁰ and ZnSO₄, respectively. The removal of nitrate from aqueous solution has been carried out⁸¹ using modified and unmodified wheat straw that passed through a fine-mesh sieve. Epichlorohydrin and dimethylamine are crosslinked with raw wheat straw to make modified adsorbent. The results showed the maximum nitrate adsorption in aqueous solution using modified wheat straw is 2.08 mmol/g. Nitrate and sulfate adsorption evaluated in water using modified sugar beet pulp⁸². Zirconium(IV) ion was used to preparation modified adsorbent. Various variables such as contact time, anions concentration, temperature and pH are used to investigation the effect on the adsorption capacity. It was reported that 114 and 63 mg/g are the maximum adsorption capacity of modified adsorbent for sulfate and nitrate respectively. Raw wheat and mustard straw charcoal were used to assess nitrate reduction from water by Mishra and Patel⁸³. The maximum calculated values of adsorbents are reported as 1.10 and 1.30 mg/g, respectively. Sugar beet bagasse was modified using ZnCl₂ for removal of nitrate⁸⁴. They reported that temperature and pH are two most influence factors on nitrate removal. The maximum nitrate reduction was 41.2 % at pH equals to 3. The adsorption capacity raised from 9.14 to 27.55 mg/g when temperature changed from 25 to 45 °C. Nitrate adsorption assessed using modified beet residue⁸⁵. The adsorbent was modified using epichlorohydrin and pyridine. The results showed adsorbent caused nitrate reduction more than 90 %.

Other adsorbents: Industrial wastes, biosorbent and miscellaneous are different adsorbents that have been studied in few cases by different researchers. The removal of nitrate studied from water by red mud in both active and non-active conditions⁸⁶. A batch experiment tests carried out to study the removal of nitrate from aqueous solution. Different conditions such as different pH, contact time between adsorbent and contamination and the dosage of adsorbent were investigated. The results showed the capacity of active red mud for nitrate adsorption was more than the original red mud which was about three times, 1.859 and 5.858 mmol/g for non-active and active red mud, respectively. Chitosan can be considered as an adsorbent because of the presence of amino and hydroxyl functional in it⁸⁷. The use of chitosan examined in nitrate removal from water⁸⁸. They reported temperature and pH are two variables which had effect on nitrate adsorption. The maximum nitrate adsorption was found 92.1 mg/g at 30 °C. According to Islam and Patel⁸⁹, layered-double hydroxides are inorganic material which tends to remove anionic contaminants from water. Layered-double hydroxides are made with various different ions *viz.*, Zn-Al-Cl⁸⁹, Mg-Al, Co-Fe, Ni-Fe, Mg-Fe⁹⁰, Mg-Al-Cl⁹¹⁻⁹⁴ and so on. Nitrate removal by this method is relatively high. For example, Zn-Al-Cl, and Mg-Al-Cl can remove upto 85.5 and 87.6 % nitrate, respectively⁹². Some other adsorbents have been proposed by researchers to adsorb nitrate from aqueous solution. Characteristics of those adsorbents are summarized in Table-2.

Conclusion

Ever increasing of nitrate in aqueous media and its adverse effect on human health and ecology has led intensive research on cost effective and efficient nitrate removal techniques. Consequently, numerous methods have been developed for nitrate removal. The review of the methods indicates that Ion exchange, reverse osmosis, Biological denitrification and electrodialysis are the most effective methods of nitrate removal from water. However, the major limitation of these methods is the disposal concentrated waste stream to environment and risk of nitrite formation (potential incomplete denitrification). Adsorption methods absorb pollutant therefore are free from some limitations; however, the full scale application of adsorption options is currently limited.

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TABLE-2
OPTIMUM PARAMETERS FOR VARIOUS ADSORBENT: USED FOR
NITRATE REMOVAL (AFTER BHATNAGAR AND SILLANPAA, 2011)

#	Adsorbent	Amount adsorbed	Concentration range (mg/L)	Contact time	Temperature	pH	Refrence
1	H ₂ SO ₄ Treated carbon cloth	2.03 mmol/g	115	60 min	25 °C	7.0	67
2	Powdered activated carbon	10 mmol/g	-	60 min	25 °C	< 5.0	68
3	Carbon nanotubes	25 mmol/g	-	60 min	25 °C	< 5.0	68
4	Untreated coconut granular activated carbon	1.7 mg/g	5-200	2 h	25 °C	5.5	69
5	ZnCl ₂ treated coconut granular activated carbon	10.2 mg/g	5-200	2 h	25 °C	5.5	69
6	Coconut shell activated carbon	2.66 × 10 ⁻¹ mmol/g	-	-	303 K	2-4	96
7	Bamboo charcoal	1.04 × 10 ⁻¹ mmol/g	-	-	303 K	2-4	96
8	Bamboo powder charcoal	1.25 mg/g	0-10	120 h	10 °C		70
9	Halloysite	0.54 mg/g	100	17 h	Room temperature	5.4	66
10	HDTMA modified QLD-bentonite	12.83-14.76 mg/g	100	17 h	Room temperature	5.4	66
11	Calcined hydrotalcite-type compounds	61.7-147.0 g/kg	12.7-236	24 h	25 °C		97
12	Layered double hydroxides	20-35 mg/g	0-1000	4 h	21 °C	8.5	93
13	Chitosan coated zeolite	0.6-0.74 mmol/g	10-3100	72 h	4 and 20 °C		98
14	Chitosan hydrobeads	92.1 mg/g	1-1000	1440 min	30 °C	5.0	89
15	Chitosan beads	90.7 mg/g	25-1000	24 h	30 °C	5.0	89
16	Conditioned cross-linked chitosanbeads	104.0 mg/g	25-1000	24 h	30 °C	5.0	89
17	Pure alkaline lignin	1.8 mmol/g	1-30	48 h	30 °C	-	99
18	Sugarcane bagasse	1.41 mmol/g	1-30	48 h	30 °C	-	99
19	Pure cellulose	1.34 mmol/g	1-30	48 h	30 °C	-	99
20	Rice hull	1.32 mmol/g	1-30	48 h	30 °C	-	99
21	Raw wheat residue	0.02 mmol/g	50-500	150 min	23 ± 2 °C	6.8	68

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