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Review on challenges and opportunities in the removal of nitrate from wastewater using electrochemical method

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Abstract

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Failure to treat waste waters from the industries before discharging it into water sources has led to contamination of water. Providing clean drinking water to the next generation will be a major challenge ahead of us. There are many different methods to treat contaminated water; one being bioelectrochemical treatment. Application of electric field to a bio-denitrification unit is considered to be a way to address these challenges and hence, improve the efficiency of separation. Recently, successful attempts have been made to demonstrate the removal of nitrate from waste water. Various metals have been employed as cathodes, which are known to electrochemically reduce nitrate and copper show better catalytic activity in comparison to other transition metals. In this review, bioelectrochemical reactors for nitrate removal was analysed and compared with conventional methods. Also, the opportunities and challenges in the bioelectrochemical reactors are discussed herein.

Key words

Bio-electrochemical, Cathode, Denitrification, Nitrate, Wastewater

Introduction

Contamination of fresh water is a growing environmental issue and according to the CA reports (2007), one out of three-person in this world is affected by water scarcity. Ever increasing population, urbanization, increased standards of living, and expansion of industrial activities involving nitrogen compounds (biochemical, petrochemical, metal finishing, fertilizers, herbicides, insecticides, pharmaceuticals, wood preservatives and nuclear industries) are contributing factors that intensify this issue (Choi et al., 2002). Contamination of water with nitrate is an issue of global environmental concern. Higher concentration of nitrate in water causes disruption in the ecosystem and health problems to living beings. For example, it leads to

eutrophication in water resources and causes blue baby syndrome in infants, when the concentration is greater than 50 mg 1⁻¹. Most of the industrial waste contains higher concentration of nitrate. For example, nuclear waste contains 1.6-2.2 M NaNO₃ (Genders *et al.*, 1993). Presence of nitrate in industrial wastewaters has necessitated its treatment before disposal. Biological denitrification and non-biological methods such as chemical reduction, ion exchange and reverse osmosis have been employed in large scale operations (Li *et al.*, 1988). At the outset, the present review on the prospects of Bio-Electrochemical Reactor system highlights the future scope of this technology which produces energy while removing nitrate from the wastewater. Out of several methods available, electrochemical method offers promising advantages such as no production of secondary

effluent, removal of specific contaminant, fast, eco-friendly and cost-effective (Iizuka, 2006). On the other hand, waste waters are being considered as an inevitable resource for energy production to meet the ever increasing demand for energy. Anaerobic digestion is the only technology which has hit the commercial scale production of energy from the waste. However, it needs sophisticated conditions and is time consuming. Combination of electrochemical method alongwith bio-denitrification accelerates the denitrification process.

Impact of nitrate contaminated water: Ground water, as well as surface water is found to be contaminated with nitrate by numerous routes. Amplified usage of fertilizers to solicit the productivity in the agriculture land has led to nitrate and phosphate contamination of water sources (Feleke, 2002). Increase in nitrate concentration in the natural water resources have disqualified such waters in many countries (USA, UK, Saudi Arabia, Europe, Japan, China etc.) and northern parts of India (Gayle et al., 1989; Ghafari, 2008). Apart from agricultural source, industrial wastes, animal wastes and domestic waste, when discharged in water sources without treatment, leads to nitrate contamination in water (Islam et al., 1998).

The necessity of removal of nitrogen compounds (NO₃- and NO₂-) is due to the fact that they can lead to eutrophication in rivers, causes human health concerns and inhibits removal of phosphorus in water purification system, such as in activated sludge treatment (Gray, 1990; Sumino et al., 2006; Terblanche et al., 1991; Kempster et al., 1997). nitrate conversion of into nitrite in gastrointestinal tract, leads to methaemoglobinemia in infants (Joseph et al., 1965; Foglar et al., 2005). In adults, nitrate may get converted into nitrosamine and cause gastric cancer (Galvez et al., 2003). Maximum contaminant level for nitrate in drinking water is 50 mg NO₃-1⁻¹ (Webster, 2000; Kross, 2002). Generally, depending on the source of wastewater, nitrate concentration varies from 200 to 50000 mg. Different sources of wastewater and its nitrate concentration is reported in Table 1. Removal of nitrogen compounds from water is complicated due to the fact that nitrogen exists in different oxidation states. Also, stability and highly solubility make it difficult to treat its adsorption or co-precipitation, leading to high energy and cost requirement to treat nitrate contaminated water (Fanning, 2000; El-Shazly, 2011).

Technologies available for denitrification: The various technologies available for nitrate removal are listed in Table 2, and a rational comparison between these processes is

tabulated therein. Catalytic denitrification and biological denitrification have been commercially used while ion exchange and reverse osmosis have been highly efficient for nitrate removal (Katta, 2000; Kapoor, 1997; Hwang *et al.*, 2012, Li *et al.*, 2015). Depending upon the cost, availability, energy requirement and performance, biological denitrification technique is chosen to perform denitrification (Shrimali, 2001).

Biological denitrification: In general, living organisms need energy for its survival and they acquire it through photosynthesis, respiration and fermentation which involve reduction, as well as oxidation of chemical compounds wherein electron transfer takes place. This electron transfer may be within the cells or outside the cells with solid material such as electrodes and minerals as substrates for their respiration which is termed as extracellular electron transfer (Weber, et al., 2006, Hernander, 2001). In the early 19th century, it was reported that disintegration of organic compounds by Saccharomyces cerevisiae is accompanied by liberation of electrical energy, and it is regarded as the first observation of microbial extracellular electron transfer (Potter, 1910). Organic materials present in waste water are oxidized at anode to which electrons are supplied by extracellular electron transfer and thereby, effecting their removal (Stams et al., 2006).

Incorporation of solid materials inside the cells is impossible and special mechanisms are necessary for such reaction, which may be indirect or direct. Details of indirect and direct mechanisms have been well documented in the literature and the same is tabulated in Table 1 (Lovley et al., 2008, Ritcher et al., 2012, Shi et al., 2007, Thrash et al., 2008, Watanabe et al., 2009). Activated sludge process, a most common biological waste water treatment method, has seen several modifications in its setup to improve its nutrient removal efficiency. For instance, single aerobic reactor has been modified into multi-reactor consisting aerobic, anoxic and anaerobic zones wherein nitrification, denitrification and phosphorus removal are effected and the developed process is called biological nutrient removal processes (Jinsong et al., 2013; Wanner et al., 1988; Lettinga et al., 1980; Gray, 1990; Wentzel, 1992; Lu, 1988). Application of Bioelectrochemical Reactor designed using software based on mathematical models has been employed world-wide (Ekama et al., 1992; Henze, 1992; Kristensen et al., 1992; Gujer et al., 1992).

Detection of microorganisms with substances acting as electron acceptors has been reported, and identification of specific microorganisms (Example: Thiobacillus

Table 1: Average nitrate concentration in industrial effluents

Effluent source	Nitrate concentration	Reference	
General	200 mgl ⁻¹ Almeida <i>et a</i>		
		Zayed and Winter. (1998)	
		Peyton et al. (2001)	
Explosives, Fertilizer	$1000\mathrm{mgl}^{-1}$	Watanabe et al. (2001)	
Pectin, Cellophane	$1000\mathrm{mgl}^{-1}$	Glass et al. (1999)	
Metals finishing industries	$1000\mathrm{mgl}^{-1}$	Glass et al. (1999)	
Metal cleaning in nuclear weapon industries	$50000\mathrm{mgl}^{-1}$	Francis and Hatcher. (1980)	
High level nuclear waste	3 M	Genders, et al. (2003)	

Table 2: Comparison of available methods for denitrification

Parameter	Chemical	Biological	Reverse osmosis	Ion exchange	Electrochemical
Applicability	Medium scale	Large scale	Medium scale	Medium scale	Large scale
Continuous operation	Good	Partial	Good	Good	Good
& automation					
Post treatment	Air stripping to	Bacterial contamination	No	chlorine rich	No
ofwater	remove NH3 formed	and residual carbon source		water	
Waste	Secondary sludge	Biomass disposal	TDS rich waste	Waste brine	No
generation	is produced		disposal	disposal	
Operation	Stable operation	Requires close monitoring	Stable operation	Stable operation	Stable
& maintenance					operation
Safety issue	Unsafe	Needs controlled	Safe	Safe	Safe
		environement			
Cost	High	High	Very high	High	Low

denitrificans, *Prolixibacter* sp. phylum 'Chloroflexi') that utilize oxidized inorganic nitrate as electron acceptors has led to use electrochemical biochemical system to treat nitrate (Iino, 2015; Kawaichi, 2013, Li *et al.*, 2006). Particularly denitrifying heterotrophic bacteria is known to remove nitrate effectively from waste water under both aerobic and anoxic environments (Kuba *et al.*, 1993; Jørgensen and Pauli, 1995; Kavanaugh and Randall, 1994; Kuba *et al.*, 1997; Osborn *et al.*, 1989). Under anoxic conditions, in place of oxygen for respiration of cells, these microorganisms utilize nitrate as electron acceptors (Ketchum *et al.*, 1988; Cappuccino *et al.*, 1992; Prosnansky *et al.*, 2002). Thus, simultaneous reduction of nitrate as well as oxidation of organic matter takes place (Wanner *et al.*, 1988).

Under aerobic environment, efficient biological denitrification reaction can be performed with identification of novel species capable of using nitrate as source (Kodama et al., 2004; Shi et al., 2013; Zhao et al., 2013). Arcobacter, as well as Clostridium have been found to be effective in nitrate removal from water (Xiaowei et al., 2015). In soil, Pseudomonas, Achromobacter, Bacillus, Flavobacterium,

Agrobacterium, Hyphomicrobium, Vibrio, Alcaligenes and Chromobacterium are responsible for denitrification (Otlanabo, 1993). Rod morphology, Gram-negative species such as Pseudomonas aeruginosa, Pseudomonas stutzeri, Achromobacter xylosoxidans, Pseudomonas pickettii and Pasteurella spp., have shown strong nitrate and nitrite reducing ability. However, rest of the species are incomplete denitrifiers, as they just convert nitrate into nitrite and fail to effect further reduction. In addition, few Gram-negative cocci as well as Gram-positive rods also significantly contribute in denitrification (Drysdale et al., 1999; Rijn, 1996). Microbial treatment of NO₃- might be the most economical reclamation method of NO₃- polluted wastewaters (Soares, 2000).

Improving the speed of biodenitrification: In general, biodenitrification process is slow and in particular, when the nitrate concentration is high it takes longer time to process it. To address this issue, several efforts have been made to increase the reaction rate by changing the configuration of the reactor or combining another technique with bio denitrification. For example, membrane biofilter reactor is a

combination of membrane techniques with biological denitrification (Terada, 2003). One such promising hybrid technology is combining the electrochemical denitrification method explained in the next section with biological denitrification, which results in increased nitrate removal efficiency.

Electrochemical denitrification: The electrochemical method offers various advantages such as removal of specific species and generation of no secondary waste (Mansour, 2007). A wide range of cathode, Pt, Rh, Al, Ni, Pb, Zn, Cu, Sn and Fe cathodes have been used as cathode (Brylev, 2007; Dima, 2005; Kerkeni, 2002; Armijio, 2002; Georgeaud, 2012; Katsounaros, 2006). Under similar conditions, Rh was found to exhibit higher nitrate reduction than Ru, Ir, Pd and Pt and Cu; among Au, Ag and Cu, Cu showed higher catalytic activity (Dima et al., 2003). Other metals complexes such as phthalocyanines have been used as cathode for nitrate reduction (Chebotareva, 1997). Pt-based nanoparticles have been demonstrated cathode for nitrate reduction (Estudillo, 2011). Generally, Pd, Ir and Rh are suitable cathode for nitrate reduction. Product composition during denitrification depends on various parameters including, cathode, pH, supporting electrolyte, potential, initial concentration, other species present and various intermediates are formed (Dima et al., 2003). For example, on a palladium electrode, various products including nitrite, ammonia and nitrogen are formed. The main advantage of electrochemical system is that it can be coupled with other techniques or it can be used to treat the waste generated by other techniques. Thus, the combination of electrochemical and biodenitrification is obviously a technology to focus on.

Bioelectrochemical reactor systems: Bioelectrochemical system is a recently developed technique for cost-effective water and wastewater denitrification. Specific interest, as well as substantial advancement has been made in enhancing this technology towards industrial applications treating various waste waters (Xiaochin et al., 2015). Recent progress in wastewater treatment has led to the development of Bioelectrochemical system which uses micro-organisms capable of extracellular electron transfer and electrochemically active, that facilitates the electron transfer to the anode where oxidation of pollutants such as ammonia occurs (Stams et al., 2006). Microorganisms act as catalysts, referred to as microbial bioanode/biocathode and connecting a counter electrode essentially makes it a complete microbial electrolysis cell (Cohen, 1931; Kim et al., 1999). Its operation is determined by Gibbs free energy (ΔG , J mol⁻¹),

$$E = \Delta G / nF \tag{1}$$

where, E is the electromotive force generated, expressed in Volts, E is negative in case of microbial electrolysis cell because ΔG is positive hence energy intensive system. In other words, if ΔG is positive Bio-Electrochemical Reactor works as Micro Chip Electrophoresis where energy is used to effect chemical reaction such as removal of contaminant nitrate from water (Rabaey, 2005; Logan, 2006). Reduction of nitrate to nitrogen is catalysed by different enzymes and presumably it follows the steps given in Eq. 2 (Feleke, 2002; Killlingstad, 2002)

$$2NO_3^- + H_2O \rightarrow N_2 + 2.5 O_2 + 2OH^-$$
 (3)

The global reaction taking place is given by Eq. 3(Ghafari, 2008).

$$No_3^- \rightarrow NO_2^- \rightarrow NO \rightarrow N_2O \rightarrow N_2$$
 (2)

The detailed reaction taking place which includes electrolysis of water and step-wise autotropic denitrification using hydrogen as electron donor has been reported (Ghafari, 2008).

Studies on bio-electrochemical reactor: Investigation on Bio-Electrochemical Reactor has provided exponentially improved performances and lab scale Bio-Electrochemical Reactors have achieved current densities as high as 10 A m² anode surface areas. Practically, as high as 1000 A m⁻³ current density (volumetric) can be achieved with Bio-Electrochemical Reactor. The challenge is to convert Bio-Electrochemical Reactor volume by 3 to 6 orders of higher magnitude from lab scale volume of 11. The optimum practical implementation of this beneficial and promising bioelectrochemical system for extraction of renewable energy lies in addressing the challenges of enhanced degradation of complex materials, the microbial reactions control, stable electrodes and ohmic loss reduction (Rene et al., 2008). A biofilm electro reactor was used for nitrate reduction, it was demonstrated that the current density directly influences the denitrification and it was shown that 0.2 mol of NO₃- was converted into N₂ gas per mol of electron. These results exhibited the simplicity and feasibility of the proposed process, especially for low-concentration NO,- water treatment. (Sakakibara, 1993). Stainless steel, activated carbon, graphite felt were used as cathode and carbon rod, graphite rod, Dimensionally Stable Anodes (DSA), amorphous carbon were used as anode for bioelectrochemical reactions carried out in different reactor setup listed in Table 2 have been reported to operate with 100% efficiency (Ghafari *et al.*, 2009, Wan *et al.*, 2009). A comparative study of biological denitrification with and without constant electric field was studied in a Continuous Stirred Tubular Reactor (CSTR). A cathode potential near to its value of nitrate/nitrite redox couple was maintained constant, and it was demonstrated that the electric field enhanced the nitrate removal efficiency and a higher flow rate of fluid was possible when an electric field was applied (Parvanova *et al.*, 2009).

In bioelectrochemical system, electrolysis of water generates hydrogen at cathode, and autotrophic denitrifying microorganisms utilize this H₂ (acting as electron donor) to produce N₂ gas by reducing nitrate NO₃. In comparison to some contaminants which demands a specific microorganism, the interesting fact about denitrifying bacteria is that they are omnipresent in nature and they convert nitrate into harm less N, (Szekeres et al., 2001; Gamble et al., 1977). Carbon nanotubes have shown positive effects on biofilm formation and good biocompatibility with bacteria. Therefore, cathode material selection affects the bioelectrochemical system performance. Some of the commonly evaluated support materials are carbon felt, graphite and carbon cloth for immobilization of Multi walled Carbon Nanotubes in the custom-made dual chamber reactor, represented as Multi walled Carbon Nanotubes/X, where X = carbon felt, graphite and carbon cloth they have shown a nitrate removal efficiency of 83.58%, 63.98% and 79.66% respectively. Carbon felt was found to be the most suitable support material for Multi walled Carbon Nanotubes for nitrate removal in bioelectrochemical Reactor (Safari et al., 2014). So far, investigations have aimed at developing factors such as cathode potential, pH or conductivity for denitrifying- bioelectrochemical system. Proteobacteria was identified during analysis for microorganism as a key microorganism responsible for denitrification. However, an exceedingly dissimilar communities were found to be responsible for nitrate removal (Clauwaert et al., 2009; Pous, 2014; Wrighton et al., 2010).

Tsuneo et al., (2000), studied the nitrate removal of the conventional sugar beet pulp and improved its performance by combining it with the boron exchange membrane, and observed that the denitrification rates were enhanced in the presence of a current supplied using packed bed activated carbon electrodes submerged into the aerobic compartment of the sugar beet pulp. They inferred that the

nitrate removal rates depend on applied potential and current. An interesting observation was made that the contribution of the current towards nitrate removal was high when the dissolved oxygen concentration in the bioelectrochemical reactor was low. Interestingly, no significant change in pH was observed after electrolysis (Tsuneo $et\ al.$, 2000). The challenge with respect to engineering perspective is to uncap the essential fundamentals of microbial extracellular electron transfer. In comparison to heterotrophic biological denitrification, under similar conditions, autotropic denitrification was more effective which utilized CO₂ as carbon substrate and hydrogen as electron donors (Ghafari, 2008).

Biological de-nitrification offers advantage of treating various contaminants at a time and reducing waste disposal cost. Primary factors in the application of biological denitrification are the need for anoxic conditions, chemical demands, the standard of operator training, the heftiness of the system, and the post-treatment requirements (Valsa et al., 2012; Mateju et al., 1992). Till date, anaerobic digestion is the only technique which was commercially successful in producing energy from waste (Pham et al., 2006, Aiyuk et al., 2006). An upflow reactor tested in the lab scale, 6 m³ and 200 m³ full-scale experiments with retention time of 4 hrs was reported with high organic loading (Chemical Oxygen Demand of 20 kg m³ per day) for denitrification (Lettinga et al., 1980). Biological treatment has the capability to afford a feasible, long term nitrate treatment option. Prediction of cost suggests that biological treatment can be economically competing with ion exchange process.

When nitrate concentration is high in wastewater, additional carbon sources such as starch, glucose, CH₂OH, C₂H₅OH and CH₃COOH needs to be added to the system. The choice of substrate depends on the cost, availability, volume and specifications of processing unit and auxiliary units required for further treatment of treated water (Kim et al., 2002; Killingstad, 2002; Kim et al., 2004, Szekeres et al., 2002). Such additional substrates, which acts as an electron donor in turn results in by-products such as turbidity, biomass and substances that reduce the taste of water. Both autotrophic and heterotrophic bacteria were used to reduce nitrate. It was found that heterotrophic denitrification has faster reaction rate and need lesser reactor volume than autotrophic denitrification, thus reducing capital cost. It uses fluidized bed reactors over packed bed reactors and requires recirculation of effluent stream for effective nitrate removal.

Table 3: List of various BER systems reported

Reactor system	Abbreviation	Reference
Combined bio-electrochemical sulfur	(CBSAD)	Wan et al., 2009
autotrophic denitrification system		
Bio-electrochemical reactor	(BER)	Linda et al., 2014
Dual chamber bio electrochemical reactor	(DCBER)	Ghafari, 2008
Three-dimensional bio-electrochemical reactor	(3DBER)	Zhou et al., 2009
Up-flow bio-electrochemical reactor	(UBER)	Linda et al., 2014

Table 4: Comparison of maximum denitrification rate reported

Reactor	Denitrification rate,	Reference
	mg NO ₃ -N cm ⁻² day ⁻¹	
UBER	0.031	Kurt et al. (1987)
UBER	0.083	Macdonald (1990)
UBER	0.023	Liessens et al. (1993)
UBER	0.038	Sakakibara and Kuroda (1993)
UBER	0.17	Park et al. (2005)

Bio-denitrification requires intensive maintenance, slow start up time, slow reaction rate, difficulty in controlling the parameters such as pH, temperature to 20 $^{\circ}$ C and continuous supply of carbon source, substrate are the hurdles in implementing it industrially. Moreover, even 0.1 to 0.2 mg of oxygen per litre is found to inhibit denitrification. It cannot be used above $1000 \, \text{mg} \, \text{l}^{-1}$ nitrate concentration.

Microbial fuel cell has been used to treat waste water as well as harvest energy (Logan, 2005, Du, 2007, Feng, 2008, Oh, 2010). A membrane Bio-Electrochemical Reactor coupled with MFC was used to treat wastewater from cheese industry and it showed a satisfactory removal efficiency in which Microbial Fuel Cell removed contaminants and recovered energy, while the Multiple Bio-Electrochemical Reactor acted as post-treatment to obtain a high quality effluent (Li *et al.*, 2014a). When a hollow-fibre membrane was incorporated in the system, it resulted in 66 % nitrate removal (Li *et al.*, 2014b).

Studies with Bio-Electrochemical Reactor using Proton Exchange Membrane for electrolysis revealed that pH gradient across the membrane is a serious issue to be addressed. As high as 5 pH units across the membrane and 0.06 V/unit pH change have been reported which significantly affect performance of Bio-Electrochemical Reactor (Prosnansky *et al.*, 2002). A comparison of nitrate removal ability per biofilm surface area per day is tabulated in Table 4. Park *et al.*, (2005), reported highest denitrification rate.

Economic feasibility: With respect to the economic feasibility of scaling up of Bio-Electrochemical Reactor into

pilot and industrial applications, there are significant challenges posed by Bio-Electrochemical Reactor systems. Performance of Bio-Electrochemical Reactor systems are governed by microorganism, solid materials, electrodes, biocatalysts, reaction configuration and kinetics, resistance for electron transfer, cost of fabrication and cost of operation. Mainly, electrode surface area as well as coverage of biofilm on it, increased reduction reaction due to enriched microorganism which essentially contributes to scale up of Bio-Electrochemical Reactors. Between autotropic and heterotrophic denitrification, it was found that in later one, reaction rate is fast and requires less reactor volume, thus reducing the cost and time. The cost of electrode which houses the biofilm needs to be brought down. Enhancement of electrochemically active bacterial community (referred to as Exoelectrogens) can be achieved by means of pretreatment of anode electrode, optimization of potential and current, chemical additions, bioaugmentation and temperatures (Sai et al., 2016).

In Bio-Electrochemical Reactors, high drop in potential because of ohmic losses due to high level of internal resistance regardless of polymer electrolyte membrane is used or not (Logan *et al.*, 2006). This overpotential limits the achievable current densities (Cheng *et al.*, 2006). Hence, it is essential to address this issue and reduce the overpotential losses. Interestingly, potential losses with bioanodes (\sim 0.1V) are consistently reported as lower in comparison to bare electrocatalysts (\sim 0.5 V) (Freguia, 2007).

Future scope : We envision that the future investigation in denitrification using Bioelectrochemical system will lead to

innovations in the development of metal cathodes with biofilms of an altogether untried variety of microorganisms. Electrochemical reactions taking place at these biocathodes, catalysed by enhanced diversified populace of microorganisms are capable of donating electrons that will be meticulously selected; and/or *Geobacter species* and *Sphingobacterium* which make effective utilization of $\rm H_2$ produced cathodically or e to perform the reduction reactions required for manufacture of products that have higher market value which in turn will vastly support to counterbalance the initial costs involved with Bioelectrical Chemical Systems.

Conclusion: Bio-Electrochemical Reactor method for denitrification has gained attention and being investigated with novel microorganisms. Bio-Electrochemical Reactors offer appropriate conditions for simultaneous H₂ production on cathodes as well as proper consumption by immobilized autotrophs on cathodes. Among heterotrophic and autotrophic denitrifications, later one is most preferred and effective one with various sources for food and energy. Studies on bio-Electrochemical Reactor will be of great importance for utilization of microorganism for waste water treatment and contributing to the progress of new biotechnologies.

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