Journal of Water Process Engineering 6 (2015) 58–63

Contents lists available at ScienceDirect

Journal of Water Process Engineering

journal homepage: www.elsevier.com/locate/jwpe

Removal of nitrate ion from water by electrochemical approaches

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ARTICLE INFO

Article history:
Received 4 October 2014
Received in revised form 24 February 2015
Accepted 27 February 2015
Available online 22 March 2015

Keywords:
Electrochemical denitrification
Electrocoagulation
Electroreduction
Fe
Al
Graphite electrodes
Sludge characterization

ABSTRACT

This study is focused on the mechanism of nitrate removal from aqueous solutions by electrochemical denitrification process (EDN) in an undivided electrolytic cell. The sacrificial (Fe and Al) electrodes and Inert (graphite (Gr)) electrode are employed for evaluation of operational parameters, namely current density, electrolysis time and sodium chloride concentration. The experimental results reveal that nitrate-N removal efficiency of 92% for Al–Fe (anode-cathode) and 80% for Fe–Fe are achieved at a current density of 25 mAcm^{-2} and 180 min electrolysis time in 100 ppm of NaCl when the initial nitrate-N concentration is 100 ppm. However, during this process approximately 20 ppm of ammonia-N is also formed. Ammonia-N generated is significantly lower compared to the amount of nitrate-N removal. It can be attributed to the fact that nitrate ion can be removed by both electrocoagulation (EC) and electrochemical reduction (ER) processes simultaneously. Experiments using Al, Fe and Gr as cathodes as well as anodes indicate that the contribution of EC increases when dissolving anodes are used. Further confirmation of nitrate-N removal by EC pathway is obtained by quantitative estimation of nitrate present in the sludge. During EDN, pH of the solution also increases with ammonia generation.

1. Introduction

Groundwater is a major source of drinking water for rural communities in India and other developing countries. Due to extensive use of nitrate fertilizer in agriculture, this valuable groundwater is being contaminated by nitrates. Elevated nitrate contamination in groundwater can cause 'methemoglobinemia' in blood leading to blue baby syndrome in children [1,2]. For these reason, a maximum desirable limit for nitrate (NO_3^-) is 50 ppm (10 ppm equivalent nitrate-N) for adults, 15 ppm (3 ppm equivalent nitrate-N) for children and 0.5 ppm of ammonia was permitted by World Health Organization (WHO) [3]. Extensive data collected by Central Ground Water Board of India (CGWB) indicated the presence of 100–800 ppm nitrate levels in many regions of India [4]. Similarly nitrate contamination in groundwater and surface water has increasingly major issues. Thus the removal of nitrate from water gains more and more attention.

Among the various methods available for removal of nitrate, the biological methods [5–7] introduce additional biological impurities and degradation products into the water during treatment. Highly water soluble nitrates require large volumes of adsorbents for their removal [8], leading to the generation of solid waste. Electro-dialysis (ED) [9], nano-filtration (NF) and reverse osmosis (RO) [10] also lead to concentrated nitrate rejects, which require further treatment and disposal. Hence, there is considerable interest in electrochemical denitrification (EDN) processes, which can be operated at a small scale to meet local community demand for drinking water in villages.

The mechanistic studies of EDN exhibit certain unsettled issuses. Electrochemical reduction (ER) and electrocoagulation (EC) processes are the two approaches being investigated for denitrification. Nitrite and ammonia are the main products formed during ER of nitrate ion [11,12]. Direct $5e^-$ reduction of nitrate to nitrogen gas has been claimed in some reports, supported by the mechanism which deals with the formation of nitrogen gas during chemical reduction of nitrate using Al and Fe powders [13–17]. The suitable

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http://dx.doi.org/10.1016/j.jwpe.2015.02.008
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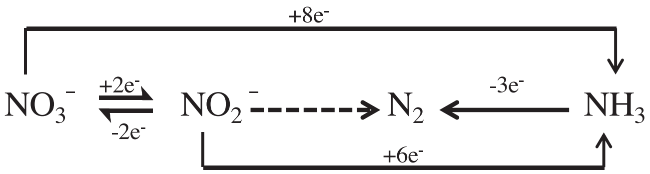

Scheme 1. Electrochemical reduction pathways for nitrate ion.

approach to understand the mechanism can be rendered as the formation of ammonia from nitrate and nitrite at cathode and subsequent re-oxidation of ammonia to nitrogen on the catalytic anode surfaces [18] as shown in Scheme 1. Detailed studies on the mechanism of ammonia oxidation on RuO_2 and IrO_2 coated electrodes [19–21] also support this mechanistic pathway. Residual ammonia-N is found to be less in the ER process when oxide coated Ti was used as the anode [13–15,22]. The complex mechanistic pathways discussed in the literature for the EC mechanism are summarized in Scheme 1.

Energy consumption for EC process is twenty times more than ER process in the faradaic EDN [23]. Hence, more attention has been given for EC pathways in EDN. Both Al and Fe anodes play a substantial role in nitrate removal by EC process. The EC process [23–26] was indeed shown to be more efficient than chemical coagulation using Al^{3+} or Fe^{3+} salts [26]. However, there are contradicting reports on the cathodic process during EC. It has been reported that ammonia is generated only on Al electrodes during initial stages of EC. Direct cathodic reduction of nitrate to nitrogen gas [25] and formation of hydrogen gas [24] has also been suggested as the predominant cathodic process. EC and ER pathways have been proposed for nitrate removal in flow reactors using mild steel [27] and zero-valent iron [28] electrodes, respectively. Further, basic studies are necessary for evaluating of the role of ER and EC pathways in EDN process. The main objective of the present research work is to identify experimental conditions under which the 100 mg L^{-1} of nitrate-N can be removed from aqueous solution containing 100 mg L^{-1} NaCl by EDN process and also to elucidate the respective role of ER and EC in nitrate removal.

2. Materials and methods

2.1. Denitrification experiments

Electrochemical denitrification experiments were performed at room temperature in an open undivided batch electrolysis cell (500 mL glass beaker) containing 300 mL electrolyte, as depicted in Fig. 1. Graphite (Gr), aluminium (Al) and iron (Fe) plates were employed as electrode materials. Anode and cathode with an inter electrode distance of 2 cm were connected to a regulated DC power supply (ApLab C3202). Synthetic nitrate contaminated water was prepared using analytical grade potassium nitrate (KNO_3, 99% Himedia, India) and sodium chloride (NaCl, 99% Merck Chemicals India). The salts were dissolved in double distilled water. Electrolyte pH was found to be in the range of 7 ± 0.1. The electrolyte was continuously stirred magnetically at a constant speed (300 rpm) during electrolysis and then allowed to settle for 2 h. The solution was filtered through Whatmann filter paper and the filtrate was used for analysis. Triplicate runs were carried out for every experiment to ensure reproducibility.

2.2. Data analysis

Nitrate-N (NO_3^--N) concentration in the solution before and after EC was analyzed by UV spectrophotometer (JASCO V-650). 1 N HCl was used for the acidification of samples to prevent the interference from hydroxide or carbonate anions. Chloride and sulfate have no effect on the estimated value [29]. Ammonia-N (NH_4-N) concentration in residual solution was estimated colorimetrically by Nesslerization method [29]. All the experiments were performed at 25 ± 2 °C. The sludge samples obtained after EDN process was washed with distilled water and dried overnight at 40 °C. The scanning electron microscope (SEM) and X-ray diffraction (XRD) pattern of the dried sludge was obtained using JEOL JSM-6390 and SHIMADZU XPERT-PRO diffractometer, respectively.

For the estimation of nitrate-N content, the sludge (100 mg) was dissolved in 3–5 mL of concentrated H_2SO_4 to obtain a clear solution. The solution was then made up to 100 mL. The nitrate-N

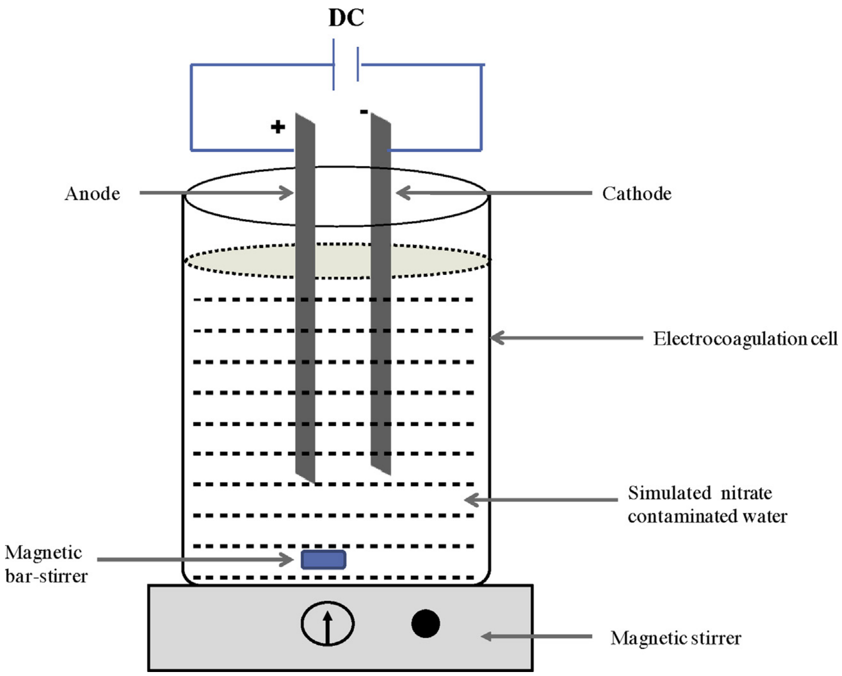

Fig. 1. Schematic representation of electrochemical denitrification (EDN) cell.

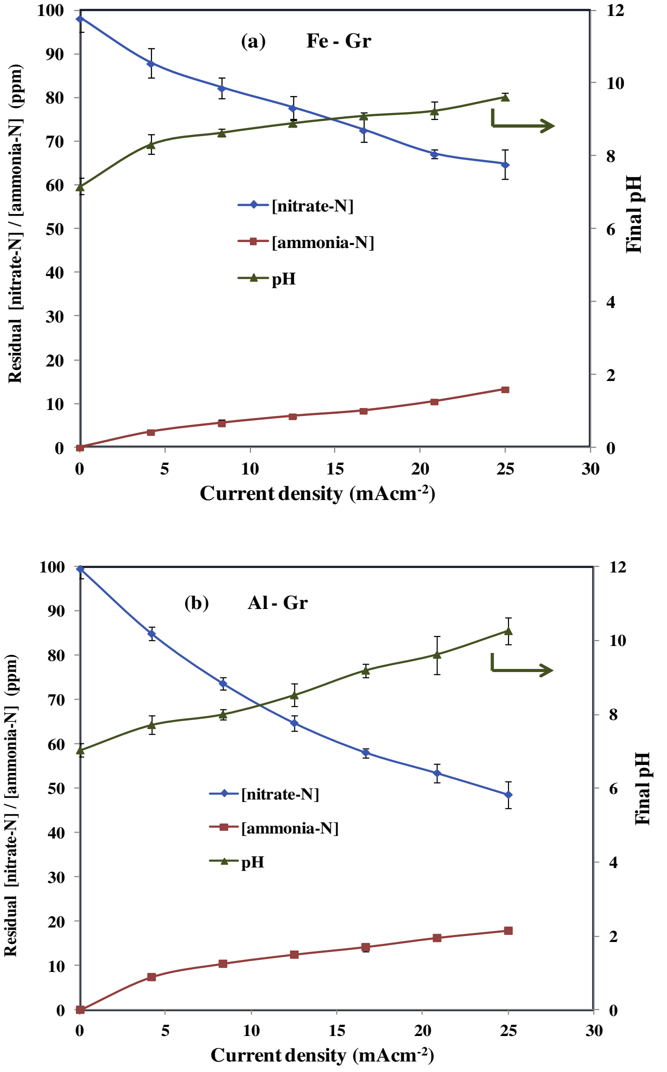

Fig. 2. Effect of current density on nitrate removal by electrochemical method using (a) Fe anode–Gr cathode and (b) Al anode – Gr cathode. (Electrolysis time = 60 min, Initial [nitrate-N] = 100 ppm, [NaCl] = 100 ppm and pH- 7). Error bars are the standard deviations of triplicate experiments.

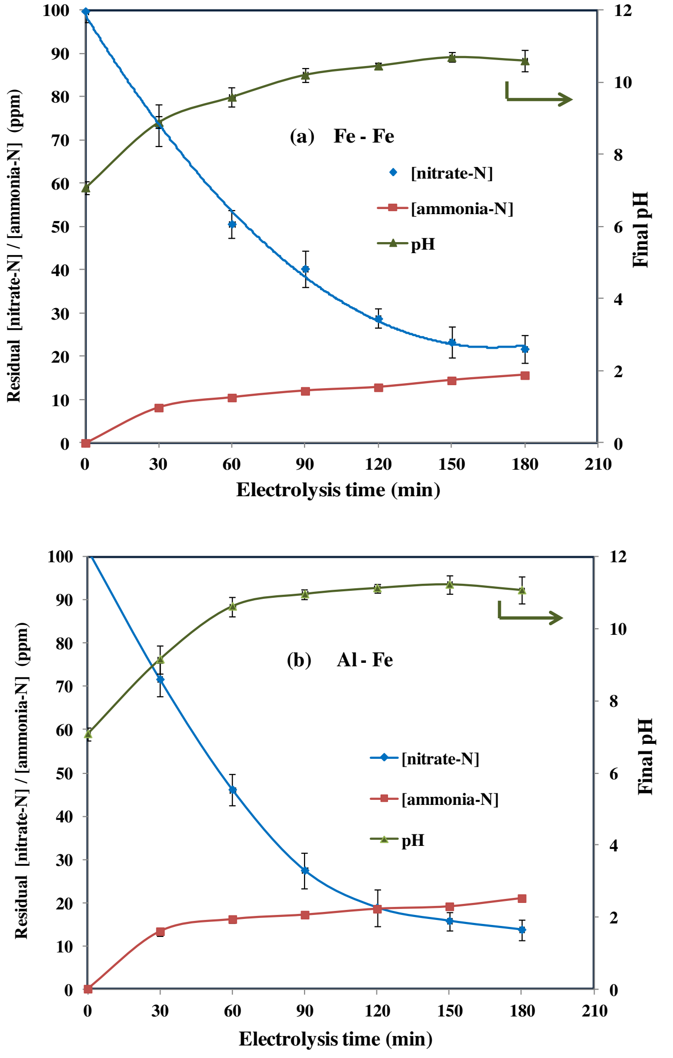

Fig. 3. Effect of electrolysis time on nitrate removal by electrochemical method using (a) Fe anode-Fe cathode and (b) Al anode -Fe cathode. (Initial [nitrate-N] = 100 ppm, [NaCl] = 100 ppm, pH-7 and current density – 25 mA cm^{-2}). Error bars are the standard deviations of triplicate experiments.

content in the solution was estimated using the same spectrophotometric method discussed above.

3. Results and discussion

3.1. *Effect of operational parameters*

The results obtained during EDN of aqueous solution containing 100 ppm nitrate-N and 100 ppm NaCl in neutral pH are presented in Fig. 2. Fe (Fig. 2a) and Al (Fig. 2b) electrodes are used as anodes with common Gr cathode. The electrolysis time is kept constant at 60 min. The concentration of residual nitrate-N, ammonia-N and final pH of the solution obtained at different current densities are compared. These results illustrate that increment of current density has a substantial effect on reducing the concentration of nitrate-N. The nitrate-N removal is also accompanied by ammonia generation which increases with increasing current density. The effect is common for both the electrode pairs. Efficient nitrate-N removal is achieved when Al is employed as the anode.

Effect of electrolysis time for nitrate-N removal efficiency on Fe (Fig. 3a) and Al (Fig. 3b) anodes (common Fe cathode) is investigated, while maintain the constant current density at 25 mA cm^{-2}. On Al anode, the residual nitrate-N concentration reaches the permissible limit (Fig. 3b) in 180 min. The residual nitrate-N concentration in the case of Fe anode is found to be around 20 ppm under identical conditions. The pH values are found to increase with electrolysis time (Fig. 3).

The experimental results presented in Figs. 2 and 3 reveals that maximum ammonia-N formed at different current densities and electrolysis times is around 10–20 ppm. The ammonia generation is expected to come from ER of nitrate ion by an overall 8e^{-} reduction as represented by the following reaction:

$$NO_3^{-} + 8e^{-} + 6H_2O \rightarrow NH_3 + 9OH^{-} \quad (1)$$

By considering 100% of electrochemical reduction (8e^{-} reduction) in cathodic region can generate around 23.3 ppm of ammonia-N in 180 min of electrolysis time and with the current density of 25 mA cm^{-2}on 14.3 cm^{2} electrode surfaces.But the measured value of ammonia-N concentration is always found to be lesser than 23.3 ppm (Fig. 3a and b). Hydrogen evolution as a competitive cathodic reaction [24] can lead to lower efficiencies for ammonia generation. Since both the cathodic processes (Eqs. (1)

and (2)) lead to the formation of hydroxide ions, the pH of the solution increases during electrolysis.

$$2H_2O + 2e^- \rightarrow H_2 + 2OH^- \quad (2)$$

The results presented in Fig. 3 also elucidate that generation of ammonia-N may reach 15 ppm in the first one hour of electrolysis. As per the faradaic process should produce only 7.8 ppm of ammonia-N in one hour. Thus it appears that there is also a non-faradaic contribution toward ammonia production, especially during the early stages of EDN. This may be due to the contribution of direct chemical reduction of nitrate by Al or Fe powder formed during anodic dissolution (Eq. (3)). Such chemical reduction of nitrate by Al [14] and Fe [17] powder is well established. For Al powder, the overall nitrate reduction is represented by the following equation.

$$8Al + 3NO_3^- + 18H_2O \rightarrow 8Al^{3+} + 3NH_3 + 27OH^- \quad (3)$$

The total nitrate-N removal efficiency is always much higher than the ammonia generation (Fig. 2 and Fig. 3). This result suggests that ER is not the only mode of EDN. But EC processes also leads nitrate removal, because nitrate ions preferentially adsorbs on surface of growing aluminum hydroxide (Eq. (5)) and iron hydroxide (Eq. (7)) sludges [26,30].

For aluminum (Al) anode

$$Al \rightarrow Al^{3+} + 3e^- \quad (4)$$

$$Al^{3+} + 3H_2O \rightarrow Al(OH)_3 + 3H^+ \quad (5)$$

For iron (Fe) anode

$$Fe \rightarrow Fe^{2+} + 2e^- \quad (6)$$

$$Fe^{2+} + 3H_2O + O_2 \rightarrow Fe(OH)_3 + H_2 \quad (7)$$

These experimental results confirm that the formation of ammonia during electrolysis, which is further stabilized by the final pH of 10–11. The residual solution obtained under experimental conditions described in Fig. 3b is found to be quite stable at room temperature.

Some reports suggested that the oxidation of ammonia by the electrochemically generated chlorine gas or hypochlorite (OCl^-) at the anode surface. The present studies indicate that the nitrate-N removal and ammonia production are quite independent of NaCl concentration (Fig. 4). The chloride (Cl^-) concentration does not have an effect on the EDN process and hence OCl^- generation does not contribute toward this process.

3.2. Effect of electrode materials

The role of EC and ER pathways is further investigated by studying the effect of electrode pairs on nitrate-N removal and ammonia generation. In addition to that the dissolving Al and Fe anodes, Gr electrode is chosen as an inert anode material. All the three electrodes are also employed as cathodes. The electrolysis is carried out at a current density of 25 mA cm^{-2} for 60 min in these experiments. The residual nitrate-N as well as generated ammonia-N values obtained for all the nine different electrode pairs are presented in Fig. 5.

These results indicate that the residual nitrate-N in solution after EDN is much higher when the non-dissolving Gr is used as the anode material. This further confirms that the additional contribution of nitrate-N removal by dissolving anodes through EC process. Among the dissolving anodes, Al leads to better nitrate-N removal efficiency when compared to Fe anode. These results also confirm that ammonia is invariably generated by ER in all the nine experiments involving Al, Fe and Gr electrodes. In the case of Gr–Gr electrode pair, the decline in the nitrate-N concentration (13.8%) is quite close to the ammonia-N generated (12.5%). It can be inferred that removal of nitrate-N occurs solely due to the ER pathway alone in the case of the Gr-Gr electrode pair.

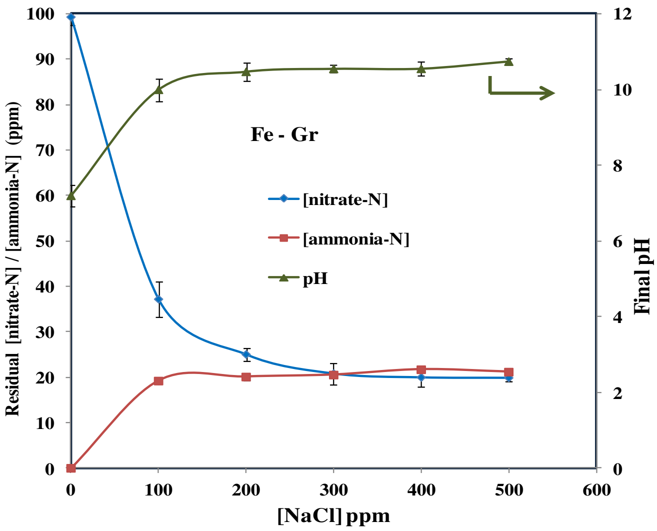

Fig. 4. Effect of NaCl concentration on nitrate removal by electrochemical method using Fe anode–Gr cathode. (Initial [nitrate-N] = 100 ppm, pH- 7, current density – 25 mA cm^{-2} and electrolysis time = 180 min). Error bars are the standard deviations of triplicate experiments.

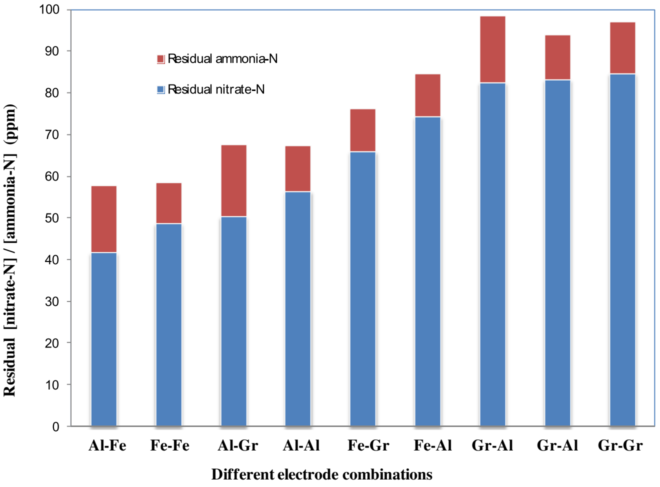

Fig. 5. Effect of electrode pairs (anode first) on residual nitrate-N and generated ammonia-N concentration with Fe, Al and Gr electrodes. (Initial [nitrate-N] = 100 ppm, [NaCl] = 100 ppm, pH- 7 and current density – 25 mAcm^{-2} and electrolysis time = 60 min).

3.3. Sludge characterization

The Fe and Al sludges generated during the EDN are found to retain individual characteristics of their respective hydroxides. The SEM images of the sludges produced by Fe and Al anodes are represented in Fig. 6a and b, respectively. Hydroxide sludges generated by Fe anode contain much smaller particles (Fig. 6a) when compared to the larger agglomers found in aluminum hydroxide sludges (Fig. 6b) produced by Al anode under identical conditions. The XRD data presented in Fig. 7a exhibits the crystalline nature of the sludge generated by EDN with Fe anode and Gr cathode. It confirms that the formation of crystalline iron oxide hydroxide (Fe_{21} HO_{32}) with 2θ = 30.27, 35.69, 43.31, 57.25 and 62.97° corresponding to (2 2 0), (3 1 1), (4 0 0), (5 1 1) and (4 4 0) crystal planes (JCPDS: 89-3850). The XRD pattern of the hydroxide sludge gen-

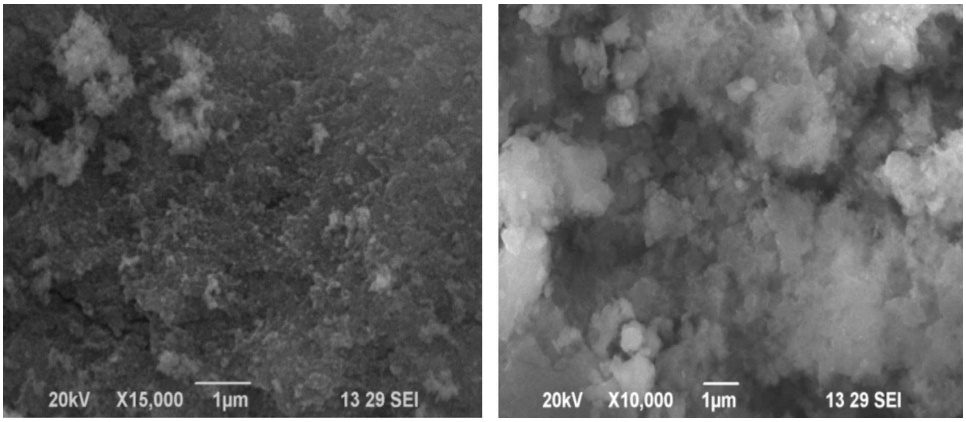

Fig. 6. SEM images of electrochemically generated sludge. Electrochemical reaction carried out by (a) Fe anode-Gr cathode and (b) Al anode–Gr cathode. (Initial [nitrate-N] = 100 ppm, [NaCl] = 100 ppm,current density – 25 mA cm^{-2}and electrolysis time = 180 min).

erated by Al anode and Gr cathode is illustrated in Fig. 7b. Strong peaks at $2\theta = 18.83, 20.35, 27.99, 40.86$ and $53.57°$ are identified to be β-Al$(OH)_3$ (JCPDS: 12-0457). No nitrate containing crystallites are identified in the XRD patterns.

However, the presence of nitrate-N in sludges generated from electrochemical process is also analyzed by dissolving the sludges in concentrated H_2SO_4. Typical nitrate-N concentration values obtained from the dissolved sludge from Fe–Gr and Al–Gr electrode pairs are presented in Table 1. As indicated in Table 1, the nitrate-N removal efficiency around 58% is achieved for Fe–Gr electrode. Whereas, around 20% of the nitrate-N reduction due to the ammonia-N generation through electroreduction of nitrate ion. As well as the presence of 31% of nitrate-N in the sludge indicate that nitrate ion can also eliminated by electrochemical coagulation. Likewise, the nitrate-N removal efficiency around 89% is obtained for Al–Gr electrode. In which, 21% of nitrate-N reduction due to the ammonia-N generation through electroreduction of nitrate ion and the presence of 66% of nitrate-N in the sludge indicate that nitrate ion can also eliminated by electrochemical coagulation. Therefore, the above experimental observation substantiate that the nitrate ion in aqueous solution is removed by both electroreduction and electrocoagulation processes.

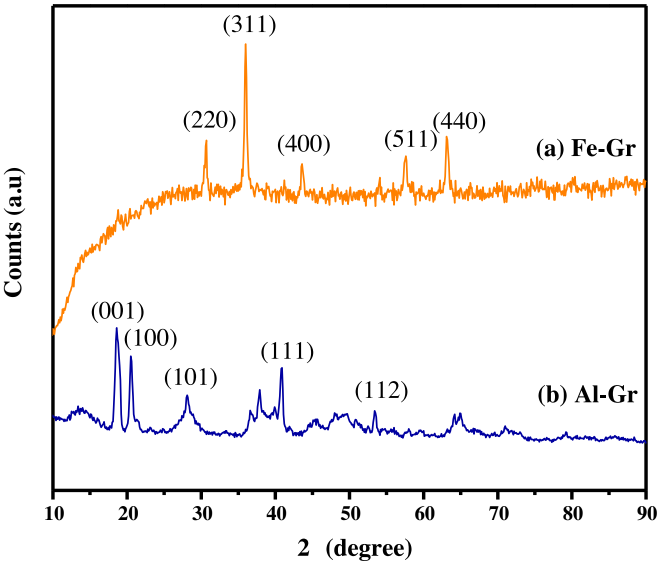

Fig. 7. X-ray diffraction patterns of electrochemically generated sludge. Electrochemical reaction carried out by (a) Fe anode–Gr cathode and (b) Al anode–Gr cathode. (Initial [nitrate-N]) = 100 ppm, [NaCl] = 100 ppm, current density – 25 mA cm^{-2}andelectrolysis time = 180 min).

Table 1
Determinations of nitrate-N and ammonia-N in residual solution and sludge. Electrochemical reaction carried out at initial [nitrate-N] = 100 ppm,[NaCl] = 100 ppm, current density – 25 mAcm^{-2} andelectrolysis time = 180 min.

Nitrate-N and ammonia-N in residual solution and sludge	Electrode materials (anode-cathode)	
	Fe–Gr	Al–Gr
Overall nitrate-N removal efficiency (%)	58	87
Nitrate-N in residual solution after electrolysis (ppm)	42	11
Ammonia-N in residual solution after electrolysis (ppm)	20	21
Nitrate-N in sludge estimated (ppm)	31	66
Total-N (ppm)	93	98

4. Conclusions

The present investigation establishes the possibility of reducing the nitrate-N concentration to permissible limits by electrochemical denitrification (EDN). The experimental results reveal that maximum nitrate-N removal efficiency of 92% for Al–Fe (anode-cathode) and 80% for Fe–Fe are achieved at a current density of 25 mAcm^{-2} and 180 min electrolysis time in 100 ppm of NaCl when the initial nitrate-N concentration is 100 ppm. However, ER invariably occurs along with EC on all the cathode materials, leading to the formation of ammonia. Adsorption of nitrate ion onto surface of growing metal hydroxide precipitates appears to be the primary mechanism behind the observed large nitrate removal efficiency.

The dissolving anodes (Fe and Al) also contribute to the nitrate-N removal by EC route. Al anode is more efficient in nitrate-N removal when compared to Fe anode. Under all experimental conditions employed in the present study, generation of around 20% ammonia was observed. The solution pH was also found to increase invariably during this process. The production of ammonia was found to be quite stable in the alkaline solution formed during EDN. The overall results indicate that electrochemical denitrification processes is an effective technologies for nitrate removal from water. However, alternative method air stripping facilities would be needed to remove the electrochemically generated ammonium ions from solution.

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