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Evaluation of Nitrate Removal from Wastewater Using Electrochemical Method

Zahra Abaspoor [1], Amin Goli [2], Amirreza Talaiekhozani [3,4]*, Benyamin Bayat [1], Poyan Ghanbarnejad [1], Marzieh Bagheri [5], Sanaz Alaee [6], Gholamreza Ziaee[7]

1- Jami Institute of Technology, Department of Chemical Engineering, Isfahan, Iran.
2- Jami Institute of Technology, Department of MechanicalEngineering, Isfahan, Iran.
3- Jami Institute of Technology, Department of Civil Engineering, Isfahan, Iran.
4- Institute of Environmental and Water Resources Management, Water Research Alliance, UniversitiTeknologi Malaysia, UTM Skudai, 81310 Johor Bahru, Malaysia.
5- Jami Institute of Technology, Department of Chemical Engineering, Isfahan, Iran.
6- Department of Reproductive Biology, School of Advanced Medical Sciences and Technologies, Shiraz University of Medical Sciences, Shiraz, Iran.
7- Jami Institute of Technology, Department of Accountancy, Isfahan, Iran.

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Abstract

Pollution of under groundwater by nitrate is a serious problem that has been widely observed worldwide. This type of pollution can be dangerous for human health and especially for children. Therefore, nitrate must be removed from contaminated under groundwater. Electrochemical method has many advantages in comparison with reverse osmosis or ion exchange, such as lower cost, requires a smaller land producing less sludge and requires fewer chemical materials. The aim of this study is to evaluate nitrate removal by electrochemical method.Carrying out this study; water samples contaminated with nitrate were prepared by adding suitable amount of $NaNO_3$ in distilled water. Then samples were treated by a batch electrochemical reactor in laboratory-scale. In this study, graphical rods were applied as electrodes. Nitrate concentration was determined by spectrophotometric method. The results showed that by using electrochemical method 15.33, 17.41, 19.48 and 21.58 % of nitrate ion could remove within hydraulic retention times of 30, 60, 90 and 120 min, respectively. These results were obtained using of voltage of 24 volt and electrode surface of 15.896 cm^3. Our results elaborated that, electrochemical method can remove nitrate from water. However, nitrate removal efficiency in this method is not efficient enough to apply in industrial scale.

Keywords: Nitrate removal, electrochemical method, water treatment, water well.

1 Introduction

The nitrate ion (NO_3) is the stable compound resulted by the chemical process between nitrogen and oxygen. Although nitrate ion is a stable compound in water, it is possible to reduce or eliminate it through microbial activities [1, 2, 3]. This ion is one of nitrogen's inorganic components and is considered as the last stage in the oxidation of ammonia by organic materials [4, 5,6].Nitrate ion is recognized as underground water pollutants releasing to environment, are growing in recent years, due to expansions in agriculture and human activities. . Under the guidance of Standards and Industrial Research organization of Iran, the maximum amount of nitrate ion in drinking water in nitrogen is equal to 10mg/L and in NO_3 is equal to 50mg/L. [7, 8]. Most people are concerned about contaminants that can have an immediate impact on water quality such as: taste, smell, darkness and water turbidity, suspended solids, water hardness, water sediments andetc. However they neglect far more dangerous pollutants such nitrate. Its effects only become apparent after at least 15 years. High levels of nitrate for livestock can cause problems such as: reduced fertility, increased stillbirths, low birth weight, slow weight gain or can even result in death. In addition, a high level of nitrate in drinking water causes diseases such as cancer, deformed babies, enlarged thyroid an inflammation of the lymph nodes [9, 10]. Although nitrate doesn't only exist in contaminated water, but also in most of the food we commonly consume on a daily basis. These compounds are used as additives and preservatives in foods such as sausages, salami and smoked salmon, added to protect them from corruption. Nitrates in food and water increase the nitric acid in saliva which increases the nitrosamine levels with combination with

Corresponding author: Amirreza Talaiekhozani, (1) Institute of Environmental and Water Resources Management, Water Research Alliance, Universiti Teknologi Malaysia, UTM Skudai, 81310 Johor Bahru, Malaysia. (2) Jami Institute of Technologies, Department of Civil Engineering, Isfahan, Iran. E-mail: atalaie@jami.ac.ir.

amino acids. In the acidic stomach environment, the synthesis rate of nitrosamines increased is harmful for human health overtime. High daily consumption of fruits and vegetables, which contain high amounts of vitamin C, can reduce the nitrate contamination risks in individuals. If the consumption of vitamin C and E is 2, or more times higher than the nitrate rates in body, the injury risk of nitrate is reduced [11, 12]. Nitrate is a natural part of human diet, therefore, meager amounts of nitrate is not hazardous to human health. But if there is a particularly high nitrate concentration above 50 mg/L, in this case, the use of such water for children (especially less than three months) is dangerous and can cause a disease called Hemoglobin Matt. The nitrate in children's gastrointestinal tract in which the acidification activity is not completely started, is of alkaline pH and is rapidly converted to nitrite by bacteria naturally present in the digestive system which is a biological restoration process. The produced nitrite is then quickly absorbed into the bloodstream and causes a bivalent iron in hemoglobin, can be converted to trivalent hemoglobin. In this case, the hemoglobin is converted into hemoglobin matt [13, 14]. When the hemoglobin matt concentration reaches to 0.51g of blood or at least 10% of adult hemoglobin, the symptoms of anemia and cyanide is caused in the individual. The most important symptom of cyanide is bluish skin especially around eyes and mouth [15]. Some studies also suggested that a high concentration of nitrate in drinking water during pregnancy increased the risk of birth defects in newborns and was also effective in the reduction of oxygen transfer to the baby through the mother's blood [16]. In the digestive tract, the nitrite is combined with amines of type 2 and type 3 by bacteria and produces nitro amines which are carcinogenic substances. To avoid fore mentioned health problems, removal of nitrate from drinking water is necessary. Many researchers have studied the organic and inorganic compounds removal using electrochemical treatment methods. For instance Jafarzadeh et al. (2010) studied removal of propylene glycol from wastewater. They could find a direct relationship between propylene glycol removal and used voltage. However, using high voltage can produce some new chemicals which are harmful for human health. Also, they reported that using copper electrodes, can release copper ions in water which is a harmful ion for human health.

2 Materials and Methods

In this study, a batch reactor with the volume of 2 L was used in laboratory-scale. The required electrical energy in this study had been supplied by an electrical transducer to convert the voltage from 220 volts to 4.5, 12 and 24 volts. Graphite electrodes were used in order to transmit the electrical power to the fluid. A view of the applied batch reactor in this study can be seen in Figure.1.

In this study, watersamples were prepared by adding suitable amount of $NaNO_3$ to distilled water for achieving nitrate concentration of 100 mg/L. In order to evaluate the effects of retention time in nitrate removal efficiency, the nitrate contaminated water sample was electrolyzed in the batch with retention times of 30, 60, 90 and 120 minutes. At the end, the nitrate concentration in the samples was measured and ultimately, the nitrate removal efficiency in various voltages was determined. Next, to investigate the effects of electrode surface in the batch removal efficiency, electrodes with an area of 11.9 and 15.9 cm^2 were used.

In this study pH was measured by using a Thermo Scientific Orion digital pH meter Model "4-Star". Electrical power was produced with a Farkhande electrical transducer. A Hack spectrophotometer model DR-5000 was used to measure nitrate ion concentration.

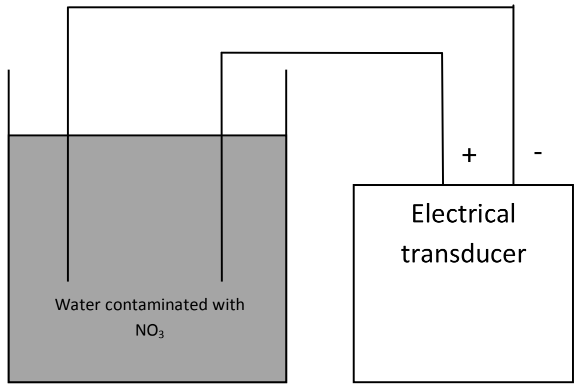

Figure.1: A view of the applied batch reactor in this study

To determine the nitrate concentration in this study, the spectrophotometric method was used according to the standard methods. For this purpose, 1 mL of normal hydrochloric acid was added to a 100 ml of sample and was well blended. Then, the adsorption rate was set at 220 nm wavelength using a spectrophotometer device. In order to generalize, the adsorption rate of nitrate concentration, samples of nitrate-contaminated water was prepared by NaNO3 and the calibration graph was drawn [18].

3 Results and Discussions

The results obtained from this experiment elaborated that the electrochemical methods can remove nitrate from aquatic solutions. Based on Fig. 2 to 4, nitrate removal has a direct relation with hydraulic retention time (HRT), applied voltage and electrode surface. As can be seen at Fig. 2 to 4, approximately 70% of nitrate removal is increased in HRT is equal 30 s when voltage is increased from 4.5 to 24 volt.

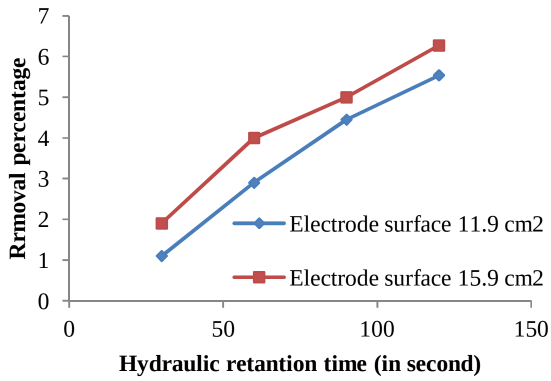

Fig. 2Nitrate removal percentage in different hydraulic retantion times by using voltage of 4.5 V

Due to finding relationship among all independent variables (HRT, Electrode Surface and Voltage) and the major dependent variable Nitrate Removal Percentage, a regression test with entry method has been done below.

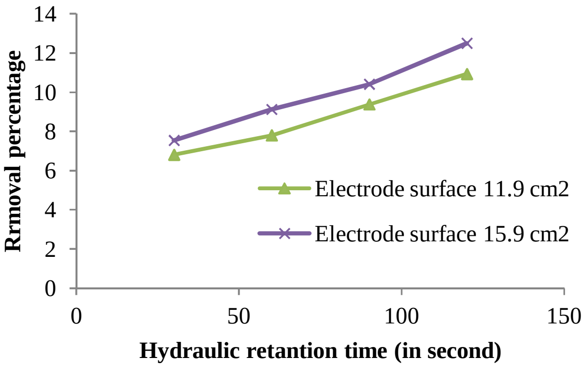

Fig. 3 Nitrate removal percentage in deferent hydraulic retantion times by using voltage of 12 V

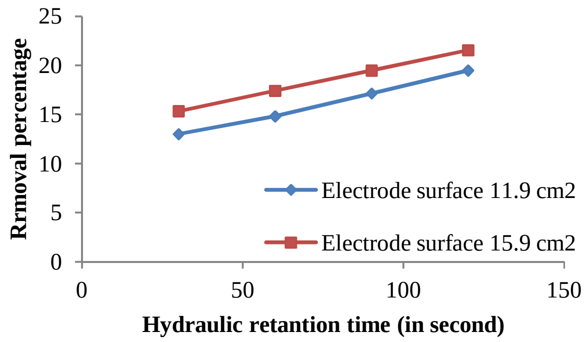

Fig. 4 Nitrate removal percentage in deferent hydraulic retantion times by using voltage of 24 V

There is a significant relation with 95% of confidence among Nitrate Removal and all of independent variables. Table1 shows some parameters like (1) sum of squares, (2) degree of freedom or df, (3) mean square, (4) Fisher exact statistic or F and (5) Probability value or sig.

Table 1: Analysis of Variances for Removal Percentage (ANOVA)

	Sum of Squares	df	Mean Square	F	Sig.
Regression	822.448	3	274.149	827.057	0.000
Residual	6.630	20	0.331		
Total	829.077	23			

A) Predictors: (Constant), HRT, Surface, Voltage
B)Dependent variable: Removal

Table 2 shows the linear multiple model among nitrate removal percentage and all of independent variables. This table is included of (1) Unstandardized coefficients of each independent variable, (2) related standard error, (3) t-student statistic or *t*and (4) value of probability or *Sig*. The table shows that all of independent variables with constant coefficient are statistically significant.

Eq.1 shows the derived regression model from Table 2 which numerically explain the relationship among all independent variables and nitrate removal percentage.

$$NRP = 0.684\,V + 0.357\,ES + 0.05642\,HRT - 8.275 \qquad (Eq.1)$$

Where, *NRP* is nitrate removal percentage, *V* is voltage, *ES* is electrode surface and *HRT* is hydraulic retention time.

Table 2: Coefficients of all independent variables of model

Model Variables	Unstandardized Coefficients	Std. Error	*t*	*Sig.*
(Constant)	-8.275	.888	-9.316	.000
Voltage	.684	.015	46.742	.000
Surface	.357	.059	6.084	.000
HRT	5.642E-02	.004	16.103	.000

A Dependent Variable: Removal

Based on Eq. 1 we are able to predict nitrate removal percentage for each optional value of independent variables *NPR*, *V* and *HRT*.In addition, the value of R square (R^2) is equal 0.992 approves a proper amount of defining nitrate removal percentage with all independent variables.

Model Summary

R	R Square	Adjusted R Square	Std. Error of the Estimate
.996	.992	.991	.5757

A Predictors: (Constant), HRT, Surface, Voltage

Figure 5 shows results of application of Eq.1 to predict nitrate removal percentage in various optional voltage values. Based on this prediction, 99.99% of nitrate removal can be achieved when reaction is conducted with voltage of 149 volt.

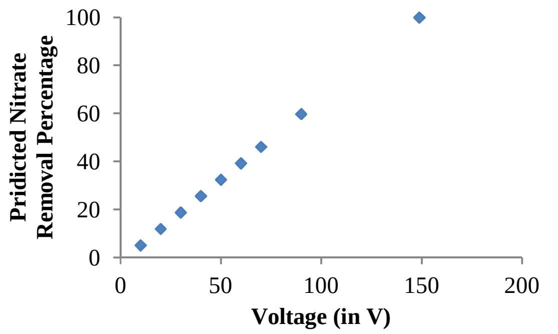

Fig. 5 predicted nitrate removal percentage with different voltage values

4 Conclusions

The adsorption rate of the sample was 0.372 mg/L According to the standard curve; the nitrate concentration of the sample was 1.1 mg/L. as proved, the standard values for nitrate is 50 mg/L, it is lower than the expected standards. This experiment elaborated that the voltage, retention time and electrode surface in the electrolyte solution are of affecting parameters in the nitrate removal efficiency by electrochemical method. The highest removal rates were observed in retention time of 120 min, voltage of 24 v and electrode surface of 15.896 cm^2 in the electrolyte solution. In the nitrate removal process, there is interplay between voltage, retention time and electrode size. In such a way which increases in voltage, retention time and electrode surface led to increments in the nitrate removal percentage from water.

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