Removal of nitrates from water by Amberlite IR-400 and economic Duolite A-378 ion exchange resins

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ABSTRACT

Water contaminated with nitrates is one of the environmental problems that Iraq suffers from as a result of sewage deposit that is discharged into rivers. In addition to agricultural fertilizer waste, old and dilapidated water pipes extend in densely populated areas throw dirt directly into the rivers. To reduce the material cost effort and time for the researcher and everyone who is concerned with purifying water from nitrates. The economic (Duolite A-378 weak base anion exchange resin) alternative to the high cost (Amberlite IR-400 strong base anion exchange resin) of removing nitrates from the water was tested, and a regression model was also found using MINITAB-19 statistical program to compare the accuracy of the practical results with what is theoretically expected. Linking the variables together and measuring the extent of their impact on nitrate removal rate. The results were summarized in the approaching efficiency of (Duolite A-378 weak base anion exchange resin) to remove nitrates, but with the increase in the quantity more than (Amberlite IR-400 strong base anion exchange resin). There was also an increased response to the removal of resin when increasing the concentrations corresponding to a decrease in the removal rate when increasing the speed of mixing and this is due to the strength of the ionic attraction.

Keywords: Ion-exchange; Resin; Nitrate removal; Amberlite IR-400; Duolite A-378

1. Introduction

In a study conducted by the World Economic Forum in 2015, water crises were listed as one of the major global challenges, even higher than wars among nations, or weapons of mass destruction. Thus, ultimately, the issue of clean water is global because it is essential to all forms of life crises and wars have affected the quality of water in the region, particularly in Iraq [1,2] where the crises and wars have led to a deterioration in the quality of water in many areas of Iraq, whether it is groundwater, rivers or drinking water. Nitrate is one of the important pollutants of water which is known as soluble nitrogen ions that produce colorless and odorless water [3,4]. It is included in sewage systems, such as the breakdown of animal and plant waste, chemical fertilizers, pesticides, etc. [5,6]. Al-Hiyaly et al. [7] selected four sites from the water of the Tigris River to study the concentration of chlorides and nitrates per month for the wastewater discharged from Baghdad Medical City Hospital during the period from October 2012 to September 2013. The findings revealed that the average nitrate levels were between 2.5 ± 0.86 and 28.8 ± 4.98 mg/L, which violated World Health Organization (WHO) and Iraqi water safety requirements for surface water protection. Al-Paruany et al. [8] demonstrated in their investigation of six Baghdad water well locations that the nitrite and nitrate content meet health guidelines. When it comes to the nitrite and nitrate levels in the Diyala and Abu Ghraib regions, in which the change in the nitrite and nitrate levels are due to the period of using nitrogen fertilizers (ammonium nitrate) in agricultural areas.

In terms of risk, children under the age of six months are at risk of hemoglobinemia (the syndrome of the blue child). In addition to several other disorders that concern adults, such as eyelid enema, digestive system and head lightness, chronic inflammation, nasal and pharynx mucous obstruction [3,9]. In order to protect human health from nitrate pollution, responsible agencies have established drinking water requirements, with a maximum pollutant level of (45 ppm) NO₃ or (10 mg/L nitratenitrogen (NO₃-N)) considered appropriate in the United States. The WHO and Iraqi standard regulations set the nitrate amount at (50 ppm) as NO₃ or (11.3 mg/L as (NO₃-N)) [10-12]. One of the characteristics of nitrate is its stability and difficult solubility, so traditional methods of removing it from water cannot be relied upon [13,14]. Ion exchange [13,15,16], biological treatments [17-21], adsorption [14,22-25], and membrane isolation [26-29] are only a few of the recent methods that have been used to safely strip nitrates from water. Since the ion exchange mechanism is one of the methods used to purify water in Iraq, so this paper found an economical alternative to the high-cost (Amberlite IR-400 strong base anion exchange resin) by testing low-cost (Duolite A-378 weak base anion exchange resin) and comparing its efficiency with the first. In addition to saving time, effort, chemicals and material cost for tests by reducing their number and comparing their accuracy. With theoretical results using the statistical technique in the MINITAB-19 program, a regression model was found linking the variables (pH, concentration of nitrates, resin amount and agitation rate) with each other to determine the percentage of removal. Because of the interest in the environment first and for the researcher secondly, especially since most of the research in Iraq is conducted at personal cost.

Table 1 Properties of resins [30]

2. Research method

2.1. Resins impregnation

Two types of resin are utilized (Amberlite IR-400 strong base anion resin and Duolite A-378 is a weak base anion resin) for the exchange of anions and are washed with deionized water to eliminate organic and inorganic impurities that may be attached to the surface. They're then immersed for 3 h in 2 M hydrochloric acid (HCl) with a purity of 36.5% and a density of 1.185 g/mL, it was made of 169.49 mL of hydrochloric acid and 1,000 mL of distilled water. To get rid of excess (Na, H) sets of resin, rinse the resins several times in distilled water and repeat the washing step until the pH exceeds 8. Then, dry for 2 h at 70°C and store in a closed container. In Table 1, the properties of resins are described.

2.2. Influent solution

To prepare the initial concentrations of nitrate solution, 0.815 g potassium nitrate (KNO_3) is dissolved in 1,000 mL deionized water and diluted in experiments to (70, 90, 100, and 120 mg/L) the size of a cup of 250 mL according to the equation [30].

$$V_o \times C_o = V_f \times C_f \tag{1}$$

where V_o and V_f = initial and final volume respectively, C_o and C_f = initial and final concentration respectively.

2.3. Nitrate analysis

A UV spectrophotometer was used to analyze the final concentrations of nitrates by selecting (215 nm) wavelength, which is a technique used to measure the intensity of light passing through an empty or reference sample, as

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	Amberlite IR-400	Duolite A-378
Resin type	Strong base anion	Weakly basic anion
Function groups	Quaternary ammonium	Tertiary amine
Ionic form	Cl-	Cl-
Theoretical capacity	≥1.40 meq/mL	1.4 mmol/mL
Particle size	0.60–0.75 mm	0.3–1.2 mm
Maximum reversible-swelling Shape		

it is a quantitative technique by which we can know the amount of light absorbed by the placed chemical, whether liquid, solid or even thin and glass films, through measuring the number of photons that passes to the detector. The amount of light absorbed by a surface at a certain wavelength is proportional to the concentration of the element being studied [31,32].

2.4. Effect of pH on adsorption

The effect of pH change (5, 6, 7, and 8) on nitrate removal was investigated for both types of resins with time stabilization at 170 min under conditions (70 ppm concentration, 0.3 g resin amount, and 100 rpm). To adjust the pH in solutions, sodium hydroxide and hydrochloric acid were used.

2.5. Effect of initial concentration on adsorption

For both types of resins, the influence of the initial nitrate concentration at 70, 90, 100, and 120 ppm was measured at the optimum pH, which was previously determined with 0.3 g resin amount, 170 min and 100 rpm conditions.

2.6. Effect of resin amount on adsorption

The effect of resin amount was examined by adding 0.05, 0.1, 0.3, 0.5 g of Amberlite IR-400 strong base anion resin and 0.05, 0.1, 0.3, 0.5, 1 g of Duolite A-378 weak base anion resin into 100 mL of nitrate solution at 170 min, 100 rpm, pH and concentration determined previously conditions.

2.7. Effect of agitation rate on adsorption

The effect of the agitation rate was studied at 0, 50, 100, 150, 250, 500 rpm with using of water bath stirring shaker under optimum pH, concentration and resin amounts that examined previously and 170 min.

2.8. Data analysis

Design of experiments statistical methodology of MINITAB-19 programming was used to achieve optimum performance analysis which involved comparing the input data and the output outcomes of tests, as well as comparing them to numerical data. As a result, a statistical model is given to describe the efficiency of each resin and its ability to remove nitrates.

2.9. Batch adsorption study

The batch test was obtained using a water bath with different amounts of resin according to the conditions and quantities by using the magnetic stirrer device. The efficiency of nitrate removal was determined by the following equation [33,34].

$$R\% = \left[\frac{C_o - C_e}{C_o}\right] \times 100\%$$
⁽²⁾

Mass balance was used to calculate the amount of nitrate adsorbed per unit mass of resin by the following equation [35,36].

$$q_e = \frac{(C_o - C_e) \times v}{w} \tag{3}$$

where R% = nitrate removal percent, C_o = the initial concentration of nitrate in (mg/L), C_e = the final concentration in of nitrate (mg/L), q_e = the adsorption capacity, V = the volume of solution in (L) and W = the weight of resin in (g).

2.10. Adsorption isotherm

Langmuir isotherm model was used to evaluate the maximum capacity of adsorption and offer the activation energy of homogeneous adsorption [37,38]. While the equilibria between solid and solution was described by Freundlich isotherm that expressed by equations respectively [35,36].

Langmuir equation:

$$\frac{1}{q_e} = \frac{1}{q_m} + \frac{1}{c_e} \frac{1}{k_a q_m}$$
(4)

Freundlich equation:

$$\log q_e = \log k_f + \left(\frac{1}{n}\right) \log c_e \tag{5}$$

where q_e = equilibrium adsorption capacity in (mg/g), q_m = a complete monolayer adsorption capacity in (mg/g), k_a = constant of adsorption equilibrium in (L/mg), c_e = concentration of nitrate at equilibrium in (mg/L), n and k_f are Freundlich constants.

2.11. Kinetic studies

Pseudo-first-order and pseudo-second-order kinetic equations were used to estimate the amount of nitrate adsorbed at the period of the time that was expressed by equations respectively [35,36].

Pseudo-first-order kinetic equation:

$$\ln(q_e - q_t) = \ln q_e + k_l t \tag{6}$$

Pseudo-second-order kinetic equation:

$$\frac{t}{q_t} = \frac{1}{kq_e^2} + \frac{1}{q_e}t \tag{7}$$

where q_{μ} , q_e = adsorption capacities at *t* time in (min) and equilibrium respectively. k_{μ} *k* = the rate constants of pseudo-first-order and pseudo-second-order models respectively.

3. Results and discussion

3.1. Effect of pH on nitrate adsorption

pH is the factor that affects the rate of adsorption and changes the surface charge of the adsorbate during ion exchange [39,40]. Studying the effect of pH, it is discovered that 6 and 7 is the optimal pH for both Amberlite IR-400 and Duolite 378, which is considered within the natural

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limits of the pH water as shown in Fig. 1. pH is descending when the pH becomes more than 7 because the emulation between NO_3^- and Cl⁻ on the surface of resin as a result of adding HCl and NaOH for setting the pH.

3.2. Effect of initial concentration on adsorption

From the experiments, it was observed that the removal percent increase as the concentration of nitrate increases. This reflects the large acceptability of nitrate removal for both resins. To compare the two types of resin used, the efficiency of the strong IR-400 anion resin is high compared to the weak A-378 anion resin, whereas the weak A-378 anion resin is equivalent to the efficiency of the strong IR-400 anion resin in case the quantity varies under the same conditions (Fig. 2).

3.3. Effect of metal solutions on adsorption

Testing the amount of resin is important in measuring its efficiency and adsorption. It was found that the percentage of removal increases as the amount of resin increased due to the availability of a larger surface area of ion exchange for the concentration of primary dissolved, noting the difference in quantity to obtain close results as shown in Fig. 3.

3.4. Effect of agitation rate on adsorption

A direct relationship was observed between the nitrate adsorption and the agitation rate until 150 rpm, which indicates the correct exchange in the sites between the solution and the sites of absorbent binding. While at a high agitation rate of more than 150 rpm, the removal efficiency would be decreased because of the ionic attraction force that is stronger at high electrolyte solution as shown in Fig. 4.

3.5. Prediction of the ion exchange isotherms model

A regression model has been designed for each Amberlite IR-400 and Duolite A-378 resin *y* using MINITAB-19 statistical software, these results have been compared with the theoretical ones, and results from MINITAB-19 have been compared with each other [41,42]



Fig. 1. Effect of pH solution on nitrate removal.

to try to determine how the factors relate to one another. The following nitrate removal % regression models have been designed on 37 runs of experiments for each (Amberlite IR-400 and Duolite A-378 resins) respectively:

$$R^{(\text{IR-400)}}_{(\text{IR-400)}} = -237.2 + 54.5x_1 + 0.372x_2 + 360.7x_3 + 0.1648x_4 \\ -4.397(x_1)^2 + 0.00194(x_2)^2 - 415.7(x_2)^2 - 0.000321(x_1)^2$$
(8)



Fig. 2. Effect of nitrate initial concentration on nitrate removal.



Fig. 3. Effect of resins amount on nitrate removal.



Fig. 4. Effect of agitation rate on nitrate removal.

$$\begin{split} R\%_{(A-378)} &= -86.0 + 18.10x_1 - 0.014x_2 + 189.37x_3 + 0.1125x_4 \\ &- 1.405(x_1)^2 + 0.00155(x_2)^2 - 97.79(x_3)^2 - 0.000216(x_4)^2 \end{split} \tag{9}$$

where $x_1 = pH$, $x_2 = initial$ concentration of nitrate (ppm), $x_3 = resin$ amount (g) and $x_4 = agitation$ rate (rpm). The (R-sq.) of regression models get (98.70%) and (99.42%) for (Amberlite IR-400 and Duolite A-378) resins respectively. This represents the fit of the models with the data. As the value of R-sq. was greater, the model was more suitable for data and the response prediction was more accurate. The normal distribution probability for nitrate removal % for (Amberlite IR-400) resin and (Duolite A-378) resin are shown in Fig. 5.

3.6. Equilibrium isotherm models

Fig. 6 represents the linear Langmuir and Freundlich isotherm plots for nitrate removal for each Amberlite IR-400



Fig. 5. The normal distribution probability of nitrate R% for: (a) Amberlite IR-400 resin and (b) Duolite A-378 resin.



Fig. 6. (a) Langmuir and (b) Freundlich isotherm for resins.

Table 2	
Summary of isotherm parameters	

		Slope	Intercept	q_m	k _a	R^2
Langmuir	Amberlite IR-400	0.07147	0.01459	68.5401	0.204142	0.89947
isotherm	Duolite A-378	0.21006	0.03861	25.90003	0.183805	0.87249
		Slope	Intercept	1/ <i>n</i>	k _f	R^2
Freundlich	Amberlite IR-400	Slope 0.37299	Intercept 1.20574	1/ <i>n</i> 0.37299	<i>k_f</i> 16.05979	<i>R</i> ² 0.98002



Fig. 7. (a) Pseudo-first-order and (b) pseudo-second-order adsorption kinetic.



Fig. 8. Amberlite IR-400 resin 3D response surfaces for: (a) pH and resin amount (g), (b) pH and concentration (ppm), (c) pH and agitation rate (rpm) and 2D contour plots interaction between data of experiments on nitrate removal %: (aa) pH and resin amount (g), (bb) pH and concentration (ppm) and (cc) pH and agitation rate (rpm).

and Duolite A-378 resins. According to the correlation coefficients and the isotherm parameters listed in Table 2 and shown in Fig. 6, it is obvious that the Freundlich isotherm model provides a better fitter than the Langmuir isotherm.

3.7. Kinetic studies

Table 3 shows the different initial concentrations by pseudo-first-order and pseudo-second-order models. The result stated that both Amberlite IR-400 and Duolite A-378

Table 3

Summary of pseudo-first-order and pseudo-second-order reversible reaction constant

		Slope	Intercept	q_e	k_1	R^2
Pseudo-first-order adsorption kinetic	Amberlite IR-400 Duolite A-378	-0.06686 -0.03912	5.07656 3.4327	160.2219 30.96012	-0.00045 -0.00026	0.90037 0.6685
		Slope	Intercept	q_e	<i>k</i> ₂	R^2
Pseudo-second-order adsorption kinetic	Amberlite IR-400 Duolite A-378	0.01454 0.03554	0.09456 0.14975	68.77579 28.13731	0.002236 0.008435	0.99485 0.99554



Fig. 9. Amberlite IR-400 resin 3D response surfaces for: (a) concentration (ppm) and resin amount (g), (b) agitation rate (rpm) and resin amount (g), (c) agitation rate (rpm) and concentration (ppm) and 2D contour plots interaction between data of experiments on nitrate removal %: (aa) concentration (ppm) and resin amount (g), (bb) agitation rate (rpm) and resin amount (g) and (cc) agitation rate (rpm) and concentration (ppm).

resins indicate a better fit for pseudo-second-order than first-order as shown in Fig. 7.

The response surfaces for them are shown in Figs. 8–11.

4. Conclusions

Among the ion exchange resins tested, it was remarked that Amberlite IR-400 strong base anion resin was more responsive and more effective to nitrate adsorption, while Duolite A-378 weak base anion resin was right on target, especially when increased the amount of it in solution where the results were satisfactory. Thus, use the Duolite A-378 because it is an economical resin that is inexpensive compared to IR-400 resin in the nitrate-removal process. The influence of PH was investigated, and it fell within acceptable bounds. Additionally, there was an increased response for both types of ion exchange resins in removing nitrate by increasing the concentration as well as increasing the amount of resins. While a decrease in the removal rate was observed at high mixing speed; this is due to the ionic attraction force that is stronger at high electrolyte solution. A statistical model was developed for the percentage of nitrate removal that the two resins can perform under. Nitrate removal result showed that the removal ratio was (98.70%) for Amberlite IR-400 resin and (99.42%) for Duolite A-378 resin. The equilibrium data analysis specified that the Freundlich isotherm model was the best for describing the adsorption. While pseudo-second-order adsorption



Fig. 10. Duolite A-378 resin 3D response surfaces for: (a) pH and resin amount (g), (b) pH and concentration (ppm), (c) pH and agitation rate (rpm) and 2D contour plots interaction between data of experiments on nitrate removal %: (aa) pH and resin amount (g), (bb) pH and concentration (ppm) and (cc) pH and agitation rate (rpm).



Fig. 11. Duolite A-378 resin 3D response surfaces for: (a) concentration (ppm) and resin amount (g), (b) agitation rate (rpm) and resin amount (g), (c) agitation rate (rpm) and concentration (ppm) and 2D contour plots interaction between data of experiments on nitrate removal %: (aa) concentration (ppm) and resin amount (g), (bb) agitation rate (rpm) and resin amount (g) and (cc) agitation rate (rpm) and concentration (ppm).

kinetic was significantly appropriate to show the adsorption kinetics of Nitrate on the surface of the sorbent.

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