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# Simultaneous Removal of Nitrate and Phosphate Ions from Aqueous Solutions Using Fume Dust from Electric Arc Furnace Industrial Waste

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The removal of nitrate and phosphate ions from wastewater has a major concern nowadays. It is necessary to remove nitrate and phosphate ions from wastewater. Adsorption becomes a promising method for phosphate and nitrate ions removal from wastewater because of their availability, low cost, stability and high adsorption capacity. Fume Dust from an Electric Arc Furnace is an industrial waste produced from El-Masryeen Steel factory which is used as an adsorbent for simultaneous removal of nitrate and phosphate ions from an aqueous solution. Surface characterization is performed for the Fume Dust from an Electric Arc Furnace waste before and after adsorption process to ensure nitrate and phosphate ions adsorption and to predict the adsorption mechanisms for each of them. The effects of changing pH, initial nitrate ion concentration, initial phosphate ion concentration and adsorbent dose on the adsorption process efficiency are studied. Response-Surface Methodology-Central Composite Design statistical method is performed to reach the best conditions of the nitrate and phosphate ions removal percentages at neutral pH using Design Expert Software. The maximum nitrate and phosphate ions removal percentages (52.74 % and 92.38 %) are achieved using Fume Dust from an Electric Arc Furnace waste under these conditions; pH ~ 7, initial nitrate ion concentration = 6 mg/L, initial phosphate ion concentration = 1 mg/L, adsorbent dose = 6.5 g/L and contact time = 90 min.

## 1. Introduction

Excessive intake of nitrate and phosphate by humans in drinking water and food can induce stomach cancer (Ali et al., 2020). The denitrification from aqueous solutions can be achieved by various methods; biological denitrification, ion exchange, chemical reduction and reverse osmosis process using semi-permeable membrane (Berkessa et al., 2019). Biological treatment usually requires biomass waste disposal which is produced in sufficient amounts. Ion exchange treatment method produces concentrated nitrate rejection and exhausted resins (Ali et al., 2020). Reverse Osmosis disadvantages are high operating costs, complexity during operation and production of significant waste streams (Ezugbe and Rathilal, 2020). Utilization of some inorganic or organic based adsorbents is becoming desirable because of their high adsorption capacity, low cost and easy operation. Fume Dust from an Electric Arc Furnace FD-EAF is an industrial waste produced from an Electric Arc Furnace is usually used as a source of some valuable metals specially Zinc then this waste is recycled again in the steel industry (Sinaga et al., 2019). The main aim of this research is to use FD-EAF waste in the adsorption of nitrate and phosphate ions simultaneously from aqueous solutions then extract the valuable metals from it like Zinc. A detailed study is performed on kinetics and isotherm models on single-component and multi-component adsorption systems using FD-EAF waste and based on this study, it is concluded that the type of adsorption in this research is physical adsorption which will facilitate the desorption process of nitrate and phosphate ions without changes in the properties of the waste itself. And thus, multiple objectives will be achieved using FD-EAF waste including; simultaneous adsorption of nitrate and phosphate ions from aqueous solutions, extraction of valuable metals from it after desorption process and finally recycling of the waste again in the steel industry.

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## 2. Materials and methods

FTIR and EDX tests are performed before and after adsorption process to ensure the ability of FD-EAF waste to adsorb nitrate and phosphate ions from aqueous solutions. Design Expert software program is used to provide a statistical analysis which illustrates the best experiment conditions of simultaneous nitrate and phosphate ions removal at neutral pH. Also, it is used to explain clearly the effects of different experimental factors including; pH, adsorbent dose, contact time, initial nitrate ion concentration and phosphate ion concentration on the adsorption process performance.

### 2.1 Chemicals, characterization methods of FD-EAF and measurement of ions concentrations

FD-EAF is an industrial waste from El-Masryeen Steel factory produced from an Electric Arc Furnace in almost amount of 12 kg/t of steel produced. The surface functional groups of FD-EAF waste are recorded using Fourier Transformation Infrared (FTIR) spectra (Vertex 70 RAM II, Germany). The surface composition of FD-EAF waste is determined via Energy Dispersive X-ray (EDX) analysis (Quattro–Thermo Fisher Scientific Company, Netherland). All chemicals used in this study are analytical grade reagents including NaOH, HCl, $Mg(NO_3)_2$ $6H_2O$ and $Na_2HPO_4$, these reagents are purchased from Alahram laboratory chemicals Company. All solutions are prepared using double-distilled water. Experiments are performed in glass conical flasks which are shaken vigorously at 180 rpm using a laboratory shaker. The concentration of nitrate ion is measured using a UV/VIS spectrophotometer (UV-5100, Shanghai Metash Instruments Company, China) and phosphate ion is measured by a Water Conditioning Photometer device with kits.

## 3. Parameters of batch experiments for FD-EAF industrial waste

The parameters of the batch adsorption experiments using FD-EAF waste are chosen based on certain criteria, as represented in this section.

### 3.1 Initial pH of the solution

Initial pH is chosen between 2 to 12 in order to study the effect of pH on the adsorption at a wide range of pH.

### 3.2 Initial nitrate and phosphate ions concentrations

Initial nitrate and phosphate ions concentration is chosen in the range from 6 - 40 mg/L and from 1 - 10 mg/L, respectively, based on the maximum limits of wastewater discharge into the Nile River, which are mentioned in the Egyptian Environmental Law number 4 of the year 1994.

### 3.3 Contact time of FD-EAF waste

The contact time range 6 - 90 min is chosen based on experiments that are performed at pH ~ 7, adsorbent dose = 3.5 g/L, initial phosphate ion concentrations that ranged from 1 - 10 mg/L and initial nitrate ion concentrations that range from 6 - 40 mg/L. Through these experiments, the maximum removal percentages are achieved at a contact time of 90 min.

### 3.4 Adsorbent dose for FD-EAF waste

Preliminary experiments are conducted to check the effect of adsorbent dose on simultaneous phosphate and nitrate ions removal efficiency. The experiments are conducted at adsorbent doses ranging from 0.5 - 6.5 g/L, at fixed initial nitrate ion concentration of 6 mg/L, initial phosphate ion concentration of 1 mg/L, contact time of 90 min and pH ~ 7. The adsorbent dose from 0.5 - 6.5 g/L gives better nutrients removal efficiency, and beyond this range the residual nutrient concentration in aqueous solution increases, leading to a decrease in the nutrient removal percentages as represented in Table 1. The reason for this can be attributed to the accumulation of adsorbent when the adsorbent dose exceeds 6.5 g/L which results in decreasing in its specific surface area and, thus, decreasing in the removal efficiency of phosphate and nitrate ions from aqueous solutions.

*Table 1: Concentrations of phosphate and nitrate ions after adsorption versus FD-EAF adsorbent dose*

| Factor | | | | | | |
|---|---|---|---|---|---|---|
| Adsorbent dose (g/L) | 3.5 | 4 | 6.5 | 7 | 8 | 10 |
| Nitrate ion concentration (mg/L) | 3.4 | 3.3 | 2.9 | 3.1 | 3.36 | 3.4 |
| Phosphate ion concentration (mg/L) | 0.14 | 0.1 | 0.075 | 0.089 | 0.11 | 0.12 |
| % Nitrate ion removal | 43.4 | 44.5 | 50.5 | 48.3 | 44 | 43.5 |
| % Phosphate ion removal | 86 | 89.2 | 92.5 | 91.1 | 88.6 | 87.8 |

# 4. Results and discussion

## 4.1 Energy Dispersive X-ray (EDX)

Figure 1(a) represents the EDX analysis of the FD-EAF waste before adsorption of phosphate and nitrate ions, where it reflects the presence of Fe, O, C, Mg, Mn, Ca, Si, Al, Zn as primary elements. Figure 1(b) shows that two peaks of phosphate appear, proving that it is adsorbed successfully using the FD-EAF waste. Figure 1(c) shows that a small peak of nitrate appears, proving it is adsorbed successfully using the FD-EAF waste.

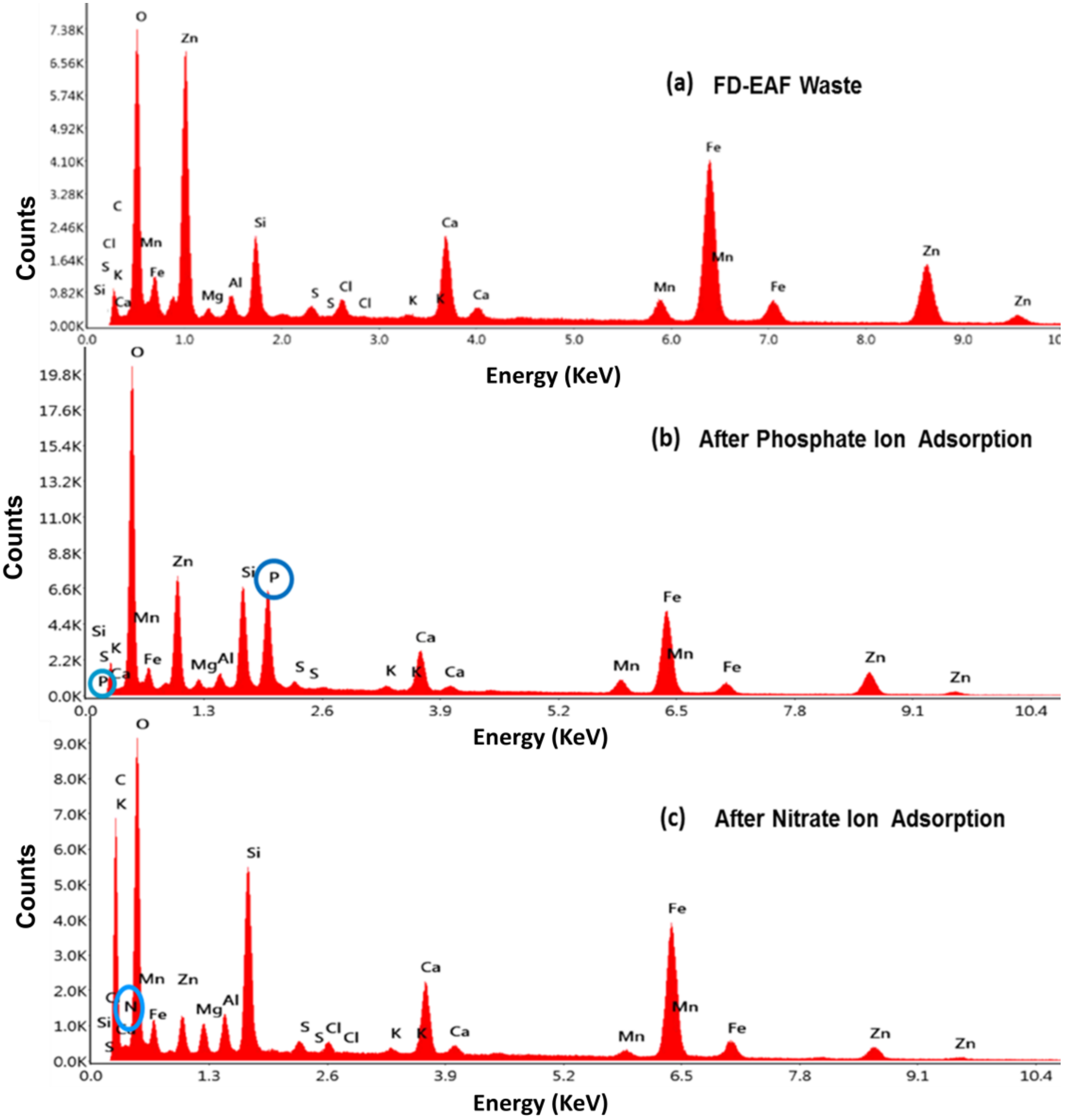

*Figure 1: EDX (a) FD-EAF waste, (b) after phosphate ion adsorption and (c) after nitrate ion adsorption*

## 4.2 Fourier Transform Infrared (FTIR)

On the basis of FTIR bands of (before) adsorption of nitrate and phosphate ions in both Figures 2(a) and 2(b); the positions of the vibrations at 3,485, 3,374.4 and 3,168.58 cm$^{-1}$ are the stretching vibrations of the (-OH) group (Coates, 2006). While vibration bands observed at 1,420.22, 1,002, and 937 cm$^{-1}$ reveal the presence of ($CO_3^-$), (Si-O) and (Al-O-OH) (Stuart, 2004). Vibration bands at 871.5 and 872.94 cm$^{-1}$ are attributed to iron oxides (Stuart, 2004). In Figure 2(a); new bands appear at 1,795.5, and 1,397.12 cm$^{-1}$ relate to nitrate ion (Bunaciu et al., 2015), which ensures, beside EDX analysis, the successful nitrate ion adsorption from an aqueous solution using FD-EAF waste. In Figure 2(b); a new strong vibration band appears at 1,031.8 cm$^{-1}$ which is related to the presence of phosphate ion (Stuart, 2004), and this ensures beside EDX analysis, the successful phosphate ion adsorption from an aqueous solution using FD-EAF waste.

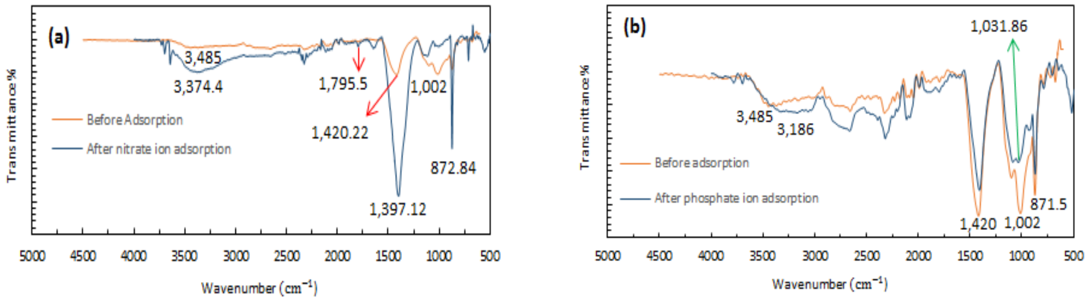

*Figure 2: FTIR analysis (a) after nitrate ion adsorption and (b) after phosphate ion adsorption*

### 4.3 Regression model equation development

The quadratic models for phosphate ion removal % and nitrate ion removal % after removal of insignificant terms to increase the model's accuracy are represented in the following Eq(1) and Eq(2):

$$Y1 = +77.44 – 14.03\ B + 4.72\ C + 3.87\ D – 5.11\ C^2 \tag{1}$$

$$Y2 = +36.77 – 12.54\ A + 5.25\ C + 3.5\ D – 6.22\ A^2 + 3.63\ C^2 – 4.44\ D^2 \tag{2}$$

Where $Y_1$ and $Y_2$ represent the removal percentages of phosphate ion and nitrate ion. A, B, C and D are the initial nitrate ion concentration, initial phosphate ion concentration, adsorbent dose and contact time.

### 4.4 Effects of initial phosphate ion and initial nitrate ion concentrations

Figure 3(a) shows that phosphate ion removal percent decreases from 90 % to 70 % with increasing phosphate ion concentration from 1.5 - 6.7 mg/L at different levels of contact time and under these fixed conditions; pH ~ 7, adsorbent dose = 3.5 g/L and initial nitrate ion concentration = 23 mg/L. The reason can be attributed to the decrease in the available active sites on the adsorbent surface while increasing the phosphate ion concentration. Figure 3(b) represents that the nitrate ion removal decreases from 40 % to 20 % with increasing initial nitrate ion concentration from 6 - 36 mg/L at different levels of contact time and under these fixed conditions; pH ~ 7, adsorbent dose = 3.5 g/L, initial phosphate ion concentration = 5.5 mg/L. The presence of nitrate ion with phosphate ion in the same aqueous solution has a negligible effect on phosphate ion removal % as it appears as an insignificant factor in the ANOVA.

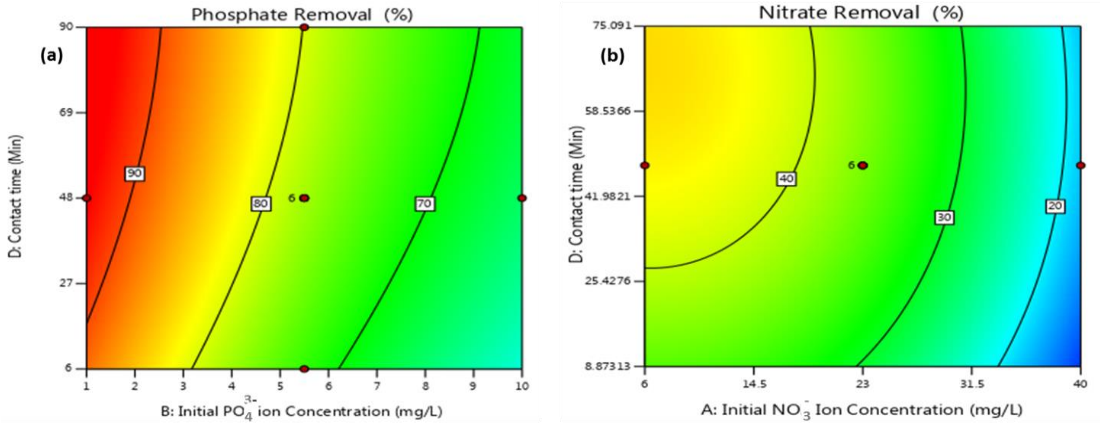

*Figure 3: Contour plots; (a) initial phosphate ion concentration and contact time versus phosphate ion removal %, (b) initial nitrate ion concentration and contact time versus nitrate ion removal %*

### 4.5 Effect of adsorbent dose

When the adsorbent dose increases, its specific surface area increases, leading to an increase in the availability of the reactive site for phosphate and nitrate ions adsorption. Figure 4(a) represents that an increase in the adsorbent dose from 0.63 - 3.51 g/L leads to an increase in the phosphate ion removal from 64 % to 76 %. While Figure 4(b) represents that an increase in the adsorbent dose from 3.3 - 6 g/L leads to an increase in the nitrate ion removal from 36 % to 44 % at different levels of time and under these fixed conditions; pH ~ 7, initial nitrate ion concentration = 23 mg/L and initial phosphate ion concentration = 5.5 mg/L.

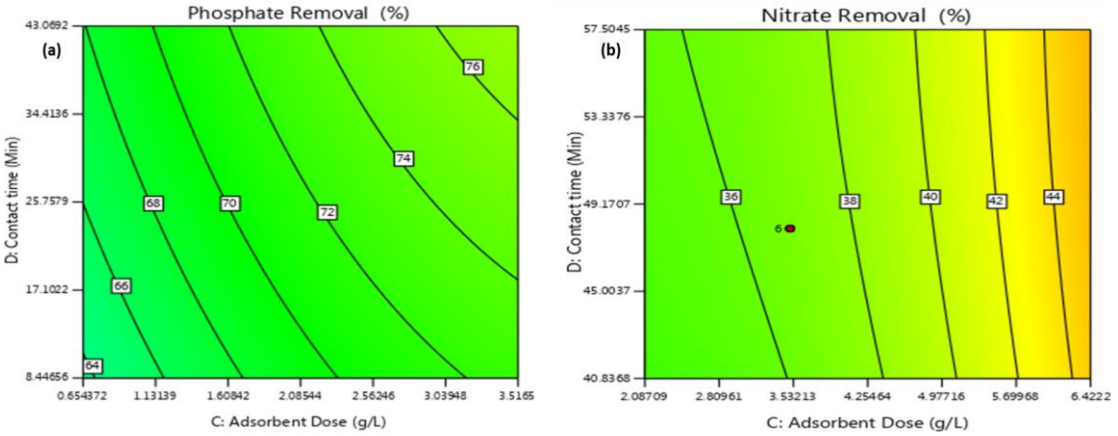

*Figure 4: Contour plots; (a) adsorbent dose and contact time versus the removal % of phosphate ion and (b) adsorbent dose and contact time versus the removal % of nitrate ion*

### 4.6 Effect of pH on simultaneous removal of phosphate and nitrate ions from aqueous solutions

As represented in Figure 5 in acidic medium; the dominant phosphate ion is $H_2PO_4^-$ which has the less negativity effect among different forms of phosphate ions. The % removal of phosphate ion increases from 57 % to 72 %. In a pH range from 6.1 to 7.5; there is a slight decrease in the phosphate ion removal % as it decreases from 72 % to 67.6 %, and the removal of nitrate ion increases from 2.1 % to 24.5 %. While in the basic medium; $HPO_4^{2-}$ ion becomes the dominant ion, and with further increase in the solution's pH the dominant ion becomes $PO_4^{3-}$ and the deprotonation rate increases. The removal % of phosphate ion decreases from 67.6 % to 12.8 % and the nitrate ion removal % decreases from 18.75 % to 2.44 %.

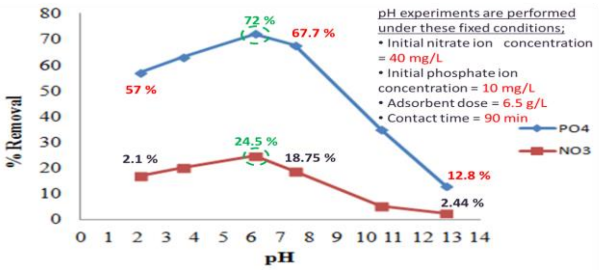

*Figure 5: Effect of pH on simultaneous removal of phosphate and nitrate ions from aqueous solutions*

### 4.7 The best experiment conditions of simultaneous nitrate and phosphate ions removal at neutral pH

The process goals are chosen to reach the best conditions for simultaneous removal of nitrate and phosphate ions from aqueous solutions using FD-EAF industrial waste at neutral pH, as represented in Table 2.

*Table 2: Experimental constraints of simultaneous removal of phosphate and nitrate from aqueous solutions*

| Factor | Goal | Lower Limit | Upper Limit |
|---|---|---|---|
| Nitrate initial concentration (mg/L) | minimize | 6 | 40 |
| Phosphate initial concentration (mg/L) | minimize | 1 | 10 |
| Adsorbent dose (g/L) | maximize | 0.5 | 6.5 |
| Contact time (min) | maximize | 6 | 90 |
| % Nitrate removal | maximize | 9.43 | 53.61 |
| % Phosphate removal | maximize | 46.6 | 93.32 |

Figure 6 (a) and (b) represent the best removal percentages for phosphate and nitrate ions from aqueous solutions 92.38 % and 52.74 %, which are achieved at these conditions; pH ~ 7, initial nitrate ion concentration = 6 mg/L, initial phosphate ion concentration = 1 mg/L, adsorbent dose = 6.5 g/L and contact time = 90 min.

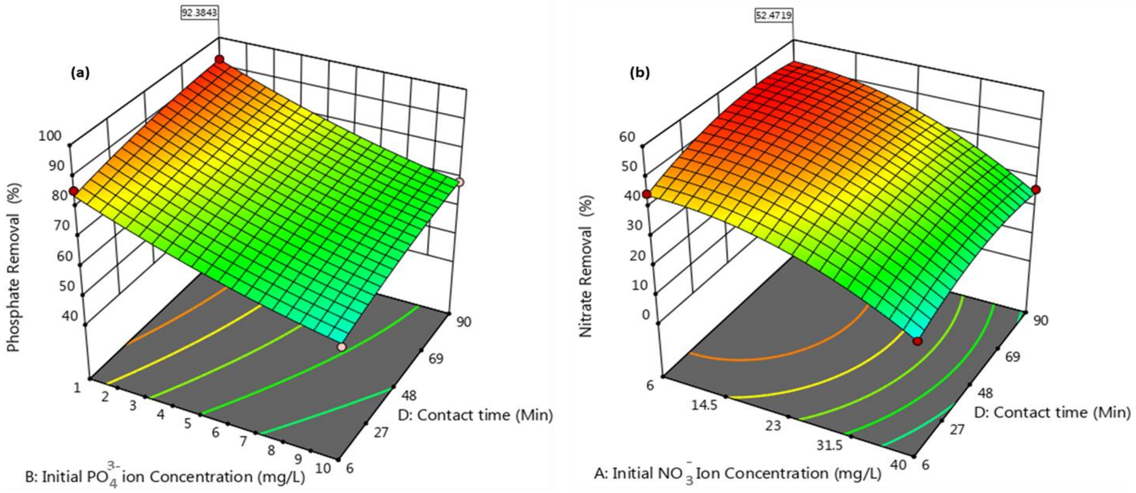

*Figure 6: Response surface for the best removal percentages for (a) phosphate ion and (b) nitrate ion*

## 5. Conclusion

Removal of phosphate and nitrate decreases their harmful effects on the human and water ecosystem. FD-EAF waste is chosen to be used for simultaneous removal of nitrate and phosphate ions from aqueous solutions based on making rough adsorption batch experiments on different ranges of pH, adsorbent dose, initial nitrate ion and initial phosphate ion concentrations. EDX and FTIR tests are performed to ensure that FD-EAF waste adsorbs phosphate and nitrate ions effectively from aqueous solutions. Phosphate ion removal decreases from 90 % to 70 % with increasing phosphate ion concentration from 1.5 - 6.7 mg/L at different levels of contact time and under these fixed conditions; pH ~ 7, adsorbent dose = 3.5 g/L and initial nitrate ion concentration = 23 mg/L. The increase in the adsorbent dose from 0.63 - 3.51 g/L has a positive effect on the phosphate ion removal % as it increases from 64 % to 76 % at different levels of time and under these fixed conditions; pH ~ 7, initial nitrate ion concentration = 23 mg/L and initial phosphate ion concentration = 5.5 mg/L. When nitrate and phosphate ions have existed in the same solution, the maximum nitrate and phosphate ions removal percentages are 52.74 % and 92.38 %, which indicates that FD-EAF waste has a higher affinity towards phosphate adsorption. A detailed study on kinetics and isotherm models for single-nitrate ion, single-phosphate ion adsorption systems and multi-component adsorption system using FD-EAF industrial waste are achieved and it will be published like a future work. Also, desorption of nitrate and phosphate ions from FD-EAF waste and studying its kinetics will be performed like a future work.

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