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# Synthesis of Magnetic Iron Oxide ($Fe_3O_4$) Nanoparticles Modified with Common wormwood Extract and Examination of its Application for Removal of Tetracycline from Aqueous Solutions

**Abstract**

The presence of antibiotics in wastewater is of the problems of advanced societies receiving great attention in recent years and using nanotechnology is undoubtedly very effective in reaching this goal. The purpose was the synthesis of modified iron oxide ($Fe_3O_4$) magnetic nanoparticles with common wormwood extract and its efficiency for removing tetracycline antibiotics from aqueous solutions. Synthesis of $Fe_3O_4$ magnetic nanoparticles was conducted to remove tetracycline by co-precipitation method. FT-IR, FESEM, and XRD TEM techniques were used to identify the synthesized nanoparticles. The parameters examined in the study were removal time, pH, and the nanoparticle value, the intervals of each of which were 15-45 minutes, 5-9, and 0.1-0.05 grams, respectively. The results obtained from the real sample indicated 70% removal and thus the removal percentage increased with the increase in nanoparticles to 0.1 g. Under standard conditions, 94.78% and real samples of up to 71% of tetracycline were removed from the environment. The results showed that a significant percentage of tetracycline could be removed with magnetic $Fe_3O_4$ nanoparticles modified with common wormwood extract turning the study into a practical goal in treatment plants.

**Keywords:** Antibiotic, Magnetic nanoparticles, $Fe_3O_4$, Iron oxide, Common wormwood

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## Introduction

Tetracycline is a class of antibiotics of natural and ancient origin. This is one of the first prescribed drugs in many cases like eye infections, acne, and so on. Thus, the more a drug is used, the more likely it is that the drug or its metabolite will be present in the wastewater, but the important thing is that tetracycline can remain active in the wastewater and in a stage of treatment called nitrification after excretion from the human and animal body and kill the necessary bacteria. Thus, one can state that the presence of this antibiotic has a significant effect on the purification process. The interdisciplinary nature of nanoscience and nanotechnology as the ability to produce new materials and tools and systems with precision atoms and molecules will sooner or later affect the medical and health departments, including the drug delivery system inside the body. Drug use is currently on a large scale so most specific cells in the body need medication. Nowadays, nanoparticles have become very popular given their extensive applications in biology, pharmaceutical, and medicine. Structurally, their size is in the range of 100 nanometers. A wide range of drugs like small hydrophilic and hydrophobic drugs, vaccines, and molecules of biological nanoparticles have been guided by these nanoparticles (8). In the new approach, the drug is directed directly to specific cells with new injection tools and will be delivered to the needed place. By the same mechanism, small and large diseases can be diagnosed and treated at the start of their development. The presence of drug compounds and residues in the environment, especially water sources given their stability and non-degradability is an important issue in the environment. Antibiotics, along with domestic wastewater, pharmaceutical wastewater, wastewater from hospitals and veterinary clinics, agricultural products, and fish ponds have significantly entered water resources and the environment. Tetracyclines are the second most common group of antibiotics produced and consumed worldwide, which are obtained naturally by the fermentation of some fungi or by semi-synthetic processes, effective against many microorganisms and indiscriminately. The presence of these antibiotics in the environment including the aquatic environment has caused different reactions from simple allergies to in some cases direct toxicity. Moreover, the presence of these drugs in the environment leads to the development of antibiotic-resistant pathogens, potentially threatening ecosystem performance and human health. Different physical, chemical, and biological methods have been used to remove these contaminants from the aquatic environment. Different studies have shown that adsorption methods, electrocoagulation process, and membrane process can be used to remove these compounds, but given their low efficiency, high investment cost, and difficult management and maintenance, these methods are not cost-effective (1). In recent years, magnetic adsorbate has been widely used to remove organic contaminants from heavy metals and biomolecules. Co-precipitation and electrochemical synthesis are the most important methods usually used for the synthesis of magnetic nanoparticles (2). In recent years, magnetic particles have been considered an adsorbent, given their magnetic properties and their separation by magnets, a large number of active adsorption sites on the surface, and high contaminant removal efficiency. Here, iron oxide nanoparticles have been considered effective adsorbents to remove contaminants from the aquatic environment, and in many cases, aqueous media have been used to remove organic contaminants and heavy metals . Scholars in different fields like physics, medicine, materials, and biology have become interested in research on iron oxide nanoparticles. This is because iron oxide nanoparticles have multiple properties, like being small, super-magnetic size, and low toxicity. Here, $Fe_3O_4$ oxide nanoparticles (magnetite), as the most important group of magnetic particles, not only given their extraordinary properties like ideal size and suitable physical and chemical properties, but also high biocompatibility and super-paramagnetic properties are of interest to researchers (2). Using as drug carriers in the core core-shell drug delivery systems as a material used in the resonant reaction due to changes in the magnetic field and the creation of gram-hyperthermia are other applications of magnetic super-paramagnetic particles. Water solubility and superparamagnetic properties of these particles, associated with high saturation magnetization (Ms), have made it possible to combine with biomolecules to target the release of these materials. Creating a suitable organic or inorganic coating on the surface of the super-paramagnetic core of magnetite increases the half-life of these particles in the bloodstream and improves their effectiveness by delaying the clearance of magnetite nanoparticles (3).

Moreover, the presence of microorganisms and especially bacteria in food are of great importance for public health and food quality control. Pathogenic microorganisms in food cause many financial and human losses annually in the world. In addition, food spoilage because of the growth of microorganisms is still considered a problem in the food industry. One way to control the growth of bacteria in food is to use preservatives and antimicrobial compounds. Chemical preservatives have been used for a long time to prevent the growth or killing of some harmful microorganisms. Nowadays, however, given the increasing level of public awareness and concern over the side effects of chemical preservatives, the tendency to consume products without natural preservatives is increasing. Hence, in recent years, many studies have been conducted on natural preservatives like essential oils and plant extracts. Extracts and essential oils of medicinal plants and their constituents have known antibacterial effects. Among these plants, one can state a kind of native plant of Iran called common wormwood. Common wormwood (scientific name: Artemisia absinthium) belongs to the Asteraceae family and belongs to the Asteroid family and is one of the native aromatic plants, especially in the northern and eastern regions of Iran. In traditional Iranian medicine, the infusion of this plant has been used to treat liver failure, sore throat, ear infections, constipation, and chronic diarrhea, to heal wounds, and as an anthelmintic and antiseptic drug (4).

Suking Wu et al. (2020) managed to degrade tetracycline using visible photocatalytic light $TiO_2$. $TiO_2$ will be used extensively as a photocatalyst for the degradation of antibiotic residues in UV-irradiated water where photocatalysis of visible light is inefficient given its large band gap. The results indicated that the decomposition of TC may be visible under light irradiation more than $TiO_2$. Moreover, 1.25 of the removal efficiency for TC was obtained even under 700 nm light irradiation (5).

implemented straight design - z 1d / 2d wo2.72 / znin2s4 hybrid photocatalysts with very low light visible photodegradation to remove tetracycline hydrochloride. Environmental concerns over the serious contamination of antibiotic wastewater are increasing. Here, a series of direct design hybrid photocatalysts (Z-WO2.72 / ZnIn2S4 WOZi) composed of one-dimensional (1D) WO2.72 (WO) nanorods and two-dimensional (2D) ZnIn2S4 (ZIS) nanoparticles have been designed and constructed. For TCH tetracycline hydrochloride, degradation was done without the presence of solid-state electron mediators.

The crystalline phase, chemical composition, morphology, optical properties, and photocatalytic activity of the prepared samples BET-XRD-XPS-SEM-HRTEM, UV-VIS DRS, and PL were identified. Photocatalytic activity can be obtained by modulating the molar ratio between WO and nanoparticles (6).

Yang Kong et al. (2020) managed to remove tetracycline by alginate-graphene by bonding two metals through an advanced Fenton reaction. Polymer hydrogels often have limited catalytic activity and stability in Fenton catalysis. For the first time, the preparation of a bimetallic alginate hydrogel with two-way bonding using graphene oxide facilitated the redox (III), Fe (II) / Fe cycle. Alginate-GO hydrogels with double bonding can degrade tetracycline much faster within the first 10 minutes. Hydrogel Alginate GO-GO-Fe-M hydrogel with two cross-metals could TC much faster during the initial 10 minutes than single-cross hydrogel alginate bonding GO-M iron (II) iron (III) La (III) sol (III) and co (AG) -M) less iron secretion during reaction Fenton. For the treatment of crude water which has a high percentage of protein, TC AG-Fe-Ce hydrogel will significantly reduce the molecular weight of the dissolved organic matter. With low cost, high catalytic activity, and easy stability and separation, alginate-based hydrogels of great potential are used as a Fenton catalyst (7).

Jacob Zidtra et al. (2020) used inter-structured cellular silica foam materials for lactase immobilization and tetracycline removal. Given its exceptional textural, morphological, and mechanical properties, the cellular foam between structured

MCf silica is a suitable support material for enzyme immobilization. Nonetheless, the simple and efficient protocol of enzyme immobilization using MCF is usually ignored (8).
Yanting Wang et al. (2020) studied the removal of tetracycline adsorbent by a stable servomite substrate from bentonite / red mud / sawdust pine. In this study, a type of CWs Ben-Rm-Ps ceramite was prepared to remove TC in dilute. Ben ceramite / Rm / Ps-op with ben conditions: Rm: Ps = 4: 1: 0.9, preheating temperature 240°C, preheating time 20 minutes, calculation temperature 1150°C, and calculation time 14 minutes, TC absorption simulated by Ben / Rm / Ps-op and the maximum adsorption capacity can reach 2.5602 mg / g (9).
Hong Cheng Wang et al. (2020) performed a green synthesis of RGO / $Fe_3O_4$ / PANI nanocomposites to increase the absorption of electromagnetic waves. In sum, a simple, green, and scalable method was developed for rGO / $Fe_3O_4$ / PANI composites. Aniline monomer was loaded on the surface of the nanocomposite ($Fe_3O_4$ rGO) as a reducing agent to reduce the graphene oxide, followed by oxidation of the polymerization. The rGO / Fe3O4 / PANI composite has a relatively strong correlation between dielectric drop and magnetic drop. The nanocomposite with multiple interfaces provides many scattering points for electromagnetic waves. The maximum reflection loss of the rGO / Fe3O4 / PANI composite is -51.5 dB at 7.4 GHz with a thickness of 3 mm and the absorption bandwidth below -10 dB is equal to 4.2 GHz. It is believed that such a composite could be a potential material for electromagnetic wave absorption with different advantages like lightweight, high-efficiency absorption, and suitable preparation (10).
Given the above, the study tries to answer the question of whether it is possible to modify iron oxide nanoparticles with wormwood extract. Can nanoparticles based on magnetic nanoparticles and their functionalization and modification with common wormwood extract be used in drug delivery or drug absorption?

**Materials and methods**

Thirty grams of wormwood were prepared to extract. Ferric chloride hexahydrate ($FeCl_3.6\ H_2O$) and ferrous tetrahydrate chloride ($FeCl_2.4H_2O$) were bought from Merck Co., Germany for the production of magnetic iron oxide nanoparticles in the form of a brown solid. Sodium hydroxide produced by Merck Co., Germany was used to producing magnetic iron oxide nanoparticles. Deionized water with 18.0153 gr / mol was prepared and used from Chem Lab Belgium for the solution. Two options are suggested for selecting an aqueous medium for testing distilled water and deionized water. Although the composition of distilled water is closer to real aqueous media and helps analyze more accurately and realistically, given the reduced accuracy of the results of atomic absorption spectroscopy in the presence of additional ions and even in some cases changing the results and reducing the accuracy of deionized water that lacks any ion species was selected for our experiments.
Tetracycline with $C_{22}H_{24}N_2O_8$ formula prepared by Sina Daru Company was used for removal from aqueous solutions in the experiment.
UV-Vis device was used to measure the absorption of the solution and the rate of degradation of the tetracycline drug from the company (Jenway Hakim Azma Tajhiz).
FT-IR spectrum of magnetic nanoparticles was taken from Shimadzu FTIR-8400S to analyze the results.
A heater stiller by Heidolph Co. was used to provide a temperature of 70-80 degrees and stir the solution.
PH meter by Sana Co. was used to adjust the pH in the designed experiments.
Deionized water as a solvent used to dissolve PVA and Fe $(NO_3)_3.9H_2O$ to produce a polymer solution was prepared from the ionized water production machine of Youngling Korea.
X pert MPD model of Philips Co. in the Netherlands was used to get the XRD spectrum of magnetic nanoparticles.

**Methodology**

The coprecipitation method was used to synthesize magnetic iron oxide nanoparticles. $FeCl_2$ and $FeCl_3$ were combined to prepare $Fe_3O_4$ iron oxide magnetic nanoparticles. During laboratory research, $FeCl_2.4H_2O$ and $FeCl_3.6H_2O$ were first added to 100 ml of deionized water and mixed at the appropriate temperature under the influence of nitrogen by a mechanical stirrer and then a proper volume of ammonium hydroxide was injected into the mixture, and later on, separated from the nanoparticles by adding base to the iron salt solution and forming a chemical precipitate. Then common wormwood extract was refluxed with synthesized magnetic nanoparticles for 6 hours. The solution was then centrifuged and the resulting precipitate was separated. This nanocatalyst was then identified using electron microscopes and other detection techniques like FT-IR-FESEM-TEM-XRD and used as a recycled and heterogeneous nanocatalyst in the reaction of tetracycline removal from aqueous media. We placed the drug and obtained it in it for the right time. The possible parameters affecting the removal of antibiotics like the value of nanoparticles, temperature, and time were examined and optimized by chemometric design to examine the removal of antibiotics by synthesized nanoparticles to reach the highest efficiency. The removal of antibiotics in an aqueous medium was examined under optimal conditions. UV-Vis spectroscopy was used to measure the degradation of antibiotic removal bonds. Super magnetic iron nanoparticles are the most widely used inorganic materials used in pharmacy that can be prepared using chemical methods like co-precipitation or biological methods.

**Optimization and designing the experiments to study the factors affecting the absorption of tetracycline**

A designed test is the one in which targeted changes are applied to process input variables so that a change in response can be observed and the reasons for it identified.

The effect of several factors on an experiment necessitates the optimization to be performed on that experiment to find which factor has the greatest effect on the experiment process.

Overall, one can state that the philosophy of chemometrics is two: examining the parameters with statistically significant effects on the results and obtaining the optimal values of significant parameters.

Ultimately, our overall goal of optimization is to be able to select the conditions that reach maximum efficiency with the least energy in terms of time, cost, and so on.

As various factors can affect adsorption and its efficiency, the study selected the possible factors according to previous studies and preliminary tests to reach maximum absorption.

The parameters affecting the adsorption of tetracycline by $Fe_3O_4$ magnetic nanoparticles are as follows:

1- The time of nanoparticle contact with the drug

2- Ambient pH

3- The value of magnetic nanoparticles $Fe_3O_4$

Other factors like temperature or the effect of salt (ion strength effect) can be effective, yet they did not have a statistically significant effect on the experiment process according to previous experiments, so there was no need to study these factors again.

Box–Behnken design for this experiment has 15 randomized experiments to determine the simultaneous effect of the gram value of adsorbent $Fe_3O_4$, pH, and contact time. All the experiments were done on 10 ccs of the standard tetracycline solution at a concentration of 10 ppm in Table 1

**Table 1: Box-Behnken test design**

| **Experiment** | **Nanoparticles in grams** | **pH** | **Contact time** |
|---|---|---|---|
| **1** | 0.05 | 9 | 30 |
| **2** | 0.05 | 7 | 45 |
| **3** | 0.1 | 9 | 30 |
| **4** | 0.075 | 9 | 15 |
| **5** | 0.1 | 7 | 45 |
| **6** | 0.075 | 5 | 45 |
| **7** | 0.05 | 7 | 15 |
| **8** | 0.075 | 7 | 30 |
| **9** | 0.1 | 5 | 30 |
| **10** | 0.05 | 5 | 30 |
| **11** | 0.075 | 9 | 45 |
| **12** | 0.1 | 7 | 15 |
| **13** | 0.075 | 7 | 30 |
| **14** | 0.075 | 7 | 30 |
| **15** | 0.075 | 5 | 15 |

**Adsorption test under optimal conditions**

The optimal conditions for reaching maximum adsorption of tetracycline on $Fe_3O_4$ adsorbent modified with common wormwood extract were determined using Box–Behnken design after performing 15 designed experiments. The adsorption experiment was repeated 3 times under optimal conditions to evaluate the reproducibility of the results.

**Examining the effect of adsorbent on tetracycline adsorption from a real sample**

Laboratory wastewater or hospital wastewater can be used as a real sample.

The first step in working on the wastewater is filtration and centrifugation to separate and suspend all suspended particles in the solution. These are very effective in obtaining the correct absorption number. Then UV absorption of the solution in tetracycline $\lambda_{max}$ is measured with a device. The adsorption number obtained from the wastewater is compared with the adsorption number of the standard 10 ppm tetracycline solution, and then, either wastewater should be diluted or the optimal conditions changed accordingly depending on the difference between these numbers.

After diluting the wastewater, 10 ccs of it is removed and poured into a beaker and then added to the nanoparticle solution according to the optimal conditions obtained from the test design, and the tetracycline removal process is done just like removal under standard conditions.

Finally, the adsorption of the solution is taken to measure the removal of tetracycline from the real sample.

## Results

### Plotting a calibration curve

Calibration curves were plotted in Excel at the desired concentrations for tetracycline. According to previous studies and results on the drug and scanning wavelength $\lambda_{max} = 264$ nm was selected in **Figure 1**

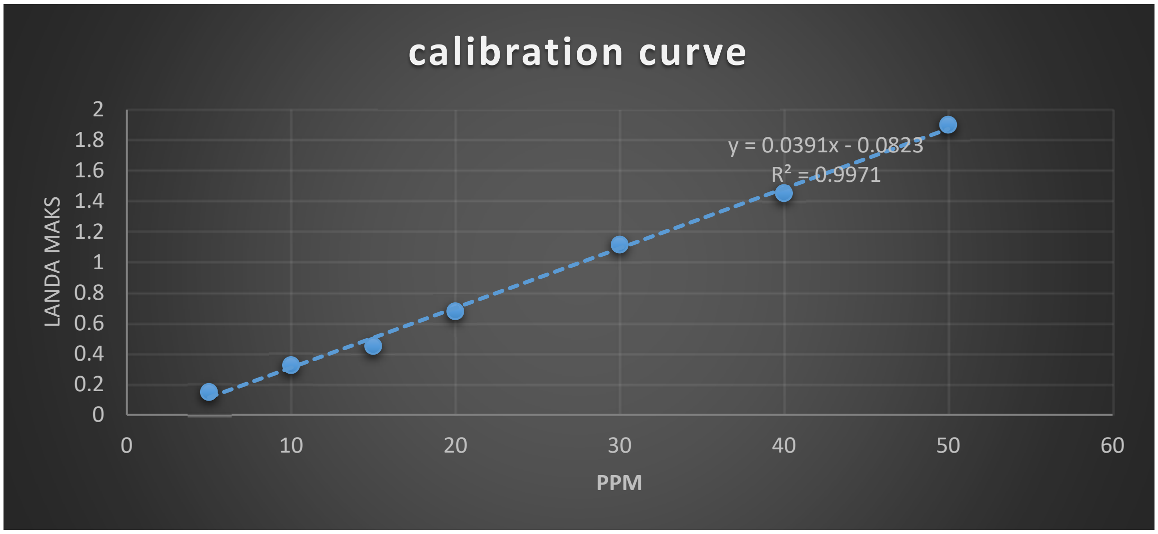



**Figure 1) Calibration curve for various concentrations of tetracycline**

Based on the results obtained from the calibration curve, a concentration of 10 ppm was considered for optimization experiments. Thus, the concentration of tetracycline remains constant in all states where the effectiveness of each parameter is examined.

All steps are examined at $\lambda_{max} = 264$ nm.

### Optimization of parameters affecting adsorption by Box–Behnken diagram

Box–Behnken design was used in the study to examine the parameters affecting the rate of tetracycline adsorption simultaneously. The parameters examined in the study were pH, a gram of nanoparticles, and time.

The design has 15 experiments performed according to Table (2).

The adsorption value of tetracycline with a concentration of 10 ppm before the use of nano adsorbent is equal to 0.326 at $\lambda_{max} = 264$ nm.

The removal percentage is obtained according to the following formula:

$$R = \frac{A_0 - A_1}{A_0} \times 100$$

R: Removal percentage

$A_0$: Initial adsorption

$A_1$: Secondary adsorption (adsorption after the effect of nano-adsorbent)

**Table 2: The results of absorption of Box–Behnken design experiments for three selected factors**

| **Experiment** | **Nanoparticle in grams** | **pH** | **Contact time** | **Removal efficiency percentage** |
|---|---|---|---|---|
| **1** | 0.05 | 9 | 30 | 74.53 |
| **2** | 0.05 | 7 | 45 | 88.34 |
| **3** | 0.1 | 9 | 30 | 87.73 |
| **4** | 0.075 | 9 | 15 | 94.78 |
| **5** | 0.1 | 7 | 45 | 92.33 |
| **6** | 0.075 | 5 | 45 | 88.95 |
| **7** | 0.05 | 7 | 15 | 86.50 |
| **8** | 0.075 | 7 | 30 | 90.96 |
| **9** | 0.1 | 5 | 30 | 92.63 |
| **10** | 0.05 | 5 | 30 | 88.65 |
| **11** | 0.075 | 9 | 45 | 88.65 |
| **12** | 0.1 | 7 | 15 | 93.25 |

| | | | | |
|---|---|---|---|---|
| **13** | 0.075 | 7 | 30 | 91.94 |
| **14** | 0.075 | 7 | 30 | 91.18 |
| **15** | 0.075 | 5 | 15 | 93.55 |

The results obtained were given to the software and the following analyses were obtained:

**Pareto Chart**

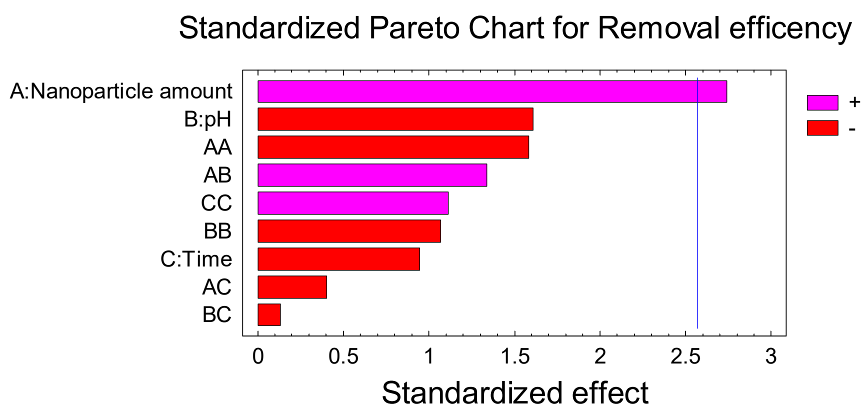



**Chart 2) Pareto Chart**

Pareto Chart is the ANOVA table, which differs in appearance and as it is in the shape of an image unrelated to numbers and is easier to understand than it is, it has received more attention these days. The blue line in this chart is the confidence level for examining the effect of factors. P = 0.05 means a 95% probability of influencing factors statistically affecting the results. Indeed, it shows which parameters are negative and which are positive. Pink shows the positive effect and the red the negative. Each of these factors crossing the blue line shows that it can affect our work, so the value of nanoparticles has a positive effect, the higher the value of nanoparticles, the better the result or efficiency of work. Moreover, the only statistically influential factor according to this figure was the value of nanoparticles, with other parameters having an effect but not statistically significant.

The positive and negative effects of the parameters on the results show whether these effects are significant or not.

**Main effects chart**

It examines the factors as a figure.

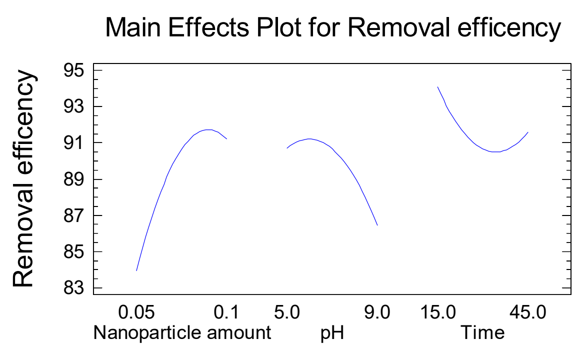



**Chart 3) Main effect chart**

According to Figure 3, one can say:

• The effect of each parameter is shown as a shape on the result. Only nanoparticles are significant and pH and time do not have much effect. As the nanoparticles increase, the efficiency increases. Indeed, a descending graph shows errors in the study. The optimal point is selected and from there on, no matter how many nanoparticles are added, it is useless because the absorption of the drug has been done to its maximum, and even if the concentrations increase, the drug may interfere with the nanoparticles and reduce absorption.

• At pH, first it increases then decreases as the best efficiency is in neutral conditions. If we increase the pH, it will decrease efficiency.

• The maximum efficiency was observed in 15 minutes and the efficiency decreased with increasing time.

**Interaction diagram**

• This diagram shows the interaction between the parameters.
• They intersect each other - 2 parameters interact with each other. However, if it is not cut, it means that they do not have a Nash diagram. Indeed, this figure shows which parameters interact. Each of these lines interacts with each other at the point of intersection if they intersect. According to Figure AB, they only interact, but this interaction is not significant according to the Pareto chart.

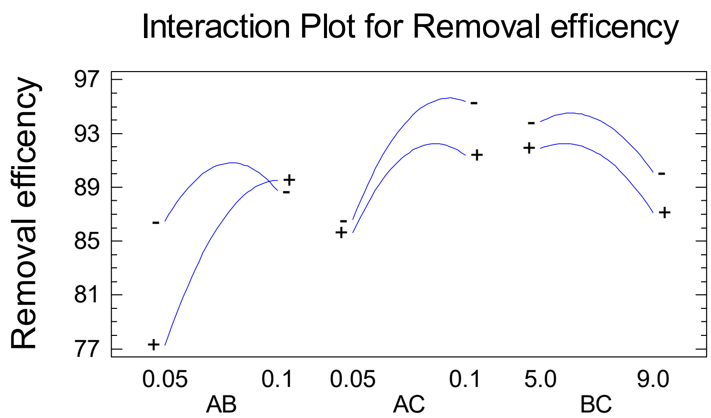



**Figure 4) Interaction plot for removal efficiency**

According to the Pareto Chart, the interaction of factors like AB, BC, and AC is below the main line and does not have statistically significant effects. In this chart, this result is expressed in another way. In the interaction diagram, if the lines intersect somewhere, it shows that the two parameters affect each other (Diagram 4).

**Normal probability diagram**

This diagram shows the errors to some extent.

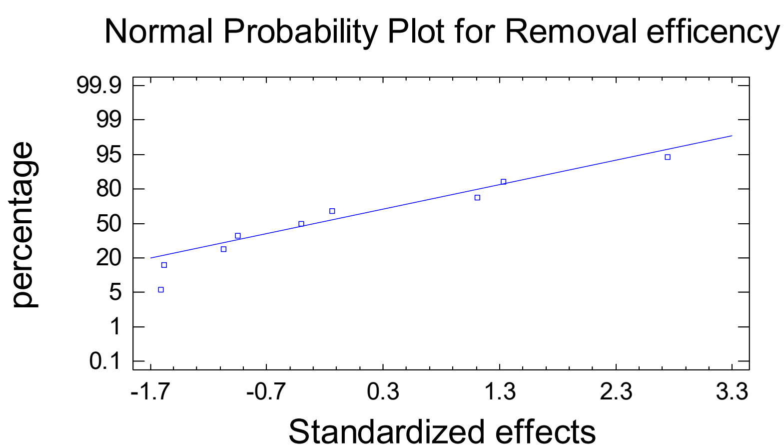



**Figure 5) Normal probability**

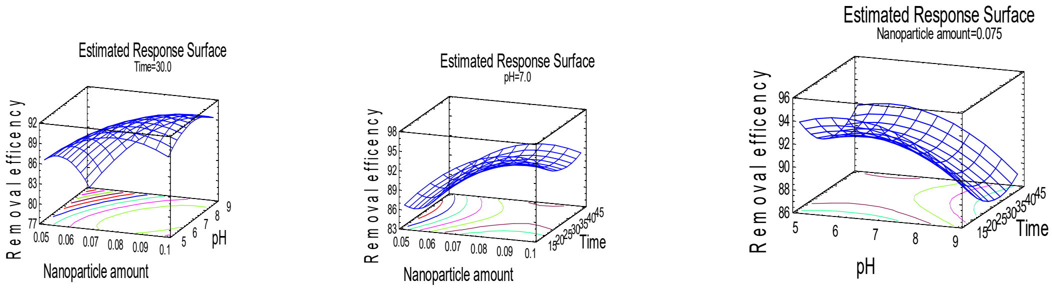
If the number of points above and below the normal line is approximately equal, it shows that there are no systematic errors and the errors that occurred are all random.

**Response surface curves**

Adsorption of the drug on the common wormwood adsorbent was examined using a pH of 5 to 9. At PH = 7, we witness the maximum adsorption. Indeed, one can state that at PH = 7, it forms the most stable complex and the highest adsorption of nanoparticles at 0.09 has caused the most stable adsorption of tetracycline. It puts the factors 2 by 2 next to each other and keeps the third factor constant, and as it examines 2 factors with one response, the shapes of the figures become 3D. One can state that the response surface curve is a 3D chart of the Main Effects.

The highest efficiency in these curves is the highest peak, which is the optimal point.

Figure 6 was plotted using Statgrafics software to illustrate the interaction and relationship between tetracycline removal and three factors (pH, @ $Fe3O4$, contact time),.




A B C

**Figure 6: a) Interaction of pH and the value of nanoparticles at constant time, b) Interaction of time and the value of nanoparticles at constant pH, c) Interaction of time PH at a constant value**

In 15 minutes, the absorption has reached the maximum value and the absorption of the drug in the value of 0.09 g has a maximum absorption and the absorption slightly decreased with increase and decrease.

Absorption reached the maximum value in 15 minutes with the highest absorption at PH = 7, so 15 minutes and PH = 7 were selected for optimal conditions.

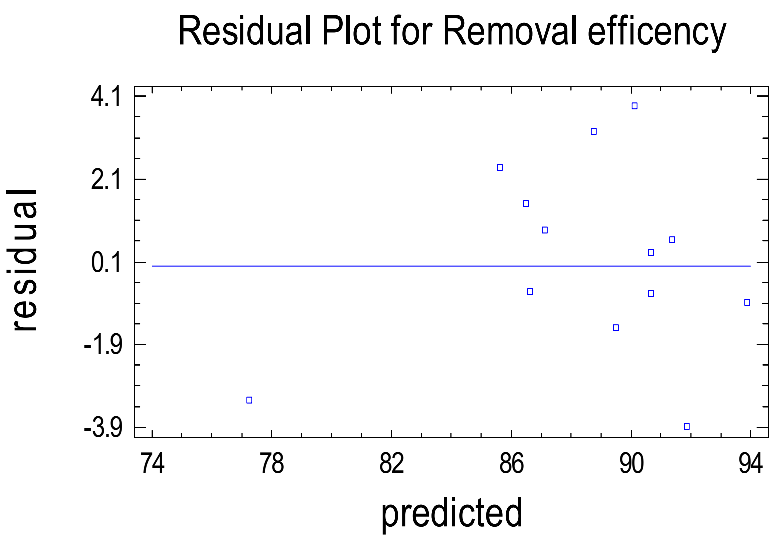



**Figure 8) Residual Plot for removal efficiency**

Figure 8 shows how much of a value that is real has a random error with the value that is predicted. There should be no real errors in statistical work. An error should be created whose cause is not clear and should be eliminated. One has to look visually at the number of points that are up and down to detect that there is no real error. The work is done correctly if they are approximately equal to each other.

The following equations show how consistent the model is with the truth, which means one can place only pH and the time and the value of nanoparticles on the following equation and observe the result if no practical work is done. If we put the value of nanoparticles, time, and pH in the removal efficiency equation, the probability percentage of the result that the equation gives is equal to the laboratory.

Formula:

Removal efficiency = 65.125 ………… 0.00962962..$time^2$

The last diagram shows that the optimized values of pH factors and the value of adsorbent and time show the best result.

$R^2$ is about 79%, statistically very valuable showing that if one substitutes the values of the parameters according to the following formula, one can be confident up to 79% of the response it gives us.

**Table 3) Absorption results of Box–Behnken design experiments for three factors**

| Experiment | Nanoparticle in grams | pH | Contact time | Removal efficiency percentage |
|---|---|---|---|---|
| **1** | 0.05 | 9 | 30 | 74.53 |
| **2** | 0.05 | 7 | 45 | 88.34 |
| **3** | 0.1 | 9 | 30 | 87.73 |
| **4** | 0.075 | 9 | 15 | 94.78 |
| **5** | 0.1 | 7 | 45 | 92.33 |
| **6** | 0.075 | 5 | 45 | 88.95 |
| **7** | 0.05 | 7 | 15 | 86.50 |
| **8** | 0.075 | 7 | 30 | 90.96 |
| **9** | 0.1 | 5 | 30 | 92.63 |
| **10** | 0.05 | 5 | 30 | 88.65 |
| **11** | 0.075 | 9 | 45 | 88.65 |
| **12** | 0.1 | 7 | 15 | 93.25 |
| **13** | 0.075 | 7 | 30 | 91.94 |
| **14** | 0.075 | 7 | 30 | 91.18 |
| **15** | 0.075 | 5 | 15 | 93.55 |

Time has not had a significant effect - an optimal time is achieved when separation is done. If the time is less than the optimal time, there is not enough time for the drug to be absorbed in the nanoparticle sites. If the time is too long, the drug adsorbed on the nanoparticle may be separated from the surface again.

**Conclusion**

The purpose of the project was to remove tetracycline from aquatic media using $Fe_3O_4$ magnetic nanoparticles. The reason for selecting this nanoparticle, besides its having easy, fast, and low-cost synthesis, was the biocompatibility of this compound and its type of structure, which can react with tetracycline antibiotics. Based on the results of the analysis and the examinations of the study, one can state that using $Fe_3O_4$ magnetic nanoparticles to remove tetracycline was very effective, and acceptable results were obtained. In this study, the efficiency of $Fe_3O_4$ magnetic nanoparticles for the removal of tetracycline drug was examined considering 3 effective parameters including pH, the hot value of $Fe_3O_4$ magnetic nanoparticles, and the contact time, and it was proved that these nanoparticles have the potential to absorb tetracycline antibiotics from aqueous solutions given their surface properties and bonding ability. The experiments have proven that:

1. $Fe_3O_4$ magnetic adsorbent with super parametric properties, high specific surface area, suitable surface active sites, high stability at different pH, ease of synthesis, and ease of separation from aqueous solutions under a magnetic field as an effective adsorbent and biocompatibility is an ideal option for removing tetracycline from wastewaters.
2. Besides the ionic bonds, the existence of hydroxyl and carbonyl functional groups sequentially in tetracycline allows the two substances to be readily adsorbed to each other.
3. The effect of the nanoparticle parameter is greater than the other two parameters, showing the different efficiency of the adsorbent in various values.
4. The increase in $Fe_3O_4$ adsorbent gram value has some good effects on efficiency, but it reduces the removal efficiency from one place to another.
5. The effect of contact time on the experiment process was positive, but the changes were insignificant compared to the changes in value, and as the desired answer was obtained in 15 minutes, it was preferred to prevent the experiment from becoming time-consuming and the optimal contact time was considered 15 minutes.
6. The highest adsorption efficiency is at neutral pH which is also because of the type of tetracycline structure and its instability at alkaline and acidic pHs.

Ultimately, it is recommended that the study of different types of drugs be considered for more examinations given the significance and concern over the presence of various types of drugs in the wastewaters.

The synthesis of $Fe_3O_4$ magnetic nanoparticles is fast, easy, and low cost. Thus, if other drugs can be absorbed with this nanoparticle, we will reach the fact that a single nanoparticle can remove a wide range of drugs from the wastewater.

It is recommended that the software calculations of the study should be done to obtain accurate information on how the two compounds react

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**Conflict of interest**

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