The International Innovations Journal of Applied Science

Journal homepage: https://iijas.eventsgate.org/iijas

ISSN: 3009-1853 (Online)

Adsorption of Fe(III) using Fe3O4@MNPs modified with Silica through a simple and time-saving method Optimization by RSM

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ARTICLE INFO

Article history:
Received 2 Apr. 2025,
Revised 19 Aug.2025,
Accepted 30 Sep. 2025,
Available online 15 Sep 2025

Keywords:
Nanocomposite;
Silica gel;
Adsorption;
Fe(III);
Response Surface Methodology

ABSTRACT

This study aims to develop a simple method for preparing modified magnetic nanoparticles. This is achieved by preparing Fe3O4 nanoparticles modified with Silica (Fe3O4@MNPs@Silica) using a rapid, straightforward, and simple solid-phase extraction approach for Fe(III) removal from aqueous solutions. This nanocomposite was characterized using FTIR, XRD, and SEM analysis. In this context, the optimization of Fe(III) adsorption by silica-coated Fe3O4 nanoparticles was studied. The effects of various parameters, including pH, adsorbent dosage, initial concentration, and time, on the adsorption process were investigated. It was found that the adsorption capacity of Fe(III) increased with increasing adsorbent dosage and initial concentration and decreased with increasing pH and time. The experimental adsorption conditions were optimized using Response Surface Methodology (RSM). The Fe3O4@MNPs@Silica adsorbent exhibits an adsorption capacity of 3.870 mmol Fe(III) g–1 at a pH of 2.5. This confirms a high affinity of Fe3O4@MNPs@Silica towards Fe(III).

1. Introduction

Water contamination is a serious issue in many regions worldwide, and conventional water treatment techniques can be expensive and ineffective. However, due to advancements in nanotechnology, magnetic nanoparticles have emerged as a potential alternative to traditional water treatment methods. The unique characteristics of these nanoparticles make them highly effective in removing contaminants from water. Particles that can be magnetized and have a size of less than 100 nanometres are known as magnetic nanoparticles (MNPs). They are effective in adsorbing pollutants from water due to their high surface area-to-volume ratio (Abdel Rahim et al., 2020a). Additionally, the nanoparticles' magnetic properties make it simple to remove them from the water after use, thereby reducing environmental contamination. The application of MNPs in water treatment is relatively simple and cost-effective. They can be removed from the water using magnetic fields, and the recovered nanoparticles can be used again, reducing the cost of water treatment. The MNPs can be used to remove heavy metals (El Shahawy et al., 2022; Rahim, 2024; Zhang et al., 2023), dyes (Moosavi et al., 2020; Perwez et al., 2023), and organic compounds (Manousi et al., 2020)from water. They can be synthesized in various forms such as ferrites, oxides, and metals. MNPs can be modified by altering their surface characteristics to perform specific functions or enhance their performance in multiple applications. The following methods are standard for modifying magnetic nanoparticles: Surface Functionalization, which involves attaching molecules or polymers onto the surface of magnetic nanoparticles. Functional groups such as carboxyl (–COOH), amine (–NH_2), hydroxyl (–OH), or thiol (–SH) groups can be introduced. These functional groups can facilitate further modification or conjugation with biomolecules, drugs, or targeting ligands (Geißler et al., 2021). Silica coating: Silica (SiO_2) coatings provide a robust and biocompatible shell around magnetic nanoparticles. Fortunately, the sialylation

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process of coating Fe_3O_4 MNPs with a SiO_2 layer enhances their chemical stability, preventing Fe_3O_4 from oxidizing and aggregating. For this reason, surface modification with a specific organic or inorganic component is necessary to improve the adsorption efficacy of these nanoparticles (Sharifianjazi et al., 2021).

Modifying magnetic nanoparticles with silica gel involves coating the nanoparticles with a silica (SiO_2) layer. This process, commonly referred to as silica coating or silica encapsulation, is employed to impart various properties to magnetic nanoparticles. (Jamasbi et al., 2022; Moradnia et al., 2021). The sol-gel method is a standard method used for coating magnetic nanoparticles (MNPs) with silica, where MNPs can be directly synthesized within a silica matrix through the sol-gel process, in which silica precursors are polymerized around the nanoparticles. (Bokov et al., 2021). Silica coating of MNPs enhances their versatility and stability, making them valuable tools in biomedical, environmental, and industrial applications. The choice of silica coating method depends on the desired properties and intended applications of the modified nanoparticles.

Heavy metals such as Fe(III) are toxic and can cause severe health problems in humans if consumed in high amounts. (Billah et al., 2021). Fe(III) contamination of water can result from various sources, including both human-made and natural sources. In conclusion, although Fe(III) is generally not harmful at low concentrations, pollution-related higher concentrations of the metal can have serious adverse effects on the environment and human health. Fe(III) in water must be adequately managed, monitored, and treated to lessen its detrimental impacts on ecosystems and public health. Several effective treatment methods have been used to remove Fe(III) from water, including electrocoagulation. (Gaalova et al., 2018) solid-phase extraction technique using various adsorbents(Khuder et al., 2022; Middea et al., 2024; Zhang et al., 2023b). Among these adsorbents, MNPs can be used to adsorb and remove Fe(III) from water. (Abdel Rahim et al., 2020a; Namdeo & Bajpai, 2008).

Hence, this work aims to fully utilize Fe_3O_4@MNPs through a simple modification method, thereby providing Fe_3O_4@MNPs@Silica. The characterization and structure were examined using scanning electron microscopy (SEM), X-ray diffraction (XRD), and Fourier transform infrared spectroscopy (FTIR). Moreover, the adsorption parameters were optimized using RSM methodology.

2. Methodology

2.1. Materials

Analytical reagent-grade $FeCl_3 \cdot 6H_2O$, HCl, NaOH, the sodium salt of EDTA, and silica gel were purchased from Sigma-Aldrich. D-64291 Darmstadt, Germany), $FeSO_4 \cdot 7H_2O$ was purchased from LOBA CHEM Pvt. Ltd., Mumbai 400005, India. Salicylic acid ($C_6H_4(OH)COOH$) was obtained from Techno Pharm Chem, Haryana, India, and double-distilled water was used throughout all experiments.

2.2. Instruments

A Fisher Scientific Accumet pH-meter (Model 825, Germany) calibrated against two standard buffer solutions at pH 4.0 and 9.2 was used for pH measurements. Wrist Action mechanical shaker model 75 (manufactured by Burrell Corporation, Pittsburgh, PA., USA) was used for shaking. A Thermo Scientific Nicolet 380 spectrometer, operating from 400 cm–1 to 4000 cm–1, was used to record Fourier transform infrared (FTIR) spectra for molecular nature analysis. An X'Pert PRO 30-40-60 diffractometer was used to examine powder X-ray diffractograms in the 20-80° range using Kα radiation from a Cu source (λ = 1.5406 Å). Scanning electron microscopy (SEM) analysis was performed using a JEOL JSM-6000 plus SEM and a ZEISS LEO SUPRA 55.

2.3. Preparation of Fe_3O_4@MNPs@Silica

Fe_3O_4@MNPs@Silica was synthesized using the co-precipitation technique. (Ba-Abbad et al., 2022). In short, two 50-ml beakers of water

https://doi.org/10.61856/bj875986

were filled with separate solutions of $FeSO_4 \cdot 7H_2O$ and $FeCl_3 \cdot 6H_2O$, and then they were mixed and continuously stirred at room temperature. $FeSO_4 \cdot 7H_2O$ and $FeCl_3 \cdot 6H_2O$ have molar ratios of 2:1 for Fe^{2+} and Fe^{3+}, respectively. After that, an appropriate weight of silica gel was added to the mixture and stirred at room temperature. The temperature of the mixture was raised to 35 ± 5°C. NaOH was added to adjust the pH of the final solution to 11.0. For two hours, the samples were vigorously agitated at 60 ± 5°C. The samples were allowed to cool for two hours. After that, unreacted precursors were removed from both samples by washing. Double distilled water (DDW) was used for the washing phase until the solution's pH reached 7.0 ± 0.2. Afterward, Fe_3O_4@MNPs@Silica was dried in an oven for 24 hours.

2.4. Software for statistical analysis and data analysis

The optimum values of four effective and independent factors on the effectiveness of Fe(III) removal from water were found in the current study using the Box-Behnken experimental design (BBD). These parameters are (A) pH, (B) the amount of Fe_3O_4@MNPs@Silica in milligrams, and (C) the concentration. of Fe(III) in mol/L, and (D) the adsorption time (min). Minitab 8.0, from the United States, was used to specify the experimental design and enhance the system through statistical parameter estimation. The number of experiments (N) required for the development of BBD can be calculated from the following equation:

$$N = 2k\,(k-1) + C_o \qquad (1)$$

where k is a combination of several factors and C_o is the number of central points (where C_o = 4).

Table 1 contains the amounts and ranges of the variables used in the BBD for the adsorption of Fe(III) using Fe_3O_4@MNPs@Silica.

Table 1: Experimental range and amounts of independent factors for Fe(III) using Fe_3O_4@MNPs@Silica

Factor	min.	max.
pH	1	4
Wt of Fe_3O_4@NPs@Silica gel (mg)	10	100
Conc. of Fe (III) (mol/L)	0.001	0.1
Time of adsorption (minutes)	5	60

2.5. Batch Adsorption Method

The following procedure was used to remove Fe(III) from the water's surface. In a 50.0 mL beaker, an appropriate amount of the nano adsorbent was added to a solution containing a specific concentration of Fe(III) (0.001 to 0.1 mol/L). Before adding the adsorbent, pH levels were adjusted between 1.0 and 4.0 by varying quantities of either 1.0 M NaOH or 1.0 M HCl solutions. After achieving equilibrium through mechanical shaking for a time ranging from 5 to 60 minutes at room temperature, the metal-ion-free solution was transferred to a conical flask, and the adsorbent was separated using an external magnetic field. A complexometric EDTA titration was used to determine the concentration of Fe(III) in the solutions.

3. Results and discussion

3.1. Characterization of the adsorbent

The different functional groups adsorbed on the Fe_3O_4-MNPs were detected by Fourier transform-infrared absorption spectrometry (FT-IR). Fig. 1a displays the resultant spectra. The vibrational bands at approximately 581 cm^{-1} and 631 cm^{-1} for the pure magnetite sample are indicative of the υ(Fe–O) lattice vibrations, as shown in Fig. 1b. The silica-coated magnetite sample exhibits faint bands at 1027 and 633–589 cm^{-1}, and a band at 1157 cm^{-1} that corresponds to the stretching vibrations of υ(Si–O–Si), υ(Si–OH), and υ(Si–O–Fe), respectively. These bands align well with coatings of Fe_3O_4-MNPs made with silica gel (Abdel Rahim et al., 2020a).

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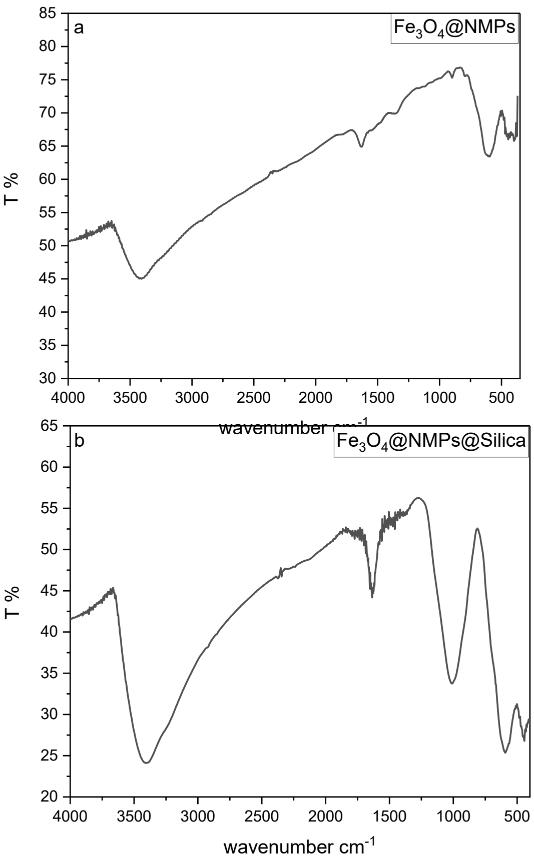


Figure 1: FT-IR spectra of (a) Fe_3O_4 MNPs, and (b) Fe_3O_4@MNPs@Silica

X-ray diffraction patterns (XRD) were used to confirm the Fe_3O_4 structure before and after modification with silica gel. As shown in Fig. 2, the powder XRD pattern of the synthesized magnetic nanoparticles was like the pattern for crystalline magnetite Fe_3O_4 (Ahmed & Soliman, 2015). The distinctive peaks of pure Fe_3O_4 MNPs at 2θ/deg. The values of 30.1, 35.4, 43.9, 53.4, 57.0, and 62.6 confirm the presence of the crystalline structure of magnetite.

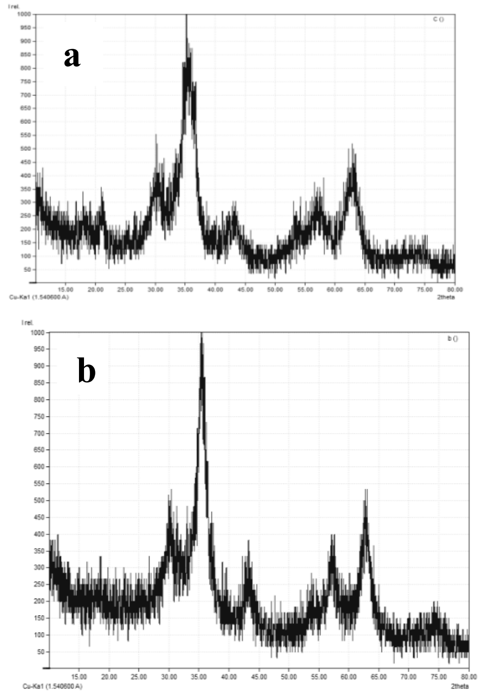
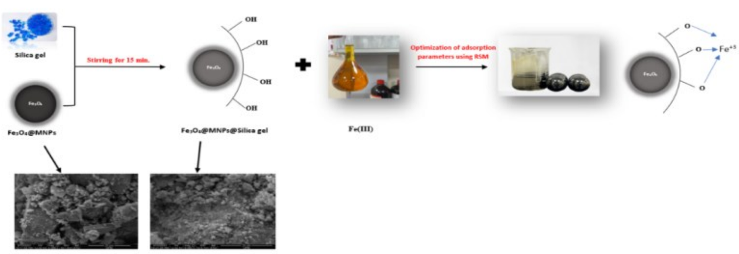


Figure 2: XRD patterns of (a) Fe_3O_4 MNPs, and (b) Fe_3O_4@MNPs@Silica

SEM was used to describe the morphology of Fe_3O_4 MNPs before and after modification (Fig. 3). Figure 3(a) shows the scanning electron micrograph of Fe_3O_4 MNPs, revealing a rough surface. Such material is suitable for adsorption research due to the presence of OH functional groups and surface porosity. The fact that the Fe_3O_4@MNPs@Silica particles aggregated after being modified with Fe_3O_4 MNPs, as seen in Fig. 3(b), verifies the effectiveness of the binding process between silica gel and Fe_3O_4 MNPs. Moreover, the particles become small after modification in Fe_3O_4@MNPs@Silica adsorbent.

All these changes support the success of fabricating Fe_3O_4@MNPs@Silica, as shown in Scheme 1.



Scheme 1: Synthesis of Fe_3O_4@MNPs@Silica through time-saving method

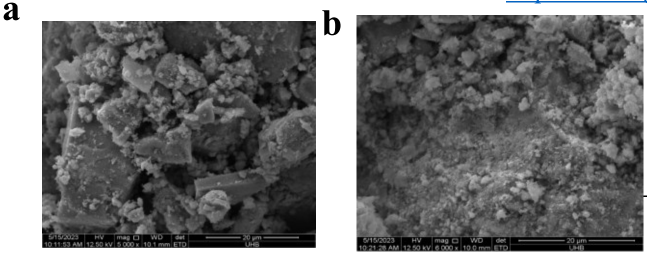


Figure 3: SEM images of (a) Fe_3O_4 MNPs, and (b) Fe_3O_4@MNPs@Silica

3.2. Optimization using Box-Behnken design

In RSM, the connections between several explanatory variables and one or more response variables are examined. To explain the process and investigate the impacts of the independent variables, this experimental methodology develops a mathematical model. (Baş & Boyacı, 2007). According to the prior equation (1), it was reasonable to employ the Box-Behnken design (BBD) in this investigation, which consisted of four independent variables and three center points, resulting in 27 experimental runs.

Factors:	4	Base runs:	27
Replicates:	1	Total runs:	27
Base blocks:	1	Total blocks:	1

These runs were performed to determine the adsorption capacity of Fe(III) using Fe_3O_4@MNPs@Silica in an aqueous solution. The obtained data and the four independen variables were used to fit a quadratic response surface model, and the fitted equations for Fe(III) adsorption using Fe_3O_4@MNPs@Silica.

Table 2: Adsorption capacity values according to the amounts of independent factors for Fe(III) using Fe_3O_4@MNPs@Silica

no	pH	Wt	Conc.	Time	Adsorption capacity (mmol/g)
1.	2.5	55	0.55	32.5	0.902
2.	2.5	55	0.10	5.0	0.131
3.	2.5	55	0.55	32.5	0.902
4.	4.0	100	0.55	32.5	1.560
5.	1.0	55	0.10	32.5	0.140
6.	1.0	55	0.55	60.0	0.556
7.	2.5	100	1.00	32.5	0.500
8.	2.5	55	0.55	32.5	0.556
9.	4.0	55	0.10	32.5	0.090
10.	2.5	55	1.00	5.0	0.956
11.	4.0	55	1.00	32.5	0.650
12.	2.5	10	0.55	5.0	2.560
13.	2.5	10	0.55	60.0	2.060
14.	1.0	55	0.55	5.0	0.353
15.	1.0	55	1.00	32.5	1.400
16.	4.0	55	0.55	60.0	1.223
17.	2.5	100	0.10	32.5	0.890
18.	1.0	10	0.55	32.5	2.260
19.	2.5	100	0.55	5.0	0.306
20.	2.5	55	1.00	60.0	1.327
21.	4.0	10	0.55	32.5	2.060
22.	1.0	100	0.55	32.5	0.306
23.	2.5	55	0.10	60.0	2.636
24.	2.5	10	0.10	32.5	0.960
25.	2.5	10	1.00	32.5	3.200
26.	4.0	55	0.55	5.0	1.101
27.	2.5	100	0.55	60.0	3.870

3.3. Interpretation of surface plot

The pH of the solution, the weight of the adsorbent, the concentration of Fe(III), and the duration of the experiment were the most significant variables influencing Fe(III) adsorption onto Fe_3O_4@MNPs@Silica in aqueous solution. Surface plots demonstrated the relationship between these factors and adsorption capacity. While the other two elements in these plots are kept constant, the function of two of the factors is investigated (Fig. 4).

Through previous forms and the values of adsorption capacity in Table 2, it is evident that the value of adsorption capacity changes in response to different factors, increasing with the pH value from 1.0 to 2.5, after which it begins to decrease. As for the weight effect, it was found to be gradually increasing by weight until it reached constant, i.e. saturation of the surface of Fe_3O_4@MNPs@Silica with all Fe(III), which meant that, with the lowest amount of Fe_3O_4@MNPs@Silica, Fe(III) could be removed, indicating their efficiency in the extraction process. Hence, we conclude that the highest value of adsorption capacity is 3.870 mmol/g, which we obtained under conditions

https://doi.org/10.61856/bj875986

2.5, 100 mg, 60 min, and 0.005 mM, respectively, from the pH, the weight of Fe3O4@MNPs@Silica, the time of adsorption, and the concentration of Fe(III).

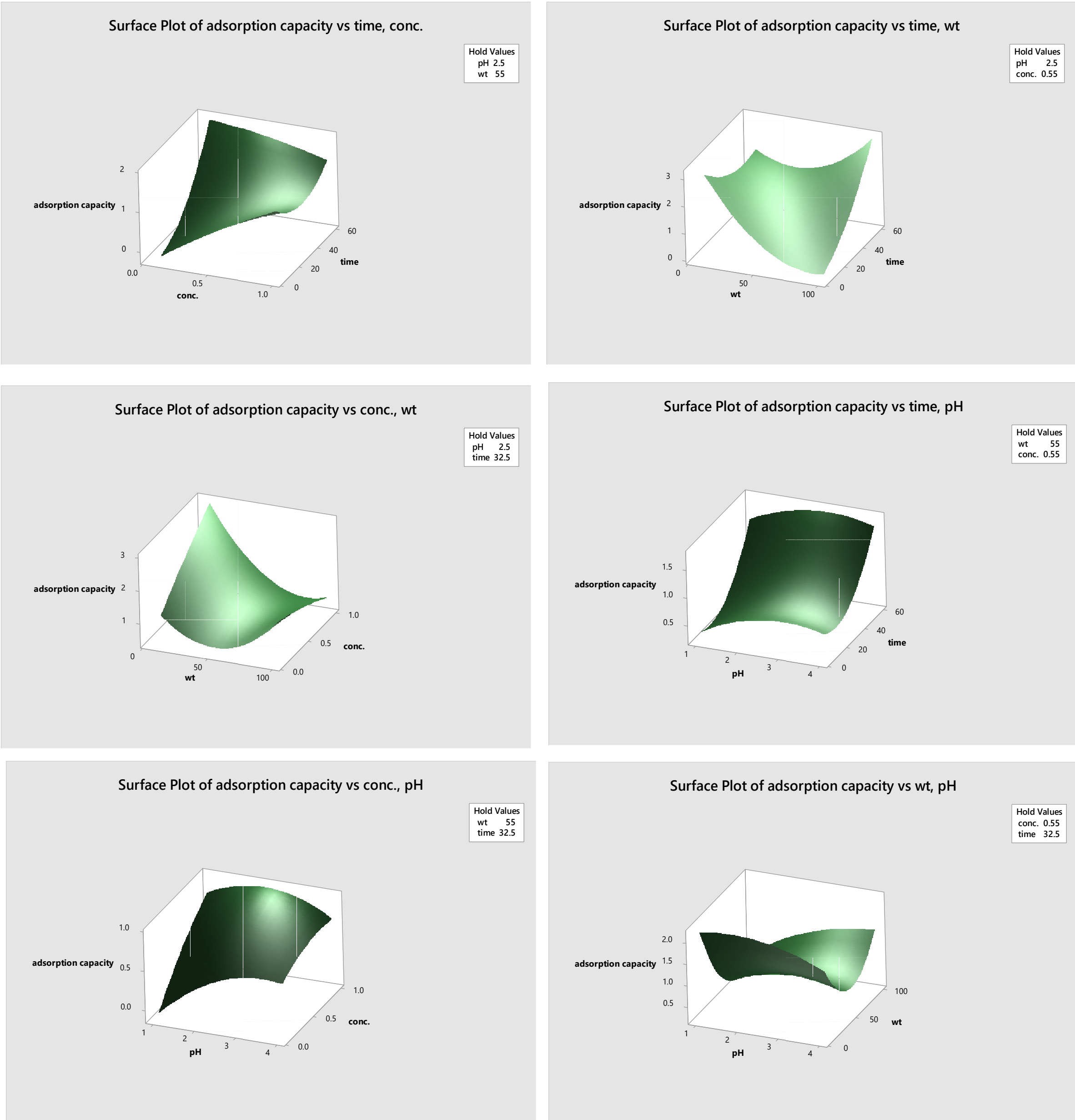


Figure 9: Response surface plots for interactions of adsorption of Fe(III) by Fe_3O_4@MNPs@Silica

4. Conclusions

This study reports the development of a nanocomposite adsorbent composed of Fe_3O_4@MNPs@Silica. This adsorbent exhibits a strong affinity for removing Fe(III) ions using external magnetic separation from aqueous solutions. The Response Surface Methodology was used to analyze the adsorption data. At 2.5, 100 mg, 60 min, and 0.005 mM, from pH, the weight of Fe3O4@MNPs@Silica, the time of adsorption, and the concentration of Fe(III), respectively, the maximum value of adsorption capacity was 3.870 mmol/g. These results demonstrate the possibility of synthesizing a nanocomposite through a simple method.

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