

Synthesis, Application, and Efficiency of the Cellulose@Fe₃O₄@ZIF-8 Carbon Aerogel Magnetic in the Removal of Gasoil from Aqueous Solutions

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16

Abstract 17

Water pollution by Gasoil seriously threatens the environment and human health. Removing 18
Gasoil from the aqueous solutions is critical to maintaining the safety and quality of the water 19
source. This research looked at the feasibility of filtering Gasoil out of water. As a first step, 20
we developed a new adsorbent from *Prosopis farcta* cellulose fibers by preparing a metal- 21
organic magnetic framework (cellulose@Fe₃O₄@ZIF-8 carbon aerogel magnetic). The design 22
of experiment method was utilized to explore how variations in pH, Gasoil concentration, and 23
adsorbent dose influenced the elimination of Gasoil. Adsorbent has an adsorption capacity of 24
7 g.g⁻¹ and can adsorb 100 % of the petroleum floating in water. Time studies demonstrated no 25
time dependence in the Gasoil adsorption process by the adsorbent. Freundlich isotherm 26
described the adsorption of petroleum onto the prepared magnetic carbon aerogel. The free 27
energy, enthalpy, and entropy changes as a part of the Gibbs thermodynamic equation were 28
determined. The proposed magnetic carbon aerogel was an effective adsorbent for extracting 29
petroleum from the aqueous solutions because of its rapid separation and high stability. Even 30
after eight times of use, this adsorbent maintained its relatively high adsorption capacity, and 31
only a 10% drop in adsorption efficiency was observed. 32

Keywords: Gasoil; *Prosopis farcta*; Metal-organic framework; Design of experiment; 39
Adsorption; Cellulosic fibrous; Cellulose@Fe₃O₄@ZIF-8 carbon aerogel magnetic. 40

1. Introduction

The presence of Gasoil in water sources is particularly detrimental to plant/animal life and human health. Removing Gasoil from aqueous solutions is vital for protecting the safety and purity of water supplies. Various traditional techniques have been employed for pollution removal, such as adsorption (Bashiri et al. 2020, Falaki & Bashiri 2021, Sarabadan et al. 2019), photocatalysts (Arabameri & Bashiri 2022, Jafarinejad et al. 2023, Simon et al. 2020), sedimentation (Chen et al. 2018, Sun et al. 2022), and ion exchange. However, these methods often suffer from restrictions such as low performance, cheapness, and complicated operation. Due to their promising adsorption capabilities and prospective uses in environmental remediation, metal-organic frameworks (MOFs) have engrossed much consideration in recent years as a unique class of porous materials. In this context, the extension of a highly efficient, profitable, and environmentally benign adsorbent based on MOFs is necessary for Gasoil removal from aqueous solutions (Karimi et al. 2021, Paz et al. 2023, Wu et al. 2019). MOFs are a porous material with a definite crystalline structure, a large surface area, and a pore size that can be modified by altering the concentration of metal ions or organic ligands. They have interesting characteristics that make them good candidates for many uses, such as gas storage, catalysis, and environmental remediation. The excellent thermal and chemical stability of Zeolitic Imidazolate Frameworks (ZIFs) has made them a prominent subclass within the larger family of MOFs (Kuwer et al. 2021).

ZIF-8, made up of Zinc ions coupled by imidazole ligands, is a water-stable MOF with an explicit microporous structure (Li et al. 2019). Heavy metal ion removal and dye removal from aqueous solutions are only two of the many uses that have prompted substantial research into the material (Zheng et al. 2014). The high surface area, broad pH stability, and feasible chemical interactions of ZIF-8 make it an ideal material for the adsorption of Gasoil and other contaminants (Bui et al. 2022b). Cellulosic fibrous materials derived from biomass have also

shown potential as adsorbents for Gasoil removal. One such biomass is *Prosopis farcta* (PF), a renewable resource with abundant cellulose, hemicellulose, and lignin content (Liu et al. 2022). The magnetic properties of the composite material facilitate its easy separation and recovery from the aqueous solution, making it an ideal candidate for practical applications in water treatment (Gorkunov et al. 2002, Oliveira et al. 2002, Zeng et al. 2022).

Recent studies have shown that CelloMOFs have multifaceted applications such as water treatment, antibacterial, gas separation, and high-performance flexible supercapacitors (Abdelhamid & Mathew 2022, Jabbar et al. 2022, Liu et al. 2021, Tao et al. 2023, Tu et al. 2022, Zhou et al. 2019). These MOFs are a type of MOF that incorporate magnetic nanoparticles into their structure, resulting in materials with unique properties and applications (Nadar & Rathod 2018, Yadav et al. 2021, Zhao et al. 2015b). Magnetic MOFs can be utilized as sorbents to eliminate organic compounds from water. This category of MOFs has possible utilizations in various areas, including water treatment, catalysis, and biotechnology (Zhao et al. 2015a).

Research (Tajari & Bashiri 2023) looked at how well a magnetic MOF adsorbent constructed from the cellulose fibers of the PF plant worked in extracting Gasoil from aqueous. Preparation of the adsorbent includes design of experiment to determine the influence of factors on Gasoil treatment, including pH, Gasoil concentration, and adsorbent dose. This adsorbent had 7.5 g.g^{-1} adsorption capacity, 100 times greater than iron-based adsorbents. Thus, the magnetic aerogel can efficiently extract gasoline from water. For removing Fosfomycin from pharmaceutical wastewater, a core-shell $\text{Fe}_3\text{O}_4/\text{PVP}/\text{ZIF-8}$ nanostructure adsorbent was generated (Abdelmigeed et al. 2022). The research evaluated the regeneration performance of the core-shell nanostructure adsorbent for Fosfomycin removal.

The adsorption efficacy of sonochemically produced $\text{Fe}_3\text{O}_4/\text{ZIF-8}$ for the elimination of copper ions from water is shown in another work (Bui et al. 2022a). It has a high maximal

adsorption capacity (345 mg.g^{-1}), rapid adsorption time (10 min), and efficient elimination (95.3%). Heydari Moghadam et al. investigated zeolite imidazolate framework-8 (ZIF-8) to remove trichloroethylene from aqueous solutions with an optimum elimination efficiency of 96.29%. The adsorption capacity was 136.33 mg.g^{-1} . Trichloroethylene was demonstrated to be successfully removed from water by the ZIF-8 platform (Moghaddam et al. 2019). Khalilifard et al. used oleic acid- and graphene oxide-doped magnetic superhydrophobic polyurethane to separate organic species from aqueous solution. The magnetic sponges can be regulated and recycled using a magnet 15 times for oil spill cleaning without losing their adsorption ability. The modified sponge is an efficient adsorbent for treating oil in polluted water (khalilifard & Javadian 2021).

This research presents a novel adsorbent, cellulose@Fe₃O₄@ZIF-8 carbon aerogel magnetic, for Gasoil removal from aqueous solutions. It combines the advantages of biomass, magnetic Fe₃O₄ nanoparticles, and ZIF-8, resulting in a highly efficient, cost-effective, and environmentally friendly adsorbent material. The composite structure effectively overcomes the inherent limitations of ZIF-8, such as particle aggregation, and enables rapid Gasoil diffusion and adsorption. Design of Experiment (DOE) method, precisely the Box-Behnken method, is employed to optimize adsorption. Additionally, we investigate the adsorption isotherms, thermodynamics, and adsorption mechanism of the cellulose@Fe₃O₄@ZIF-8 carbon aerogel magnetic.

2. Experimental section

2.1. Materials and methods

Fe (III) chloride hexahydrate (FeCl₃.6H₂O, 99.0%), ethanol (96%), sodium citrate, sodium acetate, ethylene glycol, mercaptoacetic acid (MAA), zinc nitrate, methanol and 2-

methylimidazole have been purchased from Merck Company. Fruits of PF were collected from meadows in central Iran.

2.2. Synthesis of cellulose@Fe₃O₄@ZIF-8 carbon aerogel magnetic adsorbent

2.2.1. Preparation of Cellulosic Fibrous Biomass

PF is a renewable biomass resource, which was used as the cellulose fiber material. A mechanical grinder was used to reduce the size of the material to a finer consistency. They were then given several methanol washes.

2.2.2. Synthesis of Fe₃O₄ Nanoparticles

Fe₃O₄ Particles were successfully generated via a solvothermal approach. Using magnetic stirring for 30 minutes, 80 ml of ethylene glycol dissolved 2.7 g FeCl₃.6H₂O, 1.0 g sodium citrate, and 4.8 g NaAc. This mixture was sealed in a Teflon-lined stainless steel container and autoclaved at 473 K for 10 hours. Consequently, microspheres of Fe₃O₄ were collected, washed many times in ethanol, and then vacuum dried at 50 °C. To make mercaptoacetic acid (MAA)-functionalized Fe₃O₄ nanoparticles, the following steps were taken: After shaking a mercaptoacetic acid (2.9 mM) ethanolic solution of 100 ml for 24 hours, 0.5 g Fe₃O₄ was added. The MAA-Fe₃O₄ nanoparticles were collected and washed.

2.2.3. Synthesis of cellulose@Fe₃O₄@ZIF-8 carbon aerogel magnetic

First, 6.4 g of Zn(NO₃)₂ and 14 g of prepared biomass were added to 20 ml of methanol. Then 1.0 g of MAA-Fe₃O₄ and 9.24 g of 2-methylimidazole are added and stirred for 2 hours at room temperature. After that, dry in a vacuum at 50 °C. Carbon aerogels were made by pyrolyzing cellulose@Fe₃O₄@ZIF-8 at 600°C for 1 hour in an N₂ environment in a tubular furnace. Then, magnetic cellulose@Fe₃O₄@ZIF-8 carbon aerogels were prepared.

2.3. Characterization of cellulose@Fe₃O₄@ZIF-8 carbon aerogel magnetic adsorbent 141

The cellulose@Fe₃O₄@ZIF-8 carbon aerogel magnetic adsorbent is identified using various 142
analytical methods, including: 143

- Fourier Transform Infrared (FTIR) Spectroscopy: To examine the functional adsorbent 144
groups in the sample and confirm the successful incorporation of ZIF-8 and Fe₃O₄ 145
nanoparticles. 146
- X-ray Diffraction (XRD): To verify the ZIF-8 and Fe₃O₄ nanoparticle crystalline 147
structure and to evaluate the composite material's stability. 148
- Scanning Electron Microscopy (SEM): To observe the morphology, size, and 149
distribution of the ZIF-8 and Fe₃O₄ nanoparticles within the cellulosic fibers. 150
- Vibrating Sample Magnetometry (VSM): To assess the magnetic properties of the 151
Fe₃O₄ nanoparticles within the composite material. 152
- Inductively Coupled Plasma emission spectroscopy (ICP): Analyze the amount of ions 153
lost during the adsorption process due to ion leaching. 154

2.4. Adsorption experiments 155

First, a measured amount of Gasoil was dropped into the well of the culture dish's water. 156
Afterward, a certain weight of adsorbent was carefully sprinkled over the top of the water- 157
Gasoil solution. After Gasoil was adsorbed from water, the adsorbent was magnetically 158
extracted and weighed. Adsorption of Gasoil onto an adsorbent occurred within 10 seconds, 159
which makes it a fast process. Equilibrium adsorption amount (q_e) and Gasoil removal ($R\%$) 160
were determined using the following formulas: 161

$$q_e = \frac{m_2 - m_1}{m_1} \quad (1) \quad 162$$

$$R\% = \frac{m_2 - m_1}{m_g} \times 100 \quad (2) \quad 163$$

An initial mass of Magnetic - cellulose@Fe₃O₄@ZIF-8 carbon aerogel is m_1 , a final mass of it is m_2 , and m_g is used for initial mass of Gasoil (Shi et al. 2020, Tajari & Bashiri 2023).

2.5. Design of Experiment: Box-Behnken Method

The Box-Behnken DOE approach studied the effects of variables on the Gasoil removal by cellulose@Fe₃O₄@ZIF-8 carbon aerogel magnetic in an aqueous solution. This method helps determine the optimal parameters for the adsorption process, such as the mass of adsorbent (A), the mass of Gasoil (B), and the pH value (C). By systematically varying these parameters, it is possible to identify the conditions that result in maximum Gasoil removal efficiency. After that, we followed the blueprint and coded the independent variables' levels (-1, 0, 1 for low, medium, and high values) (Table 1). Table 2 shows the Box-Behnken design values for each independent variable and Design-Expert 13.0.5.0 trial outcomes.

Table 1. Experimental range and level of independent variables for Gasoil removal by the cellulose@Fe₃O₄@ZIF-8 carbon aerogel magnetic.

Table 2. Box-Behnken design matrix for variables and removal (%) values.

2.6. Nonlinear isotherms of adsorption and Thermodynamics Studies

The adsorption isotherms of Gasoil removal by cellulose@Fe₃O₄@ZIF-8 carbon aerogel magnetics were studied at various temperatures and initial Gasoil concentrations. The Langmuir, Freundlich, Temkin, and Azizian et al isotherms were used to correlate the data to find the best adsorption model. The nonlinear Langmuir isotherm is Eq. 3.

$$q_e = \frac{q_m K_L C_e}{1 + K_L C_e} \quad (3)$$

where K_L (L.g⁻¹) is the Langmuir constant, C_e is the equilibrium concentration, and q_m (g.g⁻¹) is the maximum adsorption capacity, once a complete monolayer of adsorbent covers the

surface (Langmuir 1918). The Freundlich isotherm is represented by Eq. 4, where K_F is the
 Freundlich constant. The constant $1/n$ (with a value between 0 and 1) is related to the surface
 heterogeneity, and the closer this value is to 1 the more homogeneous the surface is (Freundlich
 1907).

$$q_e = K_F C_e^{\frac{1}{n}} \quad (4)$$

Temkin studied surface adsorption isotherm interactions. The findings have shown that
 due to these interactions, the adsorption heat of all monolayer molecules diminishes linearly
 with the surface coverage. Such an effect comes from the repulsive forces on a homogeneous
 surface or from the inhomogeneity of the surface. This isotherm is represented by Eq. 5, where
 parameters A and B are Temkin equilibrium constants (Xia et al. 2021).

$$q_e = B \ln A + B \ln C_e \quad (5)$$

However, Azizian et al isotherm suggests a revised Langmuir isotherm that better explains
 dense phase of desorption and adsorption processes. Adsorption thermodynamic parameters
 may be calculated on a global scale with the help of this method. C_S and K_{ML} parameters are
 concentration saturation and modified Langmuir coefficient (Azizian et al. 2018).

$$q_e = \frac{q_m K_{ML} C_e}{C_S + (K_{ML} - 1) C_e} \quad (6)$$

The thermodynamics of Gasoil removal by cellulose@Fe₃O₄@ZIF-8 carbon aerogel
 magnetic was investigated at different temperatures. Calculation of enthalpy, entropy and
 Gibbs free energy changes (ΔH , ΔS , and ΔG) are necessary to understand adsorption
 spontaneity, practicability, and foundations. Using the Van't Hoff equation, we can get the
 values of these parameters (Azizian et al. 2018):

$$\Delta G = -RT \ln K_{ML} \quad (7)$$

$$\ln K_{ML} = \frac{\Delta S}{R} - \frac{\Delta H}{RT} \quad (8) \quad 212$$

The dimensionless K_{ML} is the modified Langmuir coefficient that may be derived using the 213
Azizian et al isotherm. 214

3. Results and discussion 216

3.1. FT-IR Spectroscopy 217

Figure 1 presents the FTIR spectra of cellulose@Fe₃O₄@ZIF-8 carbon aerogel magnetic. The 218
absorption spectra of the sample showed peaks at 1558, 1003.2, and 872.2 cm⁻¹, which 219
correspond to C=O, N-O, C-N, Zn-O, and Fe-O, respectively. The band at 872.2 cm⁻¹ represents 220
the stretching vibration of the Fe-O bond, indicating FeO_x in the aerogel's carbonaceous phase. 221
This band also shows the formation of the ZIF-8 structure. The 2923 cm⁻¹ band represents 222
cellulose's hydroxyl and carboxyl stretching vibrations. The functional groups identified in the 223
sample can participate in the surface adsorption of Gasoil. 224

Figure 1. FT-IR spectrum of cellulose@Fe₃O₄@ZIF-8 carbon aerogel magnetic. 226

3.2. SEM and EDS analyses 228

SEM pictures of cellulose@Fe₃O₄@ZIF-8 magnetic carbon aerogel are shown in Figure 2. 229
Examining the SEM images shows that the adsorbents structure has many pores and gaps 230
suitable for adsorbing Gasoil and removing it from the aqueous solution. The formation of 231
adsorbent nanoparticles provides more access to the adsorption sites. Figure 3 shows EDS 232
analysis, which confirms the presence of Fe, O, and Zn elements in the adsorbent. 233

Figure 2. SEM images of (a) surface structures of PF-carbon and (b, c, d) cellulose@Fe₃O₄@ZIF-8 carbon 235
aerogel magnetic. 236

Figure 3. The spectrum obtained from sample analysis by the X-ray energy dispersion (EDS) method.

3.3. X-ray Diffraction (XRD)

Figure 4 displays the pre- and post-adsorption XRD patterns of cellulose@Fe₃O₄@ZIF-8 carbon aerogel magnetic, MAA-Fe₃O₄, and PF cellulose carbon. Figure 4c shows the magnetic carbon aerogel's XRD patterns are identical to those of MAA-Fe₃O₄ (Figure 4a) and PF cellulose carbon (Figure 4b). Figure 4c shows the XRD pattern of cellulose@Fe₃O₄@ZIF-8 carbon aerogel magnetic, which reveals a strong peak at $2\theta=35^\circ$. The absence of change in the XRD patterns before (Figure 4c), and after (Figure 4d) adsorption demonstrates the adsorbent's high stability in Gasoil adsorption, and its structure is unaltered throughout the adsorption process.

Figure 4. XRD patterns of (a) MAA- Fe₃O₄, (b) Cellulose carbon of PF, (c) cellulose@Fe₃O₄@ZIF-8 carbon aerogel magnetic before adsorption and, (d) cellulose@Fe₃O₄@ZIF-8 carbon aerogel magnetic after adsorption.

3.4. Vibrating-Sample Magnetometer (VSM)

The magnetic hysteresis loop tests a magnetic aerogel's reaction to a magnetic field. It is shown as a curve of change of magnetization of the material concerning the applied area. The residual loop relies on magnetic field direction, sample ratio, sample extent, material type, and magnetic return method. Figure 5 demonstrates the hysteresis loop under the field -15000 to +15000 Oe. The cellulose@Fe₃O₄@ZIF-8 carbon aerogel magnetic hysteresis curves showed super magnetic adsorption. The measured saturation magnetization (M_s) and magnetic remanence (M_r) were 5.83 and 0.7081 emu.g⁻¹, respectively. An external magnet can easily extract cellulose@Fe₃O₄@ZIF-8 carbon aerogel magnetic from the solution.

Figure 5. Hysteresis curve of cellulose@Fe₃O₄@ZIF-8 carbon aerogel magnetic

3.5. Inductively coupled plasma emission spectrometry (ICP)

During adsorption, ion leaching from the adsorbent was investigated using ICP. The findings show that the quantity of iron in the adsorbent decreases from 5.06 % before adsorption to 4.06% after adsorption. Also, the amount of zinc before adsorption is 3.88%, which reaches 1.2% after adsorption. This finding demonstrates the durability and presence of magnetic characteristics in the adsorbent following the adsorption process.

3.6. Optimizing adsorption parameters using Box-Benken design

The dependence between parameters A, B, C, and the response parameter (R%) was obtained as follows:

$$R(\%) = 44.41 + (9.98 \times A) - (31.34 \times B) + (1.38C) - (1.8 \times AB) - (0.1450 \times A \times C) -$$

$$(0.1175 \times B \times C) + (0.1888 \times A \times A) + (13.73 \times B \times B) + (0.9612 \times C \times C)$$

(9)

According to Eq. 9, the weight of the adsorbent has a favorable effect on adsorption efficiency.

This equation shows the adsorbent weight's significant impact on the adsorption reaction. The experimental and predicted Gasoil removal values are similar (%R= 98.99 and 94.2, respectively), and showing the model's power.

Pareto diagrams may also determine each factor's relevance by estimating its percentage influence on the solution (Khataee et al. 2011). The Pareto chart analysis for removing Gasoil by cellulose@Fe₃O₄@ZIF-8 carbon aerogel magnetic adsorbent is shown in Figure 6. As seen in Figure 6, the factors B (76.9476), B×B (14.7686), A (7.8029), and C×C (5.3743) are the most influential parameters on the adsorption of Gasoil by cellulose@Fe₃O₄@ZIF-8 carbon aerogel magnetic.

Figure 6. Pareto graphic analysis.

When a function has two constant parameters, response surfaces are given. The significant and interaction impacts of these two parameters may be better understood. The effect of these factors on Gasoil extraction is seen in Figure 7. Magnetic carbon aerogel adsorbs the most Gasoil at 0.1 g of adsorbent (A) and 0.5 g of Gasoil. The cellulose@Fe₃O₄@ZIF-8 carbon aerogel magnetic has successfully adsorbed 100% Gasoil under these circumstances. The projected and Gasoil removal values are shown in Figure 8. The clustering of the dots along a line provides more evidence of the robust pattern.

Figure 7. Response surface plot for the effects of the adsorbent weight (A) and initial Gasoil weight (B).

Figure 8. Actual vs. predicted values of the Gasoil removal.

3.7. Adsorption isotherms and thermodynamics studies

At 30, 40, and 50°C, the nonlinear isotherms of Gasoil adsorption by cellulose@Fe₃O₄@ZIF-8 carbon aerogel magnetic were investigated. The results of the nonlinear fitting of Langmuir, Freundlich, Temkin, and Azizian et al isotherms using the solver plugin in the Excel program are presented in Table 3. The saturated concentration (C_s) of Gasoli in water was considered to be 0.35 g.L⁻¹ (Winkler 2009). The obtained corresponding correlation coefficients (R^2) showed the process of Gasoil adsorption. The magnetic carbon aerogel exhibits nonlinear Freundlich isotherm-like adsorption. Figure 9 shows the adsorption capacity regarding equilibrium concentration at different temperatures. As the temperature rises, so does the ability for adsorption, whereas, at lower temperatures, adsorption is enhanced.

Table 3. Nonlinear fitting parameters of adsorption isotherm models.

Figure 9. Isotherm curve of Gasoil adsorption on cellulose@Fe₃O₄@ZIF-8 carbon aerogel magnetic.

Figure 10 depicts a fitting curve for $\ln K_{ML}$ vs. $1/T$. The thermodynamic parameters are shown in Table 4. Since the value of ΔH is negative, adsorption is an exothermic reaction.

When ΔS is positive, it indicates that the degree of disorder has grown. Negative ΔG at a given temperature suggests the adsorption occurs spontaneously and it has a thermodynamic benefit. This result is in line with the theory that decreasing the temperature enhances adsorption.

Table 4. Thermodynamic parameters of Gasoil adsorption using cellulose@Fe₃O₄@ZIF-8 carbon aerogel magnetic.

Figure 10. The fitting curve of $\ln K_{ML}$ versus $1/T$.

3.8. Adsorption mechanism

The adsorption mechanism involves the various physical and chemical interactions between the adsorbate and the adsorbent. The interactions or forces can be electrostatic, hydrogen, vander Waals, coordination, or covalent. Unraveling the adsorption mechanism is crucial for optimizing the adsorption performance and designing more efficient adsorbents. The cellulose@Fe₃O₄@ZIF-8 carbon aerogel magnetic exhibits excellent adsorption efficiency for Gasoil removal from water, which can be attributed to several factors. These include electrostatic interactions between the negatively charged surface of the composite material and the positively charged Gasoil molecules, hydrogen bonding between the organic linkers of the MOF and gasoil molecules, and the porous nature of the composite material, which facilitates the rapid diffusion of Gasoil.

The synthesized adsorbent is very hydrophobic and has a high affinity for adsorbing Gasoil. The Gasoil is adsorbed into the sponge-and-Aerogel -structure sorbent in a short amount of time and then separated from the water. The data show that Gasoil spontaneously and physically adsorbed onto synthetic aerogel adsorbents. To perform physisorption, Gasoil must fill the pores of the aerogel. Since low-density aerogel constantly floats on the water's surface, the whole adsorption process occurs at the bottom.

3.9. Regeneration and reusability 339

The regeneration and reusability of the cellulose@Fe₃O₄@ZIF-8 carbon aerogel magnetic composite is essential for long-term and practical applications. The composite can be regenerated using a suitable desorption agent, such as NaOH, which allows for the recovery and reuse of the adsorbent material. The regeneration process can be performed multiple times, ensuring the long-term performance and cost-effectiveness of the composite for Gasoil removal. 340 341 342 343 344 345

The adsorption performance of the aerogel was estimated after eight consecutive cycles of utilization (Figure 11). The findings show that after eight uses, the adsorbent has a strong adsorption capacity. After eight cycles, the recycled adsorbent's capacity was evaluated. After eight times of uses, the adsorbent had a high adsorption capacity, and only around 10 % of its adsorption capacity was degraded. This property of magnetic carbon aerogel is precious. 346 347 348 349 350

Figure 11. Regeneration and reusability of cellulose@Fe₃O₄@ZIF-8 carbon aerogel magnetic. 351

3.10. Comparison with other adsorbents 353

Various adsorbents can remove Gasoil from aqueous solutions. Table 5 shows some examples of these adsorbents with their adsorption capacities. Overall, while there is limited research specifically comparing the effectiveness of cellulose@Fe₃O₄@ZIF-8 carbon aerogel magnetic to other adsorbents in removing Gasoil from water, the studies suggest that ZIF-8 and cellulose-based composites can be effective adsorbents for removing Gasoil from water. 354 355 356 357 358

Table 5. Comparison of the maximum adsorption capacities of Gasoil by different adsorbents. 359 360

4. Conclusion 362

In summary, the Biomass@Fe₃O₄@ZIF-8 adsorbent was synthesized through the in-situ growth of ZIF-8 on Fe₃O₄-modified cellulosic fibers, exhibits excellent Gasoil removal efficiency from aqueous solutions. The adsorption process was optimized using the Box-Behnken method, and the adsorption isotherms and thermodynamics were studied to understand the underlying mechanism. The adsorbent demonstrated a good reusability and environmental compatibility, making it a promising candidate for Gasoil removal in real-world applications.

The resulting adsorbent is a superhydrophobic polymer that efficiently removes water from Gasoil. Under an external magnetic field, the saturated adsorbent is rapidly separated from the water without filtration or centrifugation. This adsorbent helps to save time, money, and materials. Only approximately 10% of the sorbent's adsorption ability is lost after eight reuses, so it still has plenty of life.

The removal efficiency of Gasoil was studied using DOE to determine the optimal sorbent weight, starting Gasoil weight, and solution pH. By studying the impacts of numerous parameters on Gasoil removal, the starting concentration and adsorbent weight had the most significant influence. Using 0.5 g of Gasoil weight and 0.1 g of adsorbent weight is ideal for Gasoil removal. Under ideal circumstances, magnetic aerogels composed of cellulose@Fe₃O₄@ZIF-8 can remove all traces of Gasoil from the water. After entering the sponge and aerogel sorbent, Gasoil is quickly separated from water. The Gasoil removal capability of the adsorbent is very high (7 g.g⁻¹) in aqueous solutions.

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We wish to draw the attention of the Editor to the following facts, which may be considered as	542
potential conflicts of interest and to significant financial contributions to this work. We confirm	543
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Author and which has been configured to accept email from (hbashiri@kashanu.ac.ir).	551
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Tables:

Table 1.

Factors	Name	Low	Middle	High
		-1	0	+1
A	Adsorbent weight (g)	0.05	0.075	0.1
B	Gasoil weight (g)	0.5	0.75	2
C	pH	5	7.5	10

Table 2.

Run	Coded value(A)	Coded value(B)	Coded value(C)	Removal (%)
1	0.075	1.25	7.5	45.84
2	0.1	2	7.5	34.93
3	0.075	1.25	7.5	43.24
4	0.075	2	10	27.62
5	0.075	0.5	10	93.04
6	0.075	2	5	25.4
7	0.05	2	7.5	21.55
8	0.05	1.25	10	35.76
9	0.05	1.25	5	32.41
10	0.1	0.5	7.5	98.7
11	0.075	0.5	5	90.35
12	0.05	0.5	7.5	78.14
13	0.075	1.25	7.5	44.15
14	0.1	1.25	10	58.42
15	0.1	1.25	5	55.65

Table 3.

Models Isotherm	parameter	value
Langmuir	q_{max} (g.g ⁻¹)	6.99

	$K_L (\text{L.g}^{-1})$	2.68
	R^2	0.818
Freundlich	$K_F (\text{g.g}^{-1}(\text{g.L}^{-1})^{-\frac{1}{n}})$	3.716
	n	6.377
	R^2	0.9399
Temkin	$A (\text{L.g}^{-1})$	24951.76
	$B (\text{g.g}^{-1})$	0.446
	R^2	0.9333
Azizian et al	K_{ML}	1.941
	q_m	3.041
	R^2	0.8180

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Table 4.

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T(K)	$\Delta H(\text{J.mol}^{-1})$	$\Delta S(\text{J.mol}^{-1}.\text{k}^{-1})$	$\Delta G(\text{KJ.mol}^{-1})$
303.15			-5.917
313.15	-259.98	18.66	-6.103
323.15			-6.290

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Table 5.

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Adsorbent	Maximumadsorp tion capacity (g.g ⁻¹)	References
Agar-CMC-Silver	0.895	(Ali et al. 2021)
Crab shell biochar (CSB)	0.0934	(Cai et al. 2019)
Natural mud shale	0.025	(Yu et al. 2021)
Crab shell biochar (CSB)	0.451	(Han et al. 2021)
Fe ₃ O ₄ /Bent/rGO	0.085	(Ewis et al. 2022)
Fe ₃ O ₄ /Bent/MWCNTs	0.077	(Ewis et al. 2022)
Fe ₃ O ₄ /Bentonite	0.054	(Ewis et al. 2020)
MIL-53(Fe)	4.3	(Tajari &Bashiri 2023)
Cellulose fibers of PF plant	1	(Tajari &Bashiri 2023)
cellulose@Fe ₃ O ₄ @ZIF-8 carbon aerogel magnetic	7	
This work		

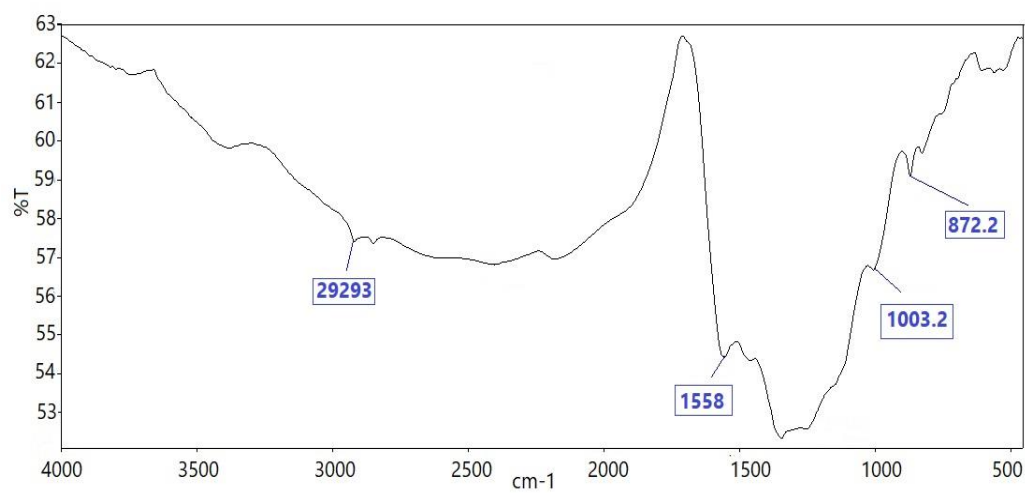
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Figures:

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Figure 1.

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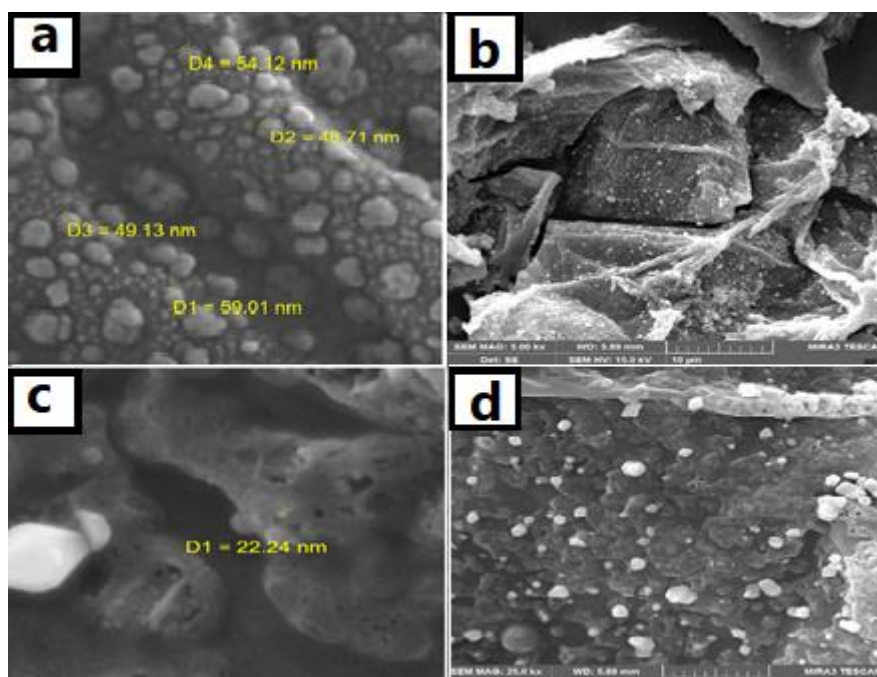


Figure 2.

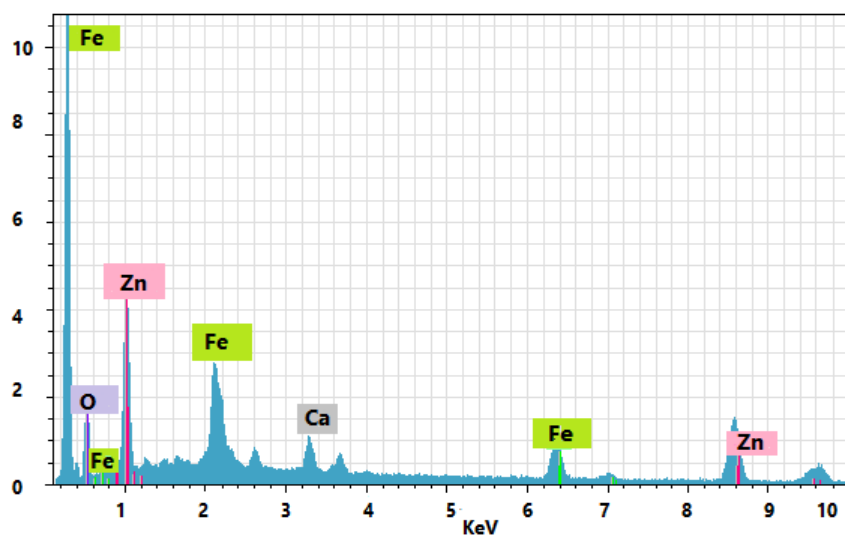


Figure 3.

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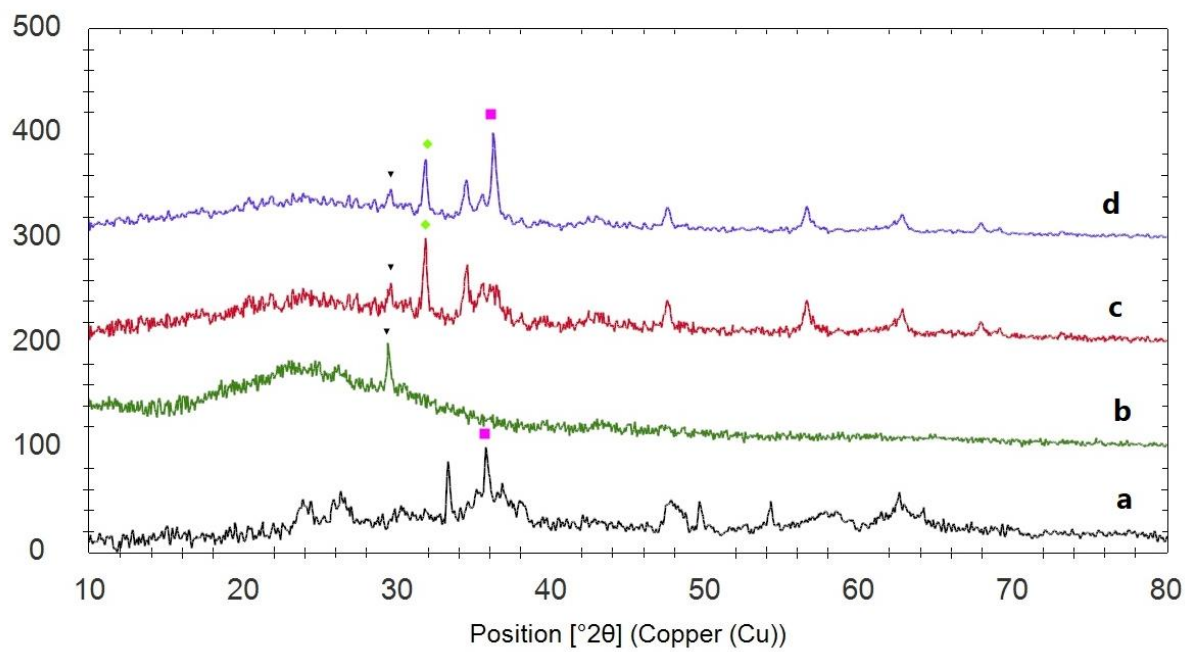


Figure 4.

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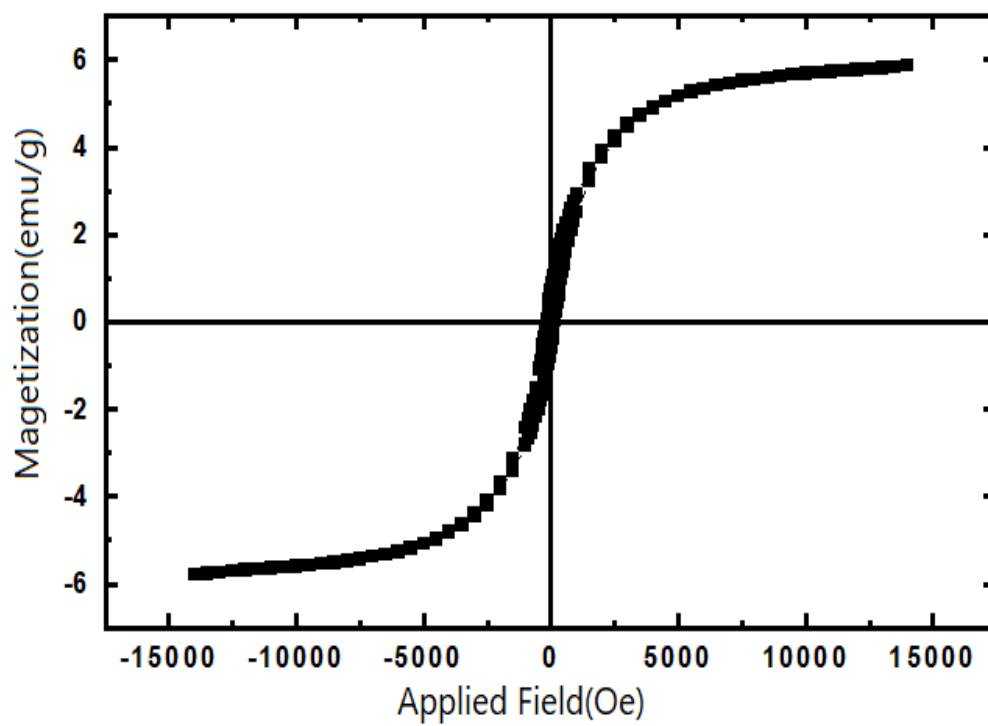
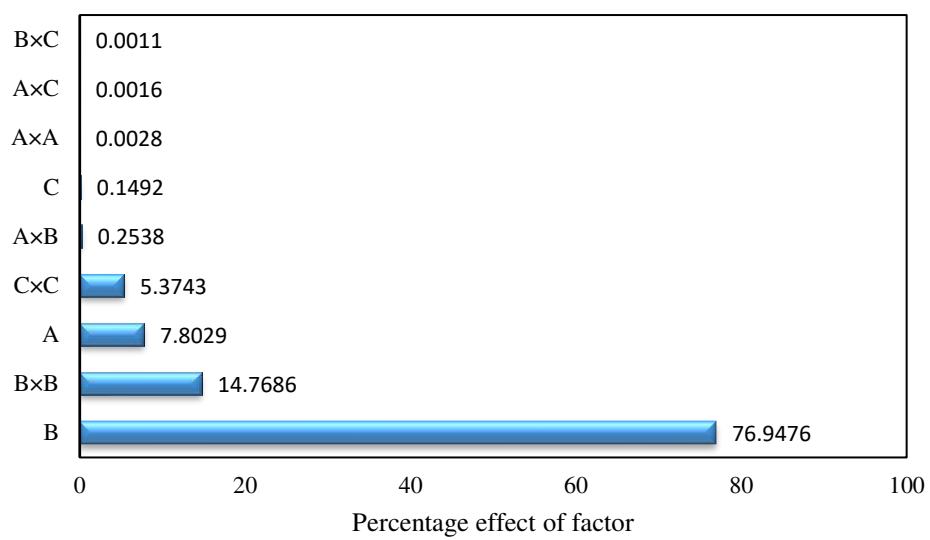


Figure 5.

**Figure 6.**

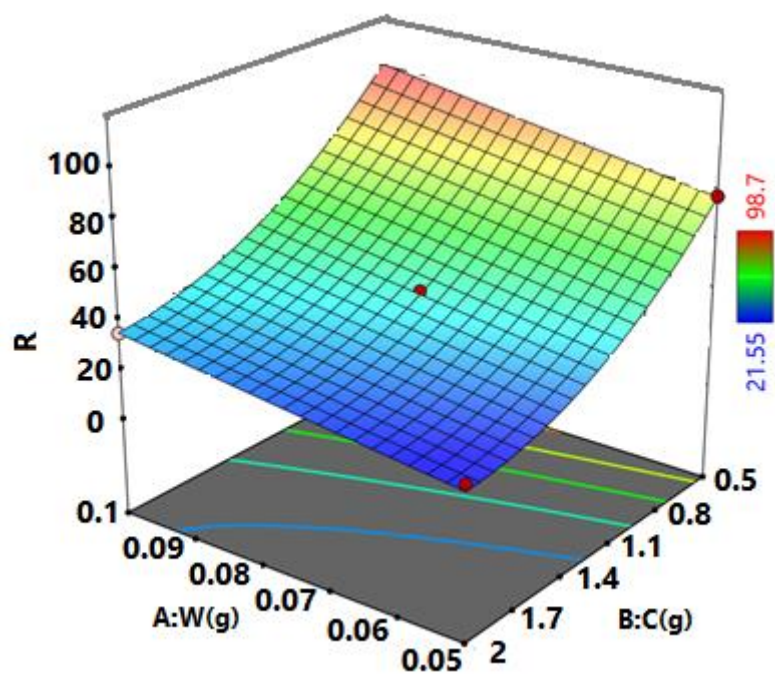


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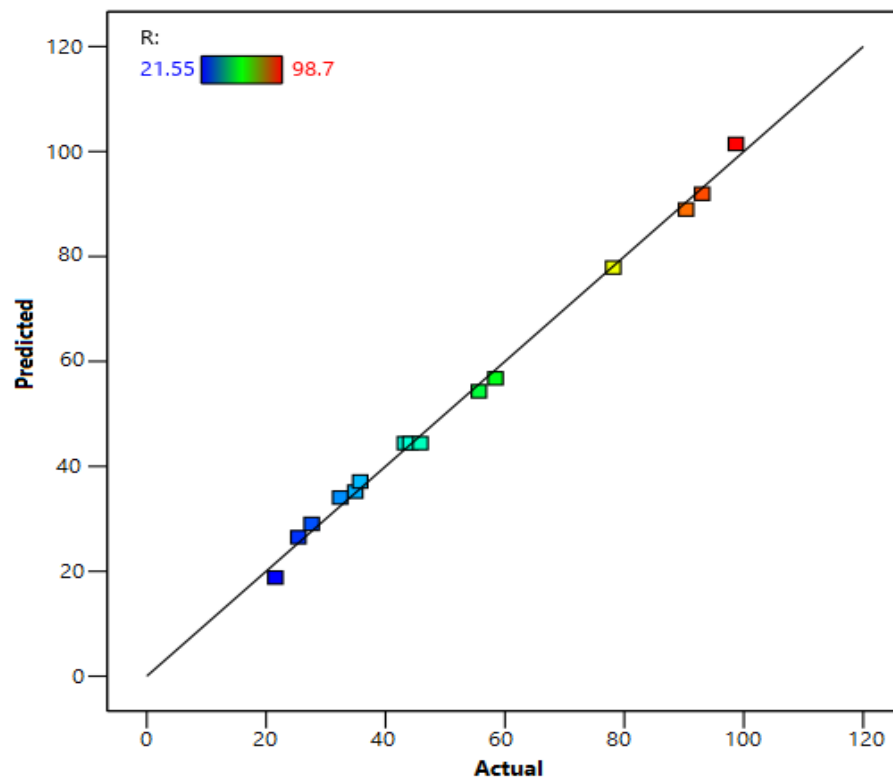


Figure 8.

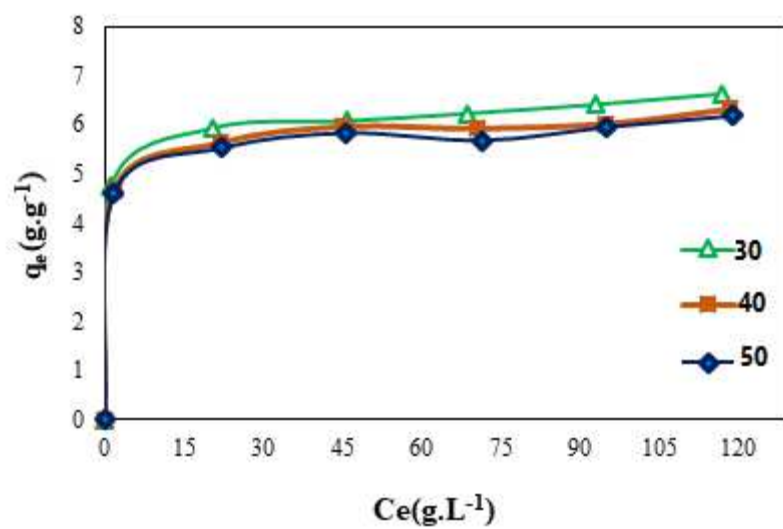


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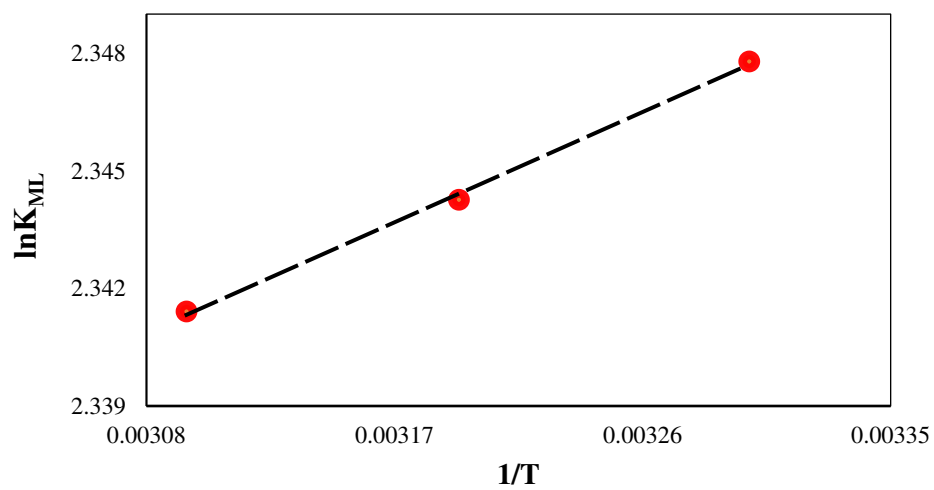


Figure 10.

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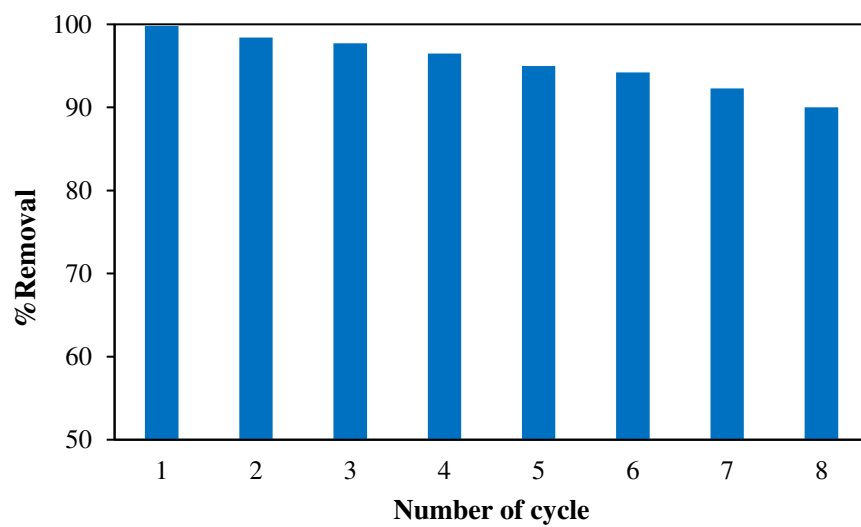


Figure 11.

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