Research Paper
Nanocomposite Fe-Co-V/Zeolite: Highly Efficient Composite for Removal of Methyl Orange Dye

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ABSTRACT

Background: In recent years, the dye industry has rapidly been developed. Dyes are one in all the foremost dangerous groups of chemical compounds found in industrial effluents, which are of considerable importance for reasons like reduced light permeability and therefore the subsequent disruption of the method of photosynthesis in water sources. This study was performed to remove dye using Fe-Co-V /Zeolite from a synthetic wastewater.

Methods: After chemical synthesis of the nanocomposite, its structure was studied by spectroscopic techniques. The experiments were performed under different pH values (3-11), contact times (5-50 min), absorbent dose (0.5-0.6g), stirring speed (240-60 rpm), and different concentrations.

Results: The results showed that the optimal and final conditions affecting the removal of methyl orange dye in the most suitable conditions for 200 mL of the solution with a concentration of 20 mg/L at pH equal to 3, contact time 20 min, adsorbent dose 0.2 g, jar speed 180 rpm and temperature 25°C, which finally with the application of the optimal data, at a concentration of 2.5 mg/L, the best efficiency was obtained. The examination of isotherm diagrams and isotherm coefficients showed that the adsorption process follows the Freundlich equation.

Conclusion: According to the obtained results and physical and chemical properties of the synthesized nanocomposite, it can be used to remove pollutants from the environment such as wastewater and air.

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1. Introduction

Dyes are one of the most dangerous groups of chemical compounds found in industrial effluents, which are of considerable importance for reasons such as reduced light permeability and the subsequent disruption of the process of photosynthesis in water sources [1, 2]. Various industries such as textiles, paper making, color images, pharmaceuticals, food industries, and cosmetics, use these dyes in abundance [3, 4]. When these dyes enter water bodies, photosynthesis and aquatic life are significantly impacted as a result of reduced light penetration. These dyes also contain metals, chlorides, and other substances that even at very low concentrations of colors (less than 1 mg/L) are toxic. Dye consumption in the world's textile industries is estimated at more than 10,000 tons per year, with this consumption, 1000 tons of dye enters the effluent of these industries annually. Also, about 100,000 dyes are commercially available with a production rate of 7×10^5 tons per year, about 2 percent of these dyes are released into water resources as waste [5, 6]. AZO dyes are the largest and most important category of colors and are widely used. The most characteristic feature of these dyes is having one or more AZO groups -N=N- that act as a bridge between the two organic parts of the dye and at least one of these groups is aromatic. The release of these substances in the environment is the biggest source of pollution for natural ecosystems. In addition, these substances are converted to aromatic amines through various processes such as hydrolysis, oxidation, and other chemical reactions, which are carcinogens [7, 8].

From an aesthetic point of view, color compounds have a negative effect on water quality for drinking and other uses, and at the same time cause allergies, skin irritation, cancer, and genetic mutations in humans. About 1 to 15% of dyes are lost during dyeing and polishing processes and enter the environment as effluents. Therefore, it is necessary to remove these pollutants [2, 9]. Numerous studies have used a variety of methods to remove and reduce color from aqueous solutions, such as physical methods (filtration/adsorption by various adsorbents), chemical methods (coagulation, flocculation, and chemical oxidation), and biological methods. A study was performed for simultaneous removal of methyl orange and hexavalent chromium from water with magnetite-based polymeric nanocomposite [10]. In another study to remove of methyl orange was conducted to synthesize the nanocomposites of TiO_2 photocatalyst by doping through graphene oxide and silver nanoparticles using sol-gel along with solvent-assisted heat treatment methods [11]. Also, the removal of methyl orange with poly (acrylonitrile-co-styrene) and carbon nanotubes (nanocomposites) [12]. Therefore, the application of a functional ferrocene firmly heterogenized over a modified nano-sized SiO_2–Al_2O_3 mixed-oxides was reported as a novel adsorbent for the removal of methyl orange from aqueous solution [13]. In the present study, the iron-cobalt-vanadium nanocomposite was synthesized on a nanometer-scale zeolite substrate and evaluated as a suitable composite for methyl orange removal.

2. Materials and Methods

Synthesis of Fe-Co-V/Zeolite

First, a certain amount of vanadium dioxide (V_2O_5) was dissolved in one liter of 3 M sodium hydroxide solution until a colorless yellow solution was obtained. The contents of the filter vessel and the solution were titrated with 6 M nitric acid to a pH of 6, at this stage, most of the vanadium was added to the decavanadite solution complex-V10O286, which is pink. Next, the calculated amount of zeolite powder was poured into a special flask containing decavanadite solution and equipped with a mechanical stirrer and the contents of the flask were stirred for 24 h at 60°C. Then the molar solution of iron nitrate and cobalt nitrate was added dropwise to the stirring flux contents and the contents stirred for another 5 h at 60°C. Finally the contents of the flask were filtered and dried. It should be noted that these materials was calcined at 600°C for 5 h using a reactor and reduced with hydrogen at 20 bar and 500°C for 5 h. At this stage, the nanocomposite was partially regenerated and activated. The structure and morphology of the nanocomposite were analyzed after the reduction step (Figure 1).

It should be noted that we stated the results and the complete analysis (details) of the synthesis method in another article [14]. The experiments were performed to optimize variables at different levels (five pH levels including acidic, alkaline and neutral conditions), five dye concentration levels (mg/L), five nanocatalyst dose levels (g), five-time levels (min), and different temperature and stirring conditions.

A calibration curve for methyl orange dye at concentrations of 5 mg/L to 30 mg/L of the dye was obtained by measuring the absorption of solutions at a maximum wavelength of 515 nm for methyl orange dye using a spectrophotometer and glass tube. The line equation corresponds to the obtained absorptions of y=21.643x - 1.2833 for the dye, where x is the dye concentration in mg/L. This calibration curve was used to determine the equilibrium concentration of methyl orange dyes in
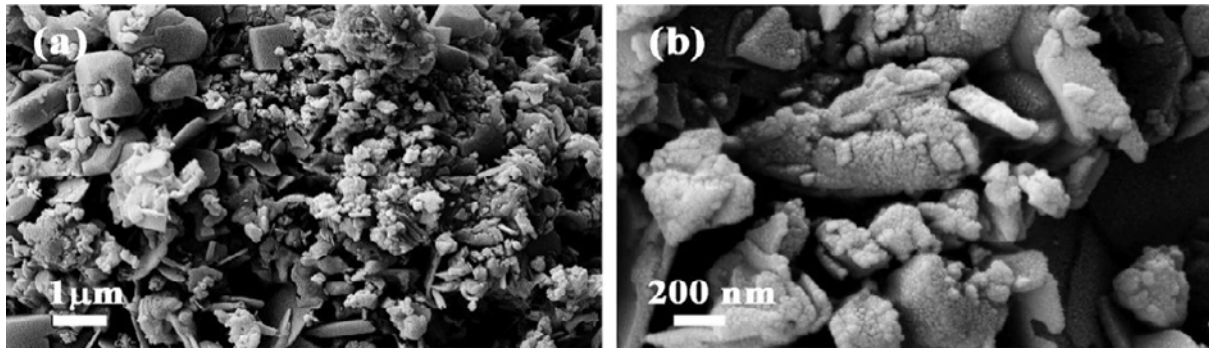
Figure 1. FESEM images of Fe-Co-V/Zeolite at different scales (a, b)

order to calculate the degree of dye output. The results have been shown in Figure 2.

To evaluate the results obtained from the optimization of various parameters, the percentage of color output was calculated using the equation calculated using Equation 1, and adsorption capacity (qt) was calculated via Equation 2 as follows:

$$1.\ R=\frac{Co\text{-}C}{Co}\times100$$

$$2.\ qt=\frac{(Co\text{-}C)V}{W}$$

where, Co and C are the initial and residual color concentrations (mg/L), respectively, and V is the volume of solution (L) and W is the mass of adsorbent (gr).

The maximum amount of monolayer adsorption capacity (qt) was obtained by fitting the Langmuir equation (Equation 3).

$$3.\ \frac{1}{qe}=\frac{1}{qm}+\frac{1}{qm.K_L.Ce}$$

where, Ce (mg/L) and q (mg/g) are the equilibrium dye concentration and equilibrium amounts of adsorption, respectively. qe (mg/g) is the maximum adsorp-

tion capacity of dye and K_L is also the Langmuir equilibrium constant. The experimental data were also fit to the Freundlich model, which assumes the heterogeneous nature of adsorption. The linear form of the Freundlich isotherm model is expressed as follows (Equation 4):

$$4.\ ln_{qe}=ln_{KF}+\frac{1}{n}\ ln\ Ce$$

where, KF and n are Freundlich equilibrium constant.

In this study, we were exposed to toxic and dangerous substances, so it was necessary to observe safety and health principles during the tests. Therefore, before starting the tests, all safety and health aspects were considered and the necessary measures were taken to reduce the potential risks.

3. Results and Discussion

To obtain the highest amount of dye removal from solution, effective parameters including pH, adsorbent (iron-cobalt-vanadium nanocatalyst) dose, initial dye concentration, contact time, jar speed (rpm), and temperature in the laboratory were examined.
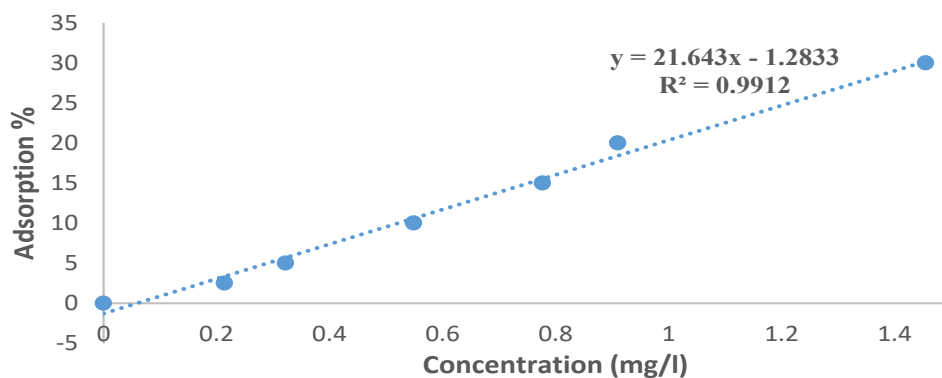

Figure 2. Calibration curve to determine the methyl orange dye concentration
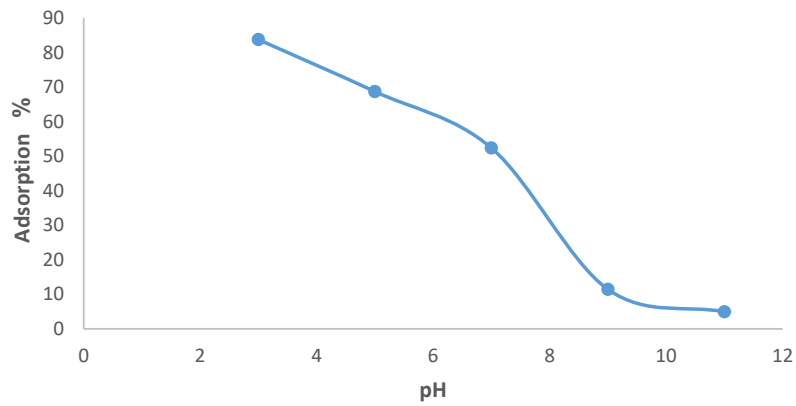
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Figure 3. Effect of pH on the adsorption of methyl orange dye at a concentration of 20 mg/L
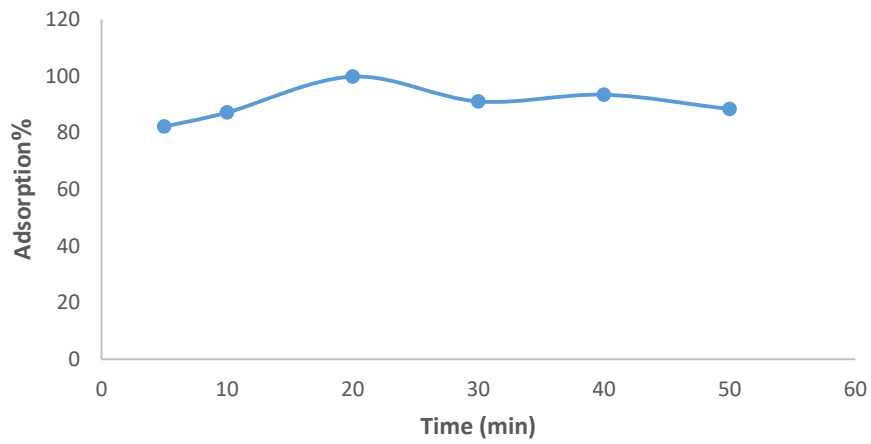

Figure 4. Effect of contact time (min) on the adsorption of methyl orange dye at a concentration of 20 mg/L
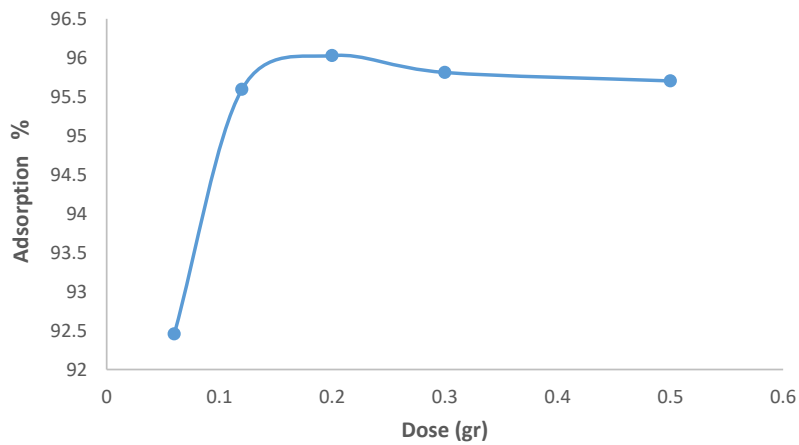
Figure 5. Effect of adsorbent dose (gr) on the adsorption of methyl orange dye at a concentration of 20 mg/L
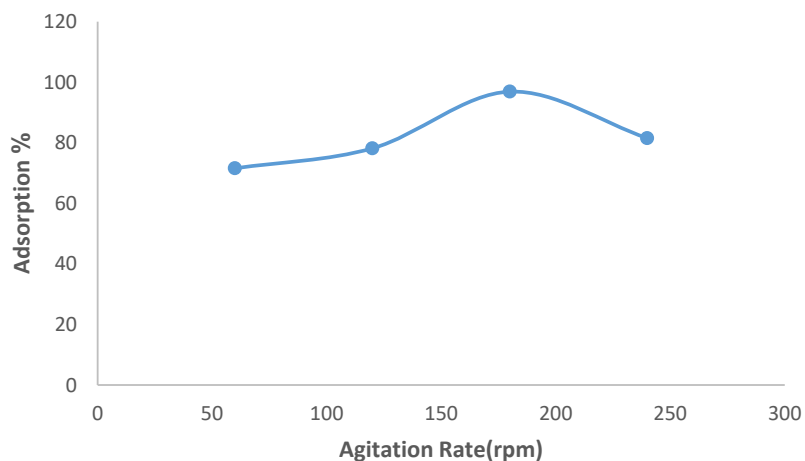
Figure 6. Effect of stirring speed (rpm) on the adsorption of methyl orange dye at a concentration of 20 mg/L

Figure 7. Effect of temperature (°C) on the adsorption of methyl orange dye at a concentration of 20 mg/L

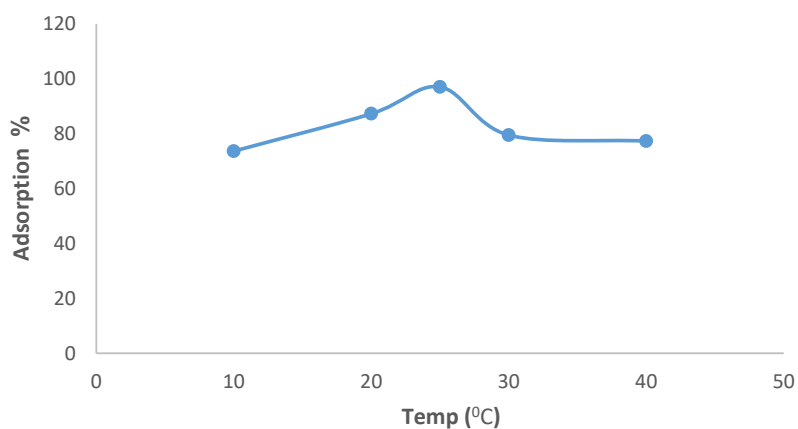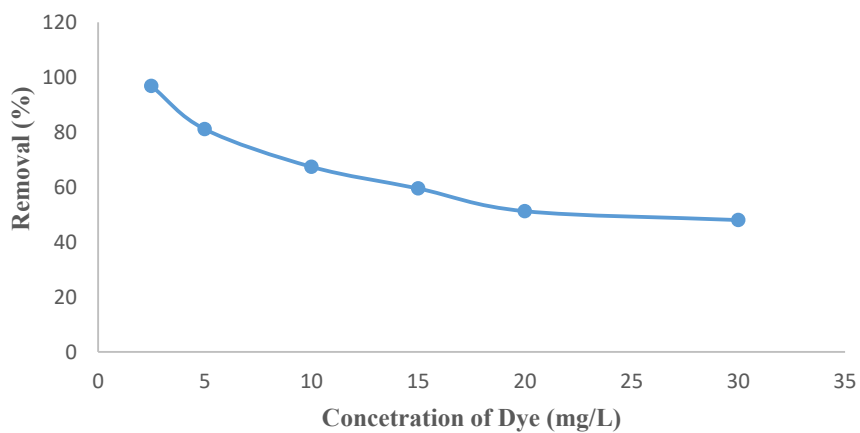**Figure 8.** Removal of methyl orange at different concentrations (optimal conditions: pH 3, adsorbent dose 0.2 g, time 20 min, temperature 25°C)

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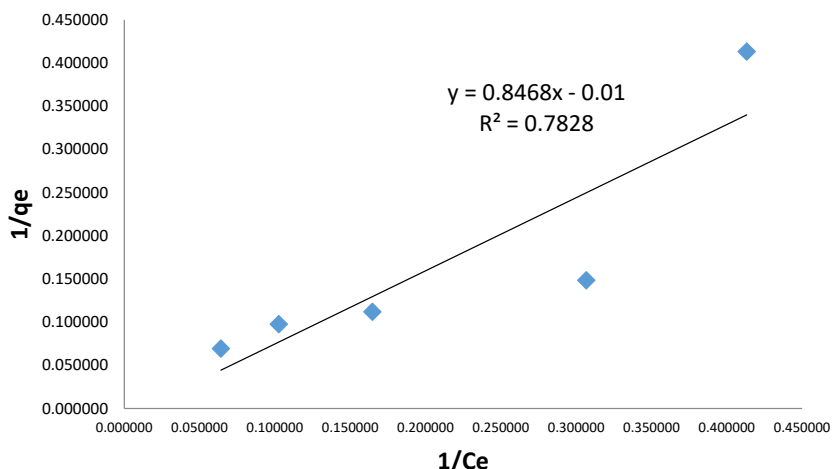
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$$y = 0.8468x - 0.01$$
$$R^2 = 0.7828$$

Figure 9. Langmuir isotherm of adsorption of methyl orange by the adsorbent

Effect of pH

The effect of pH on the adsorption of methyl orange dye using adsorbent is shown in Figure 3 (solution volume 200 mL, concentration 20 mg/L, adsorbent dose 0.3 mg, jar speed 180 rpm, and time 30 min). Among the range of pHs including acidic, neutral, and alkaline, the best pH of 3 for methyl orange dye was considered the optimal value.

Effect of contact time

The effect of contact time on the adsorption rate of methyl orange dye using adsorbent was considered as the optimal time according to Figure 4, 20 min.

Effect of adsorbent dose

The effect of adsorbent dose on methyl orange dye adsorption using adsorbent is shown in Figure 5 on the

percentage of dye output. The adsorbent value of 0.2gr was considered as the optimal adsorbent value.

Effect of stirring speed of solution

Figure 6 shows the effect of turbulence rate on the adsorption rate of methyl orange dye using adsorbent. And, 180 rpm was selected as the optimal speed.

Effect of temperature

Figure 7 shows a graph of the effect of temperature on the percentage of methyl orange dye output. The temperature of 25°C was considered as the optimal temperature.

Effect of dye concentration

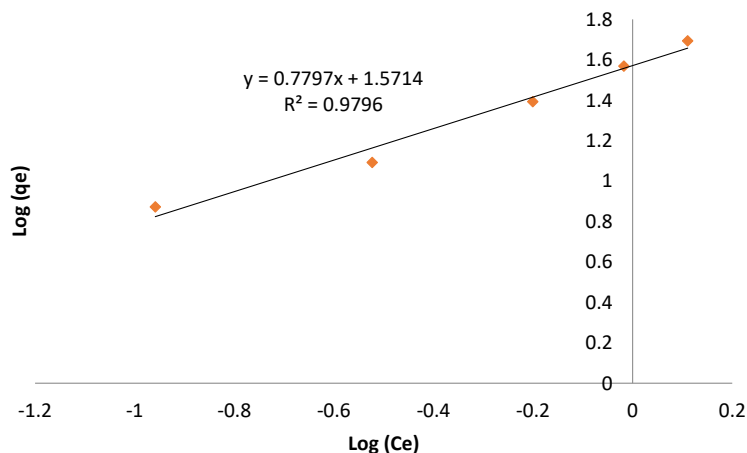To evaluate the percentage of dye output under optimal conditions at other concentrations, concentrations

$$y = 0.7797x + 1.5714$$
$$R^2 = 0.9796$$

Figure 10. Freundlich isotherm of adsorption of methyl orange by the adsorbent

Table 1. Percentage of methyl orange dye output based on initial concentration changes

Initial Concentration (mg/L)	Equilibrium Concentration (mg/L)	Removal (%)
2.5	0.080	96.79
5	0.94	81.08
10	3.26	67.38
15	6.07	59.49
20	9.75	51.22
30	15.59	48

of 2.5-30 mg/L were investigated. Thus, in 1200-mL Erlenmeyer, different amounts of methyl orange solution with a concentration of 1000 mg/L were added and after volumizing with distilled water, all solutions for methyl orange dye pH=3, 0.2 g adsorbent, for 20 min, the temperature of 25°C in the jar test machine was stirred at 180 rpm. Then, by filtering the solution from the filter paper, the absorption rate was read by UV-Vis and the amount of dye adsorption was calculated using the calibration curve (Table 1 and Figure 8).

Adsorption is a sustainable separation process and an effective way to remove contaminants (such as industrial colors) from water and wastewater. And due to its low cost, high flexibility, high sensitivity to organic compounds, it has environmental benefits. Many adsorbents are used to adsorb dyes from effluents, the most widely used of which is activated carbon, which is prepared from various materials such as worthless waste as agricultural waste. However, owing to the increase of various pollutants and organic compounds, it is necessary to prepare nanomaterial with new structures to simultaneously remove dyes and other organic compounds in effluents or water resources [15, 16]. In general, physicochemical properties (composition of nanoparticles, degree of malformability, size of crystals and nanoparticles, and specific surface area) and other properties of zeolites significantly affect the efficiency of metal nanoparticles used with zeolite. Also, the use of this method during the synthesis of nanocomposites leads to decreasing the size of crystals, increasing the number of spherical formations compared to untested samples, and their accumulation in larger forms. Zeolite substrate nanocomposites have the potential to be used as a catalyst for organic synthesis and adsorbents in wastewater treatment due to their large surface area and the presence of metals with oxidizing activity [17, 18].

Control of adsorption performance of an adsorbent depends on the nature of the adsorbent such as physical structure (degree of porosity, particle size), chemical structure, functional groups on the adsorbent surface, the chemistry of the adsorbent (polarity, molecular weight, size, etc.) and finally different conditions, such as pH, ionic strength, temperature, the concentration of solute, etc. [19].

Therefore, in this study, the effect of variables such as adsorbent dose, solution pH, temperature, mixing speed of solution and adsorbent, contact time, initial dye concentration were investigated. Optimal and final conditions affecting the removal of methyl orange

Table 2. Isotherm parameters of methyl orange adsorption by the adsorbent

Models	Parameters	Values
Langmuir	q_m(mg/g)	100
	K_L(L/mg)	0.01
	R^2	0.78
Freundlich	kf (mg/g(L/mg)1/n)	37.15
	n	1.29
	R^2	0.97

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Table 3. Efficiency of methyl orange removal by other adsorbents.

Materials	Highest Efficiency (%)	qmax (mg/g)	pH	Temp (°C)	Method qmax of Determination	Ref.
Nanocomposite Fe-Co-V/Zeolite	96.79	37.15	3	25	Freundlich	Present study (2021)
Polyvinylene fluoride/PEDOT mats	75	146.2	3	20	Langmuir	[36]
Ni–Co–S/hexadecyltrimethyl ammonium bromide	99	2355	--	25	Langmuir	[37]
Fe_3O_4/AC	--	150.4	7	25	Langmuir	[38]
Mesoporous carbon	--	9091	3		Langmuir	[39]
CuO NPs	--	217.4	6.5	60	Langmuir	[40]
Chitosan microspheres	--	207.0	3.10	--	Langmuir	[41]
Mesoporous ZSM-5 zeolite	94.3	25	1	30	Langmuir	[42]
Magnetic lignin-based CNPs	90.17	113	5	20	Langmuir	[43]
Magnetic nanoporous Fe/MCM-41	96.7	5000	2	20	Langmuir	[44]
Nanoporous carbon	100	18.80	--	25	Langmuir	[45]
Fe_2O_3/AC	80	362	3	20	Langmuir	[46]
Zeolite imidazolate framework-8	--	18	--	--	Langmuir	[47]
ZIF-67	--	16.30	--	--	Experiment	[48]
$CoFe_2O_3$/MgAl-hydrotalcite	--	1220	6	25	Langmuir	[49]
Chitosan/Fe_3O_4	95	758	7	25	Langmuir	[50]

dye in the most suitable conditions for dye removal, for 200 mL of the solution with a concentration of 20 mg/L at pH: 3, contact time: 20 min, adsorbent dose: 0.2 g, agitation: 180 rpm, temperature: of 25°C was obtained, which finally with the application of optimal data, the best efficiency was obtained at a concentration of 2.5 mg/L.

In a study, the removal of AZO dyes in water was performed using Nanosorbents. The nanoparticles used in this study were related to the nickel oxide nanoparticles. The experimental data were analyzed via the Longmuir and Freundlich isotherms. Optimal data followed the Freund Leach isotherm. In this study, the adsorption efficiency of methyl orange dye was measured at pH=6 and the adsorbent dose was 0.05 g at 95% [20]. In the present study, according to Figures 9 and 10 and Table 2, which show the diagrams and values of the methyl orange adsorption isotherm parameters by the adsorbent, the data followed the Freundlich model. Moreover, Zhu et al. in 2010 performed the adsorption of AZO anionic dyes with chitosan/kaolin/ɣ-Fe_2O_3 nanoparticles [21].

In a study, the results showed that, the Ni@ZIF-67 nanocomposite adsorbed more dye in mild acidic condition (qe=24.24 mg/g, pH=6) as compared to acidic (qe=17.69 mg/g, pH=2) and basic medium (qe=15.74 mg/g, pH=10). The Langmuir, Freundlich adsorption isotherm models were applied and results showed that the Langmuir model proved to be best fitted [22]. In different studies with nanocomposites done to remove methyl orange alone or with other dyes, different results have been obtained; removal of 87% at pH equal to 5 and followed by the Langmuir model by means of cobalt hydroxide NPs [23, 24], removal dye with NiO nanoflakes (pH=2, %removal=84.6 and Langmuir model) [25], removals using α-Fe_2O_3 NPs (pH=2, %removal=90 and Langmuir model) [26], removals using Nickel hydroxide nanocatalyst (%removal=87 and Langmuir model) [27]. In the present study, due to the simultaneous presence of iron, cobalt, and vanadium metals, the best removal efficiency was obtained in pH equal to 3 with an efficiency of 96.79%, which followed the Freundlich model. By using different methods and processes, methyl orange dye can be removed from solutions (water/

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wastewater), which has been studied in various studies, including adsorption methods [28, 29], electrochemical treatment [30], ozonation [31, 32], biological [33], nanocomposites [34], photocatalyst [35]. In this study, the experiments were performed using the adsorption process and suitable results were obtained for dye removal. Studies on the removal of methyl orange have been performed by various methods, as shown in Table 3. Adsorption, oxidation, and photocatalytic processes are common methods, respectively.

4. Conclusion

In this study, adsorption and removal of methyl orange from synthetic effluent using the iron-cobalt-vanadium nanocomposite on zeolite bed was performed. It was determined that the nanocomposite adsorbed the dye with a concentration of 2.5 mg/L in an acidic medium and separated it from the solution. Based on the nanocatalyst structure of the present study, it can be used for other dyes and other organic pollutants resistant to biodegradation in aqueous solutions. In future studies, it is suggested to investigate the photocatalytic properties of synthesized nanocomposites, synthesis of zeolite compounds and their characteristics in the removal of other dyes, application of more complex colors or structures such as sulfur dyes to remove and use recently synthesized nanocomposites. The modified nanocomposite are set to be considered by researchers in real conditions.

Ethical Considerations

Compliance with ethical guidelines

There were no ethical consideration to be considered in this research.

Funding

Authors' contributions

Conceptualization and Supervision: Reza Jalilzadeh; Abdolreza Karbul; Methodology: Reza Jalilzadeh; Mohammad Kazem Mohammadi; Investigation, Writing-original draft, and Writing-review & editing: All authors; Data collection: Abdolreza Karbul; Data analysis: Reza Jalilzadeh and Mohammad Kazem Mohammadi; Funding acquisition and Resources: Mohammad Kazem Mohammadi; Abdolreza Karbul.

Conflict of interest

The authors declared no conflict of interest.

Acknowledgments

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