Materials Research Express

PAPER

Fabrication of magnetic cobalt-nickel ferrite nanoparticles for the adsorption of methyl blue in aqueous solutions

Hezhong Ouyang, Shuyan Liu, Dandan Liu, Yan Wang, Shuping Xu and Shengying Pan

The People’s Hospital of Danyang, Affiliated Danyang Hospital of Nantong University, Zhenjiang 212300, People’s Republic of China

E-mail: dyxushuping@163.com and dypanshengying@163.com

Keywords: Co0.5Ni0.5Fe2O4 nanoparticles, combustion-calcination technique, methyl blue, adsorption kinetics, adsorption isotherm

Abstract

An innovative method of combustion–calcination of a nitrate–ethanol solution to produce magnetic Co0.5Ni0.5Fe2O4 nanoparticles was developed. The calcination temperature and the volume of ethanol were two pivotal elements that determine the properties of the Co0.5Ni0.5Fe2O4 nanoparticles in this study. When the volume of ethanol used was increased from 20 ml to 40 ml, the crystallinity of the Co0.5Ni0.5Fe2O4 nanoparticles increased; further increase of the volume of ethanol decreased the crystallinity. The smallest nanoparticle was obtained using 20 ml ethanol. As the calcination temperature increased from 400 °C to 700 °C, the saturation magnetization of the Co0.5Ni0.5Fe2O4 nanoparticles increased from 12.8 emu g−1 to 30.8 emu g−1. Co0.5Ni0.5Fe2O4 nanoparticles fabricated using 20 ml ethanol at 400 °C were used to study the removal of methyl blue (MB) by adsorption. Experimental data revealed that the adsorption was best described by pseudo-second kinetics. The adsorption isotherm applied the Temkin model, which indicated the presence of a single and multilayer associative mechanism in the adsorption of MB on the Co0.5Ni0.5Fe2O4 nanoparticles. The effect of pH and recycling on the adsorption was measured. At pH values ≥5, the adsorption was high. After eight cycles of use and recycling, the relative removal rate of MB by the Co0.5Ni0.5Fe2O4 nanoparticles was 75% of the initial adsorption value.

1. Introduction

Dyes are widely used in many industries, such as food, medicine, printing, dyeing, and cosmetic industries [1]. With the advancements made in the dye industry, an increasing amount of dye wastewater is discharged into water bodies, increasing the levels of water pollution. Most of the wastewater has intense color that blocks the incident light; this affects the aquatic flora and fauna, thereby severely damaging the ecological balance of aquatic ecosystems [2]. Moreover, most dyes are hazardous, often showing mutagenic, teratogenic, and carcinogenic effects. Many dyes in wastewater are very stable and cannot be degraded by natural methods [3, 4]. Therefore, the removal of dyes from wastewater is essential. Numerous technologies have been proposed for dye removal, including adsorption, electrolysis, precipitation, and membrane separation. Among them, adsorption is generally considered to be effective owing to its low cost, high efficiency, and negligible secondary pollution [5–7].

Various adsorbents, such as active carbon, natural cellulose, and nanomaterials, have been extensively developed for the treatment of dye wastewater [8, 9]. However, it is generally difficult to separate nanomaterial solutions, and for convenience, magnetic nanomaterials are employed to remove dyes based on the abilities of the magnetic material, such as the large specific surface area and facile separation from solutions under an applied magnetic field.

Many techniques have been applied to fabricate magnetic nanomaterials, such as coprecipitation [10], hydrothermal method [11], sol–gel [12, 13], and combustion–calcination technique [14–17]. Among them, the combustion–calcination is a newly developed facile process. It has a short processing time, short preparation cycle, and low cost, and can be used to control nanomaterials by changing the amount of the solvent and the combustion mechanism [18, 19].
We fabricated magnetic Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles using the combustion–calcination of a nitrate–ethanol solution. The removal of methyl blue (MB) by the magnetic Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles was investigated.

2. Experimental process

2.1. Fabrication and analysis of Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles

The combustion–calcination of a nitrate–ethanol solution was applied to fabricate Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles. First, for characterization, the quantity of the product was controlled at 2 g; the nitrates of Co, Ni, and Fe were added into a 100 ml beaker in the molar ratio of 1:1:4, followed by the addition of a specific volume of ethanol (20, 30, 40, 50, and 100 ml). After stirring for 1–2 h, the mixture was transferred to a melting pot and heated. Before the fire was quenched, the crucible containing the intermediate was placed in a temperature-controlled furnace to be calcined at different temperatures (400, 500, 600, and 700 °C) for 2 h. Thereafter, the furnace was cooled to or below 50 °C, and the product was ground to obtain magnetic Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles.

The morphology, composition, and phase characterizations were examined using scanning electron microscopy (SEM), energy dispersive x-ray spectroscopy (EDX), transmission electron microscopy (TEM), and x-ray diffraction (XRD) (Rigaku D/max 2500 PC). Magnetic properties were examined using a vibrating sample magnetometer.

2.2. MB adsorption performance of Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles

MB solutions with different concentrations (100, 200, 300, and 400 mg l$^{-1}$) were prepared for the adsorption experiments. The prepared MB solution (2 ml) and magnetic Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles (5 mg) were transferred into a centrifuge tube and subjected to ultrasound for dispersing the nanoparticles, and the adsorption proceeded for 10–120 min. After the adsorption was complete, the mixture was centrifuged for 5 min. Then, the absorbance of the supernatant was determined at a wavelength of 530 nm using an ultraviolet spectrophotometer (UV-2550). The adsorption and removal efficiency of MB by the Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles can be determined according to equations (1) and (2) [20].

\[
q = \frac{m_{MB}(A_{MB} - A_{mag})}{m_{mag}A_{MB}} \quad (1)
\]

\[
\eta = \frac{(C_0 - C)}{C_0} \times 100\% \quad (2)
\]

where $q$ is the amount of MB adsorbed on the Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles (mg g$^{-1}$), $m_{MB}$ is the mass of MB (mg), $m_{mag}$ is the mass of the Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles (g), $A_{MB}$ is the absorbance of the initial MB solution, and $A_{mag}$ is the absorbance of the MB solution after the adsorption. $C_0$ and $C$ (mg l$^{-1}$) represent the initial and final concentrations of MB, respectively.

At MB concentration of 200 mg l$^{-1}$, the adsorption times were controlled to the equilibrium time, and the adsorption isotherm was obtained at the indoor temperature. The pH of the MB solutions was adjusted using NaOH or HCl solutions (1 M), and the effect of pH on the adsorption of MB was determined. After adsorption, the Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles were collected and calcined, following which, the adsorption of MB onto them was measured again to investigate the cyclic utilization of Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanomaterials for MB adsorption.

3. Results and discussion

3.1. Analysis of the structure and properties of the Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles

The structural characterization of the Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanomaterials is shown in figure 1. Figure 1(A) depicts the morphology analyzed by SEM, wherein the Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles have a spherical shape with a particle size of 25 nm. Figure 1(B) shows the EDX spectrogram of the Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles, and the ratios of Fe, O, Co, and Ni agree with the proposed ratio. TEM images are shown in figure 1(C), which confirm the SEM results. Figure 1(D) shows the selected area electron diffraction (SAED) image wherein the magnetic Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles show a polycrystalline aggregate, which was determined by the presence of diffraction dots and diffraction rings. The structure and porosity of the Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles were obtained using N$_2$ adsorption–desorption. As shown in figure 1(E), the isotherm is of type IV (according to the IUPAC classification), and the hysteresis loop is of type H3 [21]. The pore size of the magnetic Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles is in the range 2–16 nm, indicating a mesoporous structure with a surface area of 112.4 m$^2$ g$^{-1}$.

Figure 2 shows the XRD patterns and hysteresis loops of the Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles fabricated using various volumes of ethanol at different calcination temperatures. The XRD patterns and hysteresis loops of the
magnetic Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles fabricated using 20 ml ethanol at 400 °C are shown in figures 2(A) and (C), respectively. With increasing calcination temperature, the peak intensity increases, suggesting that the degree of crystallinity also increases, which contributes to the increase in the crystal grain size. Simultaneously, the saturation magnetization also increased from 12.8 emu g$^{-1}$ to 30.8 emu g$^{-1}$ with the calcination temperature, as illustrated in figures 2(C). Figures 2(B) and (D) show the XRD patterns and hysteresis loops of the magnetic Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles fabricated using 20–100 ml ethanol at 400 °C. With increasing ethanol volume, the intensity of diffraction peaks increases until it reaches the maximum at 40 ml and subsequently decreases with further increase in the volume of ethanol. This tendency is explained by the fact that increasing ethanol volume increases the burning time, and a longer burning time contributes to the crystal growth, thereby enhancing crystallinity. However, further increase in ethanol volume enhances the dispersion effect of absolute alcohol, resulting in a decrease in the crystallinity and the diffraction peak intensity. The magnetic Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles fabricated using 20 ml ethanol at 400 °C, with the lowest crystallinity and the highest adsorption capacity, were used in subsequent MB adsorption experiments.
3.2. Adsorption kinetics

The adsorption kinetics research was significant for the adsorption design of MB in wastewater treated with Co0.5Ni0.5Fe2O4 nanoparticles. Three adsorption kinetics models were employed to assess the adsorption of MB on the Co0.5Ni0.5Fe2O4 nanoparticles.

The formulas of the three kinetics models are given in equations (3)–(5) [22–24].

\[
\ln(q_e - q_f) = \ln q_e - k_1 t
\]  

(3)

\[
\frac{t}{q_i} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e}
\]  

(4)

Figure 2. (A), (B) XRD patterns and (C), (D) hysteresis loops of the Co0.5Ni0.5Fe2O4 nanoparticles fabricated at various calcination temperatures with different ethanol volumes.

Figure 3. (A) Pseudo-second-order model curves and (B) removal efficiency of adsorption with diverse initial concentrations of MB.
where \( q_t \) and \( q_e \) (both in mg g\(^{-1}\)) are the quantities of MB adsorbed at the given and equilibrium times, respectively; \( k_1 \) (min\(^{-1}\)), \( k_2 \) (g mg\(^{-1}\) min\(^{-1}\)), and \( k_3 \) are the rate constants for the three models, respectively; \( x_i \) is a parameter related to the boundary layer thickness.

Figure 3(A) shows the graph of adsorption \((q_t)\) against time \((t)\) applied to the pseudo-second-order model curves at different starting MB concentrations. Accordingly, the initial MB concentration determines the adsorption values and shapes of the curves. The adsorption of MB by the Co\(_{0.5}\)Ni\(_{0.5}\)Fe\(_2\)O\(_4\) nanoparticles rapidly increases with time initially. The rate of adsorption then gradually decreases and reaches equilibrium. The approximate adsorption equilibrium time is 30 min. The efficiencies of MB removal by the Co\(_{0.5}\)Ni\(_{0.5}\)Fe\(_2\)O\(_4\) nanoparticles are shown in figure 3(B). With increasing MB concentration, the MB removal efficiency first increases and subsequently decreases. In particular, the MB removal efficiency increases from 85.3% to 93.4% as the concentration of MB increases from 100 mg l\(^{-1}\) to 200 mg l\(^{-1}\). Further increase in the MB concentration results in the decrease of the MB removal efficiency; however, even at 400 mg l\(^{-1}\), the removal efficiency is as high as 83.5%. Overall, the Co\(_{0.5}\)Ni\(_{0.5}\)Fe\(_2\)O\(_4\) nanoparticles showed excellent prospects for the removal of MB.

Figure 4 shows the simulation curves for the adsorption kinetics of MB onto the Co\(_{0.5}\)Ni\(_{0.5}\)Fe\(_2\)O\(_4\) nanomaterials at room temperature. Table 1 lists the applicable parameters of the adsorption kinetics. The goodness-of-fit \((R^2)\) values of the pseudo-second-order kinetics are the highest, suggesting that the adsorption process can be well-described by this model. The \(\delta q_t\) versus \(t\) graphs for the adsorption process at different initial MB concentrations are shown in figure 5, all of which demonstrate the goodness-of-fit of almost 1. Therefore, the pseudo-second-order kinetics can be best matched with the MB adsorption performance of the Co\(_{0.5}\)Ni\(_{0.5}\)Fe\(_2\)O\(_4\) nanoparticles.

**3.3. Adsorption isotherm**

Adsorption isotherms are required to evaluate the interaction between the adsorbate and the adsorbent surface. Therefore, the equilibrium quantities of MB adsorbed and the equilibrium MB concentration in the treated
solution were evaluated using the Langmuir, Freundlich, and Temkin models. Formulas for the above models are given in equations (6)–(8) [22, 25, 26].

\[
q_e = \frac{q_{\text{max}} K_L C_e}{1 + K_L C_e} \quad (6)
\]

\[
q_e = K_F C_e^{1/n} \quad (7)
\]

\[
q_e = B \cdot \ln (A_T \cdot C_e) \quad (8)
\]

where \(q_{\text{max}} \) (mg g\(^{-1}\)) and \(q_e \) (mg g\(^{-1}\)) are the amounts of MB adsorbed by the Co\(_{0.5}\)Ni\(_{0.5}\)Fe\(_2\)O\(_4\) nanoparticles at adsorption saturation and adsorption equilibrium, respectively; \(K_L \) (l mg\(^{-1}\)) and \(K_F \) (mg\(^{1/n}\) g\(^{-1/n}\)) are the adsorption rate constants for Langmuir and Freundlich models, respectively; \(C_e \) (mg l\(^{-1}\)) represents the equilibrium concentration of MB; the value of \(1/n\) changes from 0 to 1 and is a non-dimensional value showing the surface heterogeneity or evaluating the adsorption intensity. \(A_T \) (l g\(^{-1}\)) is the equilibrium binding constant, \(T \) (K) is the temperature of the adsorption solution, \(R \) (8.314 J/(mol K)) is the universal gas constant, and \(B \) (=RT/\(\beta T\)) is the heat constant of adsorption.

The Langmuir model assumes that the adsorbate homogeneously covers the surface of the sorbent with a monolayer of molecules and that the adsorption only occurs at the special sites of the adsorbent. Simultaneously, the adsorption energy is assumed to be invariant, and the adsorbent surface is uniform. The Freundlich model assumes that the adsorbent has a heterogeneous surface; the adsorbed molecules first occupy the binding sites, following which, the binding strength for each molecule decreases with proportionally with the number of binding sites. Therefore, the adsorption of the molecules on the adsorbent is multilayer. The Temkin model assumes that the interactions between the adsorbent and the adsorbate occur and that the adsorption heat

Table 1. Matched kinetics arguments for the adsorption of MB on the Co\(_{0.5}\)Ni\(_{0.5}\)Fe\(_2\)O\(_4\) nanoparticles at room temperature.

| Kinetic model                        | Parameter | MB concentration (mg l\(^{-1}\)) |
|--------------------------------------|-----------|----------------------------------|
|                                      |           | 100 | 200 | 300 | 400 |
| Pseudo-first-order model             | \(q_e\) (mg g\(^{-1}\)) | 33.3261 | 73.1292 | 108.1840 | 130.0785 |
|                                      | \(k_1\)   | 0.1693 | 0.2038 | 0.2029 | 0.1322 |
|                                      | Adj. R\(^2\) | 0.7869 | 0.7690 | 0.7312 | 0.8340 |
| Pseudo-second-order model            | \(q_e\) (mg g\(^{-1}\)) | 34.6323 | 75.2509 | 111.3834 | 137.1349 |
|                                      | \(k_2\)   | 0.0128 | 0.0086 | 0.0057 | 0.0021 |
|                                      | Adj. R\(^2\) | 0.9982 | 0.9970 | 0.9976 | 0.9973 |
| Intraparticle diffusion model         | \(x_i\)   | 28.9497 | 66.4132 | 98.0824 | 104.6616 |
|                                      | \(k_i\)   | 0.4699 | 0.7352 | 1.1070 | 2.6493 |
|                                      | Adj. R\(^2\) | 0.7316 | 0.7140 | 0.7207 | 0.7411 |

Figure 5. Plots of \(t/q_e\) versus \(t\) for the adsorption with diverse initial concentrations of MB.
linearly decreases with the surface coverage; a uniform distribution of binding energy is assumed, and the binding energy is maximal.

Figure 6 shows the adsorption data matched with the Langmuir, Freundlich, and Temkin isotherms, and the related simulated arguments are listed in Table 2. The Temkin has the highest isotherm $R^2$ value (0.9968) among the three considered models. According to the Temkin model, the adsorption performance of MB on Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ could be explained by the monolayer-multilayer hybrid mechanism [27].

### 3.4. Effect of pH on adsorption performance and recycle capacity of the Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles

Figure 7(A) shows the MB adsorption performance (in a 200 mg l$^{-1}$ solution) at a pH of 3–9. As the pH increases from 3 to 5, the MB adsorption gradually increases. At pH $\geq$ 5, the Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles demonstrate the highest MB adsorption, attaining 75 mg g$^{-1}$. Figure 7(B) shows the effect of recycling on the relative removal rate of MB by the magnetic Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles. With an increase in the cycle number, the relative removal rate of MB by the Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles decreases. During calcination, the pores collapse and become small [28]. After eight cycles, the relative removal rate is still as high as 75%. Table 3 displays a comparison of the MB adsorption capacities of the magnetic Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles with those of other adsorbents, and the magnetic Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles have a larger adsorption than the other adsorbents for MB adsorption. These results revealed that the magnetic Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles have good regeneration ability and good application prospects for dye adsorption.

### 4. Conclusions

The combustion–calcination of a nitrate–ethanol solution was proposed for the fabrication of magnetic Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles for the efficient adsorption of MB. The calcination temperature and ethanol volume used in the material preparation were two pivotal elements affecting the performance of the

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**Table 2.** Assessed arguments of the three isotherms for the adsorption of MB on the Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles at room temperature.

| Model     | Equation                                                                 | Adj. $R^2$ | Parameters | Parameter values |
|-----------|---------------------------------------------------------------------------|------------|------------|------------------|
| Langmuir  | $q_e = \frac{q_{\text{max}} K_L C_e}{1 + K_L C_e}$                       | 0.9639     | $q_{\text{max}}$ | 162.3493         |
|           |                                                                           |            | $K_L$      | 0.4882           |
| Freundlich| $q_e = K_F C_e^\frac{1}{n}$                                              | 0.9799     | $K_F$      | 61.7135          |
|           |                                                                           |            | $\frac{1}{n}$ | 0.3089           |
| Temkin    | $q_e = B \ln(A_T C_e)$                                                   | 0.9968     | $B$        | 30.2698          |
|           |                                                                           |            | $A_T$      | 7.3254           |

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*Figure 6.* Curve fitting of different isotherm models for the adsorption of MB on the Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles at room temperature.
Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles. The mean grain diameter of the Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles fabricated using 20 ml ethanol as the solvent was approximately 25 nm at 400 °C, and the saturation magnetization was approximately 12.8 emu g$^{-1}$. The adsorption of MB on the Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles matched splendidly with the pseudo-second-order kinetics and Temkin isotherm. The pH value affected adsorption performance significantly; at pH $\geq$ 5, the amount of MB adsorbed on the Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles was the highest. In addition, the Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles retained a relative initial removal rate of 75% after eight cycles.

**Data availability statement**

All data that support the findings of this study are included within the article (and any supplementary files).

**ORCID iDs**

Hezhong Ouyang @ https://orcid.org/0000-0002-4173-6909
Shengying Pan @ https://orcid.org/0000-0002-2015-8345

**References**

[1] Hanafy H 2021 Adsorption of methylene blue and bright blue dyes on bayleaf capertree pods powder: understanding the adsorption mechanism by a theoretical study J. Mol. Liq. 332 115680
[2] Yu L, Li Y, Huang W, Pan S and Liu R J 2020 Adsorption mechanisms and the electrochemical properties of methyl blue onto magnetic Ni$_x$Mg$_{1-x}$Fe$_2$O$_4$ nanoparticles fabricated via the ethanol-assisted combustion process Water Air Soil Poll. 231 316
[3] Liu M and Wang Z 2021 Adsorption performance of reactive red 2BF onto magnetic Zn$_{0.3}$Cu$_{0.7}$Fe$_2$O$_4$ nanoparticles Mater. Res. Express 8 025014
[4] Chen J H, Wang Z and Lv Z X 2021 Adsorption mechanism of reactive red 2BF onto Ni$_{0.5}$Co$_{0.5}$Zn$_{0.5}$Fe$_2$O$_4$ nanoparticles prepared via the ethanol solution of nitrate combustion process Mater. Res. Express 8 025014
[5] Li Y, Pan S, Yu Q M, Ding X and Liu R J 2020 Adsorption mechanism and electrochemical performance of methyl blue onto magnetic NixCo$_{1-x}$Zn$_{1-x}$Fe$_2$O$_4$ nanoparticles prepared via the rapid-combustion process Ceram. Int. 46 3614–22
[6] Xu Y Y and Zhong Z P 2020 Adsorption mechanism of reactive red 2BF onto magnetic Co$_{0.5}$Zn$_{0.5}$Fe$_2$O$_4$ nanoparticles Water Air Soil Poll. 231 392
[7] Pan S, Liu Y H, Wang Z, Huang W, Song L and Liu R J 2020 Optimization on adsorption process of congo red onto magnetic Ni$_{0.5}$Cu$_{0.5}$Fe$_2$O$_4$/SiO$_2$ nanocomposites and their adsorption mechanism J. Nanosci. Nanotechno. 20 789–801

**Table 3.** Comparison of MB adsorption capacity of the magnetic Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles with other reported adsorbents.

| Adsorbents                     | Adsorbate | $q_{\text{max}}$ (mg g$^{-1}$) | References |
|-------------------------------|-----------|-------------------------------|------------|
| Mesoporous zeolite-like       | MB        | 132                           | [29]       |
| Palygorskite                  |           | 135                           | [30]       |
| Zeolite/chitosan composite    |           | 152                           | [31]       |
| Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles | | 162                           | Present study |

CO$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles. The mean grain diameter of the Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles fabricated using 20 ml ethanol as the solvent was approximately 25 nm at 400 °C, and the saturation magnetization was approximately 12.8 emu g$^{-1}$. The adsorption of MB on the Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles matched splendidly with the pseudo-second-order kinetics and Temkin isotherm. The pH value affected adsorption performance significantly; at pH $\geq$ 5, the amount of MB adsorbed on the Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles was the highest. In addition, the Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles retained a relative initial removal rate of 75% after eight cycles.

**Figure 7.** (A) Effect of pH on the adsorption performance and (B) the recycle capacity of the Co$_{0.5}$Ni$_{0.5}$Fe$_2$O$_4$ nanoparticles.
[8] Wang H F, Li Z C, Yahyaoui S, Hanafy H, Selim M K, Bonilla-Petriciolet A, Dotto G L, Sellahoui L and Li Q 2020 Effective adsorption of dyes on an activated carbon prepared from carboxymethyl cellulose: experiments, characterization and advanced modelling Chem. Eng. J. 417 128116

[9] Liu Y H, Yu Q M, Liu X and Liu R J 2019 Adsorption characteristics of methyl blue onto magnetic Mn0.63Co0.37Fe2O4 nanoparticles prepared via a rapid combustion process Environ. Prog. Sustain. 38 5277–87

[10] Andhare D D, Patade S R, Kounsalye J S and Jadhav K M 2020 Effect of Zn doping on structural, magnetic and optical properties of cobalt ferrite nanoparticles synthesized via Co-precipitation method Physica B 583 41051

[11] Gurgel A L, Martinelli A E, Conceição O L D A, Xavier M M, Torres M A M and Melo D M D A 2019 Microwave-assisted hydrothermal synthesis and magnetic properties of nanostructured cobalt ferrite J. Alloy. Compd. 799 36–42

[12] Silva E B S D, Ferreira S R D S, Silva A O D, Matias J A L, Albuquerque A R, Oliveira J B J L D and Morales M A 2020 Cashew gum as a sol-gel precursor for green synthesis of nanostructured Ni and Co ferrites Int. J. Biol. Macromol. 164 4245–51

[13] Ansari F, Sobhani A and Salavati-Niasari M 2018 Simple sol-gel synthesis and characterization of new CoTiO3/CoFe2O4 nanocomposite by using liquid glucose, maltose and starch as fuel, capping and reducing agents J. Colloid Interf. 314 723–32

[14] Liu R J, Rong G X, Liu Y H, Huang W, He D W and Liu R Z 2021 Delivery of apigenin-loaded magnetic Fe2O3/Fe3O4@mSiO2 nanocomposites to A549 cells and their antitumor mechanism Mat. Sci. Eng. C-Mater. 120 117179

[15] Liu R J, Pan S, Liu M, Huang W, Lv Z X and He A L 2021 A label-free electrochemical biosensor with magnetically induced self-assembly for the detection of CYP2C9 3 gene Appl. Surf. Sci. 537 147868

[16] Liu R J, Huang W, Pan S, Li Y, Yu L L and He D W 2020 Covalent immobilization and characterization of penicillin G acylase on amino functionalized magnetic Fe3O4/Fe2O3 heterogeneous nanoparticles prepared via a facile solution-combustion process International J. Biol. Macromol. 162 1587–96

[17] Huang W, Pan S, Li Y, Yu L L and Liu R J 2020 Immobilization and characterization of cellulase on hydroxy and aldehyde functionalized magnetic Fe2O3/Fe3O4 nanocomposites prepared via a novel rapid combustion process Int. J. Biol. Macromol. 162 845–52

[18] Pan S, Huang W, Li Y, Yu L L and Liu R J 2020 A facile diethyl-carbonate-assisted combustion process for the preparation of the novel magnetic α-Fe2O3/Fe3O4 heterostructure nanoparticles Mater. Lett. 262 127071

[19] Huang W, Pan S, Yu Q M, Liu X, Yu Y H and Liu R J 2019 Adsorption performance of methyl blue onto magnetic Ni1-x-yCo2xZnxFe2O4 nanoparticles prepared by a novel alcohol-assisted combustion method J. Inorg. Organomet. P. 29 1755–66

[20] Mota T L R, Gomes A L M, Palhares H G, Nunes E H M and Houmard M 2019 Influence of the synthesis parameters on the mesoporous structure and adsorption behavior of silica xerogels fabricated by sol-gel technique J. Sol-Gel Sci. Techn. 92 681–94

[21] Li Y, Wang J C, Zhang S S, Zhang Y J, Yu L L and Liu R J 2021 Adsorption and electrochemical behavior investigation of methyl blue on magnetic nickel-magnesium ferrites prepared via the rapid combustion process J. Alloy. Compd. 885 160969

[22] Wang J L and Guo X 2020 Adsorption kinetic models: physical meanings, applications, and solving methods Chemosphere 258 127279

[23] Tran H V, Hoang L T and Huynh C D 2020 An investigation on kinetic and thermodynamic parameters of methylene blue adsorption onto graphene-based nanocomposite Chem. Phys. 535 110793

[24] Deng P, Liu M and Lv Z X 2020 Adsorption mechanism of methyl blue onto magnetic Co3O4-ZnO/Fe3O4 nanoparticles synthesized via the nitrate-alcohol solution combustion process AIP Adv. 10 095005

[25] Yuan J T, Wang Z, Wang L and Liu R J 2020 Preparation, surface modification, and characteristic of Fe3O4 nanoparticles J. Nanosci. Nanotechnol. 20 50351–7

[26] Yang Y H, Zhang Y F and Wang Z 2020 Adsorption characteristics and electrochemical performance of reactive red onto magnetic MgFe2O4 nanoparticles prepared via the solution combustion process J. Sol-Gel Sci. Techn. 93 535–45

[27] Xu Q, Xu Y Y, Xue J M, Zhu F H, Zheng Z P and Liu B J 2021 An innovative alcohol-solution combustion-calciation process for the fabrication of NiFe2O4 nanorods and their adsorption characteristics of methyl blue in aqueous solution Mater. Res. Express 8 095003

[28] Yin R T, Zhang S S, Xu Y Y, Xue J M, Bi J Q and Liu R J 2021 Adsorption mechanism and electrochemical properties of methyl blue onto magnetic Co3O4-Cu1-x-yFe2O4 nanoparticles prepared via an alcohol solution of nitrate combustion and calcination process J. Inorg. Organomet. Polym. Mater. 31 3584–94

[29] Tsai C K and Horng J J 2021 Transformation of glass fiber waste into mesoporous zeolite-like nanomaterials with efficient adsorption of methylene blue Sustainability 13 6207

[30] YouCEF I D, Belarouj L S and Lopez-Galincho A 2019 Adsorption of a cationic methylene blue dye on an Algerian palygorskite Appl. Clay Sci. 179 105145

[31] Khanday W A, Asif M and Hameed B H 2017 Cross-linked beads of activated oil palm ash zeolite/chitosan composite as a bio-adsorbent for the removal of methylene blue and acid blue 29 dyes Int. J. Biol. Macromol. 95 895–902