

Article

Levofloxacin Adsorption onto MWCNTs/CoFe₂O₄ Nanocomposites: Mechanism, and Modeling Using Non-Linear Kinetics and Isotherm Equations

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Abstract: In the present work, the adsorption mechanism and capacity of MWCNTs/CoFe₂O₄ nanocomposite as an adsorbent were investigated. Levofloxacin (LFX), a widely used antibiotic, was selected as a hazardous model contaminant in aqueous solutions. The surface and inner characterization of MWCNTs/CoFe₂O₄ was obtained via SEM/TEM, XRD, BET/BJH, and pH_{PZC}. These analyses indicated that MWCNTs/CoFe₂O₄ possess excellent surface and pore characteristics, e.g., specific surface area, pore volume, and mean pore diameter, which were 72 m²/g, 0.51 cm³/g, and 65 nm, respectively. The results demonstrate that by supplementing 1 g/L of MWCNTs/CoFe₂O₄ at experimental conditions of pH value of 5, temperature of 30 °C, initial LFX concentration of 50 mg/L and mixing time of 90 min, a significant outcome of 99.3% removal was achieved. To identify the phenomenon of adsorption, the thermodynamic parameters of ΔH° and ΔS° were calculated, which indicated that the nature of LFX adsorption onto MWCNTs/CoFe₂O₄ nanocomposite was endothermic and spontaneous. Nine isotherm models, including four two-parameter and five three-parameter models, were investigated. In addition, the regression coefficient as well as five error coefficient models were calculated for nonlinear isotherm models. According to the goodness of fit tests, the equilibrium data were well coordinated with the Freundlich and Sips isotherms. The kinetics study showed that the LFX adsorption data well fitted with pseudo-second-order model, and the adsorption of LFX molecules occurred through several stages from surface to intraparticle diffusion. In conclusion, the present work evinces that LFX wastewater can be efficiently treated via an adsorption process using a MWCNTs/CoFe₂O₄ nanocomposite.

Keywords: MWCNTs/CoFe₂O₄; levofloxacin; nanocomposite; adsorption; regeneration

1. Introduction

Currently, one of the most serious challenges drawing universal attention is water pollution due to its direct impact on human health and aquatic life [1–3]. One of the major water pollutants is pharmaceutical products (i.e., antibiotics), as they are common pollutants detected extensively in wastewater, surface and ground water resources, and even in samples taken from potable water [4–6]. The existence of such compounds in essential water resources is related mainly to the released effluents from hospitals, pharmaceutical

industries, and human excretion [7]. Variable antibiotics have been manufactured to face different diseases that attack both animals and humans [8]. Levofloxacin (LFX) is a type of quinolone antibiotic widely used as an antibacterial agent, anti-dysentery, and in the treating of some diseases [9].

Over time, the continuous production of antibiotics and their release with the liquid wastes of pharmaceutical industries can lead to high and dangerous levels of accumulation in living organisms [10,11]. This problem not only affects the quality of water but also the medical effect of these drugs and generates microbial resistance [12]. The existence of Levofloxacin as toxic and non-degradable antibiotic residuals causes several hazardous environmental and health impacts [13,14].

For this purpose, various methods such as adsorption [13], oxidation process [15,16], coagulation [3], membrane methods [4], and nanofiltration technology [14] have been investigated for antibiotic removal. The adsorption method is widely used for pollutant removal due to its simple operation, low requirements for equipment, low energy consumption, and high efficiency [17–19]. In recent decades, the application of nanotechnology in the removal of various contaminants due to their ease and economic use compared to physicochemical and biological methods has been further developed [20]. Toxicity, stability, and concentration of pollutants have significant environmental, economic, and health effects. One of the basic solutions to solving these challenges is the use of technologies that have low investment and operation costs and are very small in size but have a high capacity [21,22].

According to the literature, a great ability for the elimination of a wide range of pollutants has been identified for multi-walled carbon nanotubes (MWCNTs) [23,24], hence their exceptional properties, e.g., large surface area, mesoporosity nature, illustrious stability, and abundant surface groups [25,26]. Nonetheless, the issue associated with the separation of spent MWCNTs from the solutions has been reported to be a crucial weakness and limiting factor in its use [27]. From another side, the unsuitable control of spent MWCNTs in the adsorption treatment system may lead to the entry of quantities of this substance into the ecosystems and food, and this in turn may lead to serious consequences for human health. Therefore, finding a technology that ensures the separation of MWCNTs after its use in processing is an important problem to be solved at the treatment plant. One of the most suggested techniques is the combination of used adsorbent with other materials such as magnetic nanoparticles. In fact, this method has proven to have promising results for increasing the separability of many adsorbents, such as MWCNTs from aqueous solutions [28].

Over the years, the development of high-performance, highly recyclable magnetic absorbers, especially intermediate metal oxides, has been a fascinating topic for researchers [29,30]. One of the metal oxides that has been widely studied is CoFe_2O_4 ferrites; the properties such as high chemical stability, average magnetic saturation, and strong anisotropy have led to its extensive use as a microwave adsorbent [31]. Nonetheless, high specific gravity and restricted frequency bandwidth of CoFe_2O_4 ferrites are the problems associated with its use [32] and should be eliminated; for this purpose, the synthesis of composite structures through employing magnetic and dielectric materials has been considered [33]. Carbon nanotubes are among the dielectric materials with a high dielectric loss.

Therefore, this study was performed with the aim of adsorption of LFX from aqueous media using the method of stabilization of CoFe_2O_4 on MWCNTs. In addition, the effects of operational factors, such as the adsorption time, pH, amount of the MWCNTs/ CoFe_2O_4 , initial concentration of the LFX, and the temperature, on the adsorption of LFX were evaluated.

2. Materials and Methods

2.1. Chemicals and Reagents

MWCNTs (average diameter = 37 nm, length range = 1–25 μm) were purchased from the Research Institute of the Petroleum Industry (Tehran, Iran). LFX stock solution (chemical formula: $\text{C}_{18}\text{H}_{20}\text{FN}_3\text{O}_4$, molecular weight = 361.4 g/mol, purity $\geq 99\%$), acetonitrile, and distilled water were provided by Sigma–Aldrich. In addition, methanol, iodine,

$\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$, $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$, HCl , and NaOH were purchased from Merck. The structure of the LFX is shown in Figure 1.

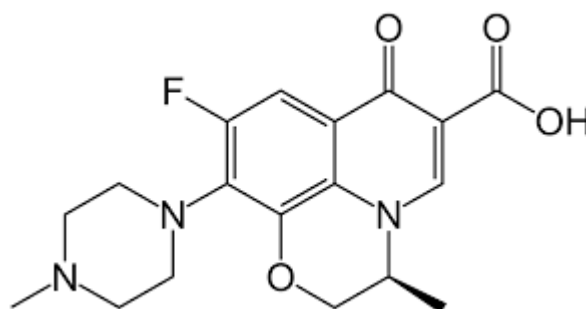


Figure 1. Chemical structure of the LFX.

2.2. Synthesis of MWCNTs/ CoFe_2O_4

The synthesis of CoFe_2O_4 nanoparticles was achieved by the employment of the solvothermal co-precipitation process, followed by the electrophoretic precipitation process to coat MWCNTs with CoFe_2O_4 nanoparticles [30]. For the first purpose, 0.25 g of $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ and 0.45 g of $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ were mixed with 100 mL of distilled water, and the resulted brown solution was further mixed using mechanical stirring for 15 min at 60 °C. During this mixing, the temperature was gradually raised to 80 °C. After that, the NH_4OH solution (30 mL, 10%) was slowly added into the brown solution until reaching a pH value of 12 where an instant black of CoFe_2O_4 nanoparticles was formed. Then, the nanoparticles were separated from the solution and were washed with both distilled water and acetone. After the CoFe_2O_4 nanoparticles were synthesized, and through the employment of a thin stainless steel sheet as an anode, coating CoFe_2O_4 nanoparticles onto MWCNTs was carried out by the electrophoretic precipitation method. First, 50 mg of synthesized CoFe_2O_4 nanoparticles was mixed with 80 mL of acetone solution to prepare a semi-stable suspension. Then, 5 mg of iodine was added under mixing into the prepared semi-stable suspension. To enhance the stability of this suspension, it was exposed to ultrasonic agitation conditions for 10 min. Next, 0.3 g of MWCNTs was poured into the prepared CoFe_2O_4 suspension using a glass holder. After setting a distance of 1 cm between the carbon fiber (cathode) and the anode surface, a direct current was applied for 10 min. The resulting solid particles, which represent the MWCNTs/ CoFe_2O_4 nanocomposites, were collected and then dried at 80 °C for 1 hr.

2.3. Adsorption Experiments

The effect of MWCNTs/ CoFe_2O_4 adsorbent on LFX removal from aqueous solution was investigated in the batch experiment. Firstly, synthetic wastewater was prepared by dissolving LFX powder in distilled water; the obtained solution contained LFX concentrations from 10 to 100 mg/L. After that, an adsorption experiment was conducted using an open glass flask at different temperatures (293 to 323 K). The conditions of pH (3 to 11), MWCNTs/ CoFe_2O_4 dosage, and mixing time (10 to 120 min) were investigated during the experiment. Mainly, the LFX solution (100 mL) and specific amount of MWCNTs/ CoFe_2O_4 were mixed in 100 mL flasks using a magnetic stirrer bar at 120 rpm for the given contact time. Finally, after completing the adsorption treatment, the absorbance of the LFX was measured by injecting samples into the HPLC injector. A mixture containing acetonitrile and 1% formic acid with a ratio of 16:84 was used as the mobile phase [8].

Both adsorption capacity (q_t , mg/g) and LFX removal efficiency (%Removal) were calculated using Equations (1) and (2), respectively [9]. It is noteworthy that the C_t and q_t in Equation (1) at equilibrium status are denoted by C_e and q_e , respectively.

$$q_t = \frac{(C_0 - C_t)}{M} \times V \quad (1)$$

$$\% \text{Removal} = \frac{(C_0 - C_t)}{C_0} \times V \quad (2)$$

where C_0 is the initial concentration of LFX (mg/L), C_t is the concentration of LFX (mg/L) after the specified time (t , minutes), of adsorption process, V is the volume of LFX solution (L), and M is the mass of MWCNTs/CoFe₂O₄ (g).

2.4. Characterization Analysis

X-ray diffraction (XRD) analysis (Lab XRD-6000 SHIMADZU Japan) was carried out to study the particle size of used materials with a 2θ range and scan rate of 8° min^{-1} . Scanning electron microscopy (SEM) was applied to provide the details of surface textures (JSM IT 100 JEOL Japan). Transmission electron microscopy (TEM) was performed to evaluate the morphology (i.e., the shape and size) of nanoparticles (JOEL 2000). The average pore diameter and specific surface area of the MWCNTs/CoFe₂O₄ were determined according to the Brunauer–Emmett–Teller (BET) and Barrett–Joyner–Halenda (BJH) methods. In addition, a high-performance liquid chromatography (HPLC Agilent 1200 series) with a C18 column (150 mm $\times$ 4.6 mm, 5 μm particle size) was used to measure the absorbance of the LFX after being subjected to treatment.

3. Results and Discussion

3.1. Characteristics Analyses

The characterization analyses of the MWCNTs/CoFe₂O₄ and their results are shown in Figures 2 and 3. The SEM images of CoFe₂O₄ nanoparticles, which are shown in Figure 2a, indicate that the CoFe₂O₄ nanoparticles are composed of spherical nanoparticles that have an average diameter of approximately 40 nm.

The FESEM image of MWCNTs/CoFe₂O₄ nanocomposite (Figure 2b,c) depicts that the CoFe₂O₄ nanoparticles were quasi spherically dispersed on the MWCNTs. Given TEM images of MWCNTs and MWCNTs/CoFe₂O₄ (Figure 2d,e), the grafting of CoFe₂O₄ nanoparticles onto the MWCNTs could be evidently perceived. In addition, according to these images, rolling CoFe₂O₄ nanoparticles by MWCNTs bundles was identified (CoFe₂O₄ nanoparticles appeared as nodes coating MWCNTs).

The XRD patterns of CoFe₂O₄ and MWCNTs/CoFe₂O₄ samples (Figure 3a) show that the diffraction peaks for the MWCNTs/CoFe₂O₄ XRD patterns were 26.8° and 47.4° . Similar diffraction peaks were obtained from the XRD patterns of pure MWCNTs [34]. The diffraction peak (002) was noted at 26.8° , and this is an indication of the hexagonal graphitic structure of MWCNTs [35,36]. Additionally, the XRD pattern of MWCNTs/CoFe₂O₄ shows diffraction angles of 30.1° , 35.4° , 43.1° , 53.4° , 56.9° and 62.4° at diffraction peaks of 220, 311, 400, 422, 511 and 440, respectively. These peaks are matched with previous results obtained by Rahimi et al. [36] and with standard cards of CoFe₂O₄ (JCPDS card number: 22-1086). These results certify the spinel CoFe₂O₄ ferrite. They also confirm the high purity of CoFe₂O₄ in the evaluated sample of MWCNTs/CoFe₂O₄. In fact, the results of the XRD study confirm that MWCNTs/CoFe₂O₄ was successfully synthesized.

Considering the results observed for the N₂ adsorption–desorption and BJH analyses (Figure 3b), the specific surface area, pore volume, and mean pore diameter, obtained for MWCNTs/CoFe₂O₄ were 72 m²/g, 0.51 cm³/g, and 65 nm respectively. As depicted, an IV-type shape was detected for the isotherms curve. This supports a mesoporous structure of MWCNTs/CoFe₂O₄ according to the classifications of IUPAC [31]. Moreover, a severe increase was detected for these isotherms at the high P/P_0 value (0.8–1.0), and that confirms the formation of extensive mesopores because of their hollow interior [32]. Furthermore, according to results related to the pore size distribution of MWCNTs/CoFe₂O₄ (inner figure in Figure 3b), the average size of our prepared adsorbent was about 32 nm. This result is consistent with Rahimi et al. [36], who revealed that a 34 nm average size of MWCNTs/CoFe₂O₄ nanocomposite was suitable for the adsorption process.

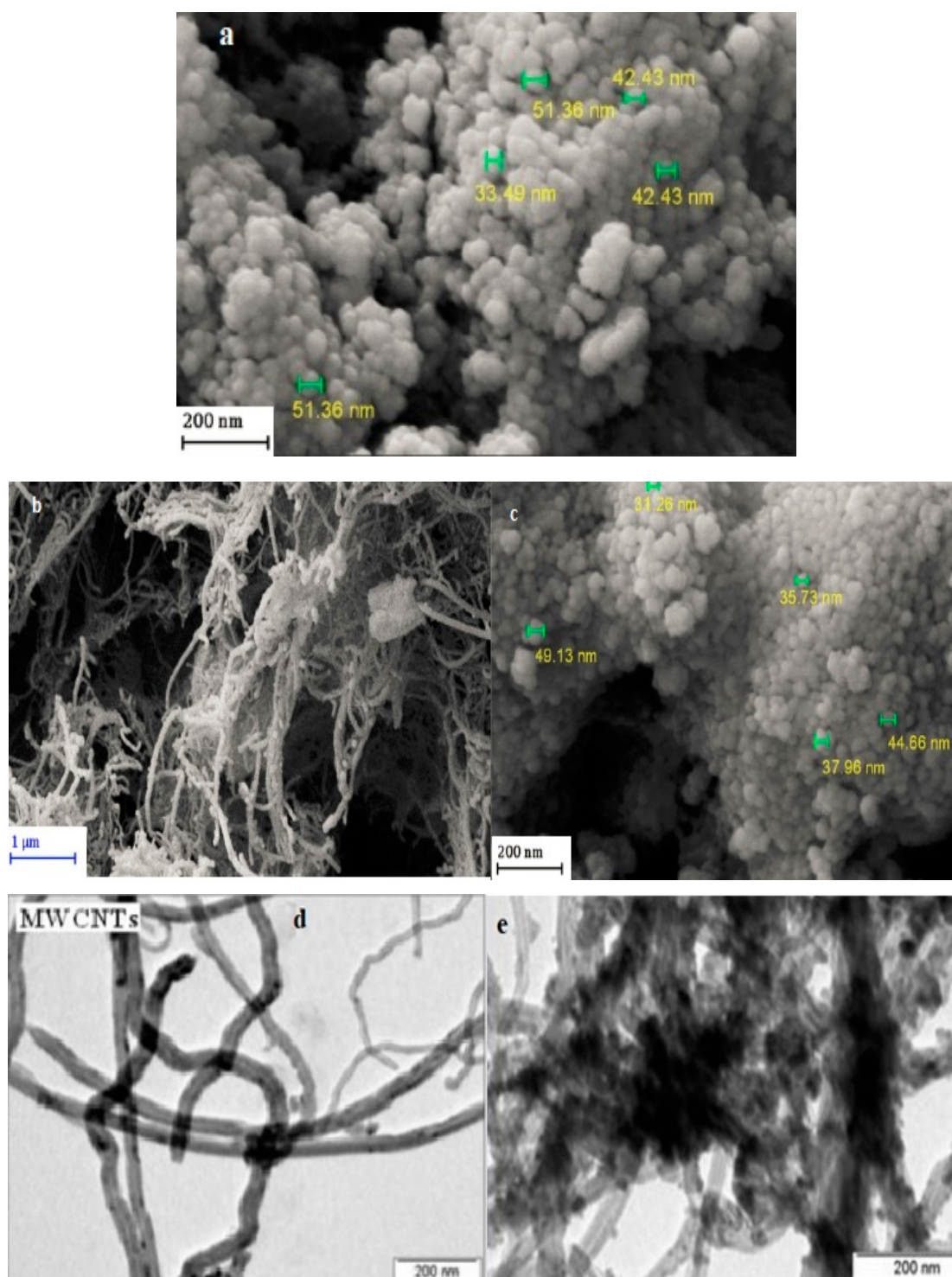


Figure 2. SEM image of CoFe₂O₄ (a), MWCNTs/CoFe₂O₄ (b,c), TEM image of MWCNTs (d), and TEM image of MWCNTs/CoFe₂O₄ (e).

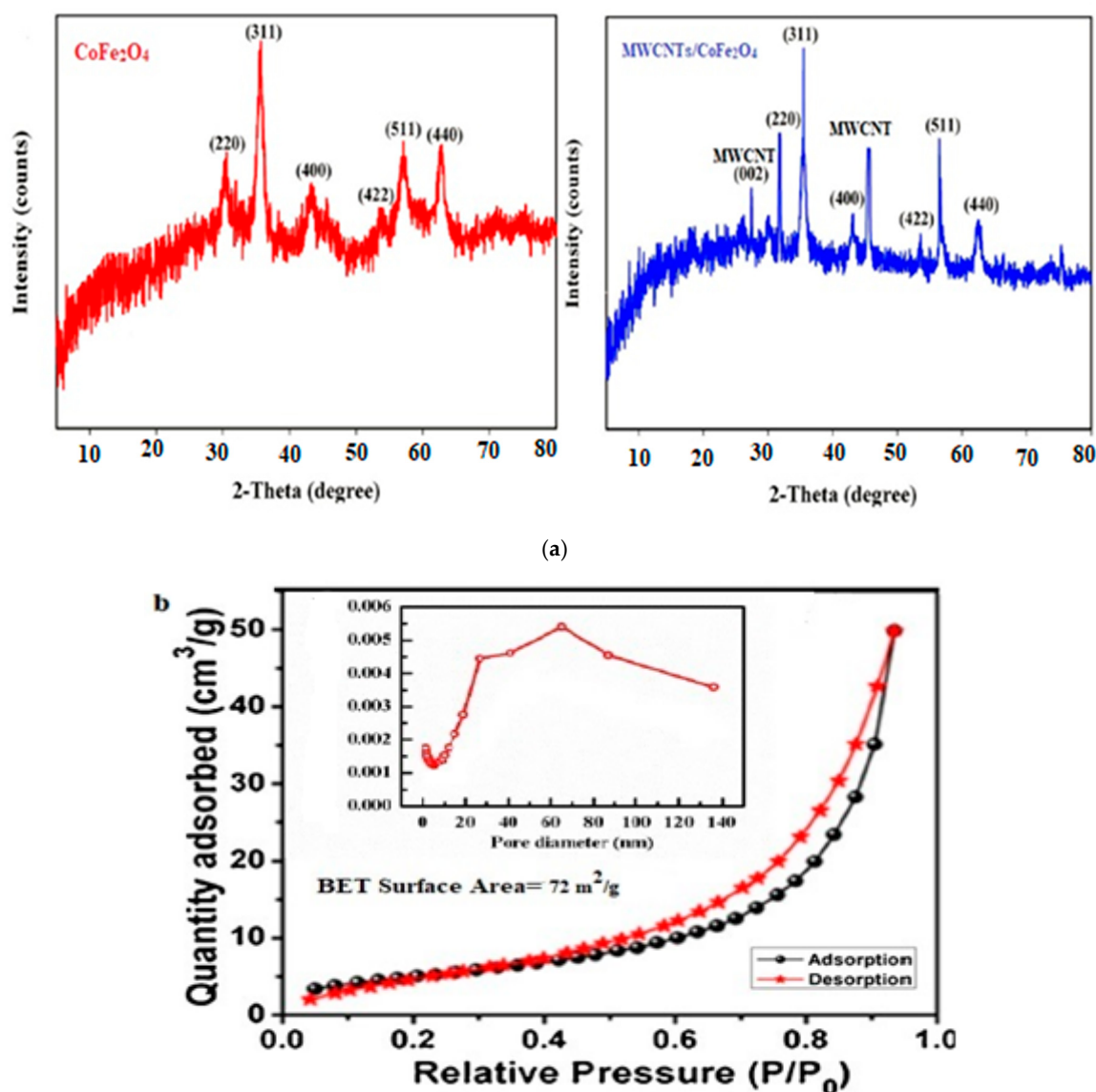


Figure 3. XRD patterns of the CoFe_2O_4 and $\text{MWCNTs/CoFe}_2\text{O}_4$ (a), and N_2 adsorption–desorption isotherms and pore size distribution (b).

3.2. Adsorption Process at Different Conditions

Experiments related to optimizing the pH value during the adsorption process was performed at a mixing time of 90 min, temperature of 30°C , $\text{MWCNTs/CoFe}_2\text{O}_4$ and LFX amount of 1g/L and 50 mg/L, respectively. The results are shown in Figure 4a. The removal efficiency decreased with increasing pH. It can be seen that the optimum pH value for LFX removal was 3–5. This can be explained by identifying the value of the point of zero charge (pH_{PZC}), which was determined to be 6.4 s, as shown in Figure 4b (The black line is the natural pH of the aqueous solution and the blue line is the measured pH after contact with the adsorbent after 24 hours of mixing, wherever the two lines intersect is known as the pH_{HPZC}). Thus, when the pH value is below the pH_{PZC} , the $\text{MWCNTs/CoFe}_2\text{O}_4$ charge is cationic, and the tendency to adsorb anions becomes higher. In addition, it is well-known that LFX antibiotic decomposition results in the release of fluoride ions and nitrogen compounds into the environment, which are anions that are absorbed at low pH [37]. When the pH value rises, the charge on the adsorbent surface and the antibiotic will be positive, and this will create a repulsive force between the $\text{MWCNTs/CoFe}_2\text{O}_4$ and the LFX molecules [38].

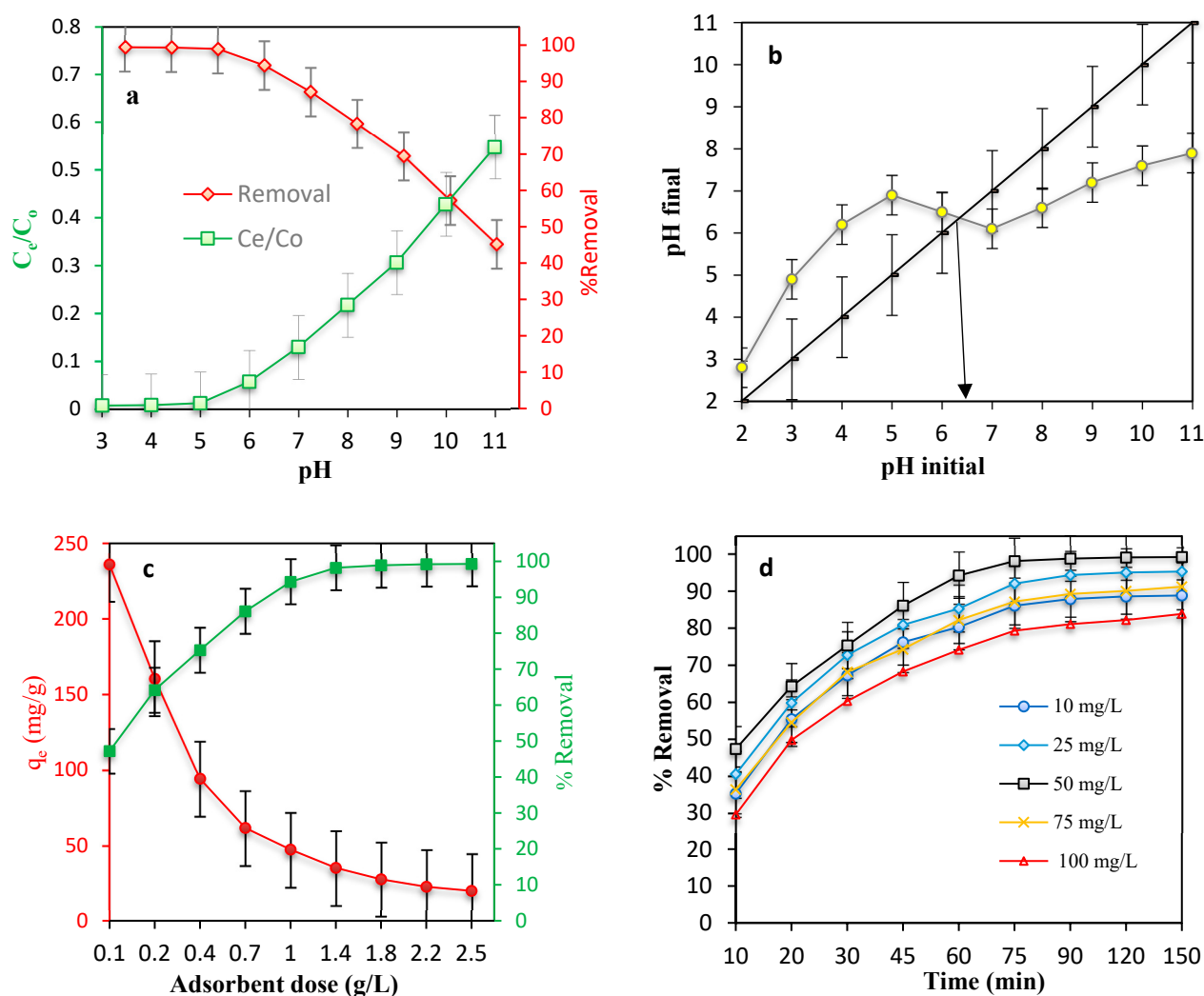


Figure 4. Results of the experiments of the effect study: solution pH (a), determination of the pH_{PZC} value (b), effect of MWCNTs/CoFe₂O₄ dose (c), initial LFX concentration and mixing time (d).

The sufficient adsorption sites that are available for a given contaminant are strongly dependent on the adsorbent dosage [39]. Thereby, in this study, the effect of the dosage of the MWCNTs/CoFe₂O₄ (i.e., 0.1, 0.2, 0.4, 0.7, 1, 1.4, 1.8, 2.2 and 2.5 g/L) on the LFX removal efficiency was investigated. Experiments were performed at a mixing time of 90 min, temperature of 30 °C, pH value of 5 and LFX concentration of 50 mg/L. As shown in Figure 4c, an increase in the percentage adsorbed was observed with an increase in the MWCNTs/CoFe₂O₄ dosage. This is attributed to increasing the available sites of adsorption on the surface of MWCNTs/CoFe₂O₄. Additionally, the early rising in the adsorbed percentage of the LFX might be because of increasing the number of receptors on the adsorbent surface [39]. However, increasing the dosage of MWCNTs/CoFe₂O₄ above 1.4 g/L resulted in reducing the adsorbed amount of the LFX [40–42]. These results indicate that the dosage of 2.5 g/L of MWCNTs/CoFe₂O₄ is optimal for removing LFX from an aqueous solution.

The influence of initial LFX concentration (10–100 mg/L) on the removal efficiency was tested at a pH of 5 and MWCNTs/CoFe₂O₄ dosage of 1 g/L, as presented in Figure 4d. This figure showed that the LFX removal efficiency was attained a maximum value of 100% at the initial concentration of 50 mg/L. Subsequently, the removal efficiency decreased with the increase in LFX initial concentration until reaching 77% at the initial concentration of 100 mg/L, and this trend is related to the identified availability of active uptake sites for increased amounts of LFX particles [43]. On the other hand, the specific amount of

MWCNTs/CoFe₂O₄ could not be able to provide enough surface area required to remove all the amount of adsorbate.

3.3. Regeneration Experiment

The recyclability performance of an adsorbent is an important parameter to evaluate the potential application of the adsorbent. In this study, the recyclability performance of the MWCNTs/CoFe₂O₄ adsorbent was investigated. For this, MWCNTs/CoFe₂O₄ was recycled six times in the adsorption of LFX under the determined optimum operating parameters. Then, the removal efficiency of the LFX ion was consecutively calculated. As shown in Figure 5, after adsorbent recycling six times, the removal efficiency was reduced by only 6%. This reduction in the removal percentage can be ascribed to the contaminant penetration into the internal pores of the MWCNTs/CoFe₂O₄ and the saturation of the active internal sites, which are not removed by surface washing [44–46]. These results provide an indication of the good potential of MWCNTs/CoFe₂O₄ recycling in the adsorption process.

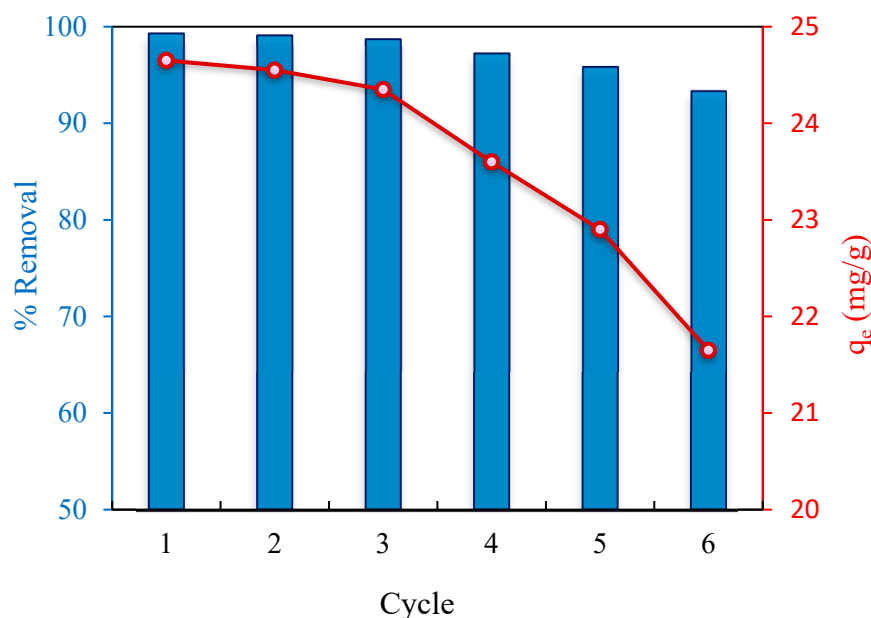


Figure 5. Reusability study of MWCNTs/CoFe₂O₄ on LFX uptake ($C_0 = 50$ mg/L, temperature 30 ± 2 °C, dose = 1 g/L, time 90 min and pH = 5).

3.4. Kinetic and Isotherm Studies

It is necessary to study the isotherms and the kinetics of the adsorption process to understand the adsorption mechanism. To achieve this, the experimental data should be carefully modeled with the relevant models. In this study, different goodness-of-fit equations were used to judge the fitting process and to determine the most suitable models (Table S1).

The kinetic data shown in Figure 4d were modeled using pseudo-first-order (PFO), pseudo-second-order (PSO), Elovich, and intraparticle diffusion model (IPD) models (Table S2) and the results are depicted in Figure 6a and Table 1. It seems that the PFO, PSO, and Elovich models are suitable for describing the LFX adsorption data (regression coefficients (R^2) > 0.97). However, the small values of χ^2 (0.112) and error functions obtained from the PSO model indicate that it was the best kinetic model to describe the LFX adsorption. For PSO, the q_e cal was close to the q_e exp, indicating the existence of a majority of π – π type interactions [47,48].

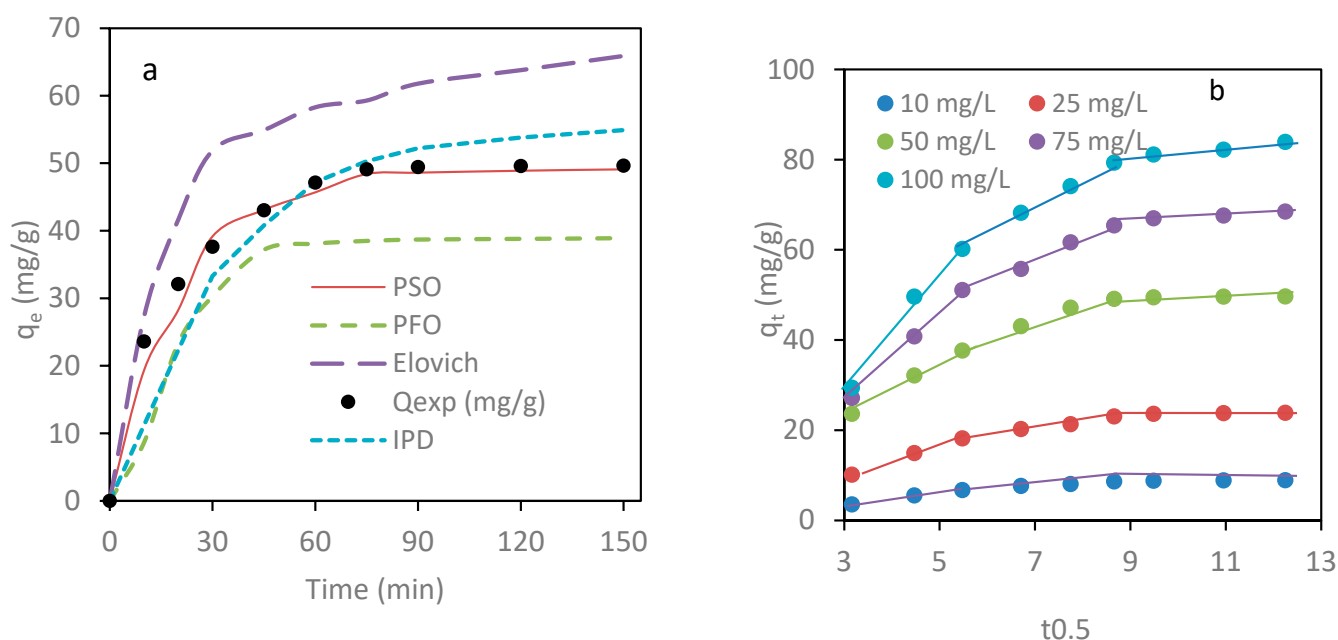


Figure 6. Nonlinear plot of kinetic models of FLX adsorption (a), IPD kinetics for FLX adsorption (b), by MWCNTs/CoFe₂O₄.

Table 1. Results of the kinetic parameters of Levofloxacin adsorption onto MWCNTs/CoFe₂O₄ nanocomposites.

Models	Parameters	R ²	χ ²	RMSE	HYBRID	ARE	EABS
PFO	$q_e = 64.2 \text{ mg/g}$, $K_1 = 0.175 \text{ 1/min}$	0.971	0.946	3.72	4.25	0.941	2.76
PSO	$q_e = 84.3 \text{ mg/g}$, $K_2 = 0.0008 \text{ g/min.mg}$	0.997	0.112	0.452	0.641	0.032	0.421
Elovich	$\alpha = 35.2 \text{ mg/g. min}$, $\beta = 0.24 \text{ mg/g.min}$	0.981	1.23	3.27	6.19	3.72	1.96
IPD	$K_{id} = 1.17 \text{ mg/g.min}^{0.5}$, $C = 18.1 \text{ mg/g}$	0.912	0.927	6.19	7.14	4.35	3.84

The results of IPD showed that the adsorption process consisted of several different stages (linear model Figure 6b). The adsorption mechanism phase began with the transfer of molecules of these LFX materials to the outer surface of MWCNTs/CoFe₂O₄. This step was very fast in the adsorption process. This may be because of the positively charged MWCNTs/CoFe₂O₄ surface in an acidic solution that can easily absorb electrons from these materials. The next step was the release of LFX molecules internally into MWCNTs/CoFe₂O₄, which was a limiting step of the process. Finally, the LFX adsorption in the MWCNTs/CoFe₂O₄ pores [49,50].

A proper understanding and interpretation of the experimental data of the adsorption process are crucial in the good design of the proposed treatment system. In fact, this is achieved by finding the best mathematical model to represent the adsorption data. In this study, the equilibrium data were modeled using nine isotherm models of two and three parameters (Table S3). All computed parameters are presented in Figure 7 and Table 2. Observing Table 2, it appears that the values of the coefficients of determination, R² were close to one for the models of Freundlich and D-R. Based on the high value of R², low value of χ², and the error functions, the Freundlich model was found to be better at describing the adsorption isotherm of LFX by the MWCNTs/CoFe₂O₄.

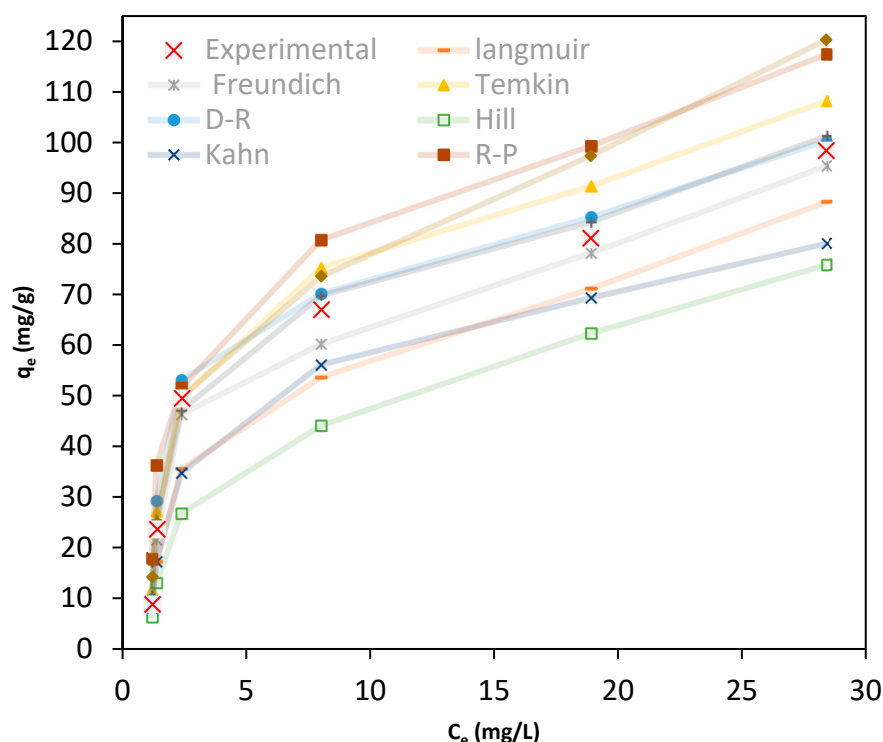


Figure 7. Nonlinear plot of isotherm models of FLX adsorption.

Table 2. Results of the isotherm parameters of Levofloxacin adsorption onto MWCNTs/CoFe₂O₄ nanocomposites.

Models	Parameters	R ²	χ ²	RMSE	HYBRID	ARE	EABS
Langmuir	Q _m = 122.2 mg/g, K _L = 0.725 L/min	0.912	0.924	1.63	2.32	2.17	2.19
Freundlich	K _F = 18.4 mg/L, 1/n = 0.271	0.995	0.254	0.21	0.74	0.27	0.29
Temkin	A _T = 556.2 L/mg, ΔQ = 117.2 J/mol	0.926	0.195	1.34	0.384	0.231	0.196
D-R	Q _m = 84.3 mg/g, K _{id} = 0.0004 L/mg, E = 39.6 kJ/mol	0.981	1.24	2.41	3.34	1.63	1.37
Hill	Q _m = 73.2 mg/g, K _H = 0.421 L/g, n _H = 1.02	0.932	0.321	2.48	3.29	0.651	0.421
Kahn	Q _{max} = 95.3 mg/g, b _K = 2.71 L/g, a _K = 0.324	0.921	0.542	3.34	2.84	0.731	0.987
Redlich-Peterson	A = 321.2 L/g, B = 18.5 L/mg, B2 = 0.812	0.971	0.473	4.17	1.65	1.72	0.761
Sips	K _S = 56.2 L/g, a _S = 0.612 L/g, β _S = 0.354	0.992	0.641	0.73	0.291	0.692	1.39
Toth	Q = 36.2 36.2, K _e = 0.352, N = 0.214	0.917	0.512	5.28	4.85	2.37	1.89

A positive adsorption energy ($\Delta Q = 117.2$ J/mol) from the Tekmin equation and a high adsorption energy of 8 kJ/mol in the D-R equation ($E = 39.6$ kJ/mol) indicate the chemical adsorption between the LFX and the MWCNTs/CoFe₂O₄ [51,52]. Furthermore, the most important adsorptive parameter, i.e., maximum adsorption capacity (Q_m) of MWCNTs/CoFe₂O₄ for LFX was high (>122 mg/g) based on the Langmuir model results. Freundlich isotherm refers to multilayer adsorption on the surface of the MWCNTs/CoFe₂O₄, with no interaction between the adsorbed molecules [53,54]. In addition, Table 2 shows that the Sips model described the equilibrium data better than the other three-parameter isotherm models based on the results of the regression coefficient and error functions. Additionally, the Sips isotherm model showed the heterogeneous surface of the MWCNTs/CoFe₂O₄, thus presenting energetically different adsorption sites [55]. In general, it can be said that two- and three-parameter isotherm models represent the heterogeneous surface of the MWCNTs/CoFe₂O₄ with the adsorption surface of the multilayer, where the energy distribution of the layers is non-uniform and non-regular.

3.5. Thermodynamic Study

The thermodynamic study of the LFX adsorption process was carried out at 293, 303, 313 and 323 K. All parameters (i.e., initial concentration of LFX, pH value, dosage of adsorbent, and contact time) were maintained according to optimal conditions. The effect of temperature on the adsorption of LFX is shown in Figure 8. These data were used in the thermodynamic study as the values of ΔG° , ΔH° and ΔS° , as shown in Equations (3)–(5) [56–59].

$$\Delta G^\circ = -RT \ln K_d \quad (3)$$

$$\ln K_d = \left(\frac{\Delta S^\circ}{R} \right) - \left(\frac{\Delta H^\circ}{R} \right) \frac{1}{T} \quad (4)$$

$$K_d = \frac{C_0 - C_e}{C_e} \times \frac{V}{M} \quad (5)$$

where ΔG° is the Gibbs free energy (kJ/mol), ΔS° is the entropy (kJ/mol·K), and ΔH° is the enthalpy (kJ/mol).

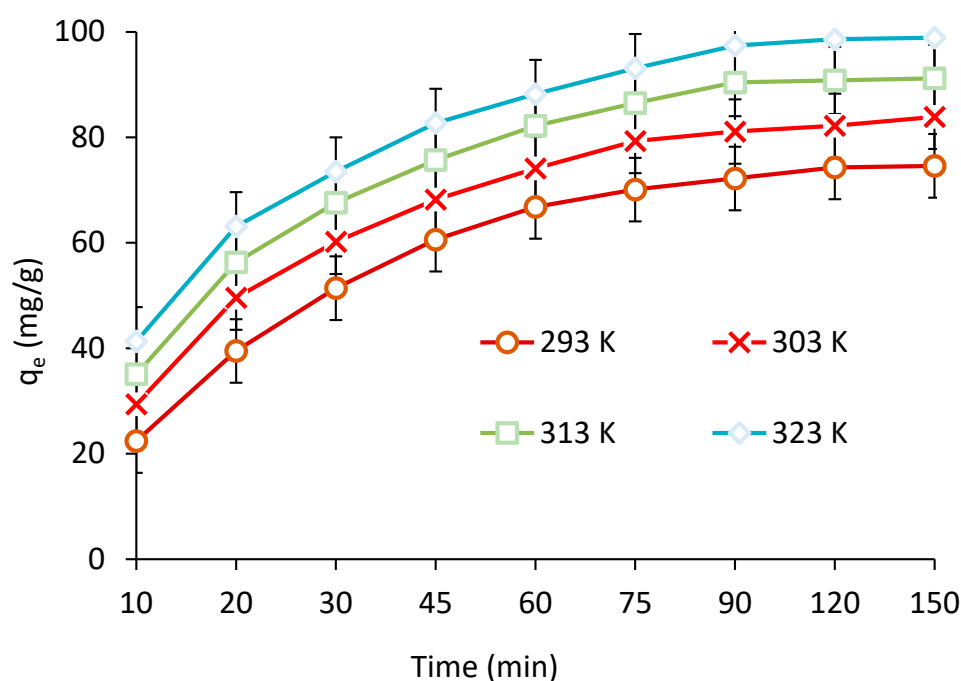


Figure 8. The effect of temperature on antibiotic uptake in the optimal number of other parameters.

The values of ΔH° and ΔS° were determined from the intercept and slope values of the straight line of plotting of $\ln K$ versus $1/T$. The obtained ΔH° was $68.65 \text{ kJ} \cdot \text{mol}^{-1}$, and this indicates that the process is endothermic [60,61]. The ΔG° values were -2.32 , -3.65 , -5.82 and -9.72 , at 293, 303, 313, and 323 K, respectively. These results showed that with increasing the temperature, the Gibbs free energy value decreased. Moreover, the negative ΔG° values indicate that the adsorption process was spontaneous, which supports the feasibility of the adsorption process. Furthermore, this indicates the desirability of adsorption at high temperatures [62]. In addition, a high enthalpy value of $40 \text{ kJ} \cdot \text{mol}^{-1}$ indicates the chemical adsorption of LFX on the MWCNTs/CoFe₂O₄, which confirms the results of isotherm studies [63]. In addition, positive entropy values ($0.24 \text{ kJ} \cdot \text{mol}^{-1} \cdot \text{K}^{-1}$) also indicate that increases in randomness, including the number of species at the solid–liquid interface during the adsorption process [64].

4. Conclusions

This study investigates the adsorption of LFX onto MWCNTs/CoFe₂O₄ under different reaction conditions such as mixing time, pH, LFX dosage, MWCNTs/CoFe₂O₄ concentration and temperature by conducting a batch experiment. The equilibrium time for adsorption of LFX from an aqueous solution was achieved after 90 min. Additionally, it is concluded that the kinetic experimental data were better explained by the PSO, while the adsorption isotherm study showed that Freundlich and Sips models were found to provide the best fit. The maximum adsorption capacity value obtained was 122.2 mg/g, according Langmuir models. Thermodynamics analyses indicate that the LFX adsorption on MWCNTs/CoFe₂O₄ is feasible, spontaneous and endothermic. From the results obtained, it can be concluded that pH is a key factor that affects the rate of adsorption of LFX. Furthermore, the results showed that LFX adsorption increased with increasing both contact time and MWCNTs/CoFe₂O₄, as well as decreasing both the pH value and LFX concentration. The results suggest that MWCNTs/CoFe₂O₄ is an efficient adsorbent for the deletion of LFX from an aqueous solution.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/magnetochemistry9010009/s1>. Figure S1. SEM image of MWCNTs/CoFe₂O₄ (a), TEM image of MWCNTs (b), and TEM image of MWCNTs/CoFe₂O₄ (c); Figure S2. XRD patterns of the MWCNTs and MWCNTs/CoFe₂O₄; Figure S3. N₂ adsorption-desorption isotherms and pore size distribution; Table S1: Errors functions used to determine the goodness of fit of the experimental data with relevant theoretical model; Table S2: Non-linear forms of kinetic models; Table S3: Non-linear forms of isotherms models.

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