

Effective Adsorption of Ciprofloxacin Antibiotic Using of Novel Magnetic Fe₃O₄/Activated Carbon Nanocomposite from Aqueous Solution

Fatemeh Zisti¹, Kethineni Chandrika², Davoud Balarak^{3,*}

¹Department of Chemistry, University of Brock, St. Chatarines, Ontario, CANADA.

²Department of Biotechnology, Koneru Lakshmaiah Education Foundation, Vaddeswaram, Guntur, Andhra Pradesh, INDIA.

³Department of Environmental Health Engineering, Health Promotion Research Center, Zahedan University of Medical Sciences, Zahedan, IRAN.

ABSTRACT

Background: Pharmaceutical residues are biologically active in trace concentrations. This has increased worries about their potential hazards to human health and living organisms and has highlighted the need to treat them once they are discharged into aquatic ecosystems. In this study, the potential of removal of Ciprofloxacin antibiotic (CIP) using novel magnetic Activated carbon nanocomposite (Fe₃O₄/AC) was studied. **Materials and Methods:** For conducting equilibrium analysis, a batch system was considered and operated under conditions including CIP initial concentration, solution pH, Fe₃O₄/AC dosage, and mixing time. Analyzing the experimental data was done using different isotherm and kinetic models. **Results:** The analysis of the physical and chemical properties of Fe₃O₄/AC was indicative of the successful preparation of Fe₃O₄/AC; the surface area, microspores volume, and average pore diameter were observed to be 129.2 m²/g, 0.512 cm³/g, and 4.6 nm, respectively. A removal percentage of 100% was obtained for CIP using Fe₃O₄/AC under optimal values of the studied parameters; the optimum values of these parameters were as follows: solution pH of 5, adsorbent dosage of 0.8 g/L, and an initial CIP amount of 25 mg/L. The Langmuir model was found to be best for labeling the equilibrium data (maximum CIP adsorption capacity of 156.2 mg/g was obtained based on the Langmuir model). The fit of experimental data kinetic with the pseudo-second-order equation was also detected. **Conclusion:** The obtained results let us suggest Fe₃O₄/AC nanocomposite as a decidedly competent adsorbent for removing CIP.

Keywords: Adsorption, Ciprofloxacin, Antibiotic, Fe₃O₄/AC, Aqueous Solution.

Correspondence:

Dr. Davoud Balarak

Department of Environmental Health Engineering, Health Promotion Research Center, Zahedan University of Medical Sciences, Zahedan, IRAN.
Email: dbalarak2@gmail.com

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INTRODUCTION

An extensive range of cytostatic drugs are detected in aquatic ecosystems, chiefly in groundwater and surface water, the sources of which are normally diverse wastewater effluents.¹ These pharmaceutical residues are linked with hospitals, animals farming, and industrial agricultural activities.^{2,3} These compounds are biologically active in trace concentrations. This has increased worries about their potential hazards to the human health and living organisms and has highlighted the need to treat them once they are discharged into aquatic ecosystems.^{4,5}

Ciprofloxacin (CIP) is recorded as a hazardous material included in the "class 1 classification of drugs and is not biodegradable and is seldom adsorbed to sediment owing to its high hydrophilicity,

low log Kow value (Kow= 0.63) and high mobility in aqueous solutions.⁶⁻⁸

Effluent and drinking water treatment operations have not been designed specifically for the removal of pharmaceuticals.⁹ Hence, plans for the elimination of pharmaceuticals from water samples have been predominantly investigated.¹⁰ Advanced wastewater treatment operations propose great removal rates for drug compounds.^{11,12} However, there are concerns in regard with the unpleasant by-products and cost-effectiveness of these treatment processes.¹³ Among above-mentioned methods, adsorption has been reported as a more preferable technique than the others owing to its high versatility and efficiency.^{14,15}

Unfortunately, activated carbon is environmentally unfriendly and a costly adsorbent because it is derived from non-renewable materials, making its use limited on industrial scales.^{16,17} Therefore, it is necessary to prepare low-cost activated carbon from materials such as waste bamboo, waste tires, waste tea, sugar cane bagasse, and rice husks. Rice is a rich source of



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carbohydrates in many countries of the world, chiefly in Asia. Each kilogram of harvested grain generates 1- 1.5 kg of straw, giving an approximately global rice straw production of 750-1120 million tons.¹⁸ Most of these rice wastes are left or burned on agricultural lands and cause possible damage to the environment due to methane emissions and airborne carbon particulate matter, which can have harmful effects on human health and climate change.¹⁹ Carbonization and activation of rice straw in inert atmosphere produces highly porous carbon with a large specific surface area and high porosity.²⁰ Although, newly porous carbons synthesized from rice straw have attracted attentions in the removal of pollutants from water, however, more research is needed on rice straw modifications such as acid treatment and the introduction of different functional groups on its surface for a better adsorption performance of contaminants.²¹

In this study, Rice was studied as feedstock for the preparation of Activated Carbon (AC) functionalized via magnetite (Fe₃O₄) and utilized for the elimination of CIP from water samples. The effect of pH, contact time, Fe₃O₄/AC dose, and initial CIP concentration were also checked. Moreover, isotherm and kinetics of CIP adsorption were considered.

MATERIALS AND METHODS

Chemicals

In this study, CIP antibiotic with the chemical formula (C₁₇H₁₈N₃O₃F), the molecular weight of 331.3 g/mol and it was produced by Sigma-Aldrich Company and was used in the preparation of the stock solution. Also, H₂SO₄ and NaOH were prepared from Merck Company, Germany (laboratory purity) to adjust the pH of the solution. Double distilled water was used in all stages of the experiments.

Preparation of Activated Carbon (AC)

The raw material used in this study for preparing the activated carbon was rice straw obtained from Rasht, Iran. After washing them, they dried in an oven, powdered using a steel blender, and sieved using a mesh 40 sieve. 15 g of the sample was then placed in Teflon-lined hydrothermal autoclave reactors. After that, it was placed in a furnace at 500°C for one hr. The sample was then rinsed with deionized water several times, and rinsing was continued until reaching the pH of the wastewater to a neutral state. It finally underwent in drying process at 110°C for 24 hr.

Preparation of magnetite-activated carbon (AC/Fe₃O₄)

This section started with the dispersion of the obtained AC in 70 mL EtOH and the homogenization of the prepared mixture via ultrasonication. After 3 hr and the addition of 1.5 g FeCl₃·6H₂O and 3.2 g C₂H₃NaO₂ to the mixture at 25°C, it was stirred for 30 min. Finally, after transferring the mixture into a 100 mL Teflon-lined stainless-steel autoclave for 10 hr at 200°C and

washing several times with ethanol and deionized water, it was exposed to a drying process in a vacuum for 6 hr at 60°C.

Adsorption tests

The static adsorption batch experiments were conducted in a 200 mL conical flask with 0.08 g of the AC/Fe₃O₄ nanocomposite and 100 mL of various primary concentrations of CIP solution, and after that the samples were kept in a water bath shaker at 180 rpm for 24 hr at 25°C. Moreover, the impacts of pH factor on the CIP adsorption were altered from 3.0–10.0, utilizing hydrochloric acid and/or sodium hydroxide solutions (0.10 mol/L). The kinetic investigations were carried out in the same way by taking 100 mL of CIP (10-100 mg/L) and 0.08 g of AC/Fe₃O₄. Finally, the AC/Fe₃O₄ were isolated at the designed adsorption time through magnetic decantation. Meanwhile, the residual concentration of CIP molecules in the bulk (C_e) was measured utilizing UV-visible spectrophotometry at 280 nm and removal efficiency and the adsorption capacity of CIP were computed in accordance with Equations (1 and 2):²²⁻²⁴

$$\% \text{Removal} = \frac{(C_0 - C_t)}{C_0} \times V \quad (1)$$

$$q_t = \frac{C_0 - C_t}{M} \times V \quad (2)$$

RESULTS

According to the results of Figure 1, the time was effective on removal efficiency, and the optimum contact time to obtain the best removal efficiency was 90 min. After the mentioned time, unchanged removal efficiency is detected, which is representative of the saturation of active sites on the AC/Fe₃O₄ surface. In Figure 1, the CIP adsorptions of AC/Fe₃O₄ as a function of CIP concentrations were represented. Based on Figure 1, for the concentration of CIP higher than 25 mg/L, a decrease in the ability of AC/Fe₃O₄ to bind to it is detected. Hence, the optimum concentration of CIP in the present study was 25 mg/L.

According to the results, there is a decline in the adsorption efficiency of the studied pollutant by reducing pH (Figure 2). Moreover, results evidently introduce a pH of 5 as the optimum pH for CIP removal.

The results show that when the dosage of AC/Fe₃O₄ increased, the CIP removal increased and, no decreasing trend was observed in their adsorption process (Figure 3).

The adsorption isotherm models which have been utilized for assessing the results of CIP adsorption onto AC/Fe₃O₄ were different models (Table 1). Accordingly, the Langmuir model was found to be the most useful model for analysis of the adsorption data; based on this, the occurrence of monolayer adsorption on the particular homogenous sites of the AC/Fe₃O₄ surface is confirmed.

In Table 2, equations related to kinetic models and results obtained from their application are brought. PSO was introduced as a superior model compared to other employed models, including pseudo-first-order and IPD, when their correlation coefficients were compared with each other. Besides, the closeness could be detected between the calculated equilibrium capacity value (q_e , cal=156.2 mg/g) using the PSO and the corresponding experimental one (q_e , exp), which approves the more applicability of this model compared to other models.

DISCUSSION

CIP adsorption might be negatively and/or positively affected by changes in contact time. As can be detected in the related Figure 1, the contact of CIP and adsorbent in the upward part of the plot has resulted in operative adsorption since there are enough unsaturated active sites yet.²⁵ After establishing equilibrium between the sorption sites and CIP molecules, there is no significant effect on adsorption due to additional reaction time; this is ascribed to saturation of the sorption sites during contact time and lack of more unoccupied space for additional CIP adsorption.^{26,27}

The pH of the solution is a factor that effectively governs adsorption effectiveness because it has a role in the determination of the surface charge of the adsorbent and adsorbate. It represents the importance of taking into consideration of the deprotonation (or protonation) of CIP.²⁸ Due to effects associated with pH, the ionization degree, the adsorbent surface charge, and the molecular structures of the adsorbates can be changed; thus, it is effective in determination of the kind of interaction between the adsorbent and CIP, which occurs due to the ionization of species in the solution.^{29,30}

The initial concentration of pollutant, i.e., CIP in the present study, is another effective factor in adsorption, and its effects should be evaluated. Previous reports have been indicative of discernible effects of this parameter up to a certain limit. Furthermore, the willingness of the adsorbent to adsorb CIP molecules has been dependent on the accessible sorption sites on the adsorbent's

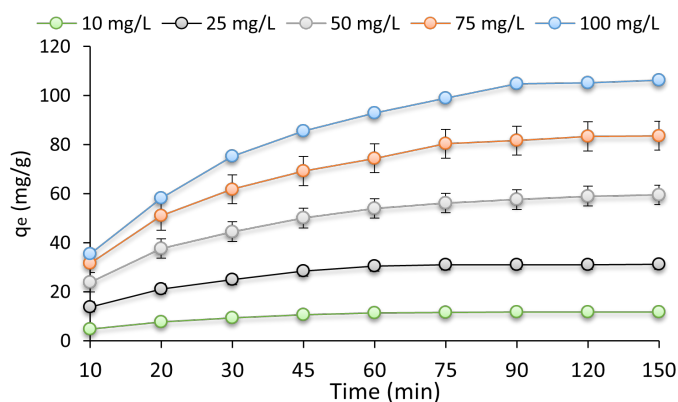


Figure 1: Effect of contact time and concentration on the removal of CIP (dosage: 0.8 g/L, pH: 5 and tem; 25°C).

surface.³¹ For values higher than the optimum value, a saturation of active sites on the adsorbent surface results in a decline in adsorption efficiency. Moreover, there is limited vacant space between the layers of AC/Fe₃O₄, and the adsorption process is carried out based on replacing interlayer anions of the adsorbent with contaminants. Therefore, this space has the capacity for accepting the optimum AC/Fe₃O₄ values and participating in efficient adsorption. Accordingly, a saturation of active sites of the interlayer of AC/Fe₃O₄ led to stopping further diffusion of contaminants onto the adsorbent.³²

K_L is indicative of the Langmuir constant. The homogeneous adsorption and single-layer coverage of the AC/Fe₃O₄ surface by CIP without any interaction between molecules is the assumption represented by the Langmuir model. Freundlich isotherm, as an experimental model, offers surface heterogeneity and an exponential distribution of the sorption sites of adsorbent and energy corresponding to them; this is a reversible adsorption model and is not restricted to monolayer formation.³³

The mean separation factors (R_L) obtained by the equation $R_L = 1 / (1 + (k_L C_0))$ are between zero and one for all temperatures, which

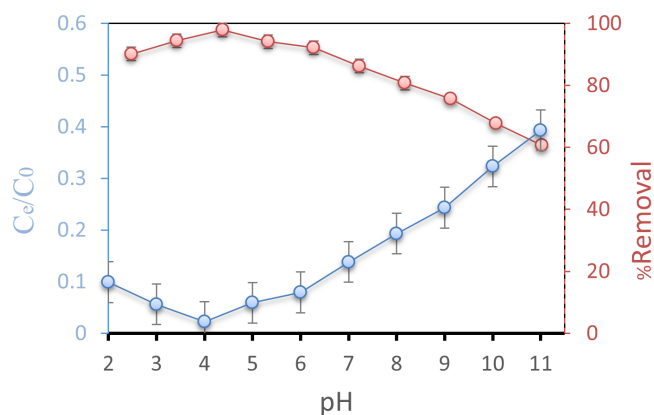


Figure 2: Effect of different pH on the removal of CIP (CIP concentration: 10 mg/L, dosage: 0.8 g/L, contact time: 90 min and tem; 25°C).

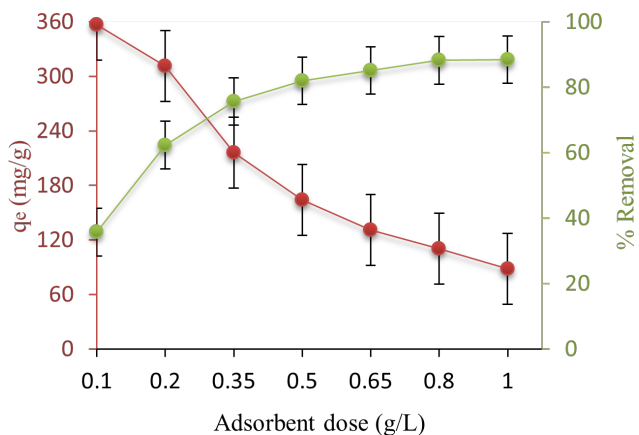


Figure 3: Effect of AC/Fe₃O₄ dose on the removal of CIP (CIP concentration: 100 mg/L, time: 90 min, pH: 5 and tem; 25°C).

Table 1: Isotherm parameters for CIP adsorption onto AC/Fe₃O₄.

Models	Langmuir $= \times +$				Models	Temkin $q_e = A + B \ln C_e$			
	288 K	298 K	308 K	318 K		288 K	298 K	308 K	318 K
Tem	288 K	298 K	308 K	318 K	Tem	288 K	298 K	308 K	318 K
q_m	117.9	132.4	141.2	156.2	A	6.11	7.23	9.16	11.2
R^2	0.995	0.996	0.999	0.997	B	0.72	0.83	0.93	1.05
R_L	0.421	0.654	0.711	0.652	R^2	0.652	0.784	0.789	0.821
Models	Freundlich $\ln q_e = \ln C_e + \ln K_f$				Models	D-R $\ln q_e = \ln q_m - K\epsilon^2$			
K_f	2.45	3.12	3.68	4.58	q_m	44.5	56.2	71.3	92.1
$1/n$	0.28	0.45	0.61	0.73	E	1.47	3.24	2.19	5.75
R^2	0.897	0.842	0.854	0.931	R^2	0.765	0.873	0.832	0.798

Table 2: The results of kinetic model studies related to the CIP adsorption onto AC/Fe₃O₄.

Model	Parameters	10 mg/L	25 mg/L	50 mg/L	75 mg/L	100 mg/L
		Q_e exp	11.2	31.5	64.2	95.5
PFO $\log(q_e - q_t) = \log q_e - t$	q_e cal	6.14	17.4	31.5	53.2	71.4
	K_1	0.051	0.061	0.065	0.069	0.071
	R^2	0.893	0.886	0.854	0.814	0.902
PSO $= +$	q_e cal	11.6	29.4	66.8	98.2	120.4
	K_2	0.009	0.007	0.004	0.002	0.001
	R^2	0.997	0.994	0.996	0.994	0.996
IPD $q_t = K t^{1/2} + C$	K	4.02	5.32	7.45	115	13.4
	C	1.19	1.34	3.02	3.11	4.23
	R^2	0.902	0.913	0.918	0.924	0.934

indicates that the adsorption processes are favorable. The n values obtained from the Freundlich model for all temperatures are between 1 and 10, indicating that CIP molecules were favorably adsorbed on AC/Fe₃O₄ adsorbent. The values of b (Temkin adsorption heat) in Table 1 indicates the amount of heat that the adsorbate needs to be adsorbed on the adsorbent surface.³⁴⁻³⁶ Therefore, the adsorption of CIP on AC/Fe₃O₄ requires more heat, which will be discussed in the section of the thermodynamics of adsorption. The values of the mean adsorption energy parameter (E) obtained from the D-R isotherm model, are less than 8 kJ/mol in all temperatures, which indicate that all of the adsorption processes are physical.³⁷⁻³⁸

Generally, electrostatic attraction, π - π stacking, hydrogen bonding, base-acid interactions, Van der Waals forces, and hydrophobic contact are involved in the adsorption of water contaminants on adsorbents. The main driving force of adsorption is the replacement of pollutants with the interlayer anion of AC/Fe₃O₄. Based on this, the adsorption phenomenon is assisted by various interactions. During the CIP removal process, the interaction between π -conjugated electrons and positively charged AC/Fe₃O₄ as π -cation interaction is possible to occur.

Since there is no π -conjugation plane in AC/Fe₃O₄, the π - π stacking interaction between them is ruled out.³⁹ The adsorption process is progressed at alkaline pH, and electrostatic interaction with CIP AC/Fe₃O₄ can be occurred due to partial deprotonation of the AC/Fe₃O₄, which is led to an acceleration of the studied process.³⁶ In the acidic pH, the CIP adsorption is decreased due to partial protonation of CIP; this diminishes its electrostatic attraction with the adsorbent. In this case, the formation of π -cation interaction is detected between CIP and AC/Fe₃O₄ due to the presence of π electrons density. Furthermore, bonding hydrogen with hydroxides of AC/Fe₃O₄ can be carried out in the case of the presence of OH groups.⁴⁰

CONCLUSION

To sum up, effective AC/Fe₃O₄ for elimination of CIP molecules from water were prepared via chemical activation. The optimum conditions for removal efficiency of CIP onto AC/Fe₃O₄ nanocomposite were obtained at CIP concentration of 25 mg/L, AC/Fe₃O₄ nanocomposite dose of 0.08 g, solution pH of 5, equilibrium time of 90 min, and temperature of 25°C, giving the highest percentage of CIP removal of 100%. The isotherm

and kinetic of CIP molecules adsorption onto nanocomposite follow the Langmuir and PSO models which suggests that the complexation chemisorption system is anticipated in the adsorption processes and AC/Fe₃O₄ nanocomposite can be swiftly and effortlessly isolated utilizing a magnet. High CIP adsorption by the AC/Fe₃O₄ suggest that AC/Fe₃O₄ have excellent utility for practical applications.

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CONFLICT OF INTEREST

The authors declare that there is no conflict of interest.

ABBREVIATIONS

AC/Fe₃O₄: Magnetic Activated carbon nanocomposite;
CIP: Ciprofloxacin; D-R: Dubinin-Radushkevich; PSO: Pseudo-second-order kinetic; IPD: Intra particle diffusion.

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