**RESEARCH ARTICLE**

# Removal of Pharmaceutical Drug by using Effective Adsorbent: Process Optimization and Isotherm Study

Fatima S. Fahim[1], Mohanad A. Mohammed[2], Noor A. Al-lamy[3], Zuhair I. Al Mashhadani[4], Emad S. Abood[5], Aseel M. Aljeboree[6]*

[1]*Department of Anesthesia Techniques, Al-Mustaqbal University College, Babylon, Iraq*
[2]*College of Pharmacy, Al Farahidi University, Baghdad, Iraq*
[3]*Al-Manara College for Medical Sciences, Amarah, Iraq*
[4]*Al-Nisour University College, Baghdad/Iraq*
[5]*Department of Medical Physics, Hilla University College, Babylon, Iraq*
[6]*Department of Chemistry, College of Sciences for Girls, University of Babylon, Hilla, Iraq*

***Received: 17th February, 2022; Revised: 08th April, 2022; Accepted: 16th May, 2022; Available Online: 25th June, 2022***

## ABSTRACT

Hydrogel can be swelling in water and is three-dimensional polymeric. Because of its ability to absorb water in large quantities, it is highly efficient in removing pollutants. The hydrogel was prepared from low-cost and available materials, and the hydrogel surface has high efficiency for removing chloramphenicol (CAP) from an aqueous solution. Many of the chemical and physical properties of the prepared surface were studied from these important techniques (fourier transform infrared spectroscopy (FTIR), x-ray diffraction analysis (XRD), transmission electron microscopy (TEM), and acute flaccid myelitis (AFM)). The adsorption capacity increased between 1-minute and 25 minutes, after which the equilibrium time for the hydrogel surface reached the equilibrium state after one hour. adsorption method is perfectly appropriate for model second-order, giving the best value of ($R^2$=1) and the $K_2$ constant of rate (0.1414 g $mg^{-1}min^{-1}$ )the temperature adsorption (25ºC).

**Keywords:** Adsorption, Chloramphenicol (CAP), Drug, Pharmaceutical, Removal.

International Journal of Pharmaceutical Quality Assurance (2022); DOI: 10.25258/ijpqa.13.2.20

**How to cite this article:** Fahim FS, Mohammed MA, Al-lamy NA, Al Mashhadani ZI, Abood ES, Aljeboree AM. Removal of Pharmaceutical Drug by using Effective Adsorbent: Process Optimization and Isotherm Study. International Journal of Pharmaceutical Quality Assurance. 2022;13(2):193-196.

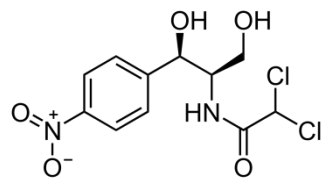
**Source of support:** Nil.

**Conflict of interest:** None

## INTRODUCTION

Personal care products and drugs are one of the most dangerous pollutants in waste water and drinking water. In 1976, it was reported that different types of drugs in drinking water and a large group of pharmaceuticals were untreated and treated in drinking water, surface water, and even groundwater.[1-5] Through the frequent and common use of drugs and pharmaceuticals in the prevention and treatment of many diseases of humans, animals, and plants, so they are abundant in drinking water and sewage. And therefore Presence of drugs in very high concentrations in water resulting from the wastes of pharmaceutical factories and hospitals. The CAP is one of the most famous and widely used antibiotic as a drug in veterinary medicine and veterinary medicine.[6-11] It has many good effects on gram-positive and gram-negative bacteria, and it is considered a low cost and widely used in the treatment. However, CAP drug has many risks and negative effects when used. The physical properties and structure of CAP appear in Table 1 . CAP is frequently detected in drinking water, wastewater, surface water and groundwater. Therefore, developing an effective, sensitive, and inexpensive technology to remove CAP drugs from water is important. Many important techniques have been used, including photocatalytic, photo-oxidation, extraction, and adsorption,[12-19] as adsorption is one of the simplest and easiest methods used to remove water pollutants due to its environmental dependence on friendly

**Table 1:** The physical properties and structure of Chloramphenicol (CAP).

| | |
|---|---|
| Chemical formula | $C_{11}H_{12}Cl_2N_2O_5$ |
| Solubility in water | 6.8 |
| Molecular weight | 323.13 g·$mol^{-1}$ |
| Molecular structure |  |

**Author for Correspondence: annenayad@gmail.com*

surfaces that are inexpensive and available. This study depended on the use of an inexpensive, environmentally friendly surface with very high efficiency in removing a drug from the aqueous solution.

## EXPERIMENTAL PART

The standard solution was prepared by dissolving a drug (0.1 g) in (100 mL) distilled water to obtain a concentration (100 mg/L), and through it, a series of concentrations (5-100 mg/L) was prepared. A calibration curve was drawn according to Lambert-Beer's law, as shown in Figure 1. The adsorption experiments were carried out at room temperature, and the equilibrium time of 60 minutes pH of drug solutions 6.7. Using a concentration from 1 to 25 mg/L of the drug and an amount of hydrogel weight of 0.05 added, the samples were quickly placed in a water bath shaker at 350 rpm for 15 minutes. The solution was separated using a centrifuge, and the drug concentration was measured before and after the adsorption process using a UV-vis spectrometer wavelength of 278 nm. The adsorption kinetics were also studied at the same optimum conditions for the experiment. Take the drug concentration of 25 mg/L by 10 mL of drug solution at room temperature and the weight of 0.05 g of the hydrogel using different times (5–120 min). The adsorption efficiency was calculated using the equation.

$$Q_e = (C_o - C_e)/C_o\ (V)/(W) \quad \text{... (1)}$$

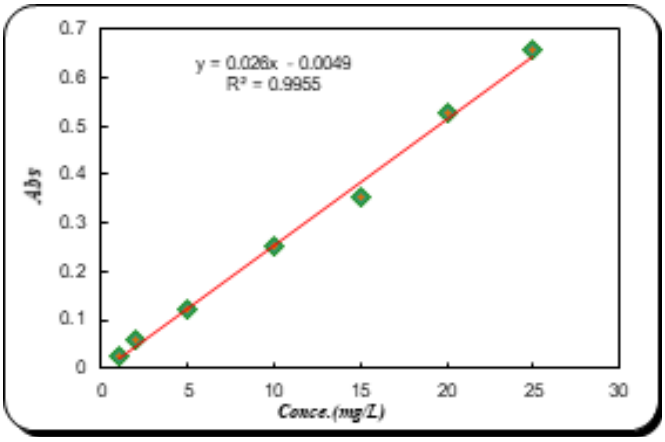



**Figure 1:** Calibration curves of the Chloramphenicol (CAP).

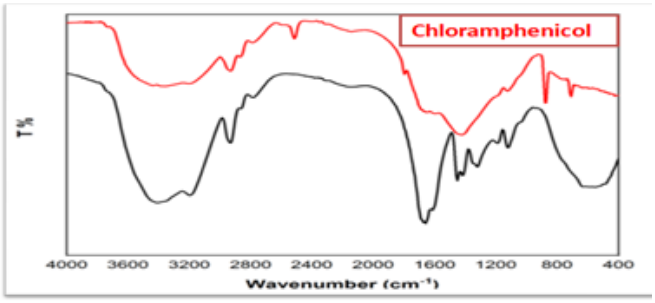



**Figure 2:** FTIR spectra of hydrogels (a) before and (b) after adsorption of drug

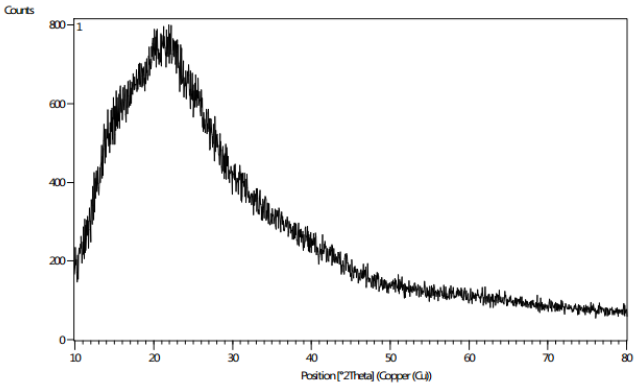



**Figure 3:** X-ray diffraction (XRD) of the hydrogel.

## RESULT AND DISCUSSION

### Characterization of the Adsorbent

*FTIR*

The FTIR was used To characterize the hydrogels before and after the surface adsorption method on the drug, as appears in Figure 2. Thus, a clear and significant difference in the intensity of adsorption between hydrogels. Before the adsorption process with the drug, the hydrogels show a clear decrease in the FTIR spectra in the intensity of the bands adjacent to the adsorption.[20] Also, the surface contains the acidic group that leads to a difference in the intensity of absorption.

*X-ray diffraction (XRD)*

The structural properties of the prepared surface, which are represented by the size and crystalline structure of the surface in its solid state, were studied using X-ray diffraction spectra. The XRD of the hydrogel in Figure 3 shows a broad main peak at position (2θ = 23.126°) and d-spacing of (d = 4.397); this refers to the non-crystalline nature of hydrogel.[21,22]

*TEM*

TEM technology mainly contributes to determining the morphology of the prepared surface where the macroscopic features were observed or affected by the interface or defects. TEM images are shown in Figure 4 of the hydrogel. It consists of aggregate and dark spots, i.e., from a dark background in color attributed to the hydrogel.

*AFM*

Atomic force microscopy (AFM) is used to give statistical information about the surface preparation and the distribution,

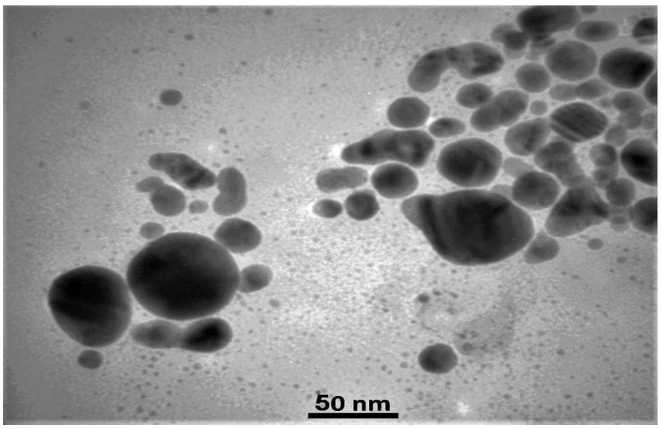



**Figure 4:** TEM image of hydrogel

homogeneity, thickness, and roughness of the surface that was observed in Figure 5 depending on the three-dimensional image of the surface. Through the results, it was shown that the surface contains many cavities, clear evidence of surface roughness.

### Kinetic Adsorption

The absorption kinetics was studied to remove a drug from the aqueous solution at a constant concentration of 25 mg/L of drug at a weight of 0.05 g. The results are shown in Figure 6. Adsorption time (1, 5, 10, 15, 20, 25, 30, 45, and 60) minutes. The adsorption capacity increased between 1 and 25 minutes, after which the adsorption reached an equilibrium state. The equilibrium time for the hydrogel surface reached the equilibrium state after one hour.[23] Kinetics adsorption was estimated in Table 2. The drug kinetics adsorption followed the model second order, as indicated via higher (R2) values Table 2). Adsorption Kinetic of the data were analyzed using the first and second order. The Lagergren –first-order calculate in eq. :

$$ln\ (q_e - q_t) = ln\ q_e - k_1\ t \qquad \ldots (2)$$

kinetic linear of the c second-order can be calculate in equation :

$$q_t = (K_2\ q_e\ t)/(1 + K_2\ q_e\ t) \qquad \ldots (3)$$

The experimental result to ln ($QE-q_t$) and t, as stated by the model first-order, found no variation among the adsorption equilibrium amount $q_e$ and adsorption amount qt at time t. consequently, enforcement of the model first-order is not potential. Thus, the absorption of the drug to hydrogel did not correspond by model first-order.[24-27]

Experimental results as t = qt and t as stated by the model second-order appear in Figure 6, by the derived parameters of the slope and intercept as in Table 2. The $k_2$ constant of rate 0.1414 $gm.g^{-1}min^{-1}$ is the temperature adsorption (25 ºC). The value of ($R^2 = 1$) gives the best. Thus, this adsorption method is appropriate for model second-order.[28]

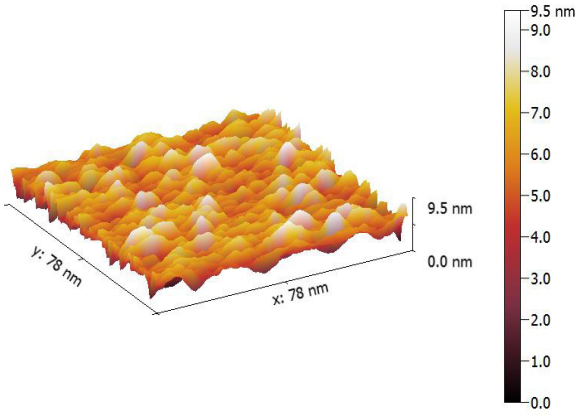



**Figure 5:** Atomic force microscopy (AFM) image of hydrogel

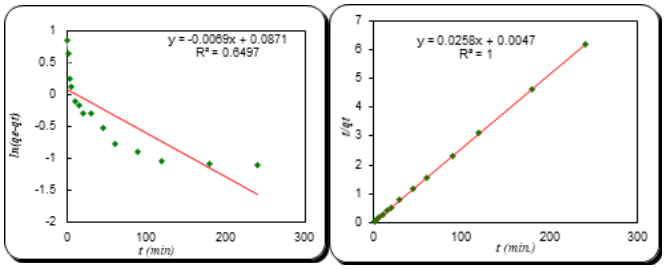



**Figure 6:** effect of kinetic adsorption of first-order (a). second-order (b) Equilibrium time

**Table 2:** Parameters of the kinetic adsorption of CAP drug.

| *First-order* | | | *Second - order* | | |
|---|---|---|---|---|---|
| $k_1$ | $q_e$ | $R^2$ | $k_2$ | $q_e$ | $R^2$ |
| 0.0069 | 1.0912 | 0.696 | 0.1414 | 38.754 | 1 |

## CONCLUSION

In this study, an environmentally friendly, high-efficiency, the inexpensive surface was prepared that can swell and retain its pollutant inside. Various chemical and physical properties of the prepared surface were studied from these important techniques (FTIR, XRD, TEM, AFM). The adsorption kinetics was studied, and it was found that it obeys a second-order model.

## REFERENCES

1. Mahmoud DK, Salleh MA, Karim WA, Idris A, Abidin ZZ. Batch adsorption of basic dye using acid treated kenaf fibre char: Equilibrium, kinetic and thermodynamic studies. Chemical Engineering Journal. 2012 Feb 1;181:449-457.
2. Bachari K, Lamouchi M. Synthesis and Characterization of Fe-JLU-15 Mesoporous Silica Via a Microwave-Hydrothermal Process. Journal of cluster science. 2009 Sep;20(3):573-586.
3. Jasim LS, Aljeboree AM. Removal of Heavy Metals by Using Chitosan/Poly (Acryl Amide-Acrylic Acid) Hydrogels: Characterization and Kinetic Study. NeuroQuantology. 2021 Feb 1;19(2):31-38.
4. Aljeboree AM. Adsorption and removal of pharmaceutical riboflavin (RF) by rice husks activated carbon. International Journal of Pharmaceutical Research. 2019;11(2):255-261.
5. Chowdhury A, Kumari S, Khan AA, Chandra MR, Hussain S. Activated carbon loaded with Ni-Co-S nanoparticle for superior adsorption capacity of antibiotics and dye from wastewater: kinetics and isotherms. Colloids and Surfaces A: Physicochemical and Engineering Aspects. 2021 Feb 20;611:125868. https://doi.org/10.1016/j.colsurfa.2020.125868.
6. Kamil AM, Mohammed HT, Alkaim AF, Hussein FH. Adsorption of Congo red on multiwall carbon nanotubes: Effect of operational parameters. Journal of Chemical and Pharmaceutical Sciences. 2016;9(3):1128-1133.
7. Konicki W, Sibera D, Mijowska E, Lendzion-Bieluń Z, Narkiewicz U. Equilibrium and kinetic studies on acid dye Acid Red 88 adsorption by magnetic ZnFe2O4 spinel ferrite nanoparticles. Journal of colloid and interface science. 2013 May 15;398:152-160.
8. Jiang W, Zhang L, Guo X, Yang M, Lu Y, Wang Y, Zheng Y, Wei G. Adsorption of cationic dye from water using an iron oxide/activated carbon magnetic composites prepared from sugarcane bagasse by microwave method. Environmental technology. 2021 Jan 28;42(3):337-350. DOI: 10.1080/09593330.
9. Aljeboree AM, Abd Alrazzak N, Alqaraguly MB, Mahdi MA, Jasim LS, Alkaim AF. Adsorption of pollutants by using low-cost (Environment-Friendly): Equilibrium, kinetics and thermodynamic studies: A review. Systematic Reviews in

Pharmacy. 2020 Dec 1;11(12):1988-1997.
10. Aljeboree AM, Alshirifi AN, Abdulrazzak FH, Abbas AS, Alkaim AF. Colorimetric Determination of pharmaceutical dyes Using 4-Aminoantipyrine: Oxidative coupling review. Journal of Pharmaceutical Sciences and Research. 2019 Mar 1;11(3):912-918.
11. ALjeboree AM, ALshirfi AN, ALkaim AF. Removal of pharmaceutical amoxicillin drug by using (Cnt) decorated clay/fe2 o3 micro/nanocomposite as effective adsorbent: Process optimization for ultrasound-assisted adsorption. International Journal of Pharmaceutical Research. 2019;11(4):80-86.
12. Al-Gubury HY, Fairooz NY, Aljeboree AM, Alqaraguly MB, ALkaim AF. Photcatalytic degradation n-undecane using coupled ZnO-Co2O3. Int. J. Chem. Sci. 2015;13(2):863-874.
13. Yongde L, Yahui S, Dongjin W, Jing C, Shuhu X. Adsorption of toxic dye Eosin Y from aqueous solution by clay/carbon composite derived from spent bleaching earth. Journal of Hazardous Materials. 2019;1:4656-5544.
14. Karim AN, Jasim LS. Synthesis and characterization of poly (CH/AA-co-AM) composite: Adsorption and thermodynamic studies of benzocaine on from aqueous solutions. International Journal of Drug Delivery Technology. 2019;9(4):558-562.
15. Aljeboree AM, Abbas AS. Removal of pharmaceutical (paracetamol) by using CNT/TiO2 Nanoparticles. Journal of Global Pharma Technology. 2019;11(01):199-205.
16. Aseel M, Aljeboree AF, loay A, algburi HM. Photocatalytic Degradation of Textile Dye Cristal Violet Wastewater using Zinc Oxide as a Model of Pharmaceutical Threat Reductions. Journal of Global Pharma Technology. 2019;11(02):138-143.
17. Ganduh SH, RQK MA, Aljeboree AM, Jasim LS. Selective Spectrophotometric Determination of 4-amino Antipyrine Antibiotics in Pure Forms and their Pharmaceutical Formulations. International Journal of Drug Delivery Technology. 2021;11(2):371-375.
18. Gamoudi S, Srasra E. Adsorption of organic dyes by HDPy+-modified clay: effect of molecular structure on the adsorption. Journal of Molecular Structure. 2019 Oct 5;1193:522-531.
19. Xu P, Zeng GM, Huang DL, Lai C, Zhao MH, Wei Z, Li NJ, Huang C, Xie GX. Adsorption of Pb (II) by iron oxide nanoparticles immobilized Phanerochaete chrysosporium: equilibrium, kinetic, thermodynamic and mechanisms analysis. Chemical Engineering Journal. 2012 Sep 1;203:423-431.
20. Banerjee S, Chattopadhyaya MC. Adsorption characteristics for the removal of a toxic dye, tartrazine from aqueous solutions by a low cost agricultural by-product. Arabian Journal of Chemistry. 2017 May 1;10:S1629-38.
21. Dai H, Ou S, Liu Z, Huang H. Pineapple peel carboxymethyl cellulose/polyvinyl alcohol/mesoporous silica SBA-15 hydrogel composites for papain immobilization. Carbohydrate polymers. 2017 Aug 1;169:504-514.
22. French AD. Idealized powder diffraction patterns for cellulose polymorphs. Cellulose. 2014 Apr;21(2):885-896.
23. Alzayd AA, Atyaa AI, Radhy ND, Al-Hayder LS. A new adsorption material based GO/PVP/AAc composite hydrogel characterization, study kinetic and thermodynamic to removal Atenolol drug from wast water. InIOP Conference Series: Materials Science and Engineering 2020 Nov 1 (Vol. 928, No. 6, p. 062023). IOP Publishing.
24. Huang R, Wang B, Yang B, Zheng D, Zhang Z. Equilibrium, kinetic and thermodynamic studies of adsorption of Cd (II) from aqueous solution onto HACC–bentonite. Desalination. 2011 Oct 3;280(1-3):297-304.
25. Wared SHH, RND, *Synthesis and characterization of sodium alginate-g-polyacrylic acid hydrogel and its application for crystal violet dye adsorption.* International Journal of Drug Delivery Technology, 2021;11(2):556 - 565.
26. Aljeboree AM, Alkaim AF. Role of plant wastes as an ecofriendly for pollutants (Crystal Violet dye) removal from Aqueous Solutions. Plant Archives. 2019;19(2):902-905.
27. Ahmad MA, Alrozi R. Removal of malachite green dye from aqueous solution using rambutan peel-based activated carbon: Equilibrium, kinetic and thermodynamic studies. Chemical Engineering Journal. 2011 Jul 1;171(2):510-516.
28. Hadi ZA, Aljeboree AM, Alkaim AF. Adsorption of a cationic dye from aqueous solutions by using waste glass materials: isotherm and thermodynamic studies. Int. J. Chem. Sci. 2014;12(4):1273-1288.