Applied Chemical Engineering (2025) Volume 8 Issue 4 ISSN: 2578-2010 (O)
doi: 10.59429/ace.v8i4.5720

ORIGINAL RESEARCH ARTICLE

Efficient adsorption of drugs from aqueous solution using a activated carbon derived from Iraqi date pits

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ABSTRACT

This research investigates the use of activated carbon from Iraqi date pits as a novel and sustainable adsorbent for removing three key pharmaceutical drugs: sulfanilamide, sulfisoxazole, and amoxicillin, from contaminated water. The study systematically evaluated crucial factors like equilibrium time, adsorbent dosage, pH, and temperature to optimize the drug removal process. To understand the underlying mechanisms, various adsorption isotherm models were tested, with the (Freundlich, Elovich and Timken) where values range between (0.931, 0.9359 and 0.8867) model proving to be the best fit. This suggests a multilayer adsorption process, where drug molecules form a single layer on the surface of the activated carbon. The investigation of thermodynamic parameters (ΔG, ΔH, and ΔS) further confirmed that the adsorption process was spontaneous where values (-1036.74,-231.775and-534.019).In addition, kinetic studies demonstrated that the pseudo-second-order model provided the most accurate description of the adsorption process for all three drugs. This was supported by exceptionally high correlation coefficients (R2) = 0.9992 for sulfanilamide, 1 for sulfisoxazole, and 0.9995 for amoxicillin. In conclusion, the findings strongly affirm that activated carbon derived from Iraqi date pits is a highly promising, cost-effective, and sustainable adsorbent for treating water contaminated with pharmaceuticals. This work paves the way for the development of new, environmentally friendly materials for water purification.

Keywords: Drugs, activated charcoal, Adsorption, Isotherms, Kinetic

ARTICLE INFO

Received: 1 August 2025
Accepted: 23 September 2025
Available online: 29 October 2025

1. Introduction

Water is a vital component of both the natural ecosystem and human life. If there is any disturbance in the physicochemical characteristics of water, It would adversely affect both [1].When a material that can affect humans and/or the ecosystem is present in excess in water, it is known as water pollution [2].Pollutants can be classified in many ways based on their chemical and physical properties, abundance, stability in the environment, effect on ecosystems, or toxicity and the among these pollutants are medicines [3].

Medicines are among the organic pollutants of water resources due to their wide use in various non-industrial areas. They are used in many different fields that include many projects for humans, animals and plants [4].It is one of the factors that cause cancerous diseases and its presence in sewage water is undesirable, so it is better to get rid of this waste before it is released into the environment [5].Because of this, several researchers have focused on developing acceptable and practical ways to eliminate these contaminants following the worsening of pollution issues in prior years. Which, particularly at low

levels and concentrations, have grown to be a serious issue. Adsorption is one of the most significant methods for treating pollutants [6].

Adsorption\\ It is the phenomenon of aggregation or adhesion of liquid molecules, ions, or atoms on the surface of another substance, whether (solid or liquid). Adsorption has several examples including the adsorption of acetic acid on animal charcoal, and also the aggregation of hydrogen gas in the form of molecules or atoms [7]. Adsorption is usually accompanied by a decrease in free energy **(ΔG)** and also a decrease in entropy **(ΔS)** [8]. because the molecules that suffer adsorption become restricted due to their association with surface atoms, and the decrease in free energy and entropy results in a decrease in enthalpy according to the thermodynamic relationship that links the three quantities together at a certain temperature T, as in the relationship

$$\Delta G = \Delta H - T\Delta S \qquad (1)$$ [9] .

The substance that suffers adsorption may be called adsorbate .The surface on which adsorption takes place is adsorbent [10].

Adsorption may be limited to the formation of a single layer, which is called monomolecular adsorption. It may include several layers, which is called multimolecular. Process usually is reversible, and the reverse process is called desorption. In this review, the definition and types of the adsorption process will be defined [10] . In addition to a brief explanation about the major adsorption isotherm models, adsorption kinetics, and adsorption thermodynamic. Physical and chemical adsorption are the two categories into which adsorption may be separated [11, 12] .

2. Experimental

2.1. Chemicals

The materials used in this research, including acetone (97%), hydrochloric acid, sodium hydroxide (98%), phosphoric acid (35%), drugs and Iraqi date pits.

2.2. Preparation the adsorbent surface

2.2.1. Activated surface of Iraqi date pits

Iraqi Date Cores (IDC) are gathered and wash with distilled water several times and leave them to dry at room temperature for two days, after which the dried date pits are placed in a ceramic pot. It is placed in the muffle furnace for (2h) at a temperature of (500 ^{O}C) in order to obtain charcoal and in the second step the burned coal is ground well to obtain a very fine powder. Then sift the powder using a sieve with dimensions (600μm) and then wash the charcoal several times with distilled water. After that, the washed coal is dried in the oven at a temperature of (80 ^{O}C) and for a period of (40 min) and this method is called physical activation. The last step is the chemical activation of the coal prepared with phosphoric acid at a concentration of (5M) and a ratio of (1:1) for 24 hours.

2.2.2. Preparation of drugs solution

Three drug from (sulfanilamide, sulfisoxazole , Amoxicillin) standard stock solutions were prepared by dissolving 0.05 g of weight in 100 ml of distilled water with a 500 ppm concentration. Next, different concentrations of three medications there were (5–50) parts per milliliter produced. To find the drug's maximum adsorption, the spectrum was recorded using an ultraviolet-visible spectrophotometer, with pure water acting as the blank. The calibration curve for the medication was then determined by testing each of the 10 samples using a UV-visible spectrophotometer. The maximum wavelength obtained at the highest absorption value (λ max) was determined by using a (UV-Vis) spectrometer. Quartz cells, which were thick

(1cm), were used, where the maximum wavelength of sulfanilamide was (268 nm), while the maximum wavelength of (sulfisoxazole) was (261nm) and the maximum wavelength of amoxicillin was (229 nm).

2.3. Effect of Contact time

Contact time affects a precision scientific shaker water bath (GCA) was used to shake 20 ml of solutions containing pharmaceuticals at a specified concentration (50 mg/l) at pH = 7 at a certain speed of 150 rpm and with the required dosage of adsorbent (0.25gm). At 298 K, the batch adsorption process was carried out. To determine how long it would take to achieve equilibrium, the solution was filtered every 30 minutes using a centrifuge (Magafuge 10, Herouse Sepatech). The solution was then examined using a spectrophotometer (Biochrom Ltd., United Kingdom). At (261,268,229 nm), or λmax. The calibration curve for the medication was used to calculate the amount of time needed to attain equilibrium, which came out to be (0.5, 1,1.5,2,2.5,3,3.5,4 h).

2.4. The effect of Surface Weight on adsorption

Weight effect of activated charcoal the effect of the weight of the activated charcoal on its adsorption capacities was examined in order to establish the optimal weight of the active surface on which three medications may be adsorbed at a temperature of 298 K. When the mixture was filtered through filter paper (110 mm) after the time of the medication in a shaking water bath with different weights of activated charcoal (0.1, 0.15, 0.2, 0.3, and 0.5 gm.), fixed volume (20 ml), and concentration (50 ppm for drugs), the absorbance of the filtrate was measured using a spectrophotometer at predetermined λmax .

2.5. Adsorption Isotherms Models

Isotherms of Adsorption numerous isotherm models may be used to represent the results of the isothermal adsorption model. The practical data are examined in the current work using Langmuir, Freundlich , Elovich, Harkin-Jura, Timken , and Dubinin isotherms [13].A fixed (0.5,0.25,0.3) weight of activated charcoal was added to 10 medicine samples for every one (sulfanilamide, sulfisoxazole , amoxiline) that had been manufactured. The prescribed duration of time is spent with the medicine samples submerged in a water bath shaker. After filtering each sample via filter paper, the absorbance of the filtrate is measured at the appropriate λmax using a spectrophotometer .The following quantitative formula was used to determine each adsorbent's concentration (at equilibrium)[13].

$$q_e = (C_o - C_e)\, V/\, m \qquad (2)$$

In this case,

q_e: stands for the quantity of adsorbate (mg/g).

$C_\circ$: Indicate the amount of primary material adsorbent in milligrams per liter. C_e: It displays the concentration under ideal circumstances (mg/L).

V sol: the adsorbent solution's volume (L), and M: Adsorbent weight (g).

3. Result and disscusion

3.1. Field Emission Scanning Electron microscopy for charcoal activated derived from the Iraqi date pits

The scanning electron microscope is a type of electron microscope that gives a sample of images using an electron beam to scan the surface, giving specific signals that provide data about the surface topography of the sample that was examined. The SEM plays a fundamental role in understanding the nature of the adsorbents. Therefore, the surface of activated carbon prepared from Iraqi date pits was studied. The images in Figure (1) showed that the prepared carbon contains a porous structure with a high degree of diffusion, making the surface

more capable of adsorbing materials, as it provides a large and sufficient surface area to accommodate a fair number of molecules. Therefore, it will be positively reflected on the possibility of using this type of activated carbon in the adsorption process [14].

3.2. Effect of conact time

The purpose of this experiment was to ascertain the optimal time for the integrated medications to react with the activated carbon layer composed of pre-existing Iraqi seeds. Based on the data, the drug has a neutral life of (0.5–4) hours, during which time its absorption capacity rises proportionately to the contact time until saturation happens [15] . Figure (2) illustrates the connection between time (h) and the quantity of activated charcoal (mg/g) made from Iraqi date pit powder, which was utilized as an adsorbent. Since there is a positive correlation between the variables, raising any one of them will raise the others. Absorption was assessed for the first drug levels at a specific contact time and a distinct reaction time of (0.5–4h). This is probably due to the fact that there was originally more adsorption surface area available for pollutant adsorption. It was found that each drug had atime in which the best adsorption process took place,which was (3.5 ,1, 0.5) according to the figure (2) for remove the bulk of the maximum amount of contaminants. Pollutant adsorption rises with longer contact times and is constant for 30 minutes after equilibrium is reached for varying initial concentrations.. The surface's unfilled pores are the cause of the high initial absorption rate. The rate of adsorption ultimately depends on the passage of electrons from the watery solution to the adsorbent surface, but it steadily declines after these gaps are filled by chemical ions [16] .

3.3. Effect of activated charcoal surface weight

Applying various weights, ranging from (0.1 to 0.5) grams, on the surfaces of activated charcoal generated from Iraqi date pits allowed for the measurement of this effect. It was discovered that the surfaces, which weighed (0.25, 0.5, and 0.3) grams, were superior. This is due to the fact that all of the adsorption surface's operational sites have achieved the saturation limit at these weights, meaning that the adsorption of pharmaceuticals has no effect on the rise in the number of adsorbed surfaces [17]. (Figure 3) illustrates the connection between the quantity of adsorption (mg/g) for (298 K) and acid functional pH = 7 and the external weight of the adsorbent. Because the active sites eventually are saturated, drug removal diminishes as adsorbent weight increases. The graphic illustrates how the quantity of adsorption rises with surface weight.[18].

3.4. Temperature Effect

The adsorbed concentration has a major impact on the effectiveness of the adsorption process on the activated charcoal surface. These experiments were carried out at a temperature of 293 K to see how much this variable influences the drug's capacity to adsorb onto the surface. The results demonstrated that drug adsorption rises with increasing concentration, and Figures (4) illustrated the magnitude of the influence of conc on drug adsorption. Increased pore size or surface activation at high concentrations might be the cause of this [19] .

3.5. Thermodynamic Parameters

The following formulas were used to calculate the enthalpy (ΔH), entropy (ΔS), and thermodynamic parameters:

$$\ln K_{eq} = \Delta S/R - \Delta H/RT \quad (3)$$

where T is the temperature in degrees Celsius (Kelvin), R is the universal constant for gases (8.314 kJ/mol. K^{-1}), K is the constant of the Vant Hoff equation, and $\ln X_{eq}$ is a natural logarithm for the largest amount adsorbed (mg/g)[20].

$$\Delta G = -RT \ln K \quad (4)$$

$$\Delta S = \Delta H - \Delta G / T \quad (5)$$

l Kc against 1/T's Van't Hoff plot's slope and slope were used to calculate ΔH◦ and ΔS◦. (Figure 5 , Table 2). Since an increase in 1 / T causes a decrease in Ln K_{eq}, the relationship between the two quantities (1 / T and Ln K_{eq}) is negative. The findings demonstrated that a positive value for ΔH indicated the endothermic character of the adsorption, and a negative value for ΔG indicated a spontaneous reaction [21], as shown in Figure (5).

3.5.1. Langmuir Isotherm Model

The Langmuir equation, with its strong theoretical foundation and ease of application, is arguably the most extensively used model for understanding the adsorption isotherm .According to this hypothesis, sorbet molecules do not interact with one another during monolayer sorption on a homogenous surface[22]. In addition, the model assumes homogeneous energies of sorption on to the surface and no transmigration of the adsorbate. The Langmuir isotherm equation is expressed as follows in its linearized version .The formula is as follows: Ce (mg/L) is the equilibrium concentration of drugs in solution

$$C_e/q_e = 1/K_L\, q_m + C_e/q_{m..} \quad (6)$$

q_e (mg/g) is the adsorbent's monolayer adsorption capacity and K_L is the adsorption energy. Figure (6) displays the linearized Langmuir equation. The findings show that, with inadequate fitting on both wastes, the Langmuir model is unable to adequately characterize the experimental data [23] [24].

3.5.2. Isotherm Freundlich

Figure (7) illustrates how the adsorption takes place on multi-layered, heterogeneous surfaces. The Freundlich adsorption equation was utilized to apply (sulfanilamide, sulfisoxazole, amoxicillin) medication to the Iraqi date activated char Coal surface at a temperature of 298 K. The adsorption intensity was expressed by the Freundlich constants, denoted by (n), which were calculated. the degree to which the surface is saturated with the curve's curvature. and (K_f), which derives from the slope and intercept and reflects the surface's adsorption capability [25]. According to the following equation Log q_e = log K_f +1/n Log C_e... (7).

When plotting log q_e versus log C_e. Where K_f and n are the system's Freundlich Constants, which correspond to the adsorption intensity and capacity, respectively

3.5.3. Elovich Isotherm

The formula that explains Elovich based on kinetic data, the model describes the connection in the following equation under the assumption that adsorption is multilayered because the number of adsorption sites grows exponentially with adsorption. The outcomes are shown in Figure (8).

$$\ln q_e / C_e = \ln K.\, q_m{}^{-1} / (q_m.\, q_e) \quad (8)\ [26].$$

3.5.4. Harkin-Jura

Adsorption Isotherm based on the idea that multilayer adsorption may occur on the adsorbents' surface when the distribution of pores is not uniform; the Harkin-Jura isotherm model may be applied to systems that use solid fuels. This is the expression for the Harkin-Jura isotherm model.

$$1 / qe^{2} = B / A - 1/ A \log Ce \quad (9).$$

In which A and B are the Harkin-Jura constants .The study of kinetics, adsorption isotherms, and the removal of drugs from aqueous solution using activated charcoal have all made use of the Harkin-Jura isotherm model [27, 28], figure (9) displays the isotherm of Harkin-Jura isothermal model for drugs (sulfanilamide, sulfisoxazole, amoxicillin) . From this figure, it is clear that the isotherm does not apply to the Harkin-Jura equation.

3.5.5. Timken Isotherm

Furthermore, the Timken parameter may also be used to characterize the isothermal work of the adsorption process using the following equation:

$$qe = B\ Ln\ Kt + B\ ln\ Ce \quad (10)$$

The Timken constants, A and B, may be calculated by plotting q_e vs. ln Ce. Together with the parameters A and B, the correlation coefficient. The results of the experiment are shown in Figure (10) which indicates that the adsorption process mostly adheres to the physical type [29].

3.5.6. Dubinin Isotherm

Because of its unrealistic asymptotic performance and inability to predict Henry's laws at low pressure, this model is only appropriate for an intermediate range of adsorbate concentrations. may also be used to calculate the average free energy of adsorption for each molecule of the adsorbate, or sorption energy. Since the Dubinin model is temperature-dependent, all relevant data may be obtained by graphing the adsorption data as a function of the logarithm of the quantity of adsorbate adsorbed versus the square of potential energy [30]. (Figure 11) presents Dubinin's results may be obtained by graphing the adsorption data as a function of the logarithm of the quantity of adsorbed vs. the square of potential energy [31].

3.6. Effect of pH

The impact of pH on the adsorption of different three drugs on surface charcoal derived activated from Iraqi date pits was investigated at various pH values between(2,4,6,8 and 10) at 293K and zero point charge value PZC=2.5. (Figure 12) shows the proportion of drugs eliminated in relation to pH fluctuation. Figure 5 illustrates how the quantity of adsorption increased as the drugs solution's pH rose to pH 6 and then decreased as the pH value increased. Activated carbon finds application in the adsorption of organic materials and non-polar adsorbents. It is also commonly employed in the treatment of waste gases and wastewater. Since the majority of its chemical properties rely on pH, it is the most often utilized adsorbent [32, 33].

3.7. Adsorption Kinetics

Using basic first-order, second-order, and pseudo-second-order equations as given, several kinetic models are used to the experimental data in order to assess the adsorption kinetics of three medicines onto charcoal activated surface generated from Iraqi date pits. Primitive simplicity Ictitious first-order in

$$\ln (qe-qt) = In\ q_e - K_T \quad (11)$$

Rewritten as :($t/q_e = 1/K_2\ q_e^2 + 1/q_e\ t$) Second-rate pseudo

Where q_e is the initial quantity of three pharmaceuticals absorbed (mg/g); K_1, K_2 and K_T (g /mg) are the basic first-order, pseudo first-order, second-order, and pseudo second-order adsorption rate constants, respectively. The model plots are shown in Figure (13, 14) table (1). The results indicate that pseudo-second order adsorption is the primary drug adsorption method on charcoal activated derived from Iraqi date pits [34].

Table 1. the values of (K_1, K_2) the correlation coefficients and q_e at initial concentration 20ºC for Pseudo 1st order and pseudo 2 st.

drug	q_e mg.g^{-1}	K_1 min^{-1}	R^2	q_e mg.g^{-1}	K_2 gm.g^{-1}.min^{-1}	R^2
sulfanilamide	0.0189	-0.4306	0.1355	476.190	7.385×10-5	0.9992
sulfisaxzole	0.00162	-1.0072	0.4549	-2500	6.262×10-7	1
Amoxicillin	0.0220	-0.5841	0.4674	285.714	4.029×10-5	0.9995

Table 2. Thermodynamic parameters for the adsorption of drugs on the surface activated charcoal derived from Iraqi date pits at different temperatures.

Drugs	ΔH (kJ/mol. K)	ΔG (kJ/mol. K)	ΔS (j/mol. K)	R^2
Sulfanilamide	-13.88	-1036.74	3.491	0.9206
Sulfisoxazole	-12.611	-231.775	0.748	0.9585
Amoxicillin	-10.135	-534.019	1.788	0.9183

Table 3. Constants of adsorption isotherms equations and correlation coefficients of adsorption of (sulfanilamide ,sulfisoxazole and amoxicillin) on to the surface of charcoal and activated charcoal derived from Iraqi date pits at temperature 293K and pH = 7.

Linear equation of isotherm model	Parameters	Value of activated charcoal Sulfanilamide	Value on to the activated charcoal Sulfisoxazole	Value on to the activated charcoal Amoxicillin
Langmuir $\frac{ce}{qe} = \frac{1}{kLqm} + \frac{ce}{qm}$	K_L(L/mg) q_m (mg/g) R^2	-0.0619 -2.219 0.5862	-0.688 -0.2328 0.4245	-0.6083 0.186 0.1507
Freundlich $lnqe = lnkf + \frac{1}{n} lnce$	1/n K_f (l/mg) R^2	0.6112 0.0735 0.931	0.16 0.4808 0.7558	-0.23 34.93 0.3264
Timken Qe = B lnkt + B lnce	K_T(L/mg) B(J/mol) R^2	0.5884 1.0495 0.8676	0.9979 10.519 0.9359	0.3486 -5.7488 0.4405
Elovich $\ln\frac{qe}{ce} = \ln kqm - 1\frac{1}{qm}$ qm	K q_m (mg/g) R^2	3.819 -1.69 0.712	1.537 -1.869 0.84	6.0557 -1.1179 0.886
Harkins-Jura $-\frac{1}{A}$ $\frac{1}{qe2} = \frac{B}{A}$ Log Ce	A B R^2	0.0154 0.738 0.5036	0.0275 0.1262 0.3152	-0.065 0.272 0.4312
Dubinin ln qe = lnqm − ßε²	E q_m(mg/g) ß(mol^2/KJ^2) R^2	1.408 0.1828 0.2523 0.8853	1.422 0.0365 0.2472 0.8892	1.347 0.00114 0.2757 0.8671

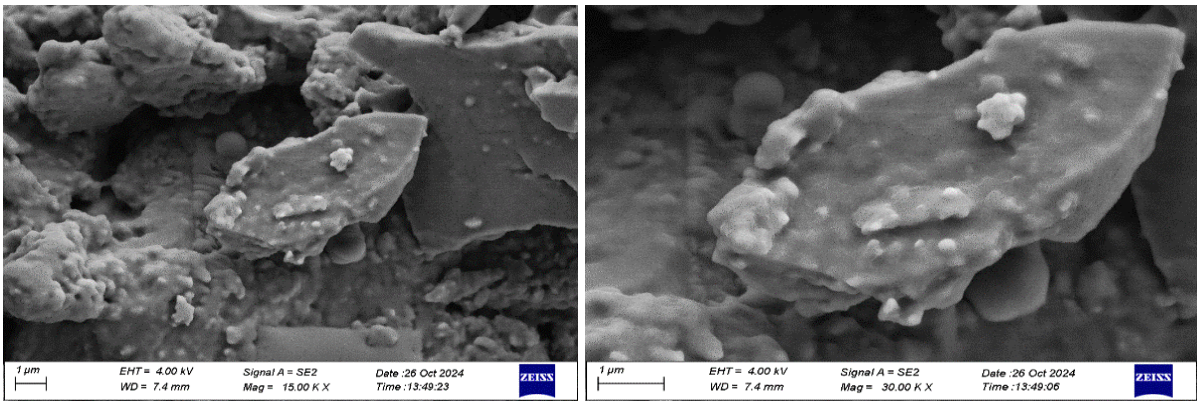

Figure 1. FESEM charcoal activated derived from Iraqi date pits

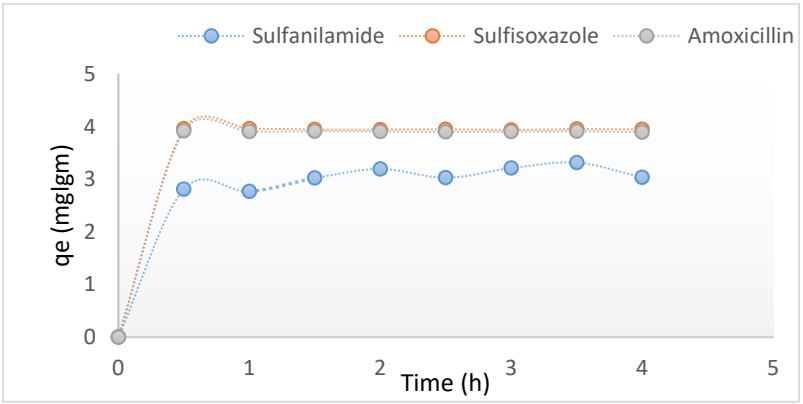

Figure 2. Contact time for different drugs on activated charcoal at pH=7 and Temp. 293K

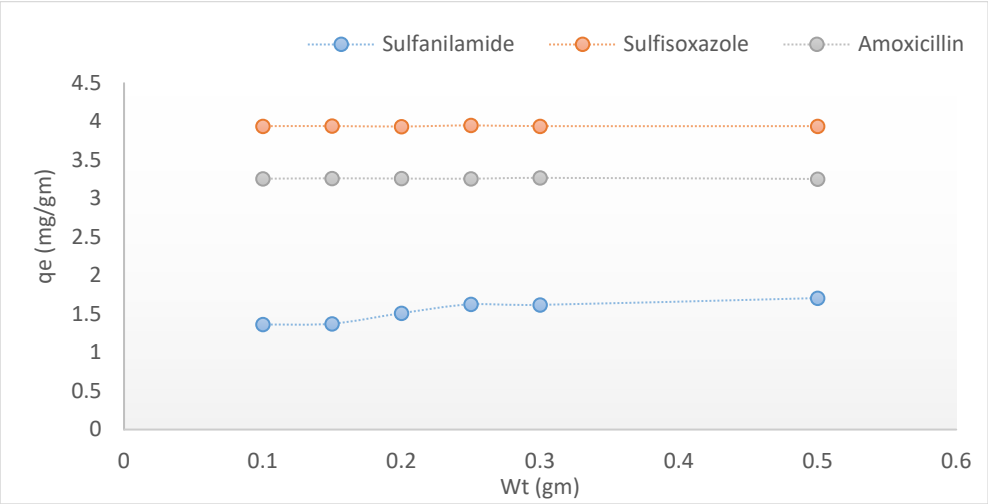

Figure 3. Effect of weight for different drugs on activated charcoal at 50ppm, pH=7, Temp.293K

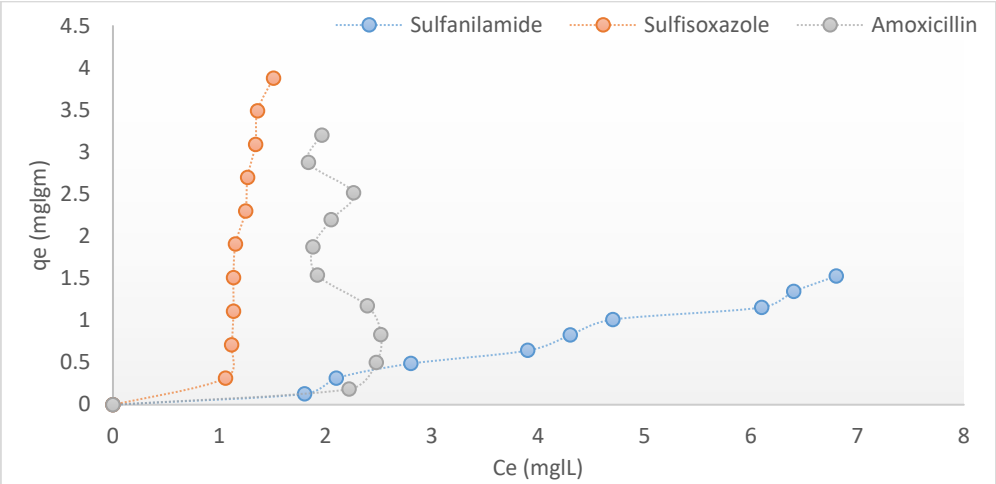

Figure 4. Adsorption isotherm of different drugs on activated charcoal at pH=7, Temp.293K

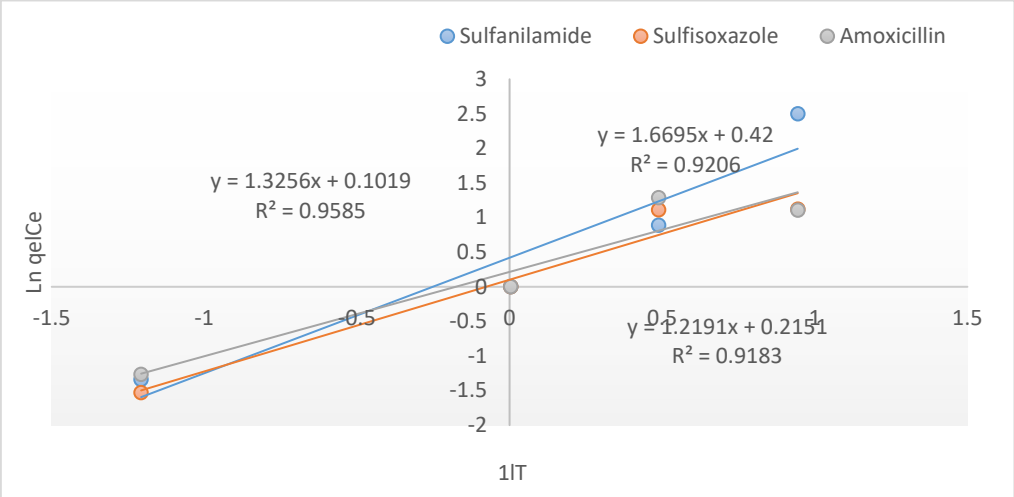

Figure 5. Equilibrium constants are related to temperature inversion of different drugs on activated charcoal at 50ppm and pH=7, Temp. 293,303,313,323 K.

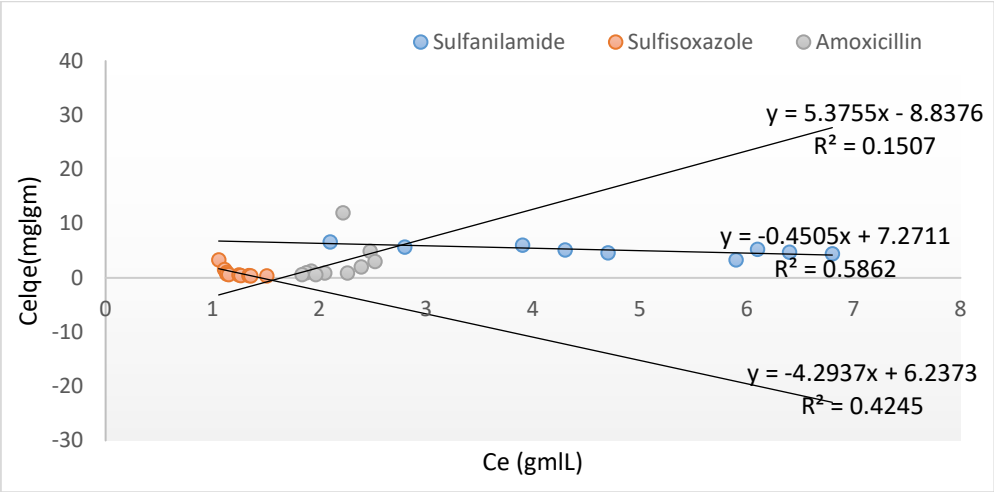

Figure 6. Langmuir adsorption for different drugs on activated charcoal at pH=7 and Temp. 293K

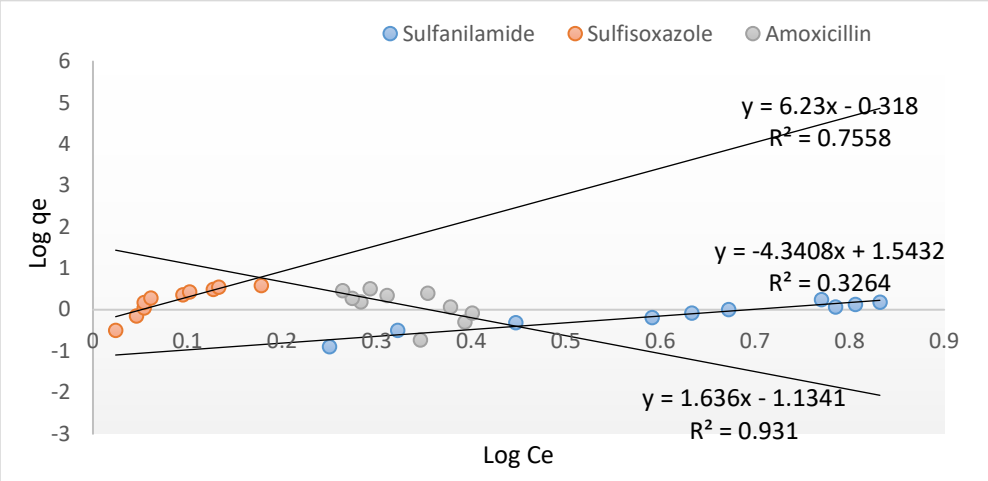

Figure 7. Freundlich adsorption for different drugs on activated charcoal at pH=7 and Temp. 293K

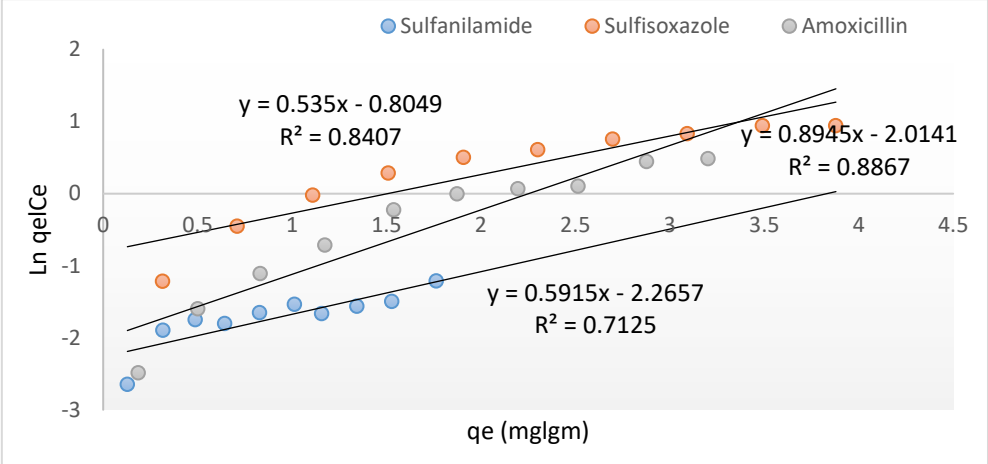

Figure 8. Eluvishion adsorption for different drugs on actvatied charcoal at pH=7 and Temp. 293K

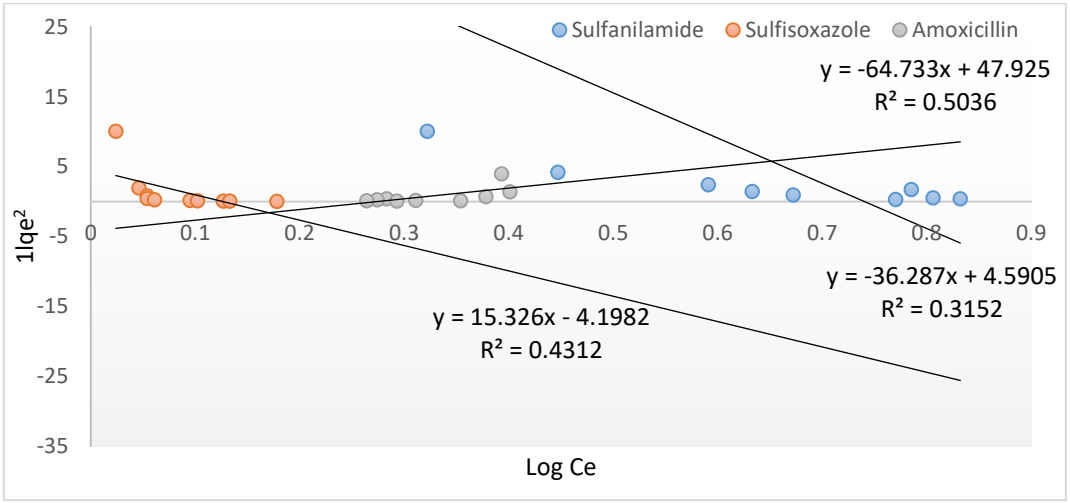

Figure 9. Hargiongour adsorption for different drugs on activated charcoal at pH=7 and Temp. 293K

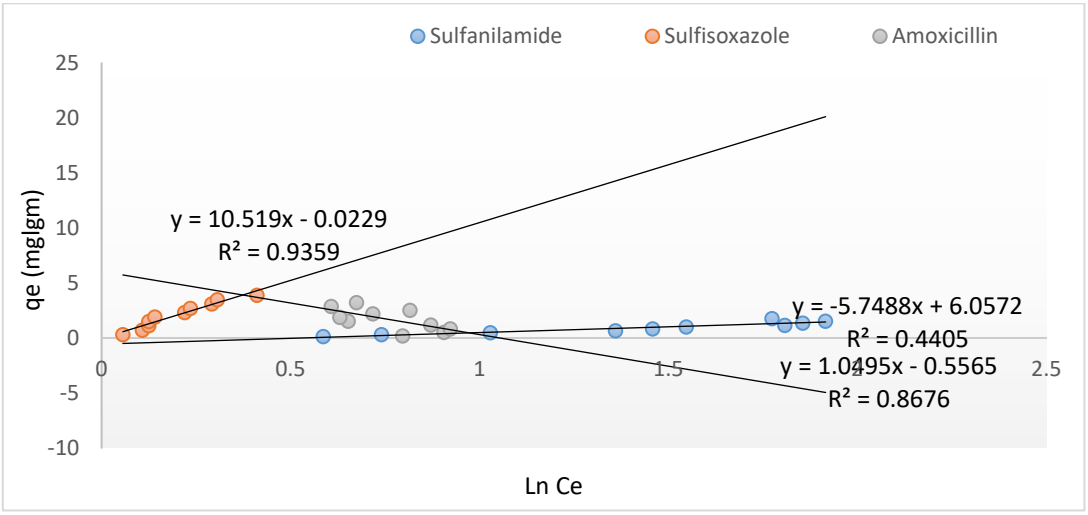

Figure 10. Timken adsorption for different drugs on activated charcoal at pH=7 and Temp. 293K

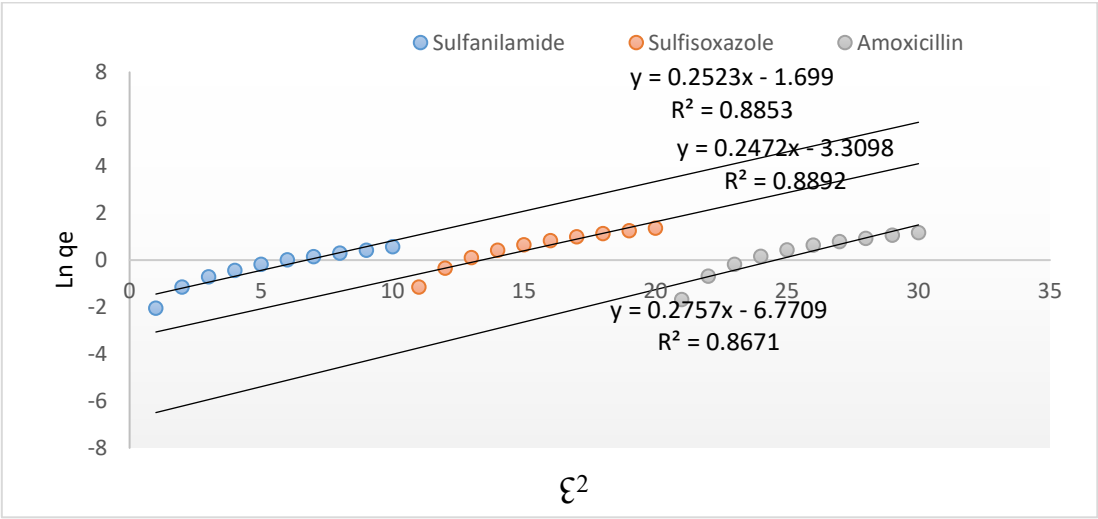

Figure 11. Dubinin adsorption for different drugs on activated charcoal at 0.25gm, Time=1h, pH=7 and Temp. 293K

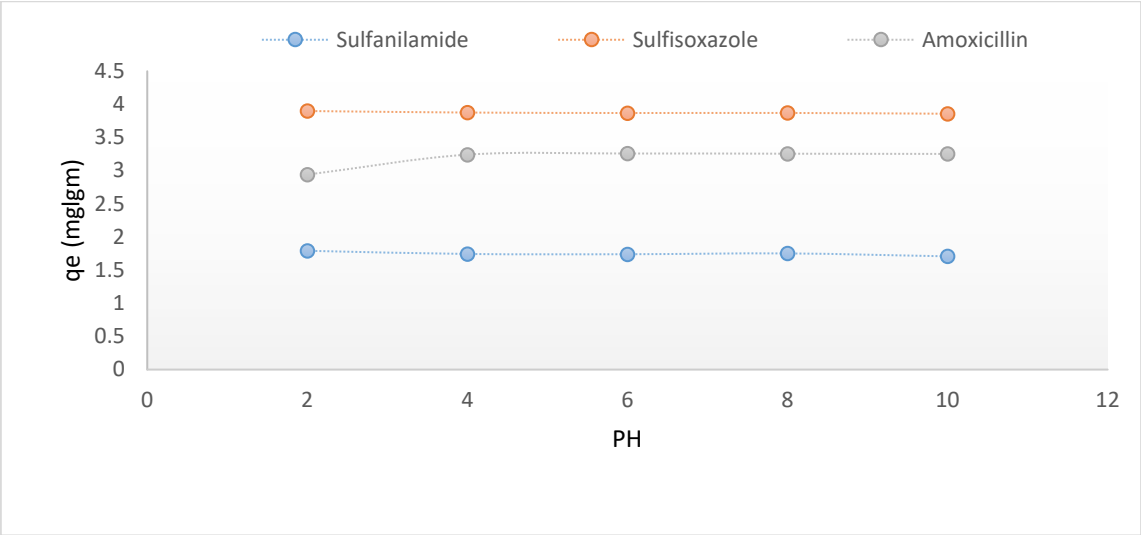

Figure 12. Effect of pH for different drugs on activated charcoal at Temp. 293K ,

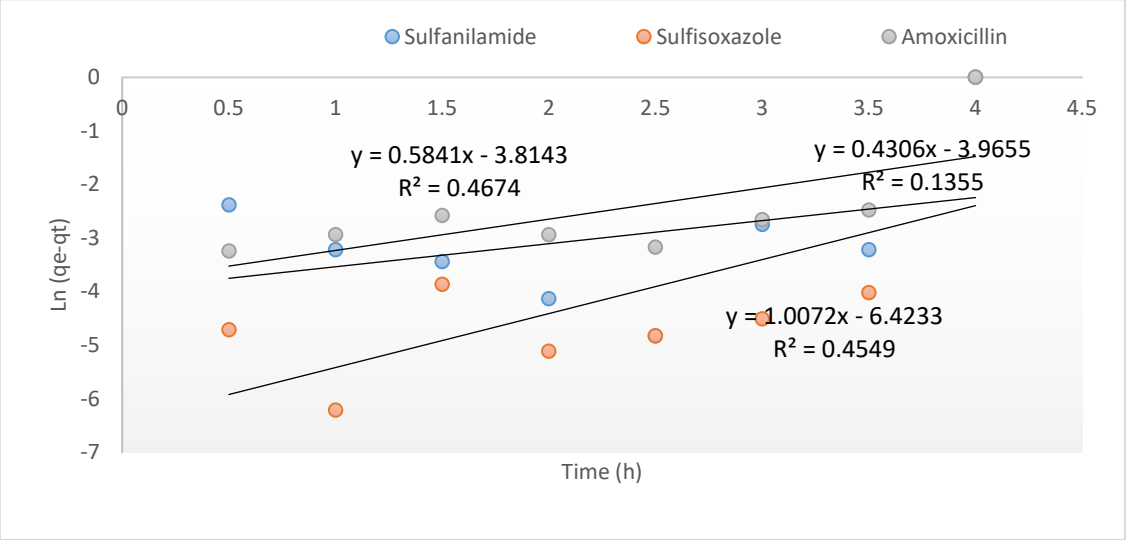

Figure 13. First order adsorption of three drugs on activated charcoal derived from Iraqi date pits in pH=7,at 50ppm,Temp 298 K

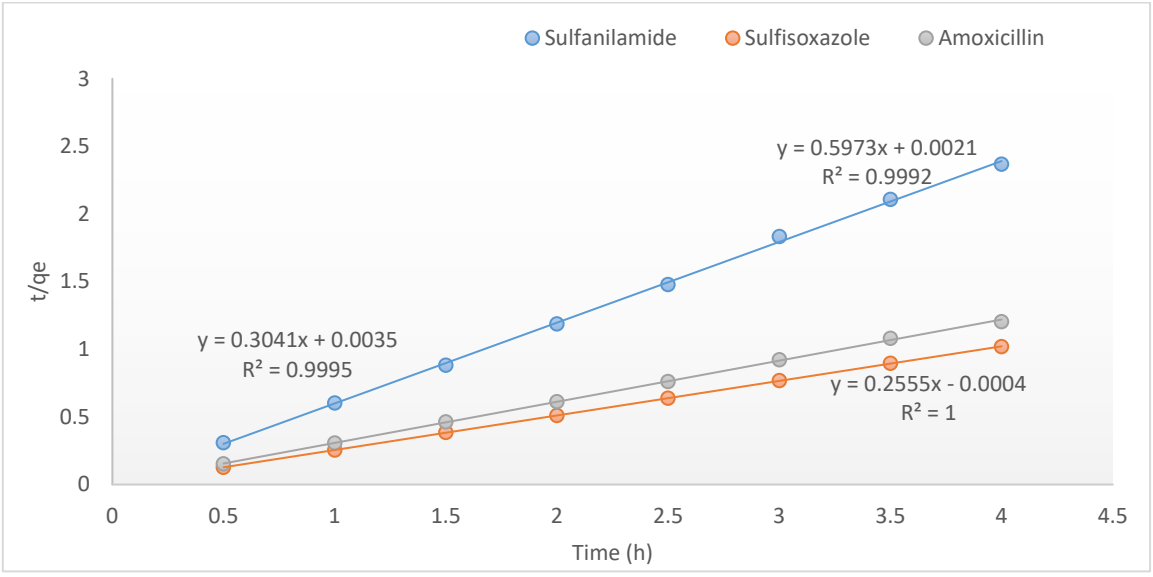

Figure 14. second- order adsorption of three drugs on activated charcoal derived from Iraqi date pits in pH=7,Temp 293K.

4. Conclusion

This study investigated the adsorption of three drugs (sulfanilamide, sulfisoxazole, and amoxicillin) onto activated charcoal prepared from Iraqi date pits. Adsorption experiments were carried out under different conditions, including temperature, sorbent dosage, contact time, pH, and initial concentration. Analysis of the equilibrium data using the Freundlich, Langmuir, Timken, and Dubinin–Radushkevich (D–R) isotherm models revealed that the adsorption equilibrium times for the drugs were (3.5, 1, and 0.5) hours, respectively. The optimum pH for adsorption of all three drugs was found to between 2, 4 .Kinetic studies indicated that the adsorption process followed the pseudo-second-order model, confirming that the adsorption is spontaneous and exothermic. Overall, the findings demonstrate that activated charcoal derived from Iraqi date pits is an efficient and promising adsorbent for the removal of these pharmaceutical compounds from aqueous solutions.

Conflict of interest

The authors declare no conflict of interest

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