

Advanced Journal of Chemistry, Section A

journal homepage: www.ajchem-a.com



Original Research Article

Highly Efficient Adsorption of Pharmaceutical Compounds on Sunflower Seed Shell Derived Porous Carbon

Aseel M. Aljeboree ¹, Ola Hamad Salah ², Usama S. Altimari ³, Ammar Ali Aljanabi ⁴, Ayad F. Alkaim ¹*¹

¹ Department of Chemistry, College of Sciences for Women, University of Babylon, Hilla, Iraq

² Department of Medical Laboratories Technology, Al-Manara College for Medical Sciences, Maysan, Iraq

³ Department of Medical Laboratories Technology, Al-Nisour University College, Baghdad, Iraq

⁴ Department of Medical Laboratories Technology, Al-Hadi University College, Baghdad, Iraq

ARTICLEINFO

Article history

Submitted: 26 November 2023 Revised: 22 January 2024 Accepted: 03 February 2024 Available online: 05 February 2024

Manuscript ID: AJCA-2401-1473 Checked for Plagiarism: **Yes** Language editor: Dr. Fatimah Ramezani Editor who approved publication: Dr. Sami Sajjadifar

DOI: 10.48309/AJCA.2024.434157.1473

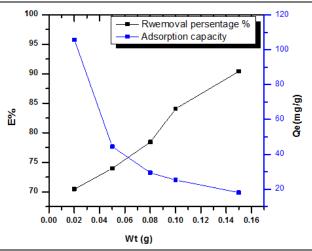
KEYWORDS

Adsorption Paracetamol drug Isotherm Thermodynamic spontaneous

A B S T R A C T

The adsorption capacity of a low-cost biosorbent, Sunflower Seed Shells (SFS) as activated carbon was prepared treatment by hydrochloric acid (HCl) and sodium hydroxide (NaOH) of 0.1 N, was investigated for removal paracetamol drug, several analyses of AC-SFS surface, like FESEM, TEM, and EDX have been done. The effect of some optimum conditions such as concentration of Paracetamol drug and equilibrium time, weight of AC-SFS, and temperature solution were investigated in the batch model. As the equilibrium time increases, the absorption efficiency increases. Likewise, the removal percentage increases and the increased weight of AC-SFS leads to an increase in the removal percentage, while the adsorption efficiency decreases. Finally, with increasing temperature, the adsorption efficiency and removal percentage increase, as a result the reaction is endothermic and spontaneous. Adsorption equilibrium studies were examined by Langmuir, Freundlich isotherm models. Freundlich model fitted the result best with an AC-SFS efficiency (Q_e = 46.567 mg/g). The thermodynamic factors presses were calculated as the maximum ΔG = -3.015 kJ/mol, ΔS = 24.47 J/mol, and ΔH = 2.222 kJ/mol, the adsorption method of the studied drug was spontaneous and endothermic. These data revealed that the use of sunflower seed shells as activated carbon to remove drugs would be an interesting option from both economic and environmental.

G R A P H I C A L A B S T R A C T



* Corresponding author: Alkaim, Ayad F.

E-mail: alkaimayad@gmail.com

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Introduction

Water contaminated with the most dangerous inorganic and organic materials, medicines, dyes, heavy metals, and radioactive materials. The wastewater generated from industrial and household waste has a significant impact on the occurrence of damage to the environment and negatively affects the lives of humans, animals, and plants. The main sources of drinking water pollution in the country are manufacturing and agriculture, as well as other environmental and global changes, where many contaminants were found [1-4]. Pharmaceuticals polluting water resources are considered pollutants resulting from very dangerous drugs because of their carcinogenic nature and have many side effects. Different methods of water treatment having wastewaters have been reported which include electrochemical method filtration, biological treatment, chemical oxidation, coagulationflocculation, electrocoagulation, flocculation, sedimentation, oxidation, and adsorption. Among these ways, adsorption has been widely used for waste water treatment. Adsorption has the ability to elimination may pollutant from aqueous solution which is difficult to break down via known biological method [5-7]. Adsorption on to activated carbon (AC) prepared from agricultural wastes is proven to be best effective removal pollutants wastes. Thus in view of the low cost and low problems for reused, like kind of adsorbents have coir pith, coconut husk, pomegranate peels, peanut hull, garlic, rice shell, kolanut shell, orang shell, bean, and bagasse. In the work, low-cost and eco-friendly agricultural wastes (sunflower seed shells) was utilized to choose best bio sorbent for the removal of drug from aqueous solutions. Adsorption study was carried out with native, acid-treated, and basetreated biosorbent [2,5-6,8-17].

Paracetamol is a name parahydroxyacetanilide or another name acetaminophen and communal kind names like Panadol and Tylenol, Paracetamol is a widely used medicine that is used to antipyretic agent, a non-opioid analgesic to treat fever, treat colds, colds, flu, and high temperature (fever) [18,19], and to reduce pain from sprains, headaches, and toothaches, its chemical formula is $C_8H_9NO_2$ with density of 1.263 g/cm³, molar mass 151.165 g.mol⁻¹, and chemical structure of Paracetamol drug, as displayed in Figure 1 [20].

In the work, treated sunflower seed shells (AC-SFS) as activated carbon was evaluated as adsorbent for elimination of drug. Several parameters like effects of pH solution, equilibrium time, drug concentration, and solution temperature were studied. Equilibrium result was analyzed via two isotherm models. Furthermore, thermodynamic analysis was carried out by the adsorption method.

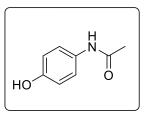


Figure 1. Chemical structure of Paracetamol drug.

Experimental

Preparation of sunflower seed shells

Sunflower seed shells (SFS) were obtained from a local market, washed to remove impurity and air dried at 25 °C for 24 hrs. The effect of treatment by acid hydrochloric acid (HCl) and base sodium hydroxide (NaOH) of 0.1 N was studied utilizing the 10 g Sunflower seed shells (SFS) to reach the pH solution of the acid and base 3.3 and 11.2, respectively, and washed several time distilled water.

The pH of the acid solution treated (SFS) was then adjusted to pH 6.8 with drops of NaOH 0.1 N while the base solution treated SSH was adjusted to pH 6.6 with drops of HCl 0.1 N. The AC-SFS was dried in the oven at 70 $^{\circ}$ C for 12 hrs to obtain constant weight. The AC-SFS was carbonized in a furnace under N₂ gas (98.98%) until 300 °C and for 3 hrs. The AC-SFS was cooled, washed, and dried in the oven. The AC-SFS are stored in a bottle and used in all experiments.

Adsorption study

Stoke solutions (0.1 g/1000 mL) of Paracetamol drug were prepared, and series concentrations of Paracetamol drug was made via dilutions with DW. The initial drug concentrations (10-100 mg/L), The influence of initial pH (3-10) of solution Paracetamol drug adsorption via AC-SFS was studied, and several weights of AC-SFS (0.02-0.15 g), of 40 mg/L, at 25 °C. Equilibrium adsorption experiment was undertaken as follows: 40 mg/L of 100 mL ranitidine drug was mixed by AC-SFS about 0.08 g, solution pH=5.5 at 25 °C for 60 min. Finally, the solution was separated in centrifuges, and then analyzed for residual concentration of Paracetamol drug by

UV-Visible spectrophotometer at the maximum wavelength of λ_{max} = 272 nm. The adsorption capacity and removal percentage were calculated in Equations (1) and (2):

Removal Percentage
$$E\% = \frac{c_o - c_e}{c_o} \times 100$$
 (1)

Adsorption capacity
$$Qe = \frac{(Co-Ce)Vml}{Wcm}$$
 (2)

Results and discussion

Characterization of adsorbent / adsorbate

Felled emission Scanning electron microscopy (FESEM) technology was used as an important method to characterize the surface morphology and determine the important physical properties of the adsorbent. The FESEM of the adsorbed surface of the preparation was taken before and after the drug was absorbed onto the AC-SFS surface. According to Figure 2a, there is a clear possibility that the drug contaminant will be trapped and absorbed within these pores.

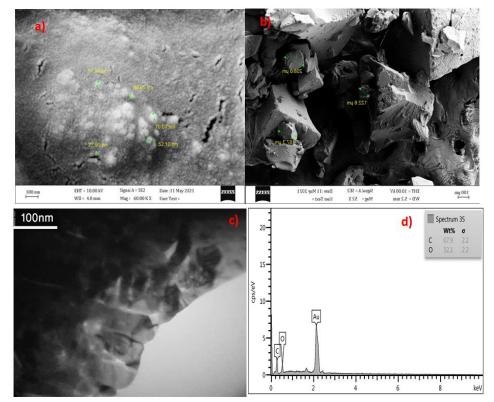


Figure 2. FE-SEM image of a) AC-SFS before adsorption, and b) after adsorption of Paracetamol drug, and c and d) E-DX of AC-SFS.

FESEM images of the surface after the adsorption process show very distinct dark spots that confirm the good adsorption of the drug molecules into the pores and cavities of the adsorbent, as shown in Figure 2 (a and b).

The transmission electron microscope (TEM) technique has a crucial and important role in controlling the properties and surface morphology and controlling the surface properties. The TEM is depicted in Figure 2c of AC-SFS consist of background bulk color attributed to AC, the average diameter of darker is 100 nm. TEM also shows the particles dispersion on the surface and there is also a little noticeable aggregation of these in the form of dark aggregates, resulting from the acid activation process. EDX is a versatile technique used for qualitative and semi-quantitative analysis, Figure 2d demosntrates the EDX patterns of AC-SFS, which verified the existence of C and O in AC [21,22].

Effect of weight AC-SFS

To study the effect of adsorbent dosage AC-SFS (0.08 g) on Paracetamol drug adsorption, experiments were conducted at concentration of Paracetamol drug 40 mg/L, whereas the amount of mass of AC-SFS additional illsutrated in Figure 3. The influence of mass of AC-SFS on the percentage removal% of Paracetamol drug [23]. It was experiential that the percentage E% of Paracetamol drug fast increased with the increase in mass of AC-SFS equal to 0.08 g, slowly increased with further rise in mass equal to 0.15 g, and then remained low changed. At equilibrium time, the percentage E% increases from 70.44-90.66% for an increase in mass of AC-SFS from 0.02-0.15 g. The increase in E% removal was due to the increase in the obtainable sorption surface [24, 25].

Effect of pH

One of the maximum significant factors affecting adsorption is the solution pH. Paracetamol drug onto AC-SFS range pH 2-10, as inidicated in Figure 3. This can be attributed to that the electrostatic attraction is not the only mechanism for the removal of AC-SFS some other mechanism might be operating for binding. On the other aspect, the presence of more number of active sites on the surface of AC-SFS may have lessened the competitive adsorption among H⁺ and AC-SFS ions resulting in a nil effect of pH at low primary concentrations. Therefore, extra experiments wear performed at solution pH 8 [26, 27].

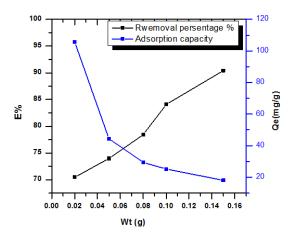


Figure 3. Effect of weight AC-SFS on the adsorption efficiency and removal percentage of Paracetamol drug.

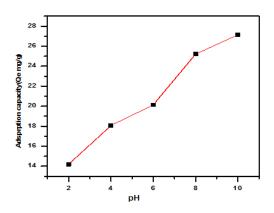


Figure 4. Effect of pH solution onto adsorption capacity by use AC-SFS.

	ricument of Sumower Seeu Shens		170		
	Hydrochloric acid		84.49		
	Sodium hydroxide		77.77		
	Table 2. Thermodynamic factors values for Paracetamol drug				
ΔH (kJ/mol)	ΔG (kJ/mol)	ΔS (kJ/K.mol)	Equilibriu	m Constant (K)	
2.22483	-2.986	24.47558	:	3.559	
	-3.015		3	3.4477	
	-3.055			3.363	

Table 1. Effect of treatment on the activity of sunflower seed shells for the adsorption Paracetamol drug

Treatment of Sunflower Seed Shells

Effect of treatment on the adsorbent surfaces

The study of the influence of acid and base treatment was essential to show the best adsorption. The sunflower seed shells were treated via hydrochloric acid and sodium hydroxide, as presented in Table 1.

Effect of temperatures and thermodynamic process

Thermodynamics parameters were studied at several temperatures rang (25-40 °C) and with several concentrations within the series (10-100 mg/L) for Paracetamol drug, where the adsorption process is greatly affected via solution temperatures, the change in solution temperature leads to an increase or decrease in the ability of the adsorbent surface to adsorption Paracetamol drug from aqueous solutions [23].

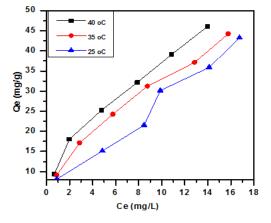


Figure 5. The influence of temperature on Paracetamol drug adsorption on the AC-SFS.

The data appear decrease in the amount of Paracetamol drug adsorbed drug with increase solution temperatures, as appeared in Figure 5, the adsorption method is an exothermic process where higher temperatures increase the solubility of the adsorbed drug particles.

E%

This leads to a decrease in the affinity of the adsorbed drug particles towards the adsorbent surface. Furthermore, increase the solution temperature leads to an increase in the entropy [28].

Table 2 lists the values of thermodynamic factors for the adsorption method of Paracetamol drug, among which the negative value of the enthalpy indicates that the Paracetamol drug adsorption method is an exothermic process, which indicates that the mutual action between the adsorbing surface and drug molecules will decrease With the increase in temperature, the reason for this is due to the separation and breaking of the bonds formed between the dye molecules and the active centers of the adsorbing surface [29,30].

Equilibrium isotherm

Adsorption equilibrium isotherm two models Langmuir and Freundlich equilibrium isotherms were applied to define the equilibrium isotherm of experimental data for the sorption of Paracetamol drug onto AC-SFS.

Isotherm Langmuir model was applied to determine the best adsorption capacity (Q_{max})

conforming to wholly monolayer coverage on the adsorbent surface [31]. A plot of adsorption capacity vs. concentration equilibrium was utilized to find isotherm Langmuir model. The $Q_{max}(mg/g)$ and K_L values were estimated from the slope and intercept. The non-linear Langmuir isotherm is described in Equation (3):

$$Q_{e} = \frac{Q_{m} K_{L} C_{e}}{1 + K_{L} C_{e}}$$
(3)

The isotherm Freundlich model was applied in heterogeneous surface adsorption and The Freundlich isotherm non-linear is provided in Equation (4):

$$Q_e = k_f C_e^{\bar{n}}$$
(4)

Where, KF (mg.g⁻¹) is the adsorption capacity and n is the intensity of adsorption. therefore, a plot of adsorption capacity (Q_{max}) *vs.* equilibrium concentration is a straight non-linear and values of K_F and n are calculated from the intercept and slope of the non-linear plot. The values of K_F and (R²) found from Freundlich model, as shown in Figure 6 and Table 3. The drug adsorption is good fitted to Freundlich isotherm model with the best R²= 0.99178.

Base on the comparison of Langmuir and Freundlich isotherm models and the obtained data, the Freundlich isotherm has exhibited the best fitness to the adsorption data than Langmuir isotherm [31,32].

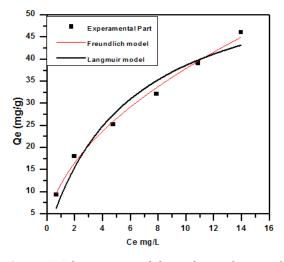


Figure 6. Adsorption models nonlinear fitness of adsorption drug onto AC-SFS, con. 40 mg.L⁻¹, mass 0.08 g, and Temperature= 25 °C).

Comparative two surface

Comparative between Sunflower Seed Shells (SFS) as nuchal without treatment and treated Sunflower Seed Shells (AC-SFS) to have ability removal drug from aqueous solution. According to Table 4, Sunflower Seed Shells (AC-SFS) as activated carbon has a very high efficiency in

<i>,</i>	•	0
Models	Parameter	AC-SFS
	K _F	11.596
Freundlich	1/n	0.513
Freununch	R ²	0.99178
	q _m (mg/g)	46.406
Langmuir	K _L (L/mg)	0.1689
	R ²	0.9234

Table 3. Langmuir and Freundlich isotherm model of parameter drug adsorbed onto AC-SFS

Absorbent	Pollutant	Removal Percentage %
AC-SFS	Paracetamol drug	84.49
AC-SFS	DY dye	89.44
SFS	Paracetamol drug	52.22
SFS	DY dye	63.33

removing the drug from aqueous solution at a higher percentage than from the Sunflower Seed Shells (SFS) without treatment. This is because activated carbon contains many active sites, which help it remove pollutants and also increase the surface area compared to non-activated plants [12,18].

Conclusion

In the present study, the results reveal that the both Sunflower Seed Shells (SFS) and treated Sunflower Seed Shells (AC-SFS) have ability to removal drug from aqueous solution; the Sunflower Seed Shells was treated via hydrochloric acid and sodium hydroxide. It was found the percentage removal increased with increase solution pH and rise with increase in weight of adsorbent and temperature solution, but decrease with rise drug concentration.

Where the experimental data fit Freundlich model which indicate to multilayer adsorption, thermodynamic factors indicated to adsorption method was spontaneous and endothermic process. Sunflower Seed Shells (AC-SFS) as activated carbon has a very high efficiency in removing the drug from aqueous solution at a higher percentage than from the Sunflower Seed Shells (SFS) without treatment.

Orcid

Aseel M. Aljeboree : 0000-0001-5397-3330 Ola Hamad Salah: 0000-0002-3194-1749 Usama S. Altimari : 0000-0002-7485-5875 Ammar Ali Aljanabi : 0000-0002-8748-3353 Ayad F Alkaim : 0000-0003-3459-4583

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HOW TO CITE THIS ARTICLE

Aseel M. Aljeboree, Ola Hamad Salah, Usama S. Altimari, Ammar Ali Aljanabi, Ayad F. Alkaim^{*}. Highly Efficient Adsorption of Pharmaceutical Compounds on Sunflower Seed Shell Derived Porous Carbon. *Adv. J. Chem. A*, 2024, 7(3), 295-302.

DOI: 10.48309/AJCA.2024.434157.1473 URL: https://www.ajchem-a.com/article 189783.html