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Original Research Article

Adsorption of Amoxillin Drug onto Activated Carbon Prepared from Cashew Nut shell by H_3PO_4 Activation: Studies on Equilibrium Isotherm by Nonlinear Equations

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ABSTRACT

Activated carbon (AC) as a source of normal plants (Cashew Nut Shell) were utilizing as the best, ecological and cost-effective adsorbent for removal of contaminants of Amoxillin (AMX) drug from aqueous solutions via sonicated. Adsorption thermodynamics and equilibrium isotherm of AMX drug onto AC-CNS. The data indicate that AC-CNS can be used as a low-cost substitute compared with commercial adsorbents removal of pollutants from aqueous solution. The effects of pH solution (2-10), initial concentration of AMX (10-100 mg/L), solution temperature (15-35 °C) and adsorbent dosage (0.01-0.1 g) on adsorption were evaluated. The structural and morphological properties of the AC-CNS were analyzed including FESEM, TEM and EDX. The non-linear model was conducted to determine thermodynamic parameters and equilibrium isotherms, where the result of equilibrium isotherm was fitted onto the Freundlich model. Freundlich suggested a determined multilayer adsorption efficiency (109.66 mg/g). The nonlinear models of thermodynamic factors appear to show that the adsorption was spontaneous (negative value of ΔG and positive value of ΔS) and endothermic (positive value of ΔH) at the same optimum conditions.

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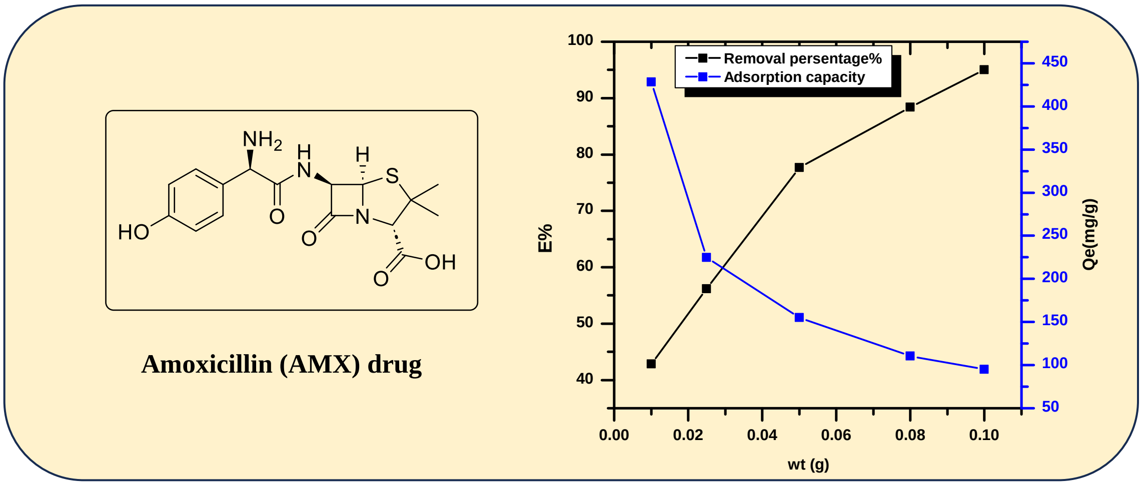
GRAPHICALABSTRACT



Introduction

Pollution of the aquatic environment is considered one of the most serious problems faced by the industrialized countries of the world due to the discharge of untreated industrial waste products that have the outputs of some dangerous chemicals that are difficult to get rid of industrially, such as organic and inorganic compounds, pharmaceuticals, drug, phenols, dyes, fertilizers dyes and pesticides. It is discharged to water sources and causes an environmental problem that threatens the life of all living organisms.

In recent years, several methods have been counted for coagulation, including reverse osmosis, biological treatments, chemical oxidation, membrane separation, flocculation, oxidative processes, electrocoagulation son catalytic degradation, membrane separation, photo degradation and adsorption [1-5]. In addition, adsorption method of drug to become economically feasible, researchers have focused on biodegradable, renewable, sustainable, simply available and ecofriendly adsorbents, like kaolin, sawdust, guava seeds, bagasse fly ash, bagasse pith, rice hull ash, palm kernel fibre, maize cob, bentonite, Organo-attapulgite, fungi, Azadirachta indicia leaf powder, [6] nucleus, Salient mud, cashew nut shell, or acorn-based, materials, clay materials, rice husk, tomato stem, date stones, biomass, zeolite and polymers. Activated carbon (AC) is considered one of the most important absorbent materials used to effectively remove contaminants, including pharmaceutical preparations, from aqueous solutions. Activated carbon, or activated charcoal, also called carob activated, or activated charcoal, is considered as a form of treated carbon that produces a small percentage of carbon, with small pore sizes and has a high surface area, thus increasing the efficiency in removing pollutants [7-18].

Amoxicillin (AMX) drug is an antibiotic belonging to class of amino penicillin of the family penicillin. The AMX is utilized to treat bacterial infections like strep throat, pneumonia, middle ear infection, odontogenic infections, skin infections and urinary tract infections. taken via mouth, or injection, its chemical formula is $C_{16}H_{19}N_3O_5S$, molar mass 365.40 $g.mol^{-1}$ and chemical stretcher [19,20], as displayed in Figure 1.

In this study, activated carbon was prepared from Cashew Nut Shell (AC-CNS) to adsorption/removal of the AMX drug from aqueous solution. Several factors affecting the adsorption process were studied, for example the weight of AC-CNS, drug concentration, pH, temperature and also the thermodynamic parameters and adsorption isotherms were studied.

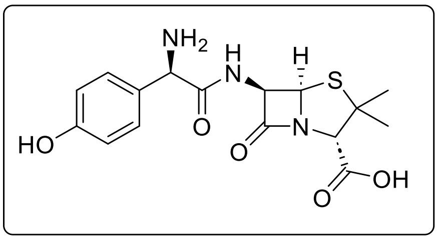


Figure 1. Chemical structure of Amoxicillin (AMX) drug

Experimental

Preparation of cashew nut shells as activated carbon

Cashew Nut Shells (CNS) were collected from the local market in Hillah, Iraq and were utilized as adsorbent and then the shells were washed in deionized distilled water for 3 hrs to get rid of unwanted substances and remove all colored extracts. Next, the Shells were dried in an oven at 105 °C for 3 hrs. After that, the shells dried, grounded and sieved using a particle size 50 nm sieve to obtain a powder. These shells are treated using H_3PO_4 (0.01 N) and heated in furnace at 300 °C for 2 h under nitrogen, they are also dried in the oven at a temperature of 65 °C for 24 h and the shells treated are stored in a bottle and used in adsorption studies.

Adsorption study

Standard solutions (1g /1000 mL) of AMX drug were prepared and range concentrations of AMX drug was made via dilutions with DW.

The initial drug concentrations (10- 100 mg/L), The influence of initial pH 2-10 of solution AMX drug adsorption via AC-CNS was studied and several weight of AC-CNS (0.01-0.1 g), of 100 mg /L, at 25 °C. The equilibrium adsorption experiment was undertaken as follows: 100 mg/L of 100 mL ranitidine AMX was mixed by AC-CNS about 0.08 g, solution pH=10 at 25 °C for 60 min. Finally, solution was separated in centrifuged and then analyzed for residual concentration of AMX 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):

$$Removal\ Percentage\ E\% = \frac{Co-Ce}{Co} \times 100 \qquad (1)$$

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

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-CNS surface. According to Figure 2a, there is a clear possibility that the drug contaminant will be trapped and absorbed within these pores. In Figure 2b, 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 [13,21].

The transmission electron microscope (TEM) technique has a crucial and important role in controlling the properties and surface morphology and controlling the surface properties.

TEM image of AC-CNS is demonstrated in Figure 2c consist of background bulk color attributed to AC, the average diameter of darker is 100 nm. TEM also shows the dispersion of particles on the surface and there is also a little noticeable aggregation of these in the form of dark aggregates, resulting from the acid activation

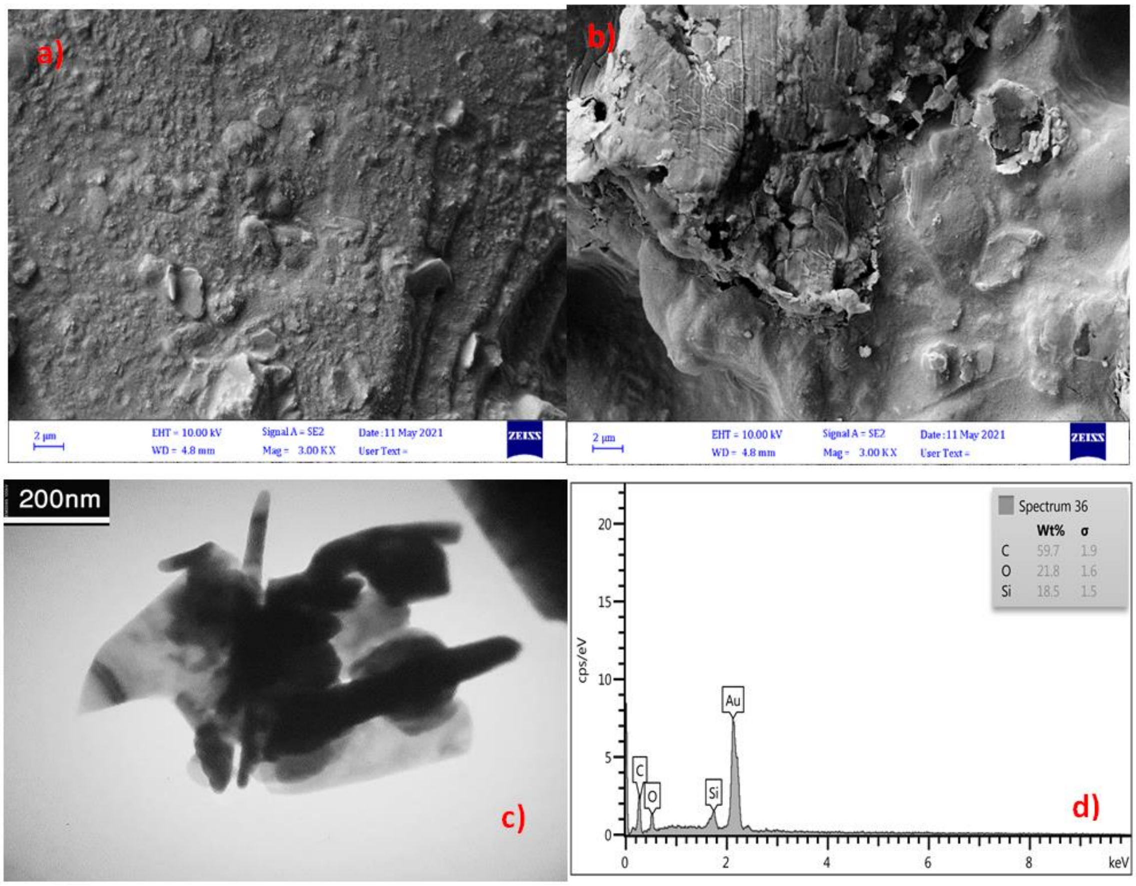


Figure 2. FE-SEM image of (a) AC-CNS before adsorption, (b) after adsorption of Paracetamol drug, (c) TEM image of AC-CNS and (d) EDX of AC-CNS.

process. EDX is a versatile technique used for qualitative and semi-quantitative analysis, Figure 2d shows the EDX patterns of AC-CNS, which verified the existence of C and O in AC. And As for the Si element, it comes from a plant source that contains silica [22-24].

Effect of weight of AC-CNS

Effect of mass of AC-CNS as a very significant parameter was studied when conducting batch mode studies. The effect of mass of AC-CNS on the removal percentage E% of AMX drug was studied via different masses of AC-CNS from 0.01-0.1 g. The top adsorption efficiency of AMX drug was experiential at 0.08 g of AC-CNS. However, the AC-CNS mass was increased due to rise in adsorption because of the ability of extra number of active sites on the surface; the better percentage E% was 88.33% [25-27]. In contrast, the adsorption capacity decreases with increasing mass AC-CNS, where the better adsorption capacity was 110.31 mg/g, as demonstrated in Figure 3.

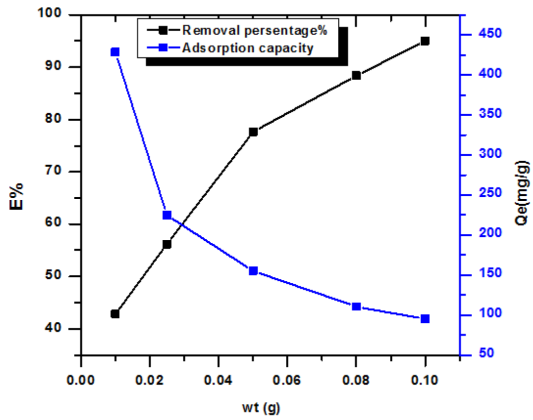


Figure 3. Effect of weight of AC-CNS on to removal percentage and adsorption capacity.

Effect of solution pH

The effect of solution pH was examined on the Uptake of AMX drug onto (AC-CNS). The solution pH significantly influences the adsorption ways. The nature of the adsorbate in solution as well as the charge on the adsorbent surface is dependent solution pH. The effects of pH on the adsorption of AMX drug onto (AC-CNS) was investigated within the pH range 2-10, the pH solution was adjusted using HCl or NaOH 0.1 N. When the pH solution increased from (2-10), adsorption efficiency increase from 28.53- 110.43 mg/g. Through data, the better removal AMX drug in pH=10 depends on the drug and charge of the reactive group inside the (AC-CNS) [25,28-29] . In other words, a competition happens among the AMX drug ions (OH-) with the (H^+) ions present in the solution and the better absorption of drug in pH 10, as appeared in Figure 4.

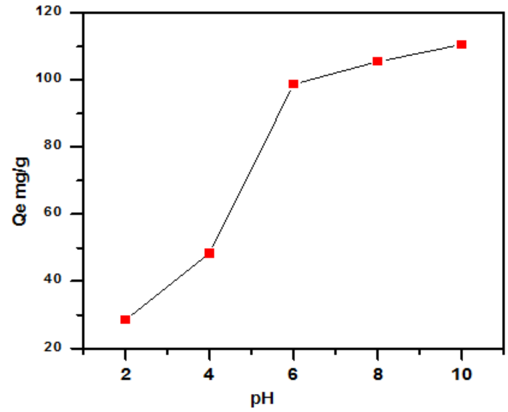


Figure 4. Effect of pH solution onto removal percentage and adsorption capacity.

Effect of temperatures and thermodynamic process

Thermodynamics factors were studied at different temperatures 15-35 ˚C and with deferent concentrations (10-100 mg/L) for AMX drug, where the adsorption method is greatly affected by solution temperatures, the variation in solution temperature leads to an increase or decrease in the ability of the surface to adsorption AMX drug from aqueous solutions [30].

The result show decrease in the quantity of AMX drug adsorbed drug with increase solution temperatures (Figure 5), the adsorption way is an endothermic process where higher temperatures increase the solubility of the adsorbed drug particles. This leads to an increase in the affinity of the adsorbed drug particles towards the adsorbent surface. Furthermore, increase the solution temperature leads to an increase in the entropy [31,32].

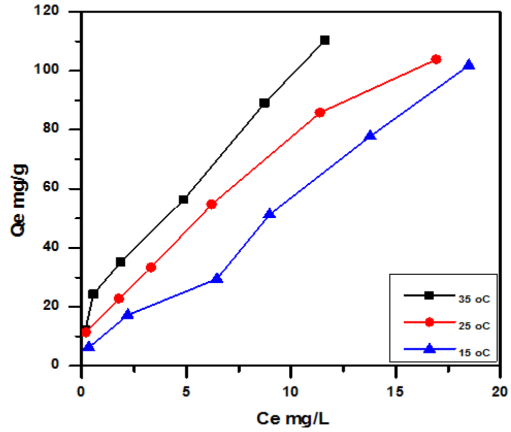


Figure 5. The influence of temperature on Paracetamol drug adsorption on the AC-SFS.

As presented in Table 1, the thermodynamic parameter for the adsorption process of AMX drug has a negative value of the enthalpy indicating that the AMX drug adsorption method is an endothermic process, which demonstrates that the mutual action between the adsorbing surface and drug molecules will increase with the increase in temperature, the reason for this is due to the separation and breaking of the bonds formed between the drug molecules and the active centers of the adsorbing surface [33].

Adsorption model

Two adsorption isotherm models including Langmuir and Freundlich equilibrium adsorption isotherms were utilized to define the

Table 1. Thermodynamic factors values for AMX drug

ΔH (kJ/mol)	ΔG (kJ/mol)	ΔS (kJ/K.mol)	Equilibrium Constant (K)
	-5.3884		**9.8768**
3.461	-5.4216	27.1618	**9.2544**
	-5.5334		**8.9945**

equilibrium isotherm of experimental result for the sorption of AMX drug onto AC-CNS [30]. Langmuir isotherm Model was used to determine the better adsorption capacity (Q_{max}) conforming to all coverage monolayer on the adsorbent surface. A plot of Q_e(mg/g) *vs.* C_e(mg/L) was used to find model Langmuir isotherm. The q_{max} (mg.g^{-1}) and KL values were estimated from the slope and intercept. The nonlinear Langmuir isotherm is given in Equation (3) [34,35]:

$$Q_e = \frac{Q_m\, K_L\, C_e}{1+K_L\, C_e} \quad (3)$$

The Freundlich model was applicable in heterogeneous surface adsorption and the Freundlich nonlinear model is calculated in Equation (4):

$$Q_e = k_f\, C_e^{\frac{1}{n}} \quad (4)$$

Where, K_F (mg.g^{-1}) is the adsorption efficacy and n is the intensity of adsorption. Thus, a plot of Q_e vs. C_e is a straight non-linear and K_F value, n is calculated from the intercept and slope of the non-linear plot.

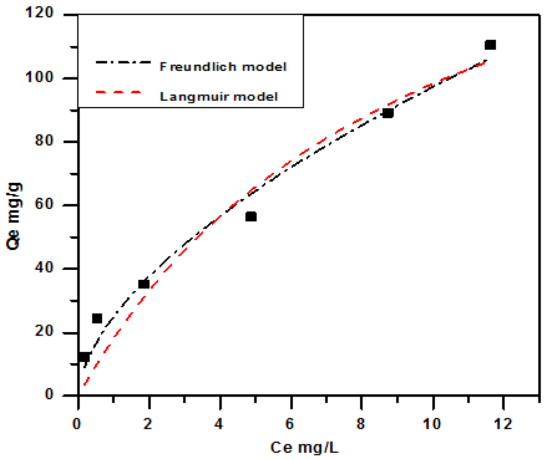


Figure 6. Adsorption models nonlinear fit of adsorption AMX drug onto AC-CNS, con. 100 mg.L^{-1}, mass 0.08 g at 25 °C).

Table 2. Langmuir and Freundlich isotherm parameter model of AMX drug adsorbed onto AC-CNS

Models	Parameter	AC-CNS
Freundlich	K_F	24.976
	1/n	0.5321
	R^2	0.9787
Langmuir	q_m (mg/g)	193.54
	K_L(L/mg)	0.1033
	R^2	0.9122

The values of R^2 and, K_F a found in Freundlich model, as shown in Figure 6 and Table 2. The adsorption of AMX drug best fitted in Freundlich model with the better R^2= 0.9787 base on the comparison in Langmuir model, the Freundlich model has exhibited the best fitness to the adsorption result than Langmuir isotherm model [36].

Conclusion

Activated carbon obtained from Cashew Nut Shell as a low-cost and eco-friendly activation utilizing H_3PO_4 (0.01N) showed a higher surface area when the initial solution pH of the drug increased, adsorption capacity increased from 28.45-110.43 mg/g t at 25 °C and the best adsorption capacity at pH 10. But, the increase in mass of AC-CNS is due to a rise in adsorption because of the extra number of active sites on the surface, the better percentage E% (88.33%). Equilibrium adsorption is described via Freundlich and Langmuir model isotherms, as well as the adsorption capacity, the adsorption of AMX drug was best fitted in Freundlich model with the better R^2= 0.9787 based on the comparison in model Langmuir. The results of thermodynamic parameter study revealed that the adsorption method was spontaneous, endothermic and governed by physisorption.

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