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Adsorption Thermodynamics of a Ciprofloxacin Drug from Aqueous Solutions on [Graphene Oxide/ poly Acrylic Acid /Microcrystalline Cellulose] Composite

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ABSTRACT

Composite hydrogels [Graphite oxide /poly (acrylic acid – microcrystalline cellulose)] [GO/ (AAC-MCC)] super absorbent composites were synthesized by free radical polymerization of (AAC and MCC) as a monomer, using N, N-methylene bisacryl amide (MBA) as cross-linker and ammonium persulfate (KPS) as initiator. GO/ (AAC-MCC) composite was characterized by (FTIR), (FE-SEM), (AFM), (XRD), and BET (Brunauer–Emmett–Teller) specific surface area and pore structure of the adsorbent was also examined. The synthesis composite was used as adsorbent for removal of a ciprofloxacin drug from aqueous solution. we obtained data about thermodynamic, we applied data on Langmuir, Freundlich and models equation to get it information on adsorption process, the study clarify effect some parameter on quantity of adsorbate such as pH at range (1.2-10), ionic strength by used three types of salts, temperature at rang (10, 15, 20 and 25°C)., contact time (1-360 min) and weight of adsorbate at range(0.01-0.1 g), it's found when increase of temperature and ionic strength will decrease of quantity the adsorbate presences under the following optimum condition at temperature 20°C, contact time 120 min, pH = 2. The adsorption isotherm appears that adsorption process applied models Freundlich and Langmuir. Thermodynamic functions process calculated and appeared the adsorption spontaneous and exothermic process, also the synthesized adsorbents showed high efficiency in removal of ciprofloxacin drug from aqueous solution and a very high adsorption capacity.

Keywords: Ciprofloxacin ,Drug, Hydrogel composites, Adsorption, Acrylic acid.

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INTRODUCTION

Hydrogel is a macromolecular polymeric gel synthesized by crosslinking of polymer chains-through physical, ionic or covalent interactions and are well known for their ability to absorb large amount of water nearly 10–20 times its molecular weight via H-bonding and become swollen[1], but remain insoluble due to chemical or physical crosslinking between polymer chains[2] . The ideal hydrogels can be designed to release drugs or other agents in response to physical stimuli like temperature , changes in pH and concentration of added salts[3], Acrylic acid is a typical pH responsive super absorbent that is commercially available. It's considered as pH sensitive material due to the ionic repulsion between its anionic groups and it can be prepared at various concentrations, fabricated into different size and shape and other materials can be incorporated into AAC prior to gel formation[4].

Ciprofloxacin Fig.(1) is an established broad-spectrum fluoroquinolone antibiotic displaying activity against both Gram-negative and Gram-positive bacteria by inhibiting DNA-gyrase and topoisomerase IV present in bacterial cells which are responsible for reproduction of DNA of bacteria , due to its inexpensiveness and its availability, it is used to treat, respiratory, abdominal infections bacterial diarrheal infections urinary tract infections, skin, ophthalmic and respiratory infection[5].

Adsorption is one of the superior physicochemical methods for sewage disposal, which has several advantages of high removal efficiency, lower costs, no chemical sludge and easy accessibility[6, 7],It considered simple, cheap, easily–handle operation, low cost and eco-friendly characters and most versatile technique for holding these pollutants adsorption is widely used in water treatment as it is easily operated with high removal efficiency. Thus, the adsorbent disposal is necessary after adsorption process[8] . This work demonstrated that the [GO/ (AAC-MCC)] super absorbent composites exhibited excellent mechanical strength and rapid drug adsorption capability. It can be expected that [GO/ (AAC-MCC)] super absorbent composites with excellent mechanical properties could play a more important role in adsorption field.

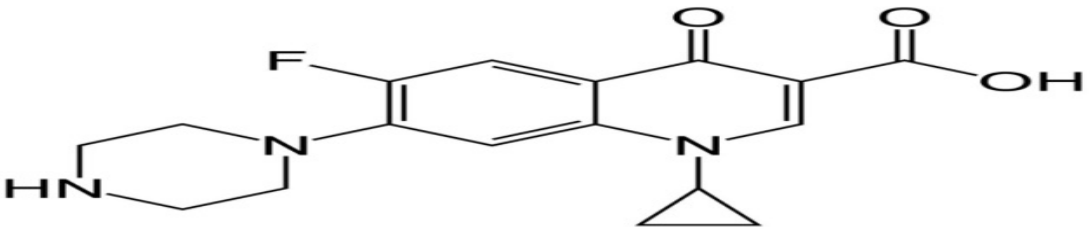

Fig. 1. Ciprofloxacin Drug

MATERIALS AND METHODS

Chemicals and Materials

Natural graphite powders, potassium permanganate ($KMnO_4$), sodium nitrate ($NaNO_3$), concentrated sulfuric acid , hydrogen peroxide (30%), were purchased from Kemiou Chemical Reagent Co, Ltd, China, Acrylic acid AAC and microcrystalline cellulose MCC were supplied by (Himedia), The initiator, potassium persulfate (KPS) and the cross linker is N,N'-methylene Bisacrylamide (MBA) were supplied by (Fluka), Sodium chloride, Carbonate Calcium and Potassium chloride was obtained from(Alpha Chimika), ciprofloxacin. HCl (Cipro)was purchase from (Samaraa company SDA /Iraq), Sodium Hydroxide and Hydrochloric acid were supplied from (B.D.H), All the reagents used were analytical grade pure with no further purification, and all the solutions were prepared with deionized water.

Preparation of adsorbent [GO (AAC) / MCC] Composite

The composite was prepared through the co-polymerization of free radicals in the aqueous solution, which includes dissolving 0.5 gm of MCC in 10 ml of distilled water in the presence of nitrogen gas with stirring for an hour at a temperature of 50°C and then added 5 ml to this solution From acrylic acid AAC and placed in a 500 mL three-neck flask,connected to a condenser and a separating funnel in the presence of nitrogen gas continuously throughout the polymerization process to remove the oxygen, then placed inside the water bath and stirred the mixture until obtaining a homogeneous solution to ensure the MCC suspended uniformly , after cooling the mixture, the crosslinking agent solution (MBA) prepared from dissolving (0.05 g in 1ml of distilled water) was added , then the KPS initiator solution that was prepared by dissolving (0.05 g in 1ml of distilled water) was gradually added to the mixture of reaction , then 1ml of graphene oxide GO prepared from dissolving (5 mg in 1mL of distilled water) by using the Sonication path for a period of 15 min was added , where it was added through a separation funnel to the reaction mixture with continuous stirring and in the presence of nitrogen gas at a temperature of 69°C for a period of 3h Then the polymeric compound of composite was formed, as it was taken and cut into small pieces and then washed in distilled water with continuous stirring for 15h, where the water was replaced every 60 min in order to remove any unreacted monomer, then it was dried in the oven at 60 ° C until a constant weight was obtained.

Characterization of Composite

FTIR infrared spectra analysis

The infrared spectroscopy (FTIR Shimadzu 8400S, Japan) was used to identify functional groups in the prepared composite. FTIR spectroscopy data for the prepared surface were obtained in the frequency range (4000-400) cm^{-1} using the standard KBr pressed pellet method.

Atomic Force Microscope

Atomic force microscopy (AFM) was performed utilizing a Park Systems XE-70 Atomic Force Microscope in non-contact style, this technique was used to study the properties of the external structure of the prepared composite, thickness, and grain size as well as obtaining three-dimensional images of the hydrogel.

X-ray diffraction

X-ray diffraction (XRD) analysis was performed using a D/Max 2550 V diffractometer with Cu Kα radiation (λ = 1.54056 Å) (Rigaku, Tokyo, Japan), and the XRD data were collected at a scanning rate of 0.03°s-1 for 2θ in a range from 10° to 80° in orderTo study the crystalline properties of the prepared hydrogel.

Emission scanning electron microscopy technique FE-SEM

FE-SEM was used as the image was taken after being coated with a thin layer of gold and low pressure, to study the properties of the external surface of the prepared surface.

Surface Area and Porosity Analysis of BET-BJH

To study the surface properties of the prepared hydrogel such as the surface area and pore diameters distribution, the BET isotherms method (adsorption-desorption) and (BJH) method were used to distribute the pore diameters.

Determination of Maximum Absorption (λ_{max}) for Ciprofloxacin Drug

To determine the maximum wavelength of the Cipro drug, the ultraviolet-visible absorption spectra of the drug solution (10 mg/L) was recorded within wavelengths of (200-800) nm by a quartz cell of 1cm thickness, The maximum wavelength of the drug solution was determined from its highest absorption in the UV-Vis spectrum found at the wavelength λmax = 276.0 nm in Fig. (2).

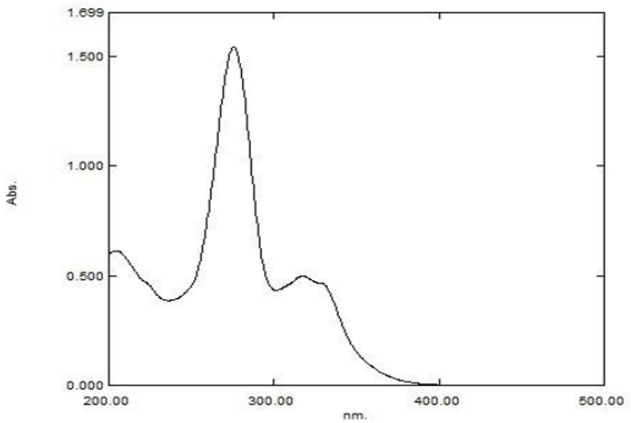

Fig. 2: UV-Visible absorption spectra for Ciprofloxacin

Calibration Curve for Ciprofloxacin

The calibration curve of the Cipro drug was determined by preparing a group of solutions by successive dilution of the standard drug solution and with a range of concentrations between (1 - 40 ppm). The absorbance of these solutions was recorded at the maximum wavelength of the drug (276 nm). As shown in Fig. (3).

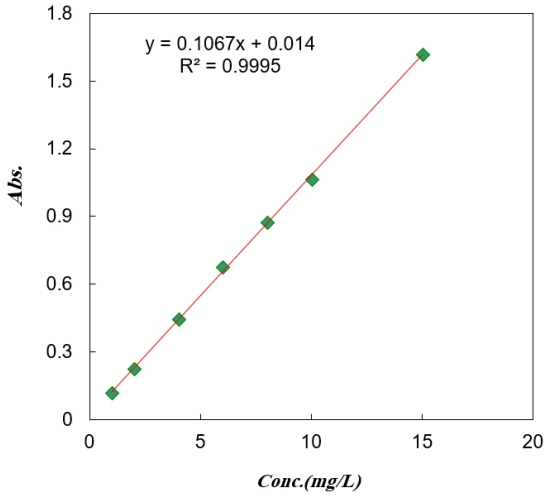

Fig. 3: Calibration curves of the Cipro drug

has been added to different concentrations of drug solution, in the same way as an experiment of adsorption.

Effect of pH on adsorption

The effect of pH on the adsorption process was studied. A weight of (0.05 g) of the composite was taken to add to a solution of Cipro drug with a concentration (100 ppm), at different pH values (1.2-10) where the pH was organized using a solution of 0.1M HCl, and 0.1M NaOH, and a pH value was measured using a pH meter.

Effect of ionic strength

The effect of ionic strength was studied by taking different weights (0.001-0.1 g) from salts (NaCl, KCl and CaCO3), a Cipro drug solution (100 ppm) was added to volumetric flasks containing (0.05 g) of the synthesized composite with the same method of the experiment of adsorption conducted.

RESULTS AND DISCUSSION

FTIR Analysis

The FT-IR infrared spectrum of [GO/P (AAC-MCC)] composites as shown in Fig. (4) at range (3438 - 2771 cm^{-1}) absorption band instructed to (νO-H) stretching of the carboxylic acid, and the absorption bands at the frequency (2929 cm^{-1}) to the C-H bands found in aliphatic compounds within the composite composition represent the symmetric and asymmetric vibration of the CH_2 groups present in the composite, also found a band at (1735 cm^{-1}) belonging to the carbonyl group C = O present in the carboxylic acid After adsorption of Cipro drug, we observe, as shown in Fig.(4), that the shifts of the bands towards lower wavenumber, as in the carbonyl group C = O, have shifted from 1735 to 1683. The absorption bands within the range 1600-1500 refer to the C = C group for the aromatic benzene ring[10, 11]

Adsorption Isotherm

The adsorption isotherms of the drug were determined by shaking (0.05g) of [GO/P (AAC-MCC)] composites with 10ml of drug solutions, having concentrations ranging from (10-100) ppm at temperature 20 ^{0} C, after 120 min. of shaking, the suspensions were centrifuged at 6000 rpm for 15 min. The drug concentration was determined spectrophotometrically. The quantity of Cipro drug adsorbed was calculated according to the following equation[9]

Q_e or

$$\frac{x}{m} = \frac{V\,(C_o - C_e)}{m}$$

…… (1)

Where:

x: the quantity adsorbed, m: weight of adsorbent (g), C_o: initial concentration (mg/L), Ce: equilibrium concentration (mg/ L), V: volume of solution (L).

Effect of Contact Time

The equilibrium time between the composite surface and the Cipro drug was determined with all conditions confirmed. With the time factor changed, the solution of Cipro drug concentration (100 ppm) was added, and the composite was added in a ratio of (0.05 g) to 10 ml of the drug. Then the solutions were placed in the centrifuge at different periods from (1-360) min, and then the residue from the drug concentration was determined.

Effect of temperature on adsorption

The effect of temperature on adsorption isotherms has been studied at different temperatures (10 - 25) ^{0}C for the drug on the composite surface, (0.05g) of composite

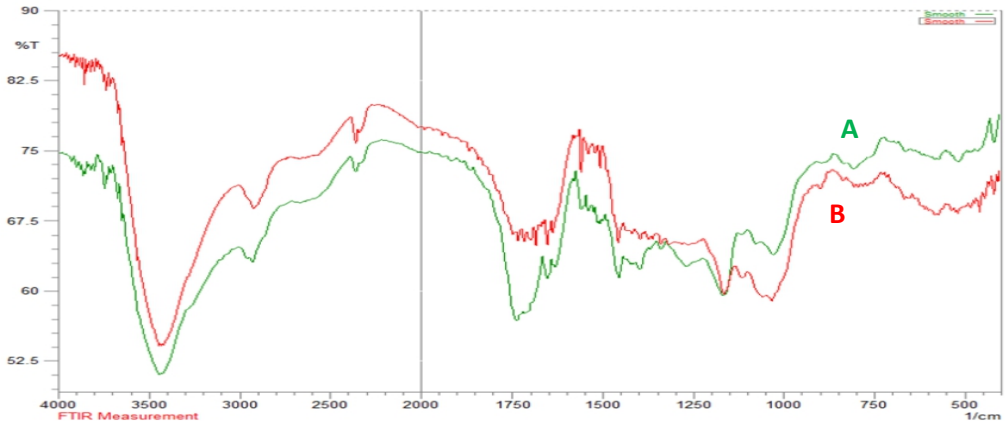

Fig. 4: FTIR spectrum of A –Composite B- Composite after adsorption drug

Atomic force microscopy (AFM)

AFM is an excellent tool that investigates the texture and morphology of composite surface, AFM images showed that the surface is porous and that the average roughness is high, and that the surface contains more declines than the tops, as well as it is bumpy nature [12], and as shown in Fig.(5) .

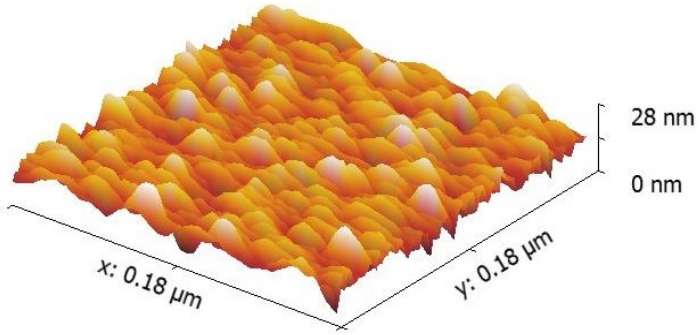

Fig. 5: AFM image three dimensional of the composite

X-ray diffraction

The XRD spectra are shown in Fig.(6) presented a broad non crystalline diffraction peak centered at (2θ = 21.192 °) corresponding to an interlayer d-spacing of (d= 4.1891A^0), which indicates the amorphous nature of the chemical composition of the composite[13, 14]

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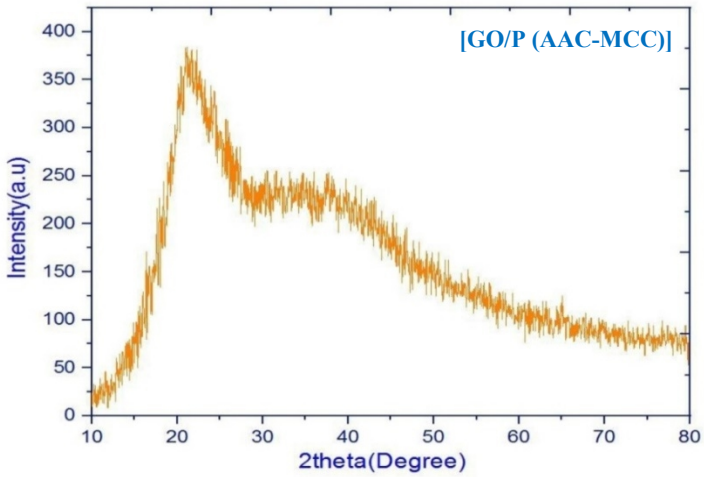

Field Emission -Scanning Electron Microscopes (FE-SEM)

[GO/P (AAC-MCC)] composites have demonstrated that its surface is rough and porous and is a Nano composite

that has a sponge-like structure and a mesh with compact layers. It contains many wrinkles that are irregularly grouped[15, 16] , as shown in Fig. (7).

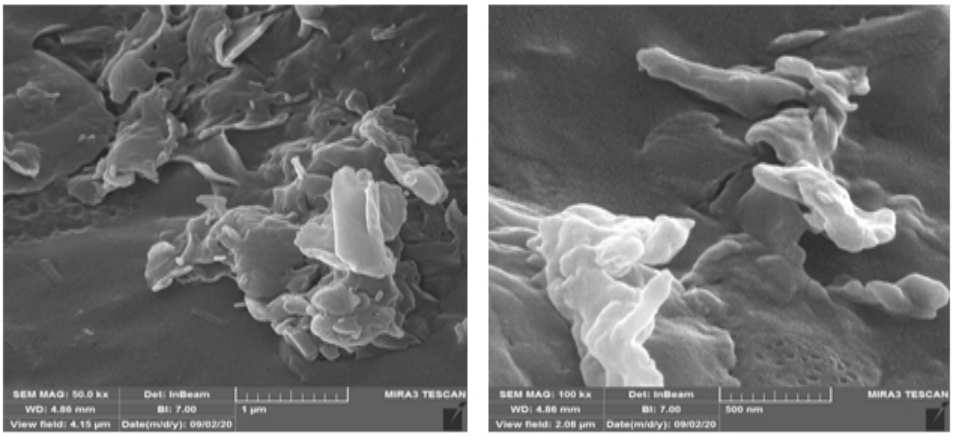

Fig. 7: FE-SEM image of [GO/P (AAC-MCC)] composites

Upon adsorption of the drug on the surface of the composite, the FE-SEM image shown in Fig. (8) showed that the surface is smoother and more coherent. Thus, the composite surface has become completely covered with the drug molecules, and this confirms the occurrence of the adsorption process[17]].

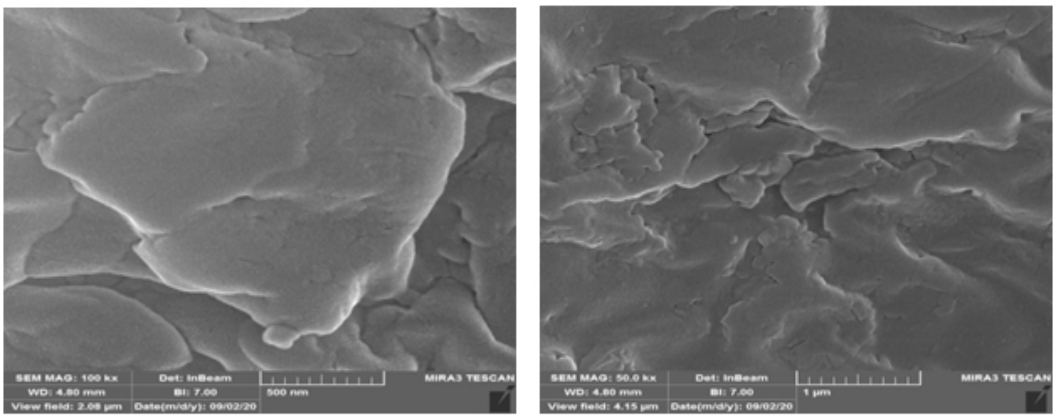

Fig. 8 FE-SEM after adsorption Cipro drug on the composite

Surface area analysis and porous surface nature

[GO/P (AAC-MCC)] composites isotherm is prepared classified by Fig. (9), (10) adsorbent isotherm - desorption of nitrogen N_2 to the prepared hydrogel is of the fourth class (IV), and this means that the multi-layer adsorption and this isotherm are hysteresis loops of H3 and this indicates that the surface pores are not regular in porous distribution[18].

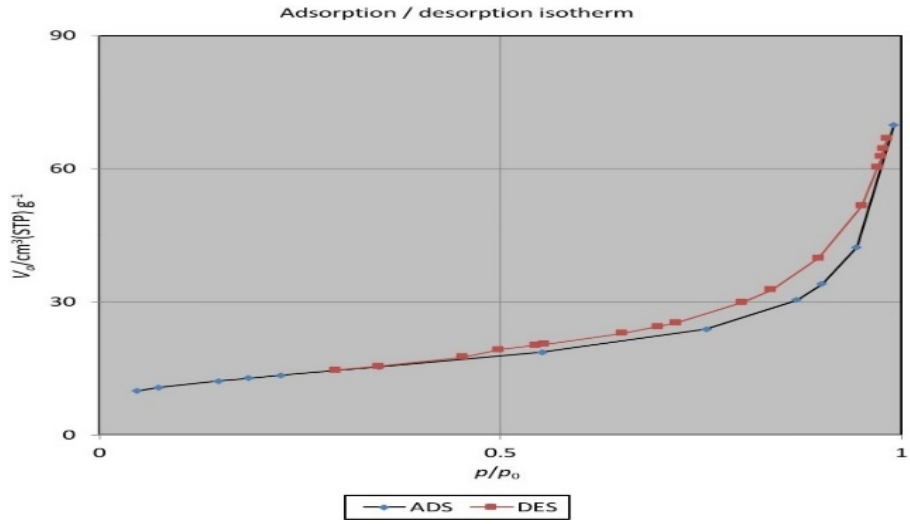

Fig 9: adsorbent isotherm - desorption of nitrogen N2 to BET

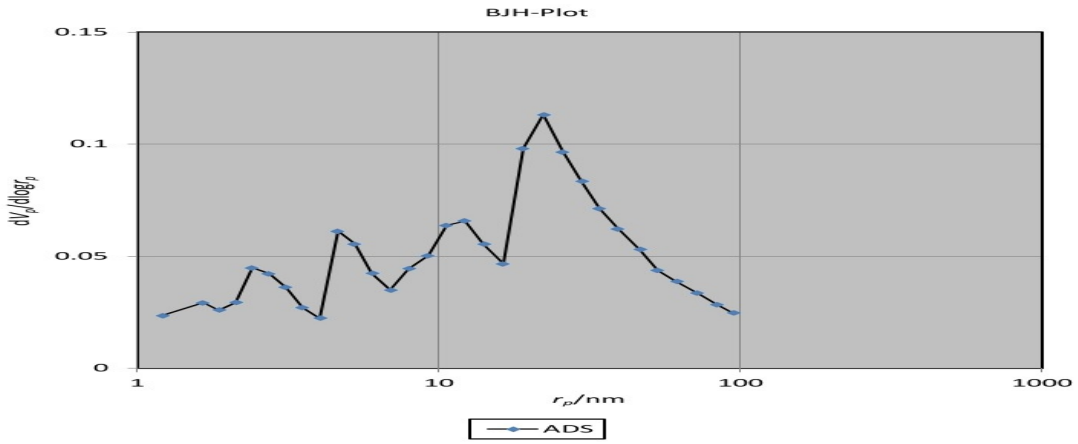

Fig. 10: adsorbent isotherm - desorption of nitrogen N_2 to BJH

Adsorption isotherms

Adsorption isotherms were calculated for the Cipro drug on the adsorbent surface, and it was observed that the adsorption process corresponds to class (L) according to the Giles classification. As this class shows, the drug particles adsorbed on the adsorbent surface will be oriented horizontally, and the adsorption will be multi-layer. It has also been observed that the application of isotherm Langmuir and Frenchlich with the adsorption process data at equilibrium to adsorb the Cipro drug, and this indicates the heterogeneous nature of the surface or surfaces that support sites of different affinity, and as shown in Fig. (11), (12) and Table (1).

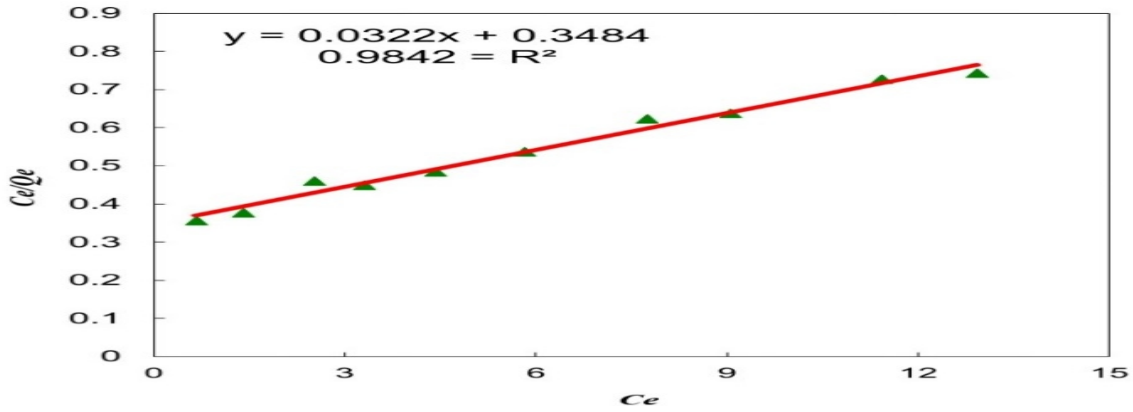

Fig. 11: The linear Langmuir isotherm for Cipro adsorption at 20 ^{0}C

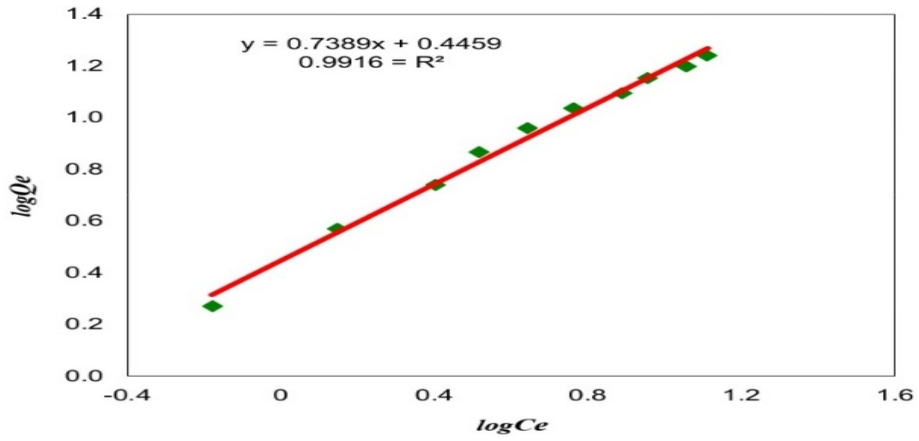

Fig. 11: The linear Freundlich isotherm for Cipro adsorption at 20 ^{0}C

Table 1: Isotherm parameters for Langmuir and Freundlich models

Langmuir equation			Freundlich equation		
K_L	q_m	R^2	K_F	n	R^2
0.092	31.055	0.9842	2.791	1.353	0.9916

Effect of Contact Time

The time required to reach the equilibrium state of the Cipro drug was studied at a concentration of 100 ppm and different periods (1-360 min) and a temperature of 20 ^{0}C and a constant weight of Composite 0.05 g and for the Cipro drug where it was found that the time required to reach the equilibrium state is (120 min), and as shown in Fig. (13), it has been observed that the adsorption process increases with time until reached to the equilibrium time at 120 min ,this state can clarification to occupy of active site by adsorbate, in the beginning, after which the increase is gradual until reaching the equilibrium time[19-21]

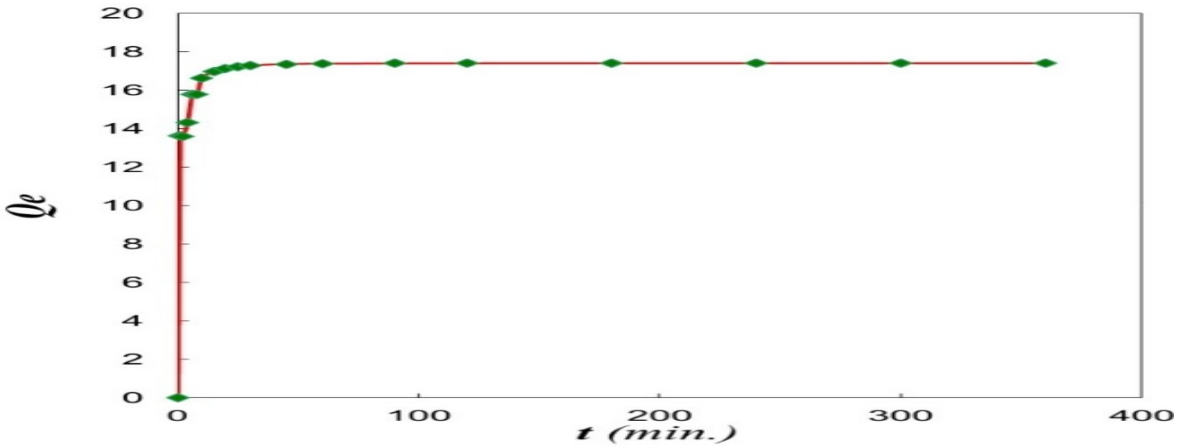

Fig. 13: Effect of contact time on Cipro adsorption

Effect of Temperature and Calculation of Thermodynamic Parameters

Cipro drug adsorption on the surface of the prepared composite has been studied at different temperatures (10,15,20 and 25) ^{0}C. The adsorption process of the Cipro drug on the composite has decreased when the temperature rises, this means that the adsorption process is an exothermic process, as it is shown in Fig. (14), and the reason for this is that the increase in temperature will increase the solubility of the minutes of the solute in the solvent, thereby reducing the adsorption affinity of the solute in the solution towards the adsorbent surface, Temperature affects the surface of the composite due to the increase in porosity of the surface as well as the increase in pore size [9, 22].

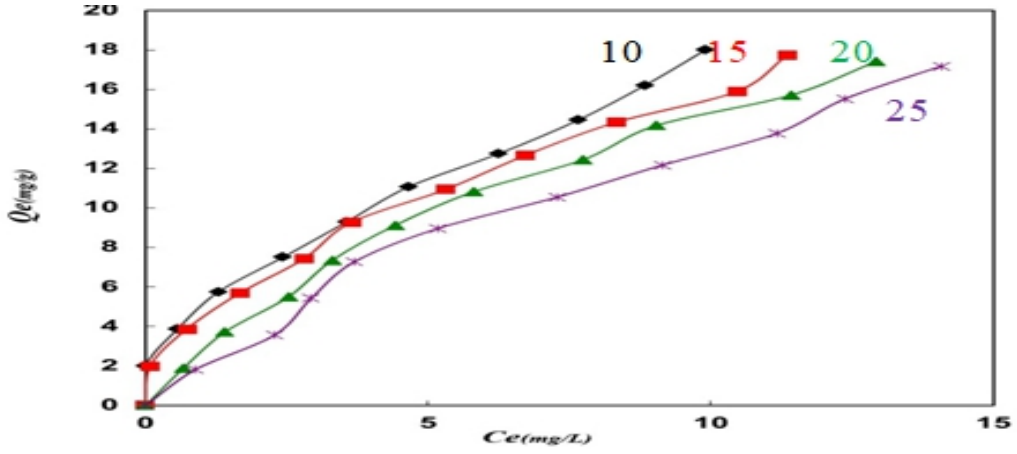

Fig. 14: Effect of temperature on the adsorption capacity of Cipro drug

The values of thermodynamic parameters have been calculated, and the results are shown in Table (2) have shown that the value of enthalpy ΔH is negative and this indicates that the process of adsorption is an exothermic process, It was also observed that free energy change was of negative value, meaning that the adsorption process was spontaneous. As for the value of the change in entropy (ΔS), it is negative, and this indicates that the adsorbed particles are in continuous motion on the surface more than in solution[23-25], as shown in Fig. (14).

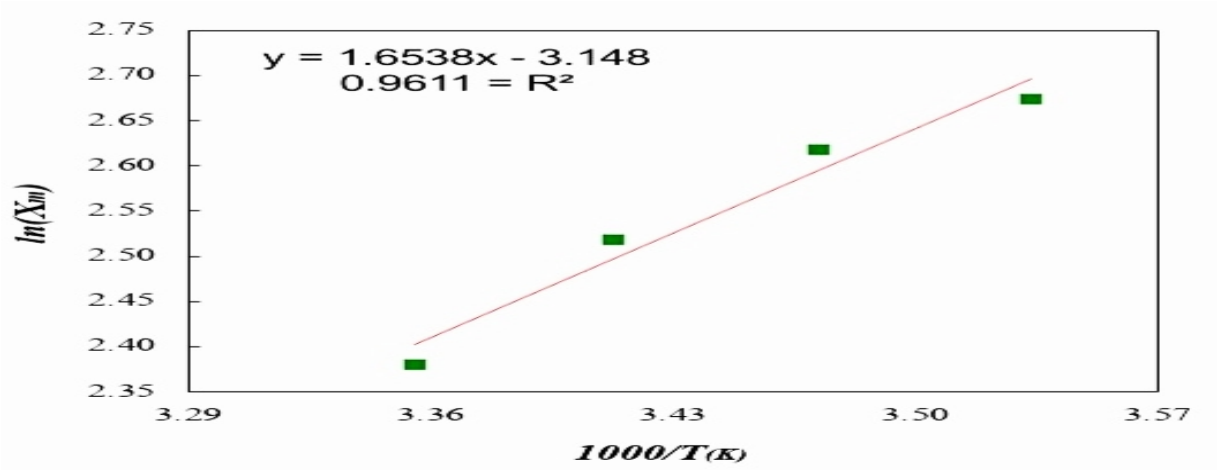

Fig. 14: Plot of ln Xm against reciprocal absolute temperature for adsorption Cipro drug on the composite surface

Table 2: Thermodynamic parameters of adsorption Cipro drug on the composite

ΔH (kJ.mol^{-1})	ΔG (kJ.mol^{-1})	ΔS (J.mol^{-1}. k^{-1})	Equilibrium Constant (K)
-13.749	-2.865	-37.148	1.296

Effect of pH

The effect of pH on the adsorption process of the Cipro drug on the adsorbent surface studies with a concentration of (100 ppm) at the values of the different pH (1.2 -10) and by adjusting the other conditions. As shown in the Fig. (15), the adsorption capacity is decrease with the increase of pH That when the pH is low, the concentration of H^+ ions is very high in the solution. Then competition will occur between it and the drug molecules with a positive charge on the active sites of the adsorbent surface, which leads to reduced adsorption, but in the case of high pH, the adsorption capacity increases when the pH rises[26].

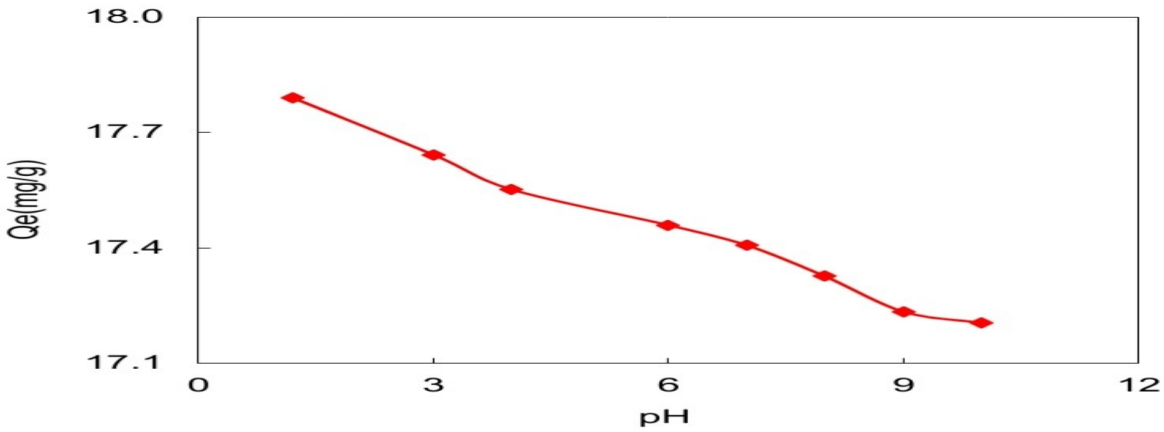

Fig. 15: Effect of pH values on Cipro adsorption on the composite

Effect of ionic strength

The effect of ionic strength in the adsorption process has been studied. The results have shown, as in Fig. (16), that the adsorption capacity of the Cipro drug on the composite increases when the salt concentration increases and this is due to the effect of competition between the positive ions of salts and the positive drug molecules on the active sites of the adsorbent surface[27-29].

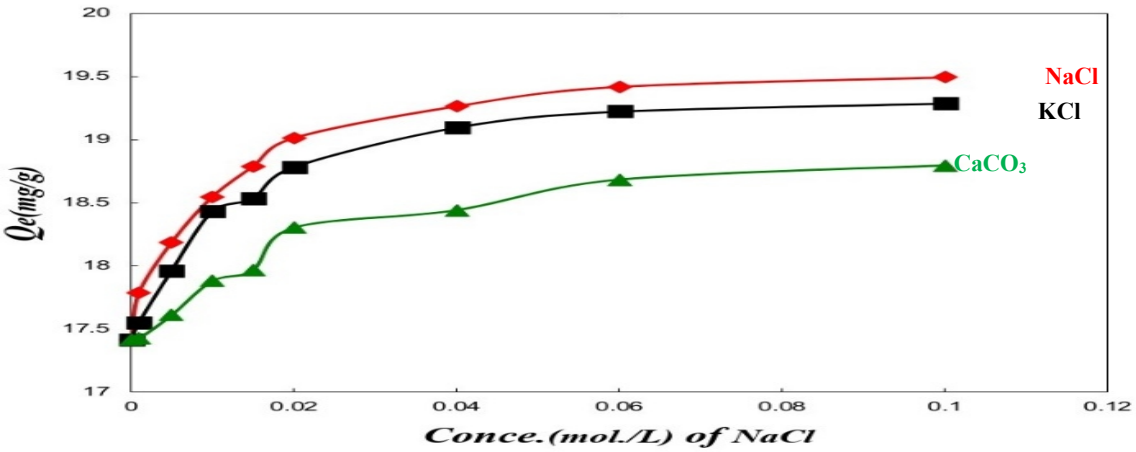

Fig. 16: Effect of ionic strength on Cipro adsorption of the composite

CONCLUSION

[(GO) /poly (acrylic acid – microcrystalline cellulose)] [GO(AAC-MCC)] superabsorbent composites were used as an adsorbent to remove a Ciprofloxacin drug from its aqueous solutions. Contact time, ionic strength, equilibrium time, temperature and pH of solution played considerable role on the adsorption capacity of composites. Langmuir and Freundlich isotherm equations model showed that it could describes the adsorption of Cipro drug on the composite surface better than Temkin model. Negative values of the free energy change, ΔG˚, showed that Cipro adsorption by composites was spontaneous. Negative values in enthalpy change ΔH˚ represent that the adsorption mechanism is found to be exothermic.

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