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Enhancement of Adsorption of Paracetamol Drug on Carbon Nanotubes concerning Wastewater Treatment

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Abstract

It is essential to eliminate antibiotics from water due to their possible harm to living creatures and the growth of resistant microorganisms. The present study focuses on paracetamol inhibition utilizing carbon nanotubes (CNTs). Fourier transform infrared, field emission-scanning electron microscopy, and transmission electron microscopy techniques were used to study the fabricated CNTs' physical and chemical properties. Several concentrations of medication (10-100 mg L^{-1}), pH solution (2.2–10.2), the weight of CNTs (0.002-0.08 g), and temperature solution (10-30 ºC) were used to estimate the adsorption studies. The obtained results indicated that a concentration of 50 mg L^{-1}, a pH of 6.6, an adsorbent amount of 0.02 g, and a contact time of 2 h are optimal conditions for removing 95.40 % of the drug from water. The results of the adsorption study indicate that the percentage of removal increases as the weight of the surfaces increases. The thermodynamic factors (ΔGº), (ΔHº), and (ΔSº) were estimated, and the negative values of ΔG indicated that the removal method was spontaneous at various temperatures. The correlation coefficient value at R^2 = 0.98881 indicates that the adsorption method has high applicability concerning the second-order model. In contrast, the applicability of the first-order model and Elkovich equation are moderate, as indicated by the correlation coefficient values of R^2 = 0.70844 and R^2 = 0.84540, respectively. Based on the findings, the prepared CNTs may serve as a promising, environmentally friendly, cost-effective, and effective material for paracetamol drugs.

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1. Introduction

Pharmaceutical preparations are among the most important chemicals used to treat, diagnose and prevent many diseases. Various medicinal groups are produced and consumed as these medicines contain antibiotics, vitamins, antitoxins, anti-inflammatory, and others. Pharmaceutical preparations differ from other chemical pollutants and are classified as molecular compounds. It differs in functions, molecular weight, and environment. It can pass through membranes, and most of these drugs can dissolve in water.[1–4]

Acetaminophen (N-acetyl-4-aminophenol) is the major active ingredient in pharmaceuticals of paracetamol. Acetaminophen is a white, odorless, crystalline powder with a bitter taste. It commonly consists of ring benzene core-substituted via amide and hydroxyl group[5] and is used as an analgesic and antipyretic drug. It is obtainable by prescription and as an over-the-counter medicine; the main physic-chemical properties and the structure of paracetamol are presented in Table 1. Micropollutants, emerging pollutants, contain extensive synthetic and normal material, including drugs, personal care products, agrochemicals and steroid hormones. Nowadays, monitoring various environmental residual drugs has been highlighted because these materials are found in waste and surface water. Utmost of these

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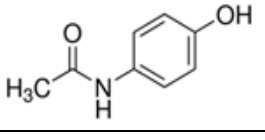
complexes are discharged into the environment incessantly by domestic sewage treatment methods.[6,7] Adsorption is a rapid phenomenon of passive sequestration separation of adsorbate from an aqueous/gaseous phase onto a solid phase.[8] Adsorption occurs between two phases in transporting pollutants from one phase to another. It is considered to be a complex phenomenon. It depends mostly on the surface chemistry or the nature of the adsorbent, adsorbate, and the system conditions between the two phases. Adsorption processes offer the most economical and effective treatment for removing pollutants from wastewater.

The process is often carried out in batch mode by adding an adsorbent to a vessel containing contaminated water. The mixture is stirred for a sufficient time before allowing the adsorbent to settle and removing the filtered water.[9] The adsorbate is the substance removed from the interface's liquid phase. The adsorbent is the solid, liquid, or gas phase onto which the adsorbate accumulates. The term adsorption is also used to describe two forces of interaction between the adsorbate and the adsorbent. The interaction forces are broadly described as physisorption (physical adsorption) and chemisorption (chemical adsorption).[10,11] The purpose of the present study was to investigate the ability of CNTs prepared by physicochemical activation with hydrogen peroxide as the activating agent to remove paracetamol from aqueous solutions.

Table 1. Physicochemical properties of paracetamol drug.

IUPAC ID	N-(4-hydroxy-phenyl)ethan amide, N-(4-hydroxy-phenyl)acet amide
Chemical Names	Acetaminophen; 4-Acetamidophenol; Paracetamol
Molecular Formula	$C_8H_9NO_2$ or $HOC_6H_4NHCOCH_3$
So. H_2O (25 °C mg mL^{-1})	14
Density g/cm^3	1.263
Molecular Weight (g mol^{-1})	151.2
Melting point	(336 °F) 169 °C
Boiling point	(788 °F) 420 °C
LogP	0.46
pKa	9.38
Chemical Structure of Paracetamol	

2. Preparation of Paracetamol Stock Solution

Paracetamol from the Samara Factory (purity 98%) was used to prepare a stock solution (1000 mg L^{-1}) by dissolving 1 g of the drug into 1000 mL of distilled water (pH 6.7). As shown in Fig. 1, the absorbance was determined spectrophotometrically using a UV-visible (UV-Vis) spectrophotometer. Solutions with five distinct concentrations are utilized to limit the relative standard deviations of the method. Table 2 displays the measured relative standard deviations (RSD) percent and standard deviation (SD).

3. Preparation of Carbon Nanotube

Carbon nanotubes (MWCNTs) were obtained from the refinery of bakery factories. The coal (Anthracite) was collected, crushed, and sieved before being subjected to a chemical process involving hydrogen peroxide, 50 g of dried CNTs were impregnated with 50 mL of 10 percent by weight of H_2O_2 solution to obtain a weight ratio of 1:5 (H_2O_2: CNTs). The carbon nanotubes were immersed in a peroxide solution with continuous stirring for one hour to activate the active sites on the surface, then filtered and washed with the distilled water several times. CNTs prepared were dried at 110 °C for two hours. Then clean biomass was mechanically ground and filtered to produce a powder with a particle size of less than 25 µm.

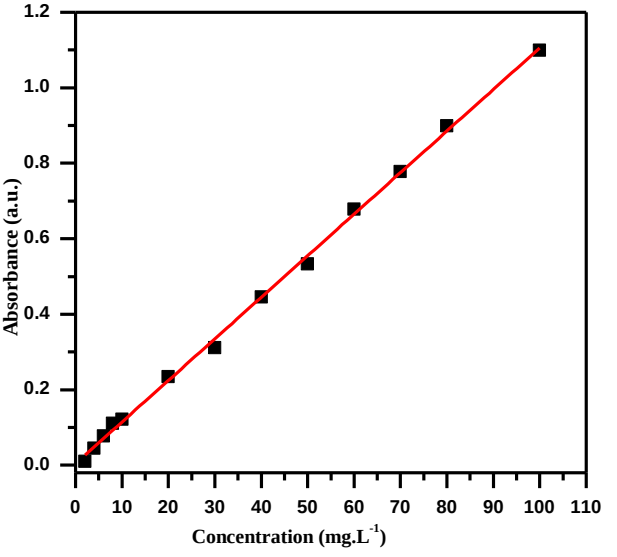


Fig. 1 Calibration curve of Paracetamol drug.

4. Batch Adsorption Experiment

Batch adsorption experiments were conducted in a 100 mL conical flask in a shaker water bath. The effect of contact time 2 h by using 0.02 g of CNTs and 100 mL of paracetamol drug solution at 200 rpm at different time intervals at 20 °C performed by varying the initial drug concentrations from (10–100) mg L^{-1}. The effect of experimental factors such as mass (0.002–0.08 g), and pH (2.2–10.5), were studied using one parameter-one-time approach. After different aliquots from the reaction mixture were analyzed for residual drug concentration using a UV-Vis spectrophotometer at 220 nm wavelength, monitoring changes in absorbance. Samples were collected from the flask at various intervals and analyzed for residual drug concentration. Adsorption kinetics investigations were carried out by agitating 100 mL of drug solution of known initial concentration with 10–100 mg/L of adsorbent at a temperature of 20 °C, by using pH metere (model HI 83141, Hanna, Romania) of 6.6 ± 0.1 and at 120 rpm for different time intervals. Adsorption efficiency (Qe) and Removal percentage (E %) were calculated using Eq. (1) and Eq. (2), respectively.

Table 2. Statistics of the calibration curve (Fig. 1) for several paracetamol drug concentrations.

Parameters	Proposed Method paracetamol
λ_{max} (nm)	342
Beer's law limit (μg ml^{-1})	2-100
Regression equation	(Y = mX + C) Y= 0.01102X + 0.000422
Slope (m)	0.01102 (± $1.28154*10^{-4}$)
Intercept (C)	0.00422 (± 0.00213)
Correlation coefficient (r^2)	0.99839
% Relative Standard deviation (RSD%)	0.52
standard deviation (SD)	0.14
Color	colorless

$$Qe = \frac{Co-Ce}{W} * V \quad (1)$$

$$E\% = \frac{Co-Ce}{Co} * 100 \quad (2)$$

Co (mg L^{-1}) is the primary concentration of the drug and *Ce* (mg L^{-1}) is the drug equilibrium concentration at time *t* (min), *W* (g) is the adsorbent weight, *V* (L) is the volume of drug solution.

5. Material Characterization Techniques

5.1 Fourier transfer infrared spectroscopy (FT-IR)

FT-IR measurements were carried out on model 8400S, Shimadzu, Japan FTIR spectrometer. The samples were diluted to a concentration of 0.1% with potassium bromide powder (KBr) from Merck. 300 mg of the mixtures were compressed for 10 minutes at 10 t (Ø 1.3 cm) under vacuum, forming a grayish-round disc.

5.2 Field Emission Scanning Electron Microscopy (FE-SEM)

FE-SEM is a powerful device used to characterize sample morphology such as grain size, particle size, particle distribution, crystal defects, and surface structure. FE-SEM model MIRA3, TESCAN, Czechia Republic, has several features like a large depth of field, higher resolution, and more control in the degree of amplification. About 50 μL of aqueous or ethanol suspension of the sample was placed on a clean silicon wafer surface.

5.3 Transmission Electron Microscopy (TEM)

TEM is a microscopy technique that uses a beam of electrons transmitted across an ultra-thin sample. The electrons are transformed into light and form an image. TEM model 912AB, Leo, Germany, provides information on phase composition, structure, and lattice defects. The mechanism for examining the sample is by adding drops from the sample as an aqueous solution to the carbon-covered TEM networks.

6. Results and Discussion

6.1 Characterization of FT-IR for adsorbent/adsorbate

FTIR was used to analyze the surface functional groups responsible for paracetamol drug adsorption. Adsorbent surfaces CNTs and drug-loaded adsorbents samples after adsorption was placed in an oven at 65 ℃ for 24 h.

Carbon nanotube was characterized via FT-IR spectroscopy spectra. FT-IR was collected in the mid -IR series about 4000 - 400 cm^{-1}. Fig. 2 depicts the FT-IR spectra of CNTs before and after paracetamol drug adsorption. The characteristic peaks are induced by S–H bonds, centered between 2327 and 3412 cm^{-1} by CNTs –OH. The FT-IR pattern appears reduced in band intensity next to the adsorption. There is a difference between CNTs before and after interaction by paracetamol drug that have been suggested that a physical-sorption phenomenon happens as a data of attractive forces among the CNTs surface and paracetamol drug in the study.[12,13]

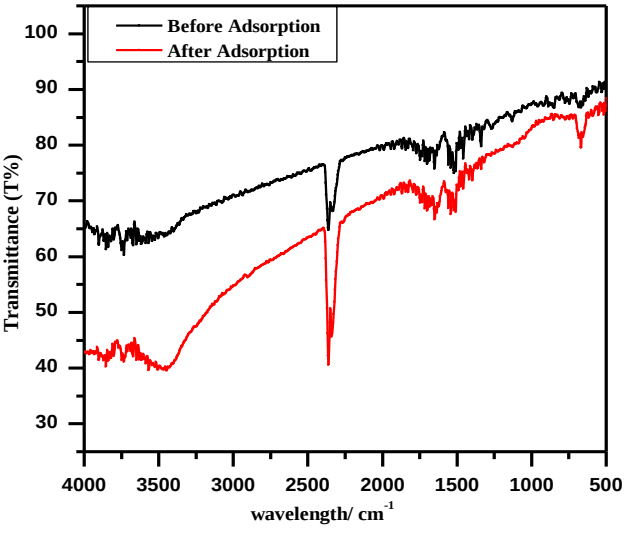


Fig. 2 FTIR spectra of Carbon Nano Tubes. (a) before absorption of paracetamol drug (b) After absorption of paracetamol drug.

6.2 FE-SEM

FE-SEM was used to characterize the surface morphology and fundamental physical properties of 200 nm CNTs before and after the divalent paracetamol drug adsorption. The FE-SEM analysis reveals that the CNTs are arranged in a flower-like pattern and that the surface is composed of numerous small, irregular clusters. Multiple layers of CNTs are compressed on top of one another by the force of Vander Walls and the cross-linking agent. As a result of adsorption,

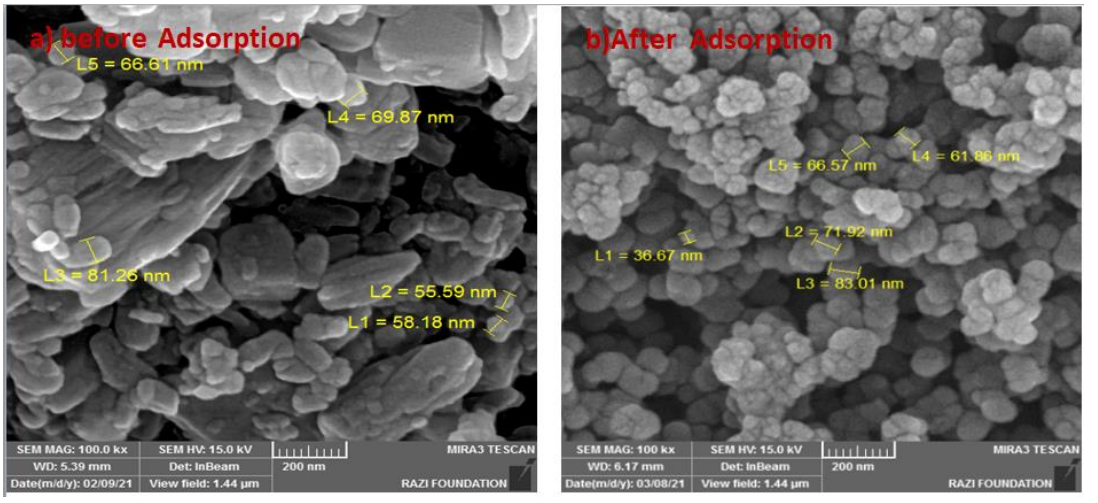


Fig. 3 FE-SEM of Carbon Nano Tubes. (a) before absorption of paracetamol drug (b) After absorption of paracetamol drug.

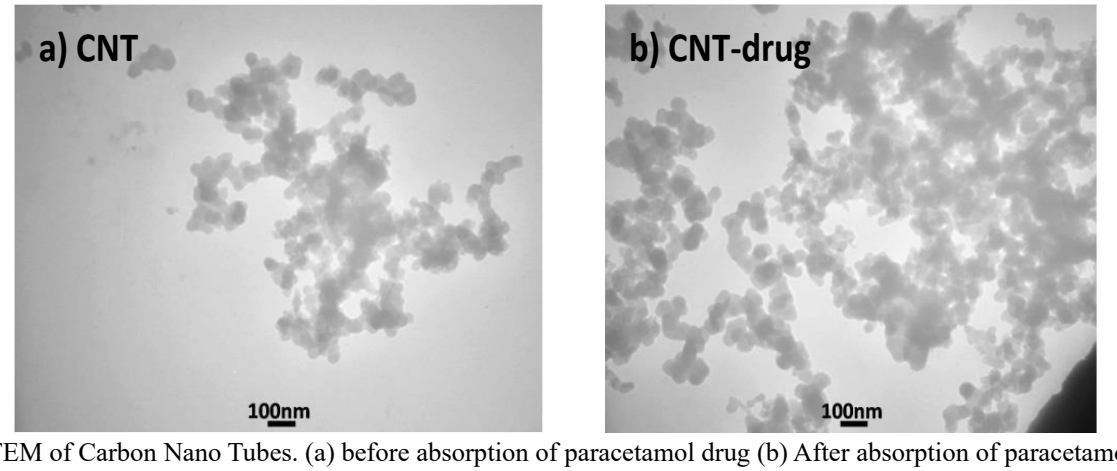


Fig. 4 TEM of Carbon Nano Tubes. (a) before absorption of paracetamol drug (b) After absorption of paracetamol drug.

the surface is characterized by ridges and humps caused primarily by the drug.[14] Observations of the physical combination of CNTs revealed that the CNTs were connected to the outer and inner surface and filled the cracks. Due to the increase in surface area of CNTs, the presence of spherical-shaped CNTs may be viewed as a means of enhancing the drug uptake of paracetamol. As shown in Fig. 3, the morphology of CNTs reveals the presence of internal pores, which facilitates the intra-particle diffusion of paracetamol.

6.3 TEM

The morphology of the CNTs surface nanocomposite was analyzed using TEM morphology surface analysis (100 nm), as shown in Fig. 4. It was evident that the cloud became more agglomerated after adsorption. A new geometry was formed, which was attributed to the loading of the drug on the surface, where the presence of clusters and the formation of subtle dark-colored[15,16] were observed.

6.4 Effect of initial paracetamol drug concentration

The amounts of paracetamol drug adsorbed "adsorption equilibrium" (Qe) and percentage removal (R%) of paracetamol drug at varied initial drug concentrations C_0 under various experimental settings are shown in (Fig. 5). Results demonstrates that the quantity of paracetamol drug adsorbed varies with initial paracetamol drug concentration and grows with increasing initial paracetamol drug concentration. In contrast, the percentage clearance decreases with increasing paracetamol drug concentrations. Increasing the initial concentration of paracetamol enhances the adsorption process by increasing the number of collisions between the drug and the CNTs. The effect of paracetamol concentration on CNTs capacity was determined to be of great value for the utilized medicine.[17]

6.5 Effect of CNTs dose

The influence of the CNT's weight on the removal (E)% of the paracetamol drug adsorbed from aqueous solutions demonstrated that the removal capacity of paracetamol drug gradually increased as CNTs increased. As shown in Fig. 6, as the mass of CNTs increases from 0.001 to 0.08 g, the proportion of adsorbed paracetamol increases from 25.83 to 94.77%. Increasing the masses of CNTs decreased the adsorption efficiency of the same paracetamol chain drug by approximately 861.212–59.193 mg/g.

This could be because increasing the amount of adsorbent

offered a greater surface area or more adsorption sites for the paracetamol medication. Increasing the amount of CNT used by an additional 0.05–0.08 g had no effect on the E percent of paracetamol drug eliminated.[18]

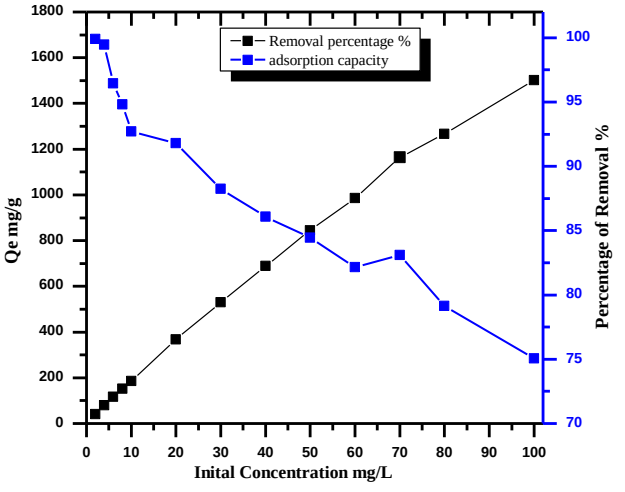


Fig. 5 Initial concentration of Paracetamol drug adsorption using CNTs at optimum condition (mass of CNT 0.02 g, pH 6.6, Temperature 20 °C).

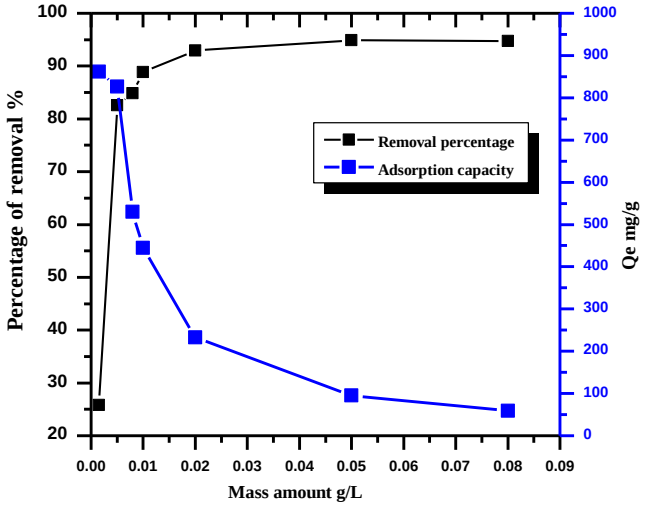


Fig. 6 CNTs surface mass effect on Paracetamol drug at optimum condition (Initial concentration 50 mg/L, pH 6.6, Temperature 20 °C).

6.6 Effect of solution pH

The pH is one of the best parameters for studying the adsorption behavior of sorbent and sorbate presses. Fig. 7 depicts the influence of a pH range of approximately 2.2 to 10.5 on the E% elimination of paracetamol from CNTs. Under optimal conditions (50 mg L-1 of 100 mL paracetamol drug mass 0.02 g CNTs, 2hr equilibrium time and shaking rate 120 rpm). The paracetamol medication exhibited the lowest adsorption capacity at pH 2.2 (829.05 mg/g) and the highest adsorption capacity at pH 10.5 (856.127 mg/g).[19]

6.7 Adsorption thermodynamics

The thermodynamic functions are among the most significant parameters that influence adsorption. The chemical or physical alterations may be dependent on the active modifications. Thermodynamics is the basic physical science responsible for determining these discrepancies, as the optimal adsorption efficiency of the substance is determined by thermodynamic functions such as (ΔG^o), (ΔH^o) and (ΔS^o). These are the important thermodynamic parameters for researching adsorption mechanisms that can establish the adsorption process's feasibility, spontaneity, and heat exchange.[20] The effect of temperature (10, 20, and 30 °C) on the absorption of paracetamol is examined (Fig. 8). A rise follows a temperature increase in CNT adsorption effectiveness (the higher the temperature, the better the adsorption).

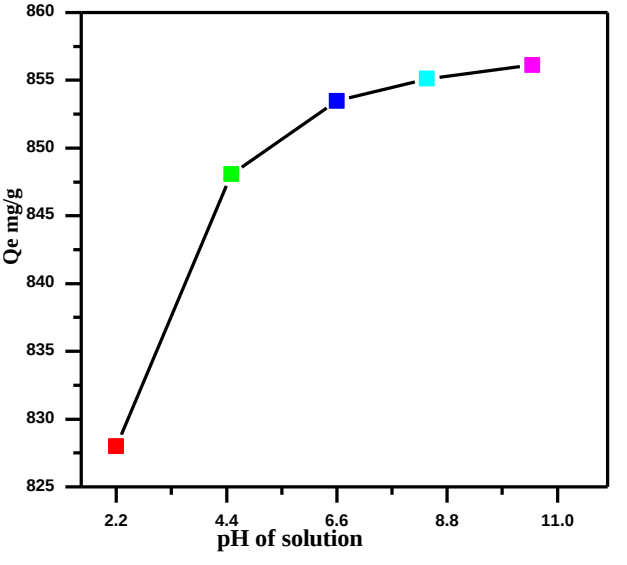


Fig. 7 Effect of solution pH on Adsorption of paracetamol drug at optimum condition (mass of CNTs 0.02 g, Initial concentration 50 mg/L, Temperature 20 °C).

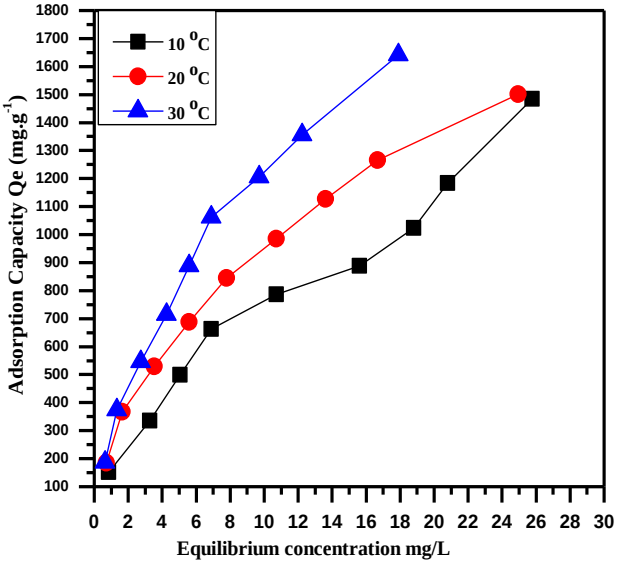


Fig. 8 Adsorption model of paracetamol drug on to CNTs at several temperatures, optimum condition (mass of CNTs 0.02 g, pH 6.6).

The change in heat that occurs when a solution comes into contact with a solid is not as easily analyzed as the heat generated by the adsorption of a single gas by a solid. The former is typically much less and can be determined by calculating the concentration required to achieve a particular quantity of adsorption at various temperatures.[21] ΔG^o may be approximated using Eq. (3) and Eq. (4). Table 3 provides the thermodynamic values estimation of paracetamol drug adsorption onto CNTs.

$$\Delta G^o = -RT \ln K \quad (3)$$

ΔS^o can be found from equation ΔG^o: Substituting ΔG^o by equation (2):

$$\Delta G^o = \Delta H^o - T\Delta S^o \quad (4)$$

Table 3. Thermodynamic parameter for paracetamol adsorption on CNTs.

ΔH (KJ mol^{-1})	ΔG (KJ mol^{-1})	ΔS (J mol^{-1} K^{-1})	Keq
	-9.344		26.536
19.05	-10.439	12.443	36.312
	-11.334		44.972

6.8 Adsorption isotherms

The isotherm model takes into account a crucial component in resolving adsorption mode. Adsorption models can discover adsorbent-adsorbate interactions in reality. The Freundlech multi-ayer and Langmuer single-layer adsorption isotherms of the adsorption equilibrium model are suitable for the solid-liquid adsorption method.[22,23]

The Langmuir isotherm [Eq. (5)] hypothesizes that adsorption occurs on a homogenous surface through monolayer coverage. Freundlech's model [Eq. (6)] is an empirical model for multilayer adsorption on heterogeneous surfaces.

$$Qe = \frac{Qm\ KL\ Ce}{1+KL\ Ce} \quad (5)$$

$$Qe = K_f\, Ce^{1/n} \quad (6)$$

K_f (L mg^{-1}) is Freundlech constant, 1/n is hetero-geneity factor.

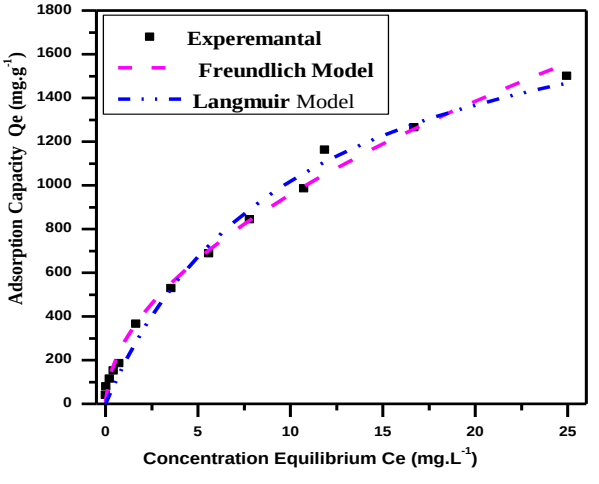


Fig. 9 Several adsorption models nonlinear fit of adsorption drug on CNTs, optimum condition (mass of CNT 0.02 g, pH 6.6, Temperature 20 °C).

A plot of Qe vs. Ce appears in Fig. 9, that values of K_F, 1/n are fixed from the intercept and slope of the linear retreating. As a look, the best regression correlation coefficient was shown by the model Freundlech (R^2 = 0.9915). This tick that the model Freundlech was appropriate for characterizing the drug's sorption onto CNT compared to model Langmuir (R^2 = 0.9851). Data are seen in Fig. 9. The studied parameters of the two-model isotherm are shown in Table 4.

6.9 Kinetic model

6.9.1 First-order model

The rate constant of adsorption is estimated from the following first-order model rate expression[22] as appears in Eq. (7):

$$ln(q_e - q_t) = lnq_e - k_1 t \quad (7)$$

where q_e is the quantity of adsorbed drug (mg/g) at equilibrium, qt is the quantity of the adsorbed drug at time t (min), and k_1 is the rate constant of adsorption of the first order model. Lagergren's usability deviates from its nonlinear form, according to the results. The new kinetic model is inadequate for forecasting pseudo-first-order sorption kinetics on CNTs. We wanted to apply the experimental data to models because the first-order kinetic model failed to represent the sorption effects.[24]

6.9.2 Second-order model

The adsorption method's kinetics model might also be described in the second-order model rate equation.[25] The nonlinear form of the t equation is expressed as Eq. (8):

$$\frac{1}{qt} = \frac{1}{K2qe2} + \frac{1}{qe}\, t \quad (8)$$

The kinetic parameter of the drug adsorption onto surface CNT appearing in Table 5 which shows the pseudo-second-order plot; it can be seen that the sorption data maintains its linear profile over the entire period; however, higher values of correlation coefficients and qe determined using the pseudo-second-order model were more compatible with the experimental data.[26]

6.9.3 Elkovich model

The equation Elkovich utilized for common application to chemisorption. The equation has been useful satisfactorily to some chemisorption processes. It has been found to cover a wide range of slow adsorption rates[27]. A similar equation is often valid for the system in which the adsorbing surface is heterogeneous and is formulated as in Eq. (9):

$$qt = \frac{1}{\beta} 1\beta \ln(\alpha\beta) + \frac{1}{\beta} lnt \quad (9)$$

Fig. 10 represent the variation of drug adsorption on CNT by time shaking (0–220 min) and different primary drug concentration solution (50 mg/L). Where note the high applicability of the adsorption method to model second-order depending on the correlation coefficient value at R^2 = 0.98881. whereas the applicability to the model first-order and equation Elkovich decrease due to the value low of the R^2 = 0.70844 and R^2 = 0.84540.[28]

Table 4. Isotherm models Freundlech and Langmuir parameter of paracetamol drug adsorbed onto CNTs optimum condition (mass of CNTs 0.02 g, pH 6.6, Temperature 20 ℃).

Isotherm models	Parameters	(SAFB)
Freundlich		282.2208 ± 17.3315
	1/n	0.53106 ± 0.0231
	R^2	0.99155
Langmuir		2080.8996 ± 156.577
	K_L(L/mg)	0.0958 ± 0.0155
	R^2	0.9851

Table 5. Adsorption kinetics factors the adsorption of paracetamol drug optimum condition (mass of CNT 0.02 g, pH 6.6, Temperature 20 ℃).

Model	Equation	Parameters	Value
First-order	$q_t=q_e[1-\exp(k_f t)]$	$K_1(min^{-1})$	0.4444 ± 0.0409
		qe(calc)(mgg^{-1})	32.8613 ± 5.5531
		R^2	0.70844
Second-order	$q_t=\frac{K2qe2t}{1+K2qet}$	K_2(g/mg/min)	0.02224 ± 0.00377
		qe(calc)(mgg^{-1})	30.794 ± 0.6991
		R^2	0.9834
Elkoveich model	$q_t=1/\beta 1\beta\ln(\alpha\beta)+1/\beta\ \ln t$	α (mg g^{-1} min^{-1})	36.0027
		β (g min^{-1})	0.0123
		R^2	0.8454

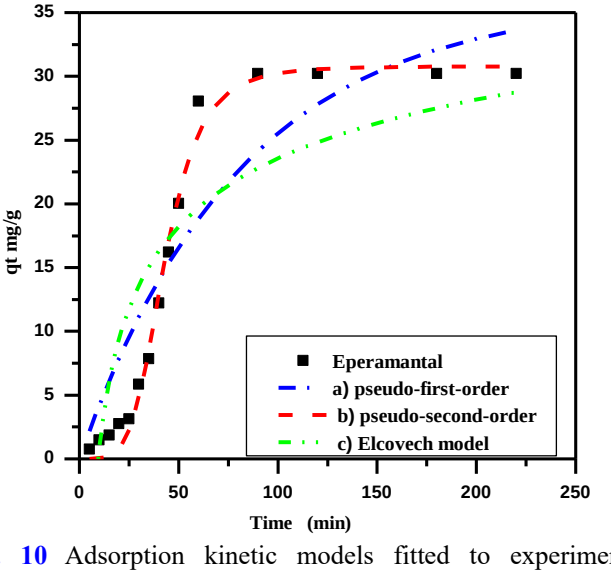


Fig. 10 Adsorption kinetic models fitted to experimental paracetamol drug adsorption onto CNTs a) pseudo-first-order b) pseudo-second-order c) Elkovech model, optimum condition (mass of CNTs 0.02 g, pH 6.6, Temperature 20 ℃).

7. Conclusions

The adsorption capacity and percentage of paracetamol drug removal increase with increasing contact time, surface area, and temperature solution. However, adsorption capacity has decreased with the increase of adsorbent dosage. The optimum contact time for equilibrium to be achieved is found to be 2 h It is basically due to saturation of the active site, which does not allow further adsorption. For paracetamol drugs on adsorbent surfaces, maximum adsorption was found at pH = 10.5. Adsorption was found to increase with an increase in the pH of the solution. The negative value of ΔG confirms the spontaneous nature adsorption process. The positive value of ΔS showed the increased randomness at the solid-solution interface during adsorption. The positive value of ΔH indicated that the adsorption process was endothermic. The interaction between dose and initial concentration significantly affected the adsorption process. Adsorbent curves showed good fits and correlations to Freundlich isotherm, which suggests that adsorption is heterogeneous.

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Conflict of Interest

The authors declare no conflict of interest.

Supporting information

Not applicable.

References

[1] V. Arancibia, J. Penagos-Llanos, E. Nagles, O. García-Beltrán, J. J. Hurtado, Development of a microcomposite with single-walled carbon and Nd_2O_3 for determination of paracetamol in pharmaceutical dosage by adsorptive voltammetry, *Journal of*

pharmaceutical analysis, 2019, **9**, 62-69, doi: 10.1016/j.jpha.2018.11.005.
[2] G. W. C. S. Perera, M. D. P. de Costa, K. R. R. Mahanama, Development of a fluorimetric method for assessing paracetamol in pharmaceuticals tablets, *Journal of Photochemistry and Photobiology A: Chemistry*, 2019, **368**, 248-253, doi: 10.1016/j.jphotochem.2018.09.039.
[3] H. Hu, H. Liu, D. Zhang, J. Wang, G. Qin, X. Zhang, pH and electromagnetic dual-remoted drug delivery based on bimodal superparamagnetic Fe_3O_4@Porous silica nanoparticles, *Engineered Science*, 2018, **2**, 43-48, doi: 10.30919/es8d136.
[4] A. M. Aljeboree, A. F. Alkaim, Removal of Antibiotic Tetracycline (TCs) from aqueous solutions by using Titanium dioxide (TiO2) nanoparticles as an alternative material, *Journal of Physics: Conference Series*, 2019, **1294**, 052059, doi: 10.1088/1742-6596/1294/5/052059.
[5] A. M. Aljeboree, N. D. Radia, L. S. Jasim, and A. F. Alkaim, Removal of Pharmaceutical by Adsorption Process; Study Effect of Various Parameters and Determination of Pharmaceutical by Oxidative Coupling Reaction, *Adsorption*, 2021, **11**, 912–916.
[6] J. Song, Y. Wang, and J. Qiu, High adsorption performance of methyl blue from aqueous solution using hyper-branched polyethyleneimine grafted MWCNTs as an adsorbent, *ES Materials & Manufacturing*, 2018, **3**, 29–37, doi: 10.30919/esmm5f193.
[7] C. Gao, W. Deng, F. Pan, X. Feng, Y. Li, Superhydrophobic electrospun PVDF membranes with silanization and fluorosilanization Co-functionalized CNTs for improved direct contact membrane distillation, *Engineered Science*, 2020, **9**, 35-43, doi: 10.30919/es8d905.
[8] E. M. Alrobayi, A. M. Algubili, A. M. Aljeboree, A. F. Alkaim, F. H. Hussein, Investigation of photocatalytic removal and photonic efficiency of maxilon blue dye GRL in the presence of TiO_2 nanoparticles, *Particulate Science and Technology*, 2017, **35**, 14-20, doi: 10.1080/02726351.2015.1120836.
[9] Y. A. Faleh, N. D. Radhy, Removal of Metformin hydrochloride from Aqueous Solutions by using Carboxymethyl cellulose-g-poly(acrylic acid-co-acrylamide) Hydrogel: Adsorption and Thermodynamic Studies, *IOP Conference Series: Earth and Environmental Science*, 2021, **790**, 012062, doi: 10.1088/1755-1315/790/1/012062.
[10] A. M. Aljeboree, S. A. Karim, and A. F. Alkaim, Removal of Amino Drug from Aqueous Solution by Clay and Study of Optimum Condition Colorimetric Determination of Amino Drug Using Reagent: Stability and Higher Sensitivity, *Journal of Pharmaceutical Quality Assurance*, 2022, **13**, 66–70, doi: 10.25258/ijpqa.13.1.14.
[11] N. Li, F. Zhang, H. Wang, S. Hou, Catalytic degradation of 4-nitrophenol in polluted water by three-dimensional gold nanoparticles/reduced graphene oxide microspheres, *Engineered Science*, 2019, **7**, 72-79, doi: 10.30919/es8d509.
[12] C. V. Jagtap, V. S. Kadam, M. A. Mahadik, J. S. Jang, N. B. Chaure, H. M. Pathan, Effect of binder concentration and dye loading time on titania based photoanode in DSSC application, *Engineered Science*, 2021, **17**, 133-141, doi: 10.30919/es8d581.
[13] M. J. Ahmed, Adsorption of quinolone, tetracycline, and penicillin antibiotics from aqueous solution using activated carbons: review, *Environmental Toxicology and Pharmacology*, 2017, **50**, 1-10, doi: 10.1016/j.etap.2017.01.004.
[14] G. Moussavi, A. Alahabadi, K. Yaghmaeian, M. Eskandari, Preparation, characterization and adsorption potential of the NH_4Cl-induced activated carbon for the removal of amoxicillin antibiotic from water, *Chemical Engineering Journal*, 2013, **217**, 119-128, doi: 10.1016/j.cej.2012.11.069.
[15] J. L. Sotelo, A. R. Rodríguez, M. M. Mateos, S. D. Hernández, S. A. Torrellas, J. G. Rodríguez, Adsorption of pharmaceutical compounds and an endocrine disruptor from aqueous solutions by carbon materials, *Journal of Environmental Science and Health, Part B*, 2012, **47**, 640-652, doi: 10.1080/03601234.2012.668462.
[16] A. M. Mohammed, A. M. Aljeboree, A. B. Mahdi, Y. S. Karim, M. A. Jawad, and A. F. Alkaim, Adsorption Features Remove a Toxic Dye from an Aqueous Solution by a Cost-effective Palm Leaf Activated Carbon (PLAC), *Journal of Chemical Health Risks (2022)s*, 2022, **12**, 509–516, doi: 10.22034/jchr.2022.691650.
[17] A. Arun, P. Malrautu, A. Laha, H. Luo, S. Ramakrishna, Collagen nanoparticles in drug delivery systems and tissue engineering, *Applied Sciences*, 2021, **11**, 11369, doi: 10.3390/app112311369.
[18] H. Venkatachalam, J. B S, A. H. Shrungeswara, Evaluation of anti-inflammatory potentials of novel flavonols and their metal complexes, *Engineered Science*, 2022, 18, 217-223, doi: 10.30919/es8e676.
[19] H. B. Quesada, A. T. A. Baptista, L. F. Cusioli, D. Seibert, C. de Oliveira Bezerra, R. Bergamasco, Surface water pollution by pharmaceuticals and an alternative of removal by low-cost adsorbents: a review, *Chemosphere*, 2019, **222**, 766-780, doi: 10.1016/j.chemosphere.2019.02.009.
[20] A. A. Ensafi, A. R. Allafchian, B. Rezaei, Multiwall carbon nanotubes decorated with FeCr2O4, a new selective electrochemical sensor for amoxicillin determination, *Journal of Nanoparticle Research*, 2012, **14**, 1244, doi: 10.1007/s11051-012-1244-3.
[21] H. Freundlich, W. Heller, The adsorption of cis- and trans-azobenzene, *Journal of the American Chemical Society*, 1939, **61**, 2228-2230, doi: 10.1021/ja01877a071.
[22] S. K. Lagergren, About the theory of so-called adsorption of soluble substances, *Sven. Vetenskapsakad. Handingarl*, 1898, **24**, 1–39.
[23] Y. S. Ho, G. McKay, Sorption of dye from aqueous solution by peat, *Chemical Engineering Journal*, 1998, **70**, 115-124, doi: 10.1016/S0923-0467(98)00076-1.
[24] M. Ghaedi, B. Sadeghian, A. A. Pebdani, R. Sahraei, A. Daneshfar, C. Duran, Kinetics, thermodynamics and equilibrium evaluation of direct yellow 12 removal by adsorption onto silver nanoparticles loaded activated carbon, *Chemical Engineering Journal*, 2012, **187**, 133-141, doi: 10.1016/j.cej.2012.01.111.
[25] J. L. Acero, F. J. Benitez, F. J. Real, G. Roldan, Kinetics of aqueous chlorination of some pharmaceuticals and their

elimination from water matrices, *Water Research*, 2010, **44**, 4158-4170, doi: 10.1016/j.watres.2010.05.012.
[26] A. Chowdhury, S. Kumari, A. A. Khan, M. R. Chandra, S. Hussain, 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, **611**, 125868, doi: 10.1016/j.colsurfa.2020.125868.
[27] D. George, P. U. Maheswari, K. M. M. S. Begum, Chitosan-cellulose hydrogel conjugated with L-histidine and zinc oxide nanoparticles for sustained drug delivery: Kinetics and in-vitro biological studies, *Carbohydrate Polymers*, 2020, **236**, 116101, doi: 10.1016/j.carbpol.2020.116101.
[28] M. A. J. Kadhum, A. J. Kadhim, and A. M. Aljeboree, Enhanced removal of phenylephrine hydrochloride using eco-friendly surface: optimization, isotherm, kinetics, and regeneration studies, *Quality Assurance*, 2021, **12**, 206–210, doi: 10.25258/ijpqa.12.3.8.