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Boosted Removal of Sulfadiazine Drug Using Multiwall Carbon Nanotubes and Comparative with Other Adsorbents

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ABSTRACT

Multiwall carbon nanotubes (MWCNTs) treated and oxidized with sulfuric acid were utilized as a higher adsorbent for fast removal of sulfadiazine hydrochloride (SFD) drug from aqueous solutions. The effect of different important parameters like equilibrium time (5-60 min), temperature 10-40 °C, pH (3-10), adsorbent dosage (0.001-0.05 g), and concentration of drug (10-100 mg/L) were well studied and optimized. As a result of the value optimization of different factors such as equilibrium time 30 min, temperature 30 °C, solution pH 3, concentration of drug 50 mg/L, and weight of MWCNT 0.03 g. The adsorbent MWCNT was characterized *via* FE-SEM, TEM, and EDX analyses. The development of MWCNT shows a better potential (removal percentage 97.29% and adsorption capacity Qe 162.15 mg/g) within 1 hr for best drug removal from aqueous solution. The isotherm result was found fitted and in best agreement with the isotherm Freundlich model. The higher and fast removal of drugs was done using MWCNTs in a very short period of time, and the best adsorption efficacy of the developed adsorbent in comparison with developed adsorbent establishes the importance of this research.

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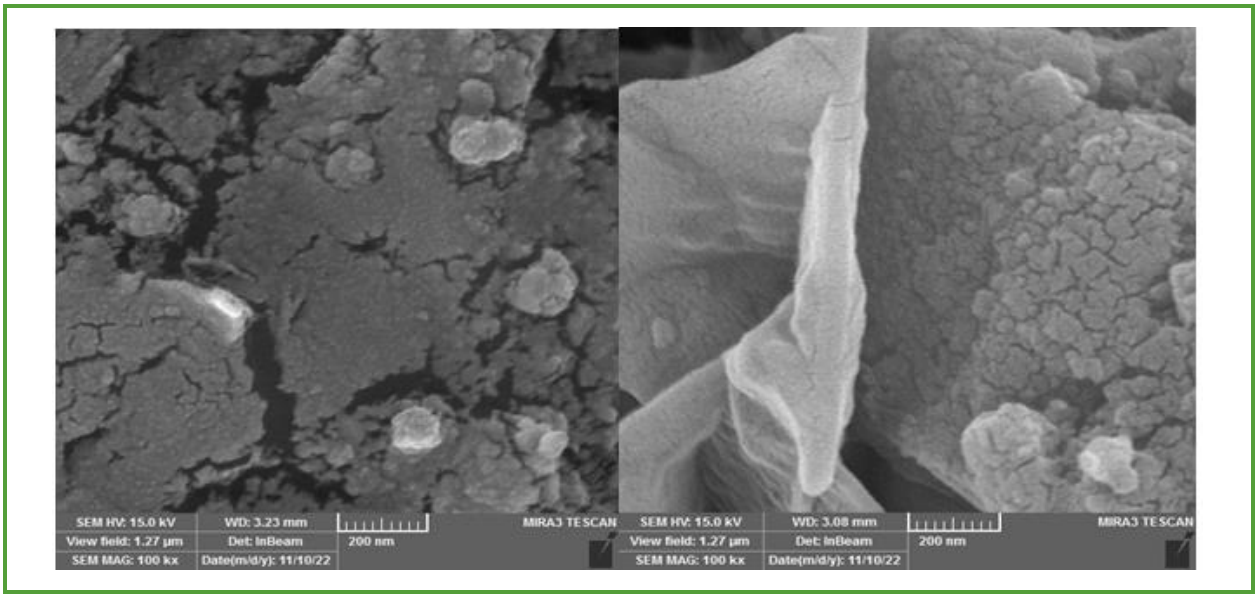
Graphical Abstract



Introduction

Multiple scientific studies conducted both in the United States and around the world have consistently found that pharmaceuticals are widely present in various sources of water, including treated drinking water [1, 2], surface water [3], groundwater, wastewater treatment plant effluent, and sludge [4, 5]. The presence of pharmaceuticals in drinking water sources, including surface water, groundwater, and treated drinking water, is raising significant worries among scientific communities and public health agencies. This is because pharmaceuticals are purposely created to produce specific physiological and biological impacts, and their unregulated consumption may create harmful health consequences for both humans and animals [6].

One of the most important and dangerous environmental pollutants is wastewater. This water pollution is considered as the result of factory waste, which is produced due to industrial expansion and population increase, especially in developed countries. Among the most dangerous of these pollutants are pharmaceuticals, which constitute a very dangerous source and cause great concern for public health and the aquatic environment [7-9].

Considering removal of contaminants as pharmaceuticals from wastewater is very essential for the life of humans as well as vital animals and plants; several techniques have been used to quickly remove harmful pollutants, such as photo-oxidation, adsorption, reduction, precipitation, and oxidation [10-12]. Therefore, the adsorption process is the most suitable for the disposal of pharmaceutical contaminants, due to very economical considerations and its high efficiency. Some adsorbents like zeolites, biomaterials, activated carbon, MWCNT, hydrogel, and polymers. Carbon nanotubes (CNTs) are considered as one of the most important types of carbon materials and have many applications in various fields in the removal of liquid waste, wastewater, etc. Multiwall carbon nanotubes (MWCNTs) are characterized by large surface areas, small pores, and a hollow structure, thus possessing

high efficiency in removing organic and inorganic pollutants [13-21].

Sulfadiazine Hydrochloride (SFD), (4-amino-N-2-pyrimidinylbenzenesulfonamide, chemical formula $C_{10}H_{10}N_4O_2S$, chemical stretcher, Figure 1) is an antibiotic sulfonamide grope utilized a several of infections, like trachoma, urinary tract infections, a systemic antibacterial agent, instance, and ocular toxoplasmosis and cancroid, treatment of trachoma. Sulfadiazine considers pharmacopeia and nonpharmacopeial impurities [22].

The objective of this study was to estimate the potential and efficiency of MWCNTs to removal drugs from aqueous solutions. The influence of several factors are the effect of the adsorption method and optimization. The main aims of this work are as follows: To study the viability of utilizing MWCNTs as adsorbents to remove drugs, regulate the application of two models (Langmuir, and Freundlich isotherms), and find best-fit the equation isotherms, and also to compare it with other adsorbents.

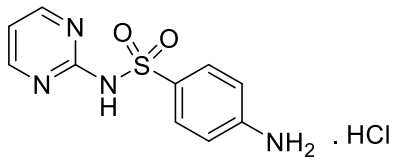


Figure 1. Chemical stretcher sulfadiazine hydrochloride (SFD) drug

Experimental

Preparation of multiwall carbon nanotubes (MWCNTs)

Black coal, which is made up of multiwall carbon nanotubes (MWCNTs), was obtained from bakery factory refineries. Initially, the coal was collected, crushed, and sieved before chemical treatment. It was washed several times in deionized distilled water to get rid of unwanted substances. After that, it was dried in the oven at 70 °C for 2 h, and then the MWCNTs (100 g) are impregnated with 100 ml of acid H_2SO_4 (0.01 M), the MWCNTs are immersed in the acid solution with continuous stirring for 3 h. To activate, all active sites were located on the MWCNTs surface, and then filtered and washed several times with deionized distilled water. It was dried at 60 °C for 24 h. After that, the powder was ground and filtered to produce a nanoparticle with a particle size of less than 25 μm.

Adsorption experiments

Effect of adsorbent dosage

The influence of weight of MWCNTs on the amount adsorbed was found *via* agitating SFD drug solution of 100 mL/ 50 mg/L in conical flasks 100 mL with MWCNTs of 0.001-0.05 g at 30 °C for contact time 30 min.

Effect of sulfadiazine hydrochloride (SFD) drug concentration

100 mL of SFD drug in 100 mL flasks was prepared in different concentrations (10-100 mg/L) and adsorbent 0.03 g at 30 °C for contact time 30 min.

Effect of solution temperature

The effect of temperature solution on the amount adsorbed was found *via* agitating 100 mL of SFD drug solution of 50 mg/L in conical flasks 100 mL with 0.03 g MWCNTs at temperature (10-40 °C) for contact time 60 min.

Effect of pH

100 mL of 100 mL solution was prepared in a 100 mL flash with a concentration of drug 50 mg/L and adsorbent dosage of 0.03 g in conical flasks 100 mL and solution pH (3-10). The pH of drug solutions was adjusted with HCl or NaOH (0.01 N) by utilizing a UV-Vis

spectrophotometer. The percentage removal and adsorption capacity of drugs was determined using Equations (1) and (2).

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

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

Results and Discussion

Characterization

Morphological analysis of MWCNTs was performed using SEM technique. As observed in Figure 2a, MWCNTs have a homogeneous, smooth, interconnected porous internal morphology that is randomly distributed. In general, this type of morphology results from the preparation method, where the surface was activated by acid washing, whereby the porosity increased. Also, after the adsorption process, MWCNT showed a rough, heterogeneous, porous, and trapezoidal shape resembling flower petals. It is likely caused by active aggregates covering a surface, which changes the final surface appearance of the preparation, as depicted in Figure 2b.

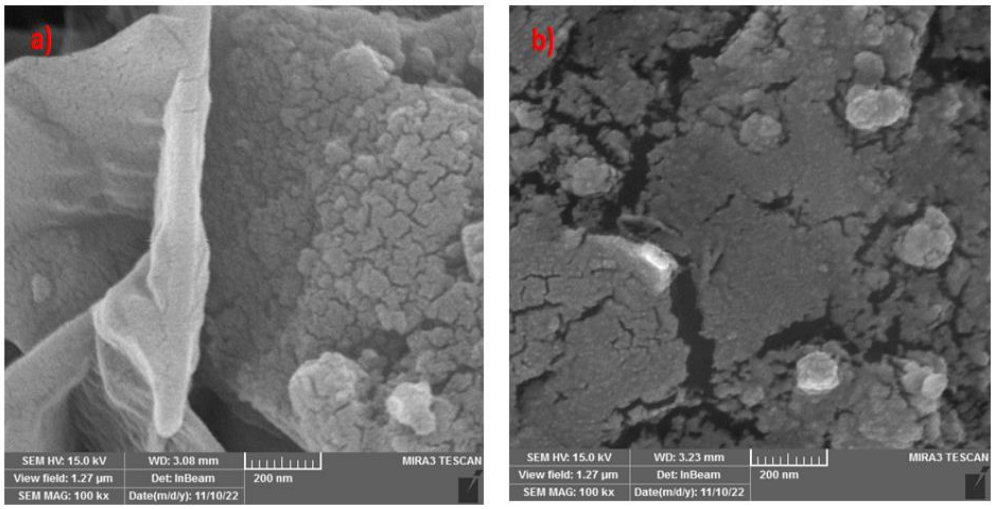


Figure 2. SEM image a) before adsorption and b) after adsorption

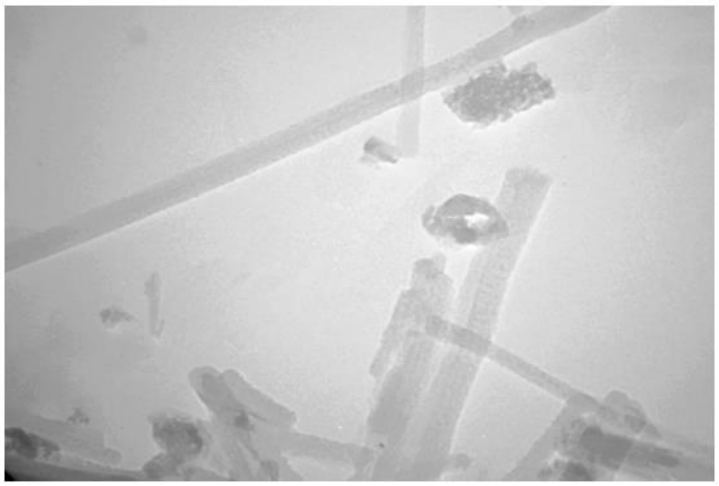
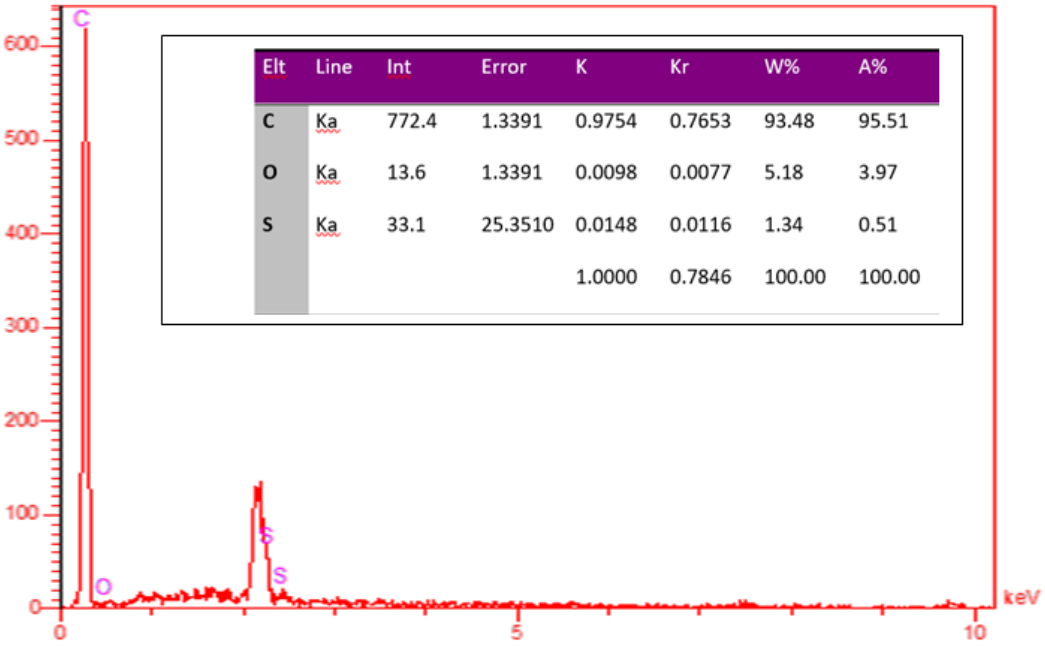


Figure 3. TEM image of MWCNTs

TEM analysis shows large aggregates in MWCNTs which may be caused by treating the prepared surface with acid. It led to an increase in the particle size, and therefore, upon surface treatment and activation, the particles tend to form large agglomerates. Thus, it is expected that larger aggregates will be formed in the results [23-25]. The results show that MWCNTs were successfully prepared and that treating them with acid increased the porosity of the MWCNTs, as shown in Figure 3

The composition and purity of the results reveal that the peaks are for C and O samples. EDX analysis indicated that the desired phases of O and C are present in the MWCNTs with good purity. Also, we found a small amount of S resulted from being activated by sulfuric acid (Figure 4) [14, 15, 23].



Elt	Line	Int	Error	K	Kr	W%	A%
C	Ka	772.4	1.3391	0.9754	0.7653	93.48	95.51
O	Ka	13.6	1.3391	0.0098	0.0077	5.18	3.97
S	Ka	33.1	25.3510	0.0148	0.0116	1.34	0.51
				1.0000	0.7846	100.00	100.00

Figure 4. The EDX analysis of MWCNTs

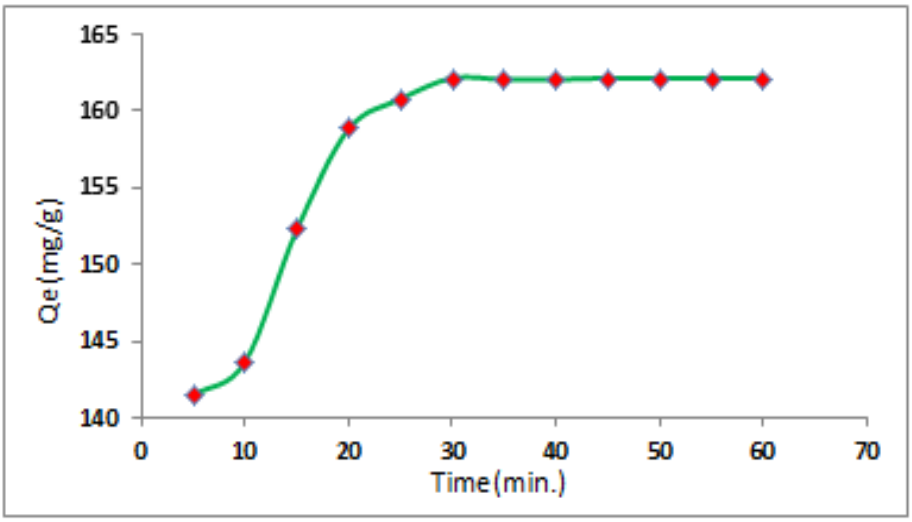


Figure 5. Effect of equilibrium time on SFD drug adsorption

Effect of equilibrium time on drug adsorption

The influence of equilibrium time on the concentration of SFD drug (50 mg/L) appears in Figure 5. It was observed that the quantity of drug adsorbed was fast for the first 10 minutes and afterwards it proceeded at a slower rate (15-60 min) and lastly reached equilibrium. The contact time of adsorption capacity rises from 141.2 mg/g - 162.15 mg/g, with the concentration of SFD drug as 50 mg/L. Accordingly, the equilibrium time needed for drug solution with drug concentrations of 50 mg/L to reach contact at 30 min. The concentration of a drug provides a significant driving force to overcome all mass transfer resistances of the drug between solid phases and aqueous solution. Though, the experimental result was measured at 30 min to be sure that full equilibrium was attained [26].

Effect weight of MWCNTs

While conducting batch mode investigation, the weight of MWCNTs is also considered as one of the significant factors. The influence of the MWCNTs weight on removal of SFD drugs was studied via changing the weight of MWCNTs 0.001-0.05 g /100 Ml (Figure 6). The removal percentage % increased with the rise of the weight of MWCNTs, due to the large number of active sites on the MWCNTs obtainable for adsorption, and hence the elimination of SFD drug was increased. With a further rise, it will lead to saturation and this might be due to the non-availability of active sites at a greater weight of MWCNTs. Therefore, the best adsorption of SFD drug for MWCNTs was found adsorption efficiency (141.1 mg/g - 162.15 mg/g) with increased weight of MWCNTs from (0.001-0.05 g) [27, 28].

Effect of pH

The influence of pH solution was studied at optimum conditions. The pH solution was adjusted with NaOH or HCl (0.01 N) before investigation (Figure 7). The pH solution of the samples was maintained utilizing a pH meter. The pH meter was calibrated by three buffers: 2.0, 7.0 and 10.0. The concentration of SFD drug was maintained at 50 mg/L. The influence of the pH solution on the removal removes the percentage of the SFD drug that uses MWCNTs. It is observed that elimination of SFD drugs via MWCNTs was best pH 3-5 in acid medium and the maximum adsorption capacity (166.54 mg/g - 162.15 mg/g) [29].

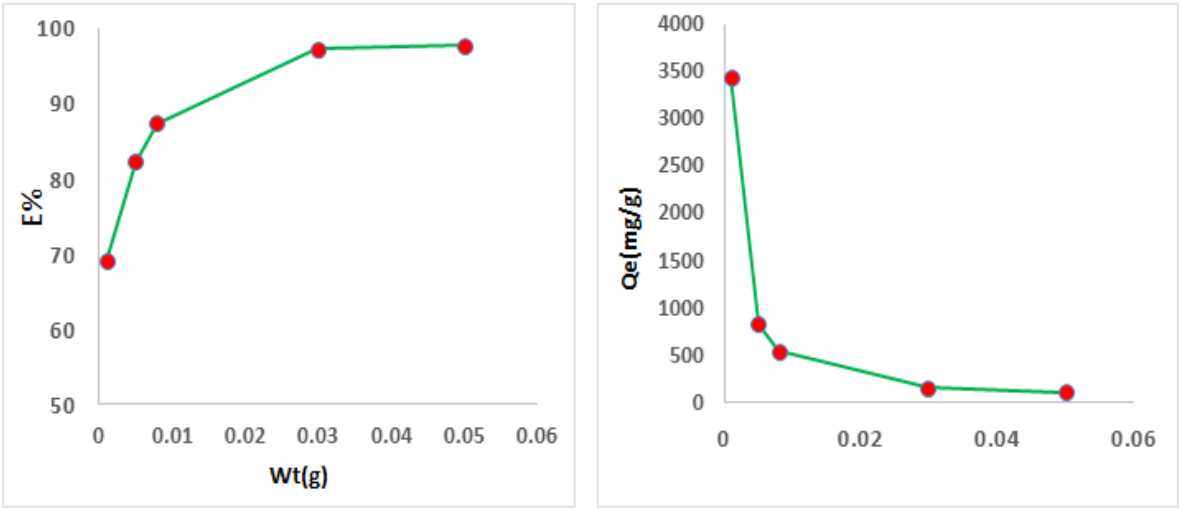


Figure 6. Effect weight of MWCNTs onto removal SFD drug

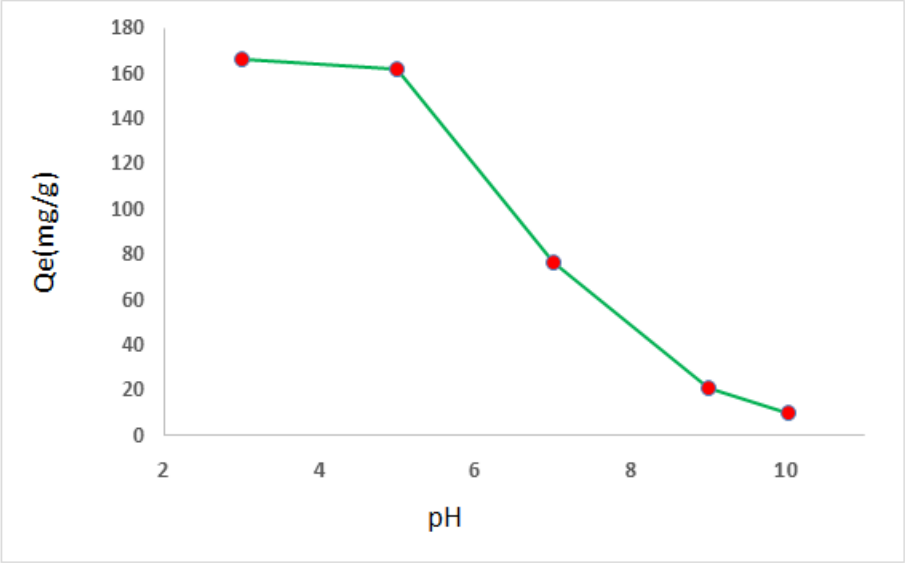


Figure 7. Effect of solution pH onto removal SFD drug

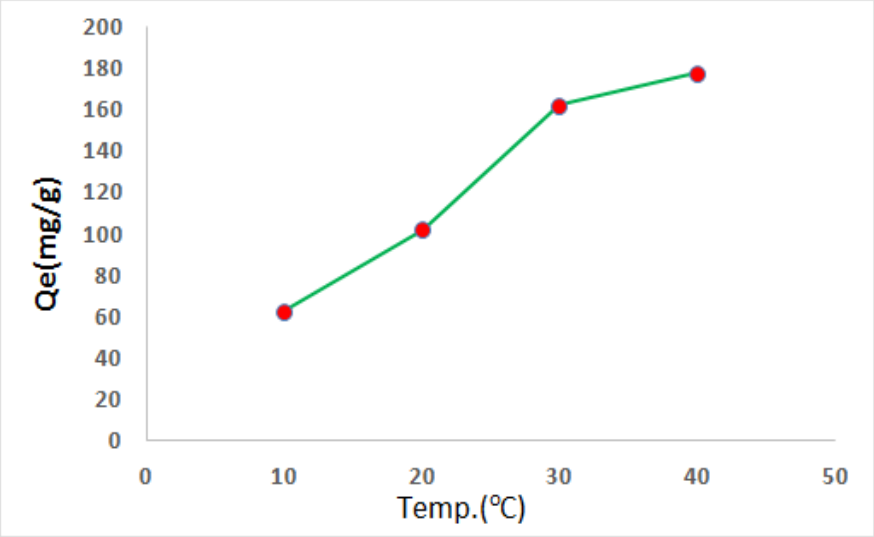


Figure 8. Effect of solution temperature onto removal SFD drug

Table 1. Langmuir model and Freundlich isotherm parameter SFD drug adsorbed onto MWCNTs

Models	Parameter	MWCNTs
Freundlich	KF	1.48
	1/n	0.0059
	R2	0.9599
Langmuir	qm (mg/g)	72.233
	KL(L/mg)	0.12
	R2	0.0431

Table 2. Effect of adsorption efficiency (mg /g) of several bio-sorbents for the sorption of SFD drug

Adsorbent	q_{max}	Conc. mg/L	pH	Ref.
CNTs	2080.8	50	6.6	[35]
Date palm fibers	24.5	10	7.6	[36]
RP-AC	329.49	20	8.0	[37]
Banana stalk-AC	141.7	30	8.0	[39]
Activated carbon (AC)	75.5	10	10	[9]
coffee residue	222.2	50	6.0	[39]
Apricot stone	123.3	100	6.0	[31]
Palm Leaf	255.5	100	6.6	[40]
Borassus bark carbon	20.7	15	6.0	[27]
pine fruit shells	112.3	50	8.0	[41]
MWCNTs	162.15	50	5	This study

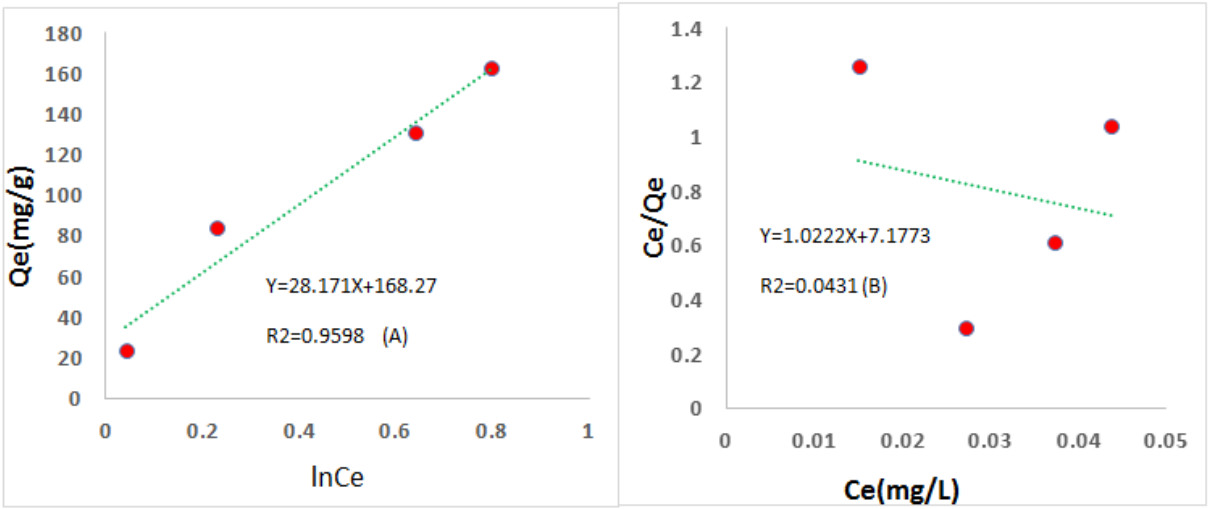


Figure 9. Adsorption isotherm (A) Freundlich model and (B) Langmuir model

Effect of solution temperature

Temperature solution increased the removal percentage (%) and adsorption capacity Qe increased. The temperature improvement increase could be explained as a result of raising the pore size, which proceeds to stir up the rate of diffusion and large collisions of particles moving into the external layer and moving inside pores. This could occur as a result of reducing the viscosity of the mixture, and a rise in the movement of ions between the liquid/ solid interfaces [30-32]. Under these circumstances, the adsorption capacity of MWCNTs will increase correspondingly as data of diffusion within internal sites as shown in Figure 8.

Adsorption isotherm

The values of adsorption isotherm model constants and the respective of R2 are indicated in Table 1. The calculated correlation coefficient R2 suggested by the isotherm Langmuir and isotherm Freundlich isotherm fitted well with the result of isotherm adsorption of drugs. The best mono-layer adsorption efficiency (qmax) according to the isotherm Langmuir isotherm is 162.23 mg/g at 30 °C [33, 34].

A comparative list of values adsorption efficiency q max of several bio-sorbents is given in Table 2. In the model Freundlich isotherm, the value of n (0.058) indicated favorable adsorption of drug on to MWCNTs *via* physico-chemisorption on the surface hetero-genous of MWCNTs. It has been suggested that a value of n less than 1 represents a beneficial sorption method. The determination of K_F value in the Freundlich model 1.48 suggests efficient adsorption of drugs *via* MWCNTs [32, 34] , as shown in Table 1 and Figure 9.

Conclusion

The study shows that Multiwall carbon nanotubes (MWCNTs) were utilized successfully to remove SFD drugs from an aqueous solution. The adsorption equilibrium of SFD drugs onto MWCNTs has been described via the isotherm Langmuir model and on the isotherm Freundlich model was fitted as a result of the Freundlich model. The adsorption equilibrium isotherm result is dependent on the correlation coefficient R^2. When the initial concentration of drug increased 10-100 mg/L, adsorption capacity increased 141.14 mg/g - 162.15 mg/g, whilst removal percentage from removal decreased 69.70%-97.60%. The best removal percentage of drug is at pH 3 and solution temperature of 30 °C.

Disclosure Statement

No potential conflict of interest was reported by the authors.

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Authors' Contributions

All authors contributed to data analysis, drafting, and revising of the article and agreed to be responsible for all the aspects of this work.

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