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RESEARCH ARTICLE

In-depth study of adsorption mechanisms and interactions in the removal of pharmaceutical contaminants via activated carbon: a physicochemical analysis

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Abstract

This study presents a theoretical analysis of the adsorption process of pharmaceutical pollutants, specifically acetaminophen (ATP) and diclofenac (DFC), onto activated carbon (AC) derived from avocado biomass waste. The adsorption isotherms of ATP and DFC were analyzed using a multilayer model, which revealed the formation of two to four adsorption layers depending on the temperature of the aqueous solution. The saturation adsorption capacities for ATP and DFC were 52.71 and 116.53 mg/g, respectively. A steric analysis suggested that the adsorption mechanisms of ATP and DFC involved a multi-molecular process. The calculated adsorption energies (ΔE_1 and ΔE_2) varied between 12.86 and 22.58 kJ/mol, with the highest values observed for DFC removal. Therefore, the adsorption of these organic molecules was associated with physisorption interactions: van der Waals forces and hydrogen bonds. These findings enhance the understanding of the depollution processes of pharmaceutical compounds using carbon-based adsorbents and highlight the potential of utilizing waste biomass for environmental remediation.

Keywords Adsorption · Biomass-based adsorbent · Pharmaceutical depollution · Multi-layer model · Statistical physics modeling

Introduction

Pharmaceutical pollution is increasingly recognized as a significant threat to both environmental and human health. The discharge of pharmaceutical compounds into the environment can have detrimental effects on ecosystems and human populations alike (Samal et al. 2022). Despite the growing awareness of the risks associated with pharmaceutical pollutants, their environmental presence remains largely unregulated and unchecked. These pollutants are characterized by their resistance to chemical decomposition and biodegradation, leading to potential long-term impacts on the health and stability of ecosystems (Okoye et al. 2022; Ighalo and Adeniyi 2020). This resilience can be attributed to their physicochemical properties, which include polarity, chemical stability, and water solubility (Rasheed et al. 2021).

Acetaminophen (ATP) and diclofenac (DFC) are pharmaceutical compounds commonly used for alleviating pain and reducing fever. Given their widespread use, there has been considerable research into their potential harmful effects on human and animal health, as well as their environmental

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impact. Some studies have reported the environmental and human health risks generated by these pollutants due to their toxicological profiles (Dos Santos et al. 2019; Rossitto et al. 2019). As a result, a variety of remediation methods, including oxidation, biological degradation, ultrasonic irradiation, and adsorption, have been explored to remove these pollutants (Dotto et al. 2015a; Anandan et al. 2020; Theerthagiri et al. 2021).

Adsorption technology is recognized for its extensive use in water treatment, owing to its numerous benefits such as simplicity in design (Dhaouadi et al. 2020; Diao et al. 2024), user-friendly operation, reliability, the potential for adsorbent regeneration, and cost-effectiveness (Rathi et al. 2021; Ali et al. 2023). A review of the literature reveals that a variety of adsorbents, including activated carbon (Mariana et al. 2021), zeolite (Pérez-Botella et al. 2022), spirulina platensis (Dotto et al. 2012), biochar (Jha et al. 2023), and clays (Shamsudin et al. 2023), has been utilized to decrease the concentration of these pollutants in water. Activated carbon (AC) is a popular choice from the available commercial adsorbents (Anastopoulos et al. 2020; Khalil et al. 2020), because of its textural properties and surface functional groups, and this adsorbent exhibits a high affinity for the separation of a wide array of substances.

In the initial stages of elucidating the mechanism of pollutant adsorption, classical models such as Freundlich and Langmuir are commonly employed to analyze the equilibrium of adsorption across various systems (Wang and Guo 2020; Lu and Na 2022). However, these models offer limited insights into the adsorption mechanism. Traditional adsorption models are often based on insufficient hypotheses and fundamentals, leading to limited scientific conclusions (Dotto et al. 2015b). For instance, the Langmuir model assumes that each functional group on the adsorbent surface adsorbs only a single adsorbate ion or molecule. Additionally, the Langmuir model ignores the presence of adsorbate–adsorbate interactions, which can significantly impact the adsorption behavior, especially at higher adsorbate concentrations (Wang and Guo 2020). This assumption limits the microscopic interpretation of the adsorption process, especially in scenarios where multiple adsorbates can simultaneously interact with the same active site, thereby influencing the interpretation of the adsorption mechanism. To address the limitations of these classical interpretations, advanced statistical physics models have been introduced. These models offer a new physicochemical perspective and theoretical insights into the mechanism of water pollutant removal (Wang et al. 2023). For example, Guedidi et al. conducted a study on the adsorption of ibuprofen on activated carbon using a classical isotherm equation (Guedidi et al. 2013). This equation was used to analyze the performance of the adsorbent and describe potential interactions during the adsorption process (Guedidi et al. 2013). The same adsorption system was further analyzed using a multilayer model to provide additional insights into the adsorption mechanism at the molecular level (Sellaoui et al. 2021a). In summary, the limitations of traditional adsorption process modeling underscore the need for more realistic and comprehensive fundamentals that can more accurately describe the mechanisms involved in the removal of water pollutants.

This paper investigates the adsorption processes of ATP and DFC, two relevant pollutants, using AC prepared from avocado biomass residues. The theoretical analysis was conducted at the molecular level, interpreting the physicochemical parameters derived from a multilayer adsorption model. The primary focus of this article is to provide an accurate characterization of the adsorption mechanisms of these two pollutants, utilizing the principles of statistical physics.

Determination of adsorption isotherm of pharmaceuticals on AC

The adsorption equilibrium of ATP and DFC on AC was experimentally studied under batch operating conditions. These equilibrium studies were conducted at temperatures ranging from 20 to 40 °C and a pH 7, using an AC dosage of 2 g/L and a contact time of 24 h. Initial pollutant concentrations of up to 600 mg/L were employed in these experiments. AC was produced by pyrolyzing avocado seed biomass at 950 °C for 3 h, followed by CO_2 activation at 800 °C for 1 h. This biomass was used as AC precursor due to its wide availability and low-cost, which also contributed to minimize its generation as an agricultural waste. The quantification of pollutants was performed using high-performance liquid chromatography (HPLC), following the methodology outlined in previous studies (Gabriela Elvir-Padilla et al. 2023). The molecular structures of these pharmaceuticals are shown in Fig. 1. A mass balance for the batch adsorber facilitated the calculation of adsorption capacities for these pollutants.

ATP and DFC adsorption isotherm description

Figure 2 presents the experimental isotherms for the adsorption of these pollutants. The experimental data suggest that the adsorption of ATP and DFC involves a mass transfer phenomenon, leading to the saturation of the adsorbent surface. This is evidenced by the asymptotic value of the adsorption capacity. It is important to note that this trend originates from the occupation of the main active sites on the AC during the removal process of ATP and DFC. This surface saturation could be due to the formation of a certain number of layers of adsorbed pollutant molecules on the AC

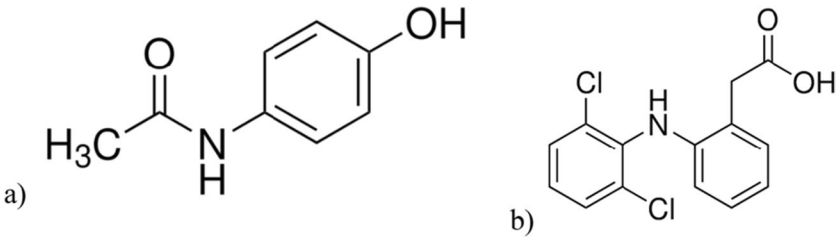

Fig. 1 Molecular structures of **a**) ATP and **b**) DFC

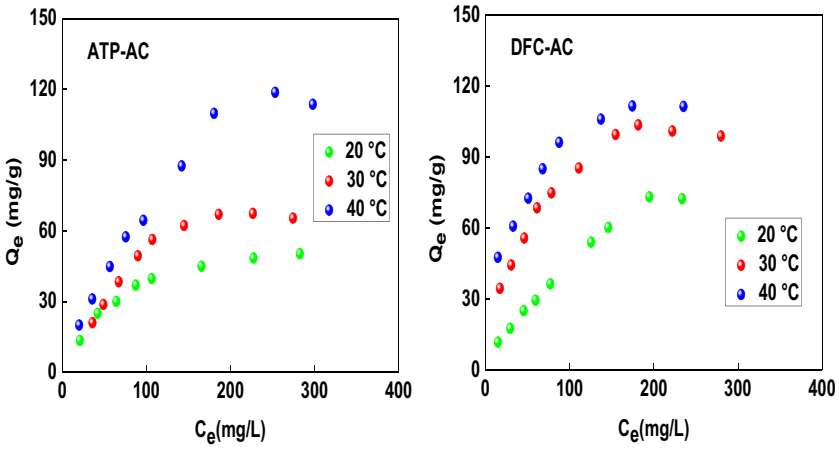

Fig. 2 Experimental isotherms of ATP and DFC adsorption on AC

surface. The theoretical evaluation of the pollutant adsorption data was performed using statistical physics models, which will be discussed in the subsequent sections.

Advance statistical physics adsorption models and data fitting procedure

Three sophisticated adsorption models, derived from the principles of statistical physics, were utilized and juxtaposed to elucidate the adsorption of ATP and DFC on AC. Detailed descriptions of these models follow.

Monolayer model (M1) The Monolayer Model (MM) establishes that the removal of ATP and DFC molecules via AC implies the formation of a single adsorbate layer, facilitated by an interaction energy. Therefore, the interactions between the adsorbate molecules and the functional groups on AC surface were described by a singular adsorption energy value ΔE. The equilibrium adsorption capacity as per this model is expressed as follows (Dhaouadi et al. 2020):

$$Q_{eq} = \frac{nD_{ph}}{1 + \left(\frac{C_{1/2}}{C_{eq}}\right)^{n}} \quad (1)$$

where the number of adsorbed ATP and DFC molecules per surface functional group is represented by n, D_{ph} is the adsorption functional group density for AC surface, and $C_{1/2}$ is the half-saturation concentration of adsorbed PCs layer formed on AC surface.

Double layer model (M2) The Dual Layer Model (DLM) postulates that the adsorption of PCs results in the formation of two layers of PC molecules on the AC surface, which contrasts with the first model where a single layer is formed. The interaction between the PC molecule and the AC functional group is characterized by the first energy term, ΔE_1. Conversely, the second adsorption energy, ΔE_2, delineates the interactions between PC molecules. As a result, the adsorption capacity is expressed as follows (Sellaoui et al. 2015):

Table 1 Determination coefficient (R^2) for the modeling of PCs adsorption on AC using different statistical physics models

R^2				
Adsorption system	**Temperature, °C**	**M1**	**M2**	**M3**
ATP-AC	20	0.996	0.995	0.994
	30	0.987	0.985	0.996
	40	0.974	0.971	0.972
DFC-AC	20	0.986	0.985	0.992
	30	0.968	0.963	0.980
	40	0.967	0.959	0.997

$$Q_{eq} = nD_{ph}\frac{\left(\frac{C_{eq}}{C_1}\right)^n + 2\left(\frac{C_{eq}}{C_2}\right)^{2n}}{1+\left(\frac{C_{eq}}{C_1}\right)^n + \left(\frac{C_{eq}}{C_2}\right)^{2n}} \quad (2)$$

where the half-saturation concentrations associated with the first and second layers are given by C_1 and C_2, respectively.

Multilayer model (M3) The Multi-Layer Adsorption Model (MLAM) assumes that the adsorption of PCs results in the formation of multiple adsorbate layers, denoted as $N_t = 1 + N_2$, where N_2 represents the number of successive layers formed after the initial layer. MLAM employs two energy terms to characterize the interaction bindings: DFC-AC, ATP-AC, DFC-DFC, and ATP-ATP. These energy terms correspond to specific interactions between the adsorbate molecules and the adsorbent surface. DFC-AC and ATP-AC interactions may involve different types of binding energies between the adsorbate and the adsorbent, while DFC-DFC and ATP-ATP interactions could represent interactions within the adsorbate layers themselves. The equation for determining the adsorption capacities is as follows (Sellaoui et al. 2015):

$$Q_{eq} = nD_{ph}\frac{\frac{-2\left(\frac{C_{eq}}{C_1}\right)^{2n}}{1-\left(\frac{C_{eq}}{C_1}\right)^n} + \frac{\left(\frac{C_{eq}}{C_1}\right)^n\left(1-\left(\frac{C_{eq}}{C_1}\right)^n\right)}{\left(1-\left(\frac{C_{eq}}{C_1}\right)^n\right)^2} + \frac{2\left(\frac{C_{eq}}{C_1}\right)^n\left(\frac{C_{eq}}{C_2}\right)^n\left(1-\left(\frac{C_{eq}}{C_2}\right)^{nN_2}\right)}{\left(1-\left(\frac{C_{eq}}{C_2}\right)^n\right)} - \frac{N_2\left(\frac{C_{eq}}{C_1}\right)^n\left(\frac{C_{eq}}{C_2}\right)^n\left(\frac{C_{eq}}{C_2}\right)^{nN_2}}{\left(1-\left(\frac{C_{eq}}{C_2}\right)^n\right)} + \frac{\left(\frac{C_{eq}}{C_1}\right)^n\left(\frac{C_{eq}}{C_2}\right)^{2n}\left(1-\left(\frac{C_{eq}}{C_2}\right)^{nN_2}\right)}{\left(1-\left(\frac{C_{eq}}{C_2}\right)^n\right)^2}}{\frac{\left(1-\left(\frac{C_{eq}}{C_1}\right)^{2n}\right)}{\left(1-\left(\frac{C_{eq}}{C_1}\right)^n\right)} + \frac{\left(\frac{C_{eq}}{C_1}\right)^n\left(\frac{C_{eq}}{C_2}\right)^n\left(1-\left(\frac{C_{eq}}{C_2}\right)^{nN_2}\right)}{\left(1-\left(\frac{C_{eq}}{C_2}\right)^n\right)}} \quad (3)$$

where C_1 represents the half-saturation concentration of the initial layer on AC surface, and C_2 represents the half-saturation concentration associated with the formation of subsequent N_2 adsorbate layers.

DLM, MLAM and MM models were utilized to fit the PCs equilibrium data for predicting the values of n, D_{ph} and N_2 that characterize the removal process. The determination coefficient (R^2) values obtained from the nonlinear data regression for the tested models and adsorption systems are reported in Table 1.

The coefficient of determination, R^2, varied between 0.959 and 0.997, suggesting a satisfactory alignment between the experimental data and the evaluated adsorption models. However, upon closer examination, it was observed that certain physicochemical parameters of MM and DLM lacked a comprehensive physical rationale, which could potentially lead to erroneous interpretations of the PCs adsorption mechanisms. MLAM emerged as the most suitable model for deciphering the ATP and DFC adsorption mechanisms. Table 2 provides the parameters of MLAM, and Fig. 3 presents the results of the data fitting.

Results and discussion

n and D_{ph} parameters for PCs adsorption on AC

The influence of the adsorption temperature on the parameters "n" and "D_{ph}" is depicted in Figs. 4 and 5, respectively. The "n" parameter facilitates the identification of the adsorption configurations of ATP and DFC molecules on the AC surface. PCs molecules (ATP and DFC) can adhere to AC functional groups via different configurations, contingent upon their molecular structures and the temperature employed during their removal. These configurations are delineated as follows: 1) when the value of "n" is less than 0.5, AC active sites can interact with a fraction of ATP or DFC molecules, culminating in a parallel configuration on the AC surface. 2) A mixed adsorption configuration

Table 2 MLAM parameters for ATP and DFC adsorption on AC

T (°C)	R^2	n	D_{ph}(mg/g)	C_1 (mg/L)	C_2 (mg/L)	N_2	Q_s (mg/g)
DFC-AC system							
20	0.992	1.576	13.88	15.038	119.005	2.856	84.349
30	0.980	2.279	18.663	10.039	72.793	1.455	104.418
40	0.997	3.069	18.171	8.397	66.631	1.022	112.760
ATP-AC system							
20	0.994	2.068	12.466	21.067	100.721	1.045	52.710
30	0.996	2.33	12.87	32.48	84.13	1.310	69.270
40	0.972	4.076	10.66	22.4568	120.534	1.682	116.533

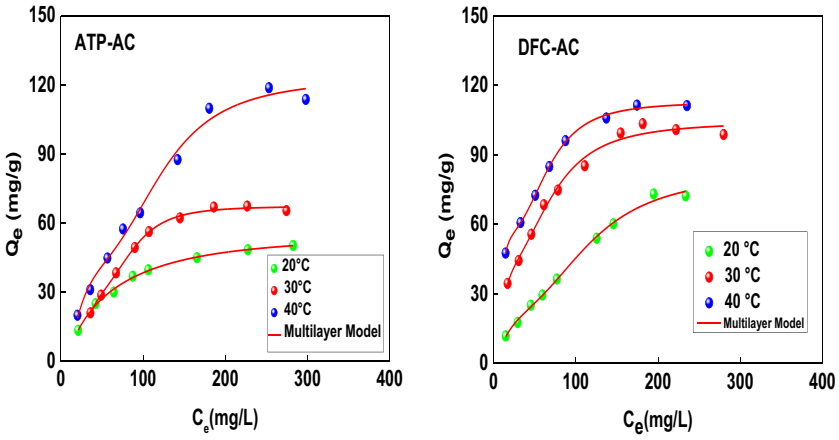

Fig. 3 Results of ATP and DCF isotherm data fitting using a multi-layer statistical physics model

(both parallel and non-parallel) can occur if the "n" parameter ranges from 0.5 to 1. 3) A non-parallel configuration is anticipated during the adsorption process when "n" > 1 (Dhaouadi et al. 2020, 2021).

Table 2 reveals that the computed "n" values were > 1 for both ATP and DFC molecules across various adsorption temperatures, implying a non-parallel configuration on the AC surface. It can be inferred that each functional group on the AC surface has the potential to bind multiple molecules, thereby suggesting that the adsorption of ATP or DFC is a multi-molecular process. However, the number of ATP and DFC molecules bonded to AC functional groups was influenced by the adsorption temperature. In general, the quantity of adsorbed ATP and DFC molecules improved with an increase in the temperature of the aqueous solution. The rise in solution temperature played a pivotal role in facilitating the removal of a greater number of pollutant molecules from the treated fluid. Furthermore, it was deduced that both ATP and DFC formed molecular aggregates during adsorption. For instance, the estimated values of the number of DFC molecules adsorbed per AC functional group were 1.57, 2.27, and 3 at 20, 30, and 40 °C, respectively. The degree of DFC aggregation amplified with the temperature of the aqueous solution, transitioning from a monomer to a trimer.

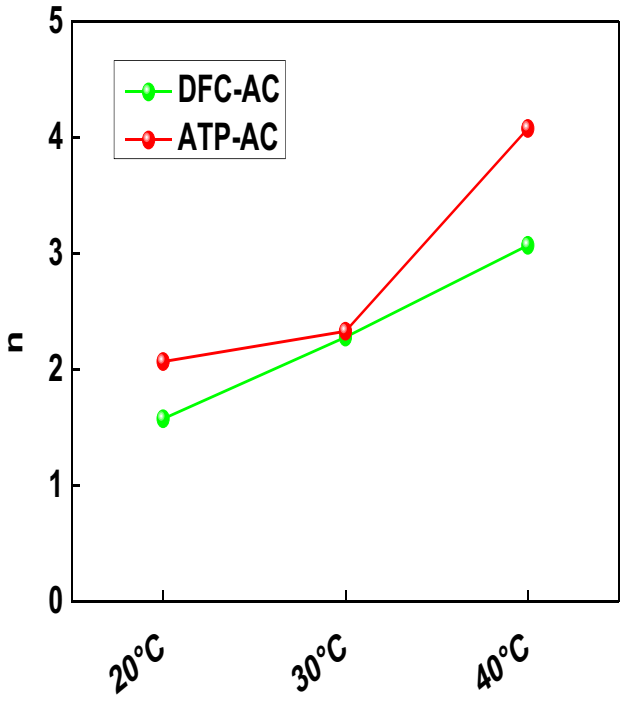

Fig. 4 Influence of temperature on the n parameter for ATP and DFC adsorption on AC

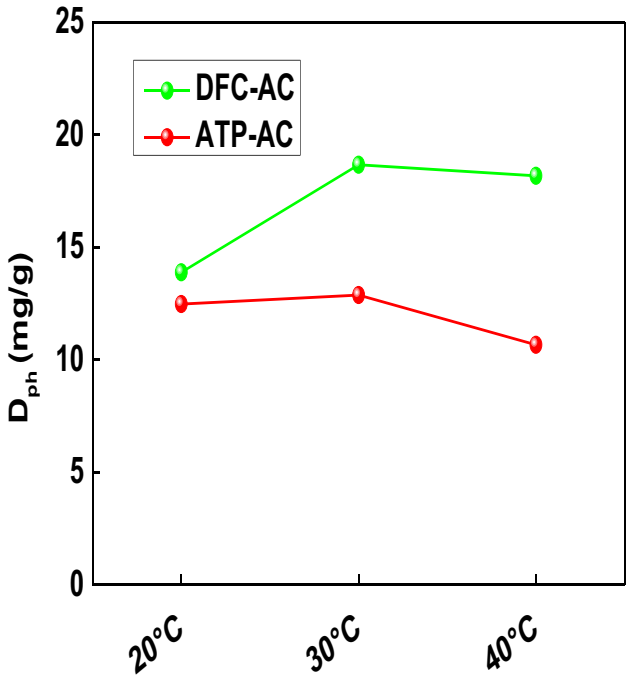

Fig. 5 Influence of temperature on the D_{ph} parameter for ATP and DFC adsorption on AC

Figure 5 delineates the influence of the solution temperature on "D_{ph}." It was discerned that the densities of the AC functional groups, which were involved in the removal of both PCs, exhibited a marginal increase as a function of the temperature of the aqueous solution. This increment is likely attributable to the activation of additional functional groups by the increase in adsorption temperature, thereby facilitating the removal of ATP and DFC pollutants.

N_2 parameter for PCs adsorption on AC

MLAM provided insights regarding the total number of adsorbed layers of ATP and DCF on the AC surface. This parameter was derived from the relationship $N_t = 1 + N_2$, and its variation as a function of the adsorption temperature is depicted in Fig. 6. The total number of formed PCs layers varied from 2.02 to 3.86 for DFC-AC, and from 2.06 to 2.68 for ATP-AC. An increase in temperature resulted in a decrease in the number of adsorbed DFC layers, while an inverse trend was observed for ATP layers. For instance, the total number of formed DFC layers on AC was 2.45 at 30 °C. This value suggested that the adsorption process involved the formation of 2 and 3 layers on the adsorbent surface. The distribution of these layers for this specific case can be determined using y = 2.45, where "y" represents the percentage of DFC molecules forming 2 layers and (1-y) denotes the percentage of adsorbed molecules via three layers (Sellaoui et al. 2017). It was determined that 55% of DFC molecules were incorporated into 2 layers, while the residual 45% constituted 3 layers. This outcome indicated the heterogeneous formation of PCs layers on the AC surface. Analogous results were observed for other adsorption systems under the experimental conditions tested.

Q_{sat} parameter for PCs adsorption on AC

The parameters n, D_{ph}, and N_t were employed to compute the saturation adsorption capacities of ATP and DCF, per the established relationship: $Q_{sat} = n \times D_{ph} \times N_t$ (Sellaoui et al. 2021a). The findings are illustrated in Fig. 7. The increase in the aqueous solution temperature corresponded to a rise in both ATP and DCF saturation adsorption capacities. This pattern can be linked to the augmentation of molecular mobility. Higher temperatures supply more kinetic energy to the adsorbate molecules and stimulate the thermal motion of ATP and DCF molecules, thereby promoting their interaction with the adsorbent surface (Ebelegi et al. 2020; Al-Harby et al. 2021). AC demonstrated superior DFC adsorption capacities compared to ATP. This can be attributed to several factors, including the molecular size and the affinity of the adsorbent for these molecules of persistent organic pollutants. The performance of AC was evaluated and compared with the results from literature for the removal of these compounds using other adsorbents (refer to Table 3). This particular AC outperformed the adsorption capacities of various other adsorbents, as documented in previous studies. Note that this avocado-based AC showed adsorption capacities similar or higher than other carbon-based adsorbents prepared from different biomass precursors (Ajiboye

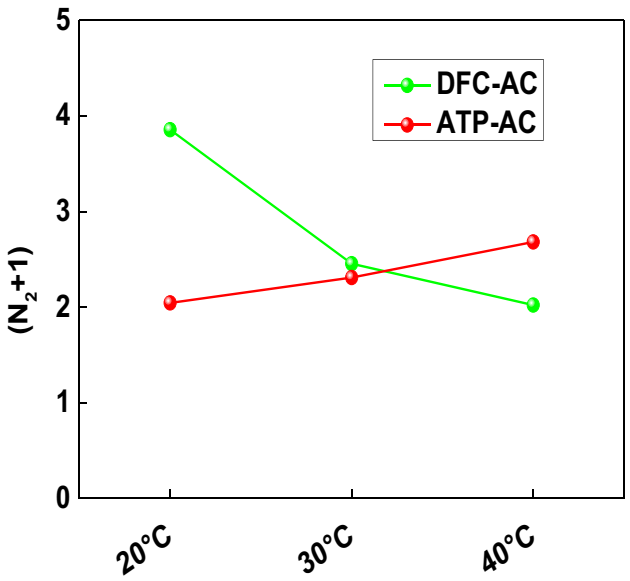

Fig. 6 Influence of temperature on the $1 + N_2$ parameter for ATP and DFC adsorption on AC

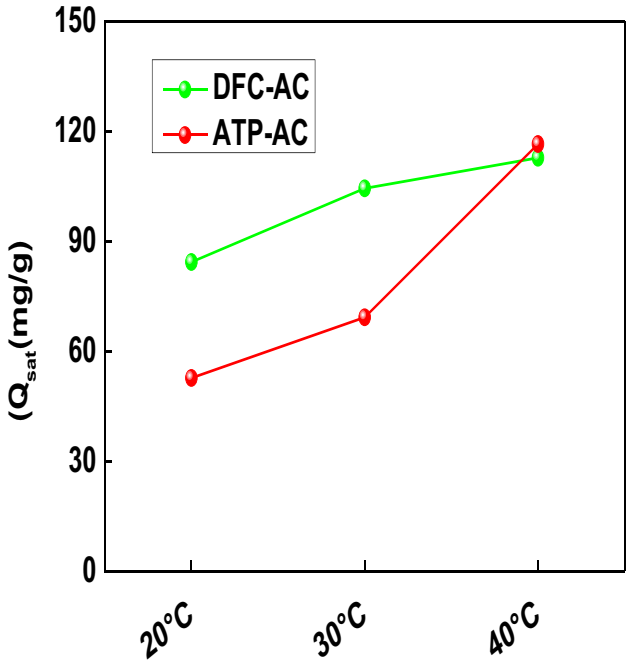

Fig. 7 Influence of temperature on the saturation ATP and DFC adsorption capacities using AC

Table 3 Performance comparison of the maximum adsorption capacities of different adsorbents to remove pharmaceutical molecules

Pharmaceutical molecule	Adsorbent	Q_{max} (mg/g)Q_{max} (mg/g)	Reference
ATP	AC	116.53	**This study**
DCF	AC	112.76	**This study**
ATP	Silica gel	95.00	(Spaltro et al. 2021)
ATP	Carbon nanotube	58.82	(Malhotra et al. 2018)
ATP	Granular Zeolites	90	(Fu et al. 2021)
ATP	Moringa oleifera Lam. biomass	17.48	(Quesada et al. 2019)
DCF	Cellulose fiber	65.19	(Pires et al. 2017)
DCF	Tea waste	74.60	(Malhotra et al. 2018)
DCF	Isabel grape bagasse	76.98	(Antunes et al. 2012)
DCF	Reduced graphene oxide	59.67	(Jauris et al. 2016)
DCF	Granular carbonnanotubes/alumina composite	33.86	(Wei et al. 2013)

Table 4 Adsorption energies calculated for ATP and DFC adsorption on AC surface

Temperature, °C	ΔE_1 (kJ/mol)	ΔE_2 (kJ/mol)
DFC-AC		
20	19.720	14.690
30	21.410	16.427
40	22.580	17.199
ATP-AC		
20	16.672	12.867
30	16.152	14.213
40	17.644	13.279

et al. 2024). Consequently, this material holds promise as a potential adsorbent for the removal of DFC and ATP in practical applications.

Energy assessment for ATP and DFC adsorption on AC

MLAM model provides further understanding of the adsorption process by estimating the adsorption energies. These energies were computed at various temperatures to characterize potential interactions between these organic molecules and AC surface. This also includes the interactions among the adsorbate molecules themselves, forming what is known as an adsorbate–adsorbate aggregate. The formulas utilized to calculate these energies are as follows (Sellaoui et al. 2021b):

$$\Delta E_1 = RT \ln\left(\frac{C_s}{C_1}\right) \quad (4)$$

$$\Delta E_2 = RT \ln\left(\frac{C_s}{C_2}\right) \quad (5)$$

where $R = 8.314$ J/mol·K denotes the ideal gas constant, C_s represents the solubilities of DFC and ATP in mg/L. Table 4 displays the calculated adsorption energies, ΔE_1 and ΔE_2, for the investigated systems. Calculated ΔE_1 and ΔE_2 varied between 12.86 to 22.58 kJ/mol. These adsorption energy values can be associated with physisorption, in which van der Waals forces and hydrogen bonds are expected to be the main contributors to pollutant removal. Note that $\Delta E_1 > \Delta E_2$, indicating that the adsorbent-adsorbate interaction was stronger than the interaction between the adsorbate molecules forming aggregates.

Conclusions

A comprehensive analysis of statistical physics was conducted to study the adsorption of two pharmaceutical compounds on activated carbon derived from avocado biomass. This activated carbon demonstrated experimental adsorption capacities ranging from 112.76 to 116.53 mg/g for the removal of these water pollutants. Statistical physics modelling revealed a multilayer adsorption mechanism for these pharmaceutical molecules, with molecular aggregation also observed. This activated carbon displayed superior adsorption properties for the removal of diclofenac. The process of water depollution for diclofenac and acetaminophen using this activated carbon was linked to physisorption. Here, hydrogen bonding and van der Waals interactions are likely to play a key role in pollutant adsorption. This study has contributed valuable insights towards understanding the removal of emerging pollutants, such as pharmaceuticals, using carbon-based adsorbents.

Author contribution All co-authors (Rihab Ghorbali, Lotfi Sellaoui, Houcine Ghalla, Adrian Bonilla-Petriciolet, Radames Trejo-Valencia, Alejandro Sánchez-Barroso, Shuguang Deng, Abdelmottaleb Ben Lamine) contributed in all parts of the manuscript.

Data availability Note applicable.

Declarations

Ethics approval Not applicable.

Consent to participate Not applicable.

Consent for publication Not applicable.

Competing interests The authors declare no competing interests.

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