



Adsorption Ability of Cephalexin onto the Straw-Based Activated Carbon: Performance and Mechanism

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A straw-activated carbon has been successfully synthesized with the high BET surface area, at 494.9240 m²/g, which is perfectly suitable for the adsorption of cephalexin antibiotic from aqueous water. It is noted that the adsorption capacity of straw-activated carbon is demonstrated by the effect of initial concentration, contact time, pH solution and dosage. The straw-activated carbon exhibited improved decontaminant efficiency towards cephalexin antibiotics. Quick and improved sorption could be attributable to the distinctive structural and compositional merits as well as the synergetic contribution of functional groups to surface material. Most interestingly, the adsorption capacity achieved at pH 6 was ~98.52%. A mechanism adsorption has been proposed to demonstrate adsorption of the straw-activated carbon (AC-S). By comparison with other studies, it is confirmed that AC-S in this study obtained a higher removal efficiency than other adsorbent materials, suggesting that straw-activated carbon may be an appropriate candidate to treat cephalexin from wastewater media.

Keywords: Straw biomass, Activated carbon, Chemical activation, Cephalexin.

INTRODUCTION

Antibiotics have been widely used in husbandry and agriculture; however, the fate and occurrence of these compounds in aqueous systems *i.e.* streams or rivers can result in a wide range of severe environmental pollution issues [1,2]. Among which, many have been identified as carcinogens and are difficult to remove from wastewater even at low concentration [3,4]. Cephalexin, which is an antibiotic that was prevalent among home use, has been extensively studied for removal from the aqueous solution, but the performance and effectiveness are limited [5,6]. Of many treatment techniques, adsorption by porous material has been considered as the optimal method due to its low cost and energy, excellent removal efficacies and environmental friendliness [7-9]. Indeed, the adsorptive removal of cephalexin molecules has paid much attention from many scientists. Liu *et al.* [10] synthesized Cu(II)/Fe(III) impregnated activated carbons developed from lotus stalks to eliminate cep-

alexin from aqueous solutions. Wu *et al.* [11] also fabricated a novel nanomaterials derived from modified biochar supported Ag/Fe nanoparticles for the treatment of cephalexin. Moreover, liquid membrane combined with strip dispersion have been developed and this technique was expected to have a favourable advantage to treat cephalexin molecules [12].

Activated carbons are an amorphous carbonaceous source, which can be regarded as versatile adsorbents [13-17]. The excellent adsorption properties of activated carbon are attributable to their porosity and the presence of graphite lattice microcrystallites, generally prepared in tiny pellets or powder [18, 19]. Recently, a great deal of attention has been paid to the manufacture of activated carbon from agricultural wastes, especially straw, a cheap, renewable and abundant waste in tropical countries [20]. As a result, there have been many studies reported the application of activated carbons for environmental remediation. Kim *et al.* [21] reported the use of hierarchically structured activated carbon for ultracapacitors, which helped to

hamper the transport of the electrolyte ions. At the same trend, Li *et al.* [22] modified activated carbon by nitrogen doping, obtaining very high surface areas ($2900 \text{ m}^2 \text{ g}^{-1}$) for a high energy hybrid supercapacitor. Activated carbon with chemical modification prepared from sucrose spherical carbon ($1534 \text{ m}^2 \text{ g}^{-1}$) has been proved as a very efficient adsorbent to remove methylene blue with 704.2 mg g^{-1} [23]. Maneerung *et al.* [24] reported the application of activated carbon from biomass gasification, which obtained activated carbon has high surface area (776.46 mg g^{-1}) and abundance of hydroxyl and carboxyl groups (phenol, carboxyl, lactone, base), and could be used for removing rhodamine B through kinetics, isotherms and thermodynamic studies.

Despite numerous studies that have been conducted to synthesize activated carbon from various materials and evaluate its adsorption towards toxic synthetic colors and heavy metals in the aqueous solution [25,26]. For example, Demiral *et al.* [27] removed Cu(II) ions efficiently onto activated carbon prepared from grape bagasse. Despite of high surface area and pore volume ($0.876 \text{ cm}^3/\text{g}$) of activated carbons, the maximum capacity for copper adsorption was obtained to be very low, at only 43.47 mg/g at 45°C , which was assumed by the Langmuir and Dubinin-Radushkevich isotherm equations. With the use of phosphoric acid as an efficient activation, Yakout and El-Deen [28] proved that modified activated carbon from olive stones could enhance the porosity, towards high surface area and pore volume up to $1218 \text{ m}^2 \text{ g}^{-1}$ and $0.6 \text{ cm}^3 \text{ g}^{-1}$, respectively by changing phosphoric acid dilute solutions (60, 70 and 80%). There is, however, a comparative absence of data in the literature on the option of generating the straw and implementation of activated carbon in antibiotic treatment. Therefore, a study describing the surface structure of activated carbons, the adsorption phenomenon onto the activated carbon of organics and inorganics from gaseous and aqueous stages is considered appropriate.

In present study, factors that are influential to adsorption capability of activated carbon have been studied. The variables consisted of contact time, cephalexin concentration, pH solution and dosage straw-activated carbon (AC-S). In addition, Boehm's titration method was employed to determine the amount of functional groups adsorbed onto the surface material. Moreover, a mechanism has been proposed to prove adsorption ability of this potential material. To our best of knowledge, the utilization of straw-based carbons for cephalexin treatment was not previously reported; and hence, more investigations and optimization assessments need to be conducted to elucidate the adsorption process of cephalexin.

EXPERIMENTAL

The straw biomass was obtained from Ben Tre Province, Vietnam. All chemicals including cephalexin, KOH pellets, and HCl (36%) were obtained commercially from Merck. The N_2 adsorption/desorption isotherm and pore size distribution data were recorded on the Micromeritics 2020 volumetric adsorption analyzer system. The SEM images with the magnification of 7000 were captured with the S4800 instrument (Japan) with an accelerating voltage source (15 kV). The infrared FT-IR spectra obtained by the Nicolet 6700 spectro-photometer were

used to explore characteristics of chemical bonds and functional groups. Cephalexin concentration was determined with UV-vis spectrophotometer at wavelength of 268 nm.

Synthesis of straw shell based activated carbon: Typically, the straw was gleaned from fields in Ben Tre, Vietnam. The precursor was then washed with water for many times. This was to remove the dirt, and impurities from them. Next, the samples were dried in an oven at 105°C during 2 days to make sure that the water in the straw body was vaporized completely.

The procedure for the pyrolysis of straw could be described as follows: The dried sample (10 g) was put on a glass tube, which was connected with a furnace and the nitrogen was allowed to flow continuously ($400 \text{ cm}^3 \text{ min}^{-1}$). The glass tube was gradually heated up at 500°C from the room temperature (5°C min^{-1}) during 4 h and then was cooled overnight. The black char sample was then unloaded and with 50 mL KOH solution (biochar: solid KOH = 1:1 by wt.%).

After impregnation process of 1 day, the KOH-modified char was activated *via* carbonization for 4 h. Again, the sample put on a glass tube of the same pyrolysis system, gradually heated up at 500°C from the room temperature (5°C min^{-1}) during 4 h, and then was cooled. Carbonized sample after being soaked in nitrogen at 500°C (with a heating rate of 10°C/min). The sample was then washed by distilled water and HCl to neutral pH and dried at 105°C . The final sample of activated carbon (AC) was referred to as AC-S and used for the next steps including structural characterization, point of zero charge experiment, Boehm's titration for identifying the surface functional groups, adsorption experiments and mechanism.

Adsorption batch: The AC-S's adsorption capacity was studied with regard to different factors including adsorbent concentration, contact time, pH solution and dosage. Herein, into an Erlenmeyer flask containing 100 mL of cephalexin aqueous solution (10-60 mg/L), the as-prepared AC-S (0.5-2 g/L) were introduced. For these experiments, the pH was adjusted to a range from 2 to 8, by adding solutions of 0.1 M HCl or 0.1 M NaOH and measuring with a pH meter. The spectrophotometer UV-VIS was used to determine the concentration of cephalexin at 268 nm.

$$\text{CFX Removal (\%)} = \frac{C_o - C_e}{C_o} \times 100 \quad (1)$$

where, C_o and C_e are initial and equilibrium cephalexin concentrations (mg/L), respectively.

Boehm's titration: To gain more evidence of functional groups on the surface of AC-S and determine the quantity of oxygenated compounds (*e.g.* phenol, lactone and carboxyl) an acid-base neutralization method, we performed the Boehm titration. The procedure of Boehm titration has been reported in a recent study [29]. In a typical procedure, AC-S materials 1.0 g was added into 250 mL beakers containing 50 mL each of the following solution (0.1 mol/L): NaHCO_3 , Na_2CO_3 and NaOH. It is assumed that NaHCO_3 is only used to neutralize carboxyl groups, Na_2CO_3 is used to neutralize both carboxyl and lactone groups, and NaOH is used to neutralize all carboxyl, phenol, lactone. Since this neutralization process finished within 1 day, the solids were removed and the aqueous residues filtered

and titrated using a standard HCl solution (0.1 mol/L) and NaOH (0.1 mol/L) with methyl orange as an indicator.

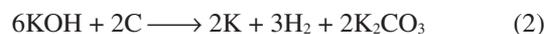
Determination of pH_{pzc} (pH point of zero charges): The pH_{pzc} determination was performed based on a simple procedure as follows. The AC-S ($m_o = 50$ mg) was added a flask containing 25 mL of KCl 0.1 mol/L at the different pH 2, 4, 6, 8, 10, 12, which was initial pH points (pH_i). The solutions were maintained stable within 1 day. After that, the solids were extracted and their final pH points (pH_f) were measured by a pH meter. By plotting the (pH_i) against (pH_f), the pH_{pzc} could be identified at $pH_i = pH_f$.

RESULTS AND DISCUSSION

Characterization of straw-shell activated carbon: BET results of the obtained AC-S are shown in Fig. 1a. Evidently, the measured BET surface area of 494.9240 m^2/g is consistent with previous reports [30] and is explained by the pore size distribution results (Fig. 1b). Similar pattern was also observed for the pore volume. The active carbon's featured characteristics is a random arrangement of microcrystallites and with a strong cross-linking between them have a well-developed porous structure. Their density is relatively low (less than 2 g/cm^3) and their graphitization is low. This porous structure created during the carbonization process is further established during the activation process when are cleared of tar and other carbonaceous material clear the gaps between the elementary crystallites. This leads to increasing surface areas and pore diameter. The improved region of the surface was therefore regarded the reason for its increased specific capacity value [31]. Beside,

as it is shown in Table-1, the SBET obtained in this study is comparable with those reported in the previous literature [32-36].

From Table-1 and Fig. 1, a large quantity of micropores and some mesopores were generated when KOH was used as the activation agent. However, a larger number of mesopores were produced when K_2CO_3 was used. The porosity and surface groups of the activated materials can be managed using various activation agents. Metallic potassium formed during the gasification process was assumed to diffuse the existing pores into the internal structure of the carbon matrix and create new porosities, according to this following reaction (eqn. 2):



These reactions explain the reasons for KOH activation prepared by high surface area and high pore volume carbon structure compared to that obtained by K_2CO_3 activation [36].

To sum up, KOH is the best activator to activated as well as make increase surface area of carbon materials in this study.

Evaluation of adsorption efficiency of cephalexin onto AC-S: Prior to testing the cephalexin adsorption by AC-S, an experiment was carried out to investigate the solution pH according the previously reported method [37] to determine pH_{pzc} . indeed, pH_{pzc} is the point at which the experimental curve ($pH_{initial}$ vs. pH_{final}) intersects the 45° line. This is an important parameter for any activated carbon due to it shows the adsorbent acidity and basicity, and the net surface charge carbon load in solution [38]. Herein, using HCl and/or NaOH 0.1N solutions to adjust the pH value of solution. For this, 0.1 g of

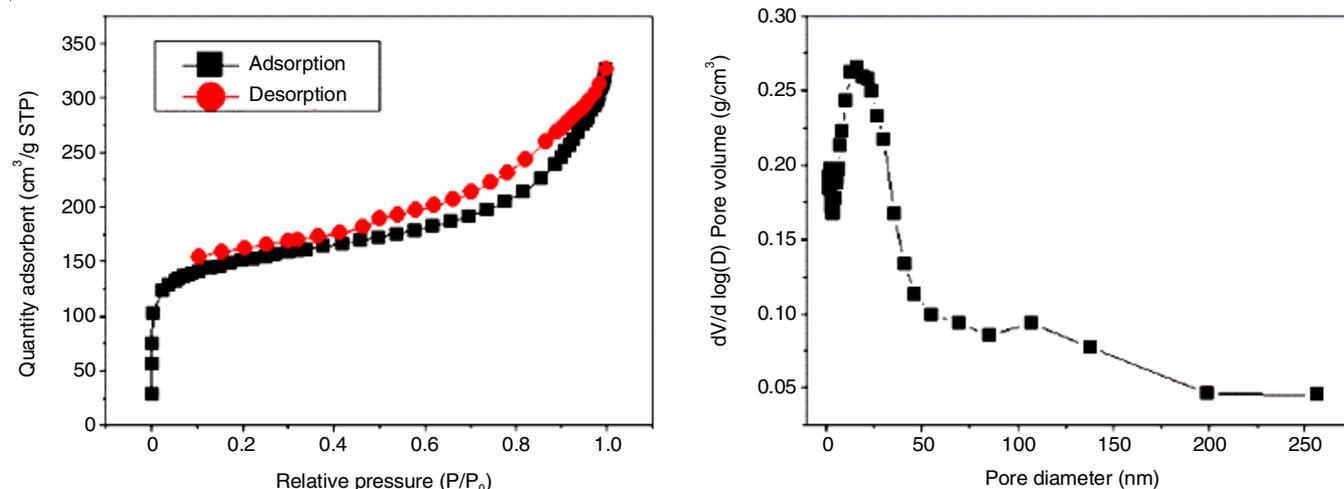


Fig. 1. N_2 adsorption/desorption isotherm (a) and pore size distribution (b) of AC-S

TABLE-1
COMPARISON OF S_{BET} OF ACTIVATED CARBON BASED STRAW AND OTHER
ACTIVATED CARBON USING DIFFERENT KINDS OF RAW MATERIALS

Material	Activator	BET surface area (m^2/g)	Ref.
Activated carbons from <i>Albizia lebbbeck</i> seed pods	KOH	1824.88	[32]
Rice husk	KOH	750	[33]
Activated carbon prepared by different types of activation	K_2CO_3	160.925	[34]
Pine apple	K_2CO_3	680.000	[35]
Cotton stalk	K_2CO_3	621.470	[36]
Activated carbon based straw	KOH	494.9240	This work

AC-S is added to 100 mL of 0.1N KOH solution with varying pH from 2 to 12 and stirred for 24 h. The obtained pH_{pzc} was 7.05 (Fig. 2a). In this regard, AC- Straw at pH values less and more than 7.05 demonstrates cationic and anionic behavior, respectively. The variation of removal values of cephalixin with respect to pH is shown Fig. 2b. In this study, the pH was allowed to vary from 2 to 8 while the concentration and contact time was kept at 10 mg/L and 90 min, respectively. From the line graph shown in Fig. 1b, the highest range of antibiotic removal was established when the pH increases from 2 to 6. Conversely, the antibiotic removal is reduced significantly when the pH rose past 6 to 7 and 8. This may be explained based on depends on pH_{pzc} value. Specifically, at $pH < pH_{pzc}$, the activated carbon straw surface has a net positive charge, contrary to at $pH > pH_{pzc}$ the surface has a net negative charge [39]. Under the acidic adsorption condition, increased positive charged environment could raise the density of positive ions (H^+) around the activated carbon molecules [40].

From activated carbon surface infrared research, it has been shown that the hydroxyl surface groups can be coordinated into one, two, or three cephalixin molecules, resulting in very distinct reactivity locations. Moreover, increasing pH promotes deprotonation of loaded amino and enol groups on the molecule of cephalixin and on AC-S carboxyl group. This leads to suppressed cation- π bonding and π - π stacking with AC-S, and in turn, reduced electron-acceptor capacity of these moieties. In addition, it was revealed that cephalixin with AC-S was considerably complexed only at $pH > 9$, which is well beyond the pH range in the study. This may explain for the highest adsorption capacity achieved at pH 6 about 98.66%. Thus, neutral condition is favorable for cephalixin seeds to perform its adsorption process.

According to Fig. 2c, with increasing adsorption time, the high activity sites on the activated carbon surface were first saturated, followed by the diffusion of the adsorbed quantity into the activated carbon interior afterwards.

One of the extremely important variables responsible for the proportion of antibiotic removal is the initial concentration. The initial concentration was manipulated from 10 mg/L to 60 mg/L, while the initial solution pH study was carried out by contacting 0.1 g of AC-S with 100 mL of 10 mg/L cephalixin solutions for 4 h. As shown in Fig. 2d, it can be seen that the increase in the initial concentration decreased the dye removal percentage of cephalixin. This phenomenon could relate to available surface area. According to Contescu *et al.* [41], the antibiotic removal will drop when the antibiotic concentration reaches a saturation point. At this point, the number of active surface sites can decrease, limiting the adsorption on the adsorbent or to the cephalixin diffusion rate. The results showed that the adsorption capacity of dye decreases from 98.52% down to 53.54% as the initial dye concentration increases from 10 mg/L up to 60 mg/L (Fig. 2d).

Beside, one consequence of changing the dosage have been presented clearly in Fig. 2e. Herein, an initial solution pH study was kept at 6 and carried out by contacting of AC-S with 100 mL of 10 mg/L cephalixin solutions at different values of initial dosage ranging from 0.1 to 0.25 g for 4 h. Interestingly, when increasing dosage AC-S, the yield of removing cephalixin significant increase. This is possibly due to the large surface area and high porous diameter size. However, on the chemical basis, the initial dosage of AC-S provides a specific adsorption site, which can absorb a set quantity of adsorption. Furthermore, another study demonstrated the existence of a point which at

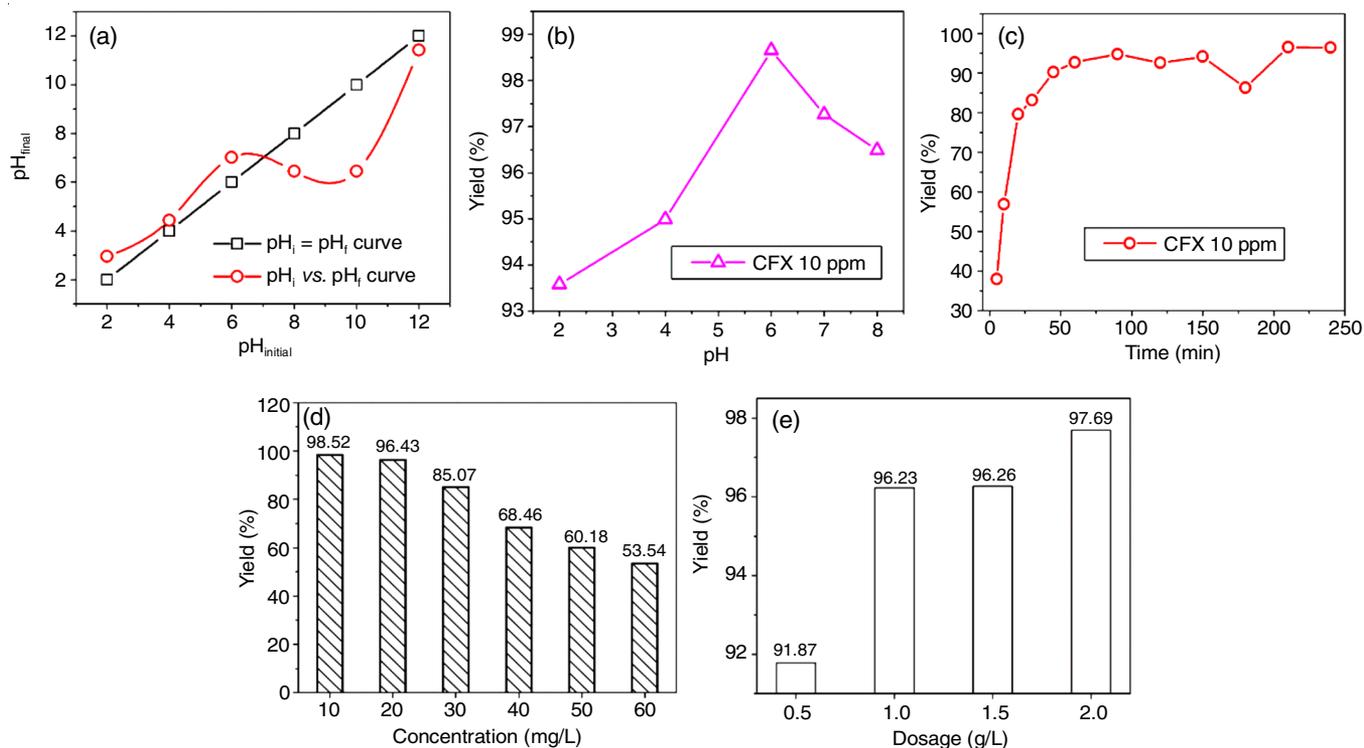


Fig. 2. Effects of the factors toward adsorption capacity of CFX onto AC-S: (a) pH_{pzc} , (b) pH solution, (c) contact time, (d) initial concentration, (e) dosage AC-S

the rise in cephalixin intake did not substantially improve because of the saturation adsorption sites onto the surface AC-S.

Chemical structure of the activated carbon surface strongly determines its adsorption capability. According to Contescu *et al.* [41], Boehm titration method could be used to evaluate the surface acidity of several activated carbons, permitting identification of acidic group in the form of carboxylic, lactones, phenols and basic species. Experimental conditions for the Boehm titration were similar to another procedure [42] Following this, 0.25 g AC-S was immersed for 48 h in HCl, NaOH, NaHCO₃ and NaCl solution at 0.1N, respectively. Observing from the obtained results (Table-2), the number of acidic groups far outstripped the that of basic groups, suggesting that the activated carbon material has acidic properties. The presence of phenol functional group serves two main purposes. First, phenol functional groups, introduced by the coverage of amorphous carbon on AC-S, plays the role as extra accessible complexation sites for the boron species. Second, abundant phenol groups in the prepared amorphous carbon improved cephalixin removal capacity. Current results indicate AC-S as a promising adsorbent materials, being capable of efficient boron remediation by the dehydration of carbohydrates.

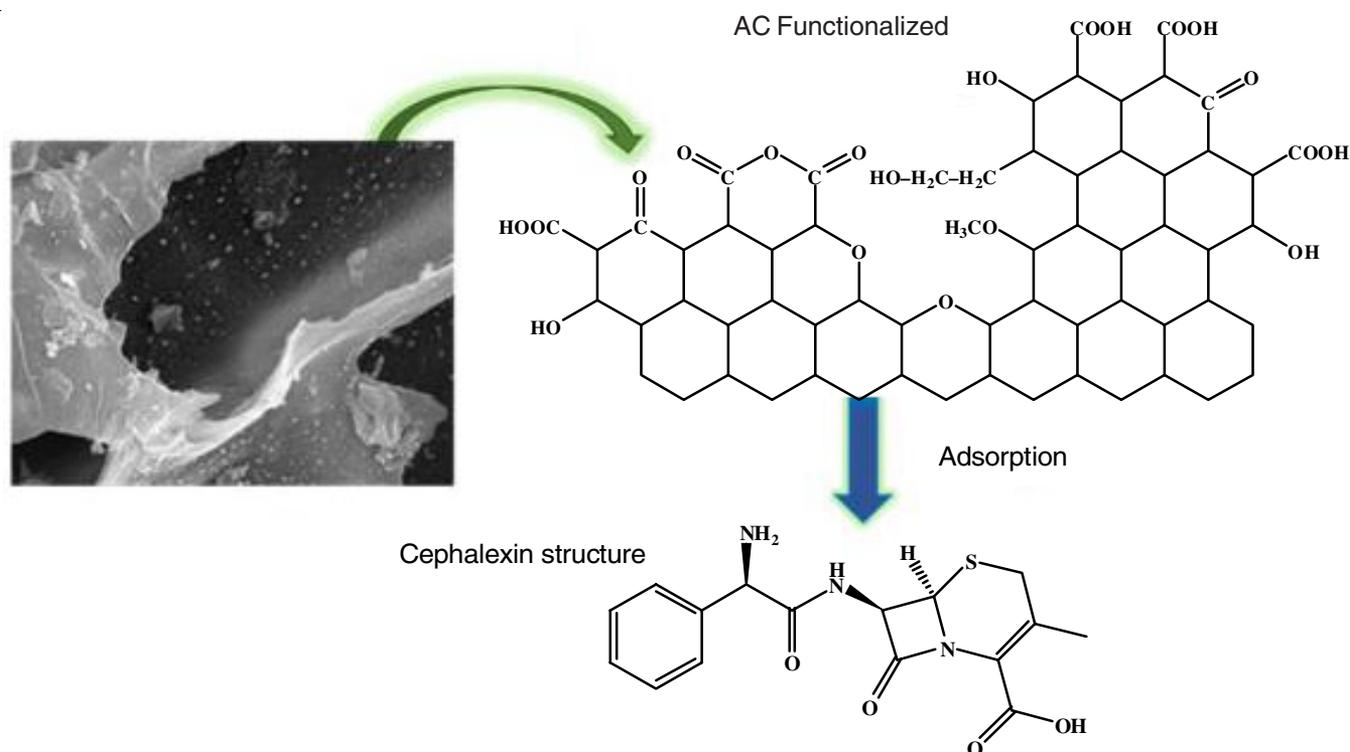
Plausible adsorption mechanism: In order to evaluate and have an in-depth knowledge of the mechanism of adsorption

process between cephalixin molecules and AC-S surface, a propose mechanism was simulated in **Scheme-I**. As observed, the electrostatic interaction between antibiotic molecules and AC-S occurring on the surface AC-S involves a large amount of functional groups and could be explained following three steps: (i) Molecules of antibiotic spread from the liquid to the liquid-solid interfaces ; (ii) molecules of antibiotic move from the liquid-solid interfaces to the solid surfaces; and (iii) molecules of antibiotic diffuse into from the solid surfaces to the particle pores.

As the result, cephalixin molecules were easily captured on the surface of AC-S. As Observed from **Scheme-I**, several types of plausible mechanisms, including formation of H-bonding and the existence of interaction type such as π - π , n- π and pore filling could be elaborated. The presence of aromatic rings, amines and carboxylic groups in structure of cephalixin and the subsistence of two main functional groups containing H-donors (hydrogen atoms belonging to groups such as -OH, -NH₂, -C₆H₄OH) and H-acceptors (electron-rich oxygen or nitrogen-rich atoms such as -CHO, N=N, -COO-) into the surface of AC-S are the reasons explaining the improvement in adsorption capacity. Besides, the creation of hydrogen bonding has helped to increase the ability to adsorb surface material. This bonding took shape between the hydroxyl group surface bonds on the surface of the AC-S and the cephalixin molecules atoms. This results completely suitable with Boehm titration's result and the theory of Matson *et al.* [43] originally proposed in 1969.

In summary, adsorption mechanism is related to surface group interaction/hindrance and surface area where cephalixin can be involved in electronic interactions with activated carbon-based straw.

Carbon material	Basic groups (mmol/g)	Acidic group (mmol/g)			
		Total	Carboxyl	Lactone	Phenol
Straw shell AC	0.032	0.581	0.084	0.195	0.302



Scheme-I: Process for adsorption cephalixin antibiotic drug onto the straw-activated carbon

TABLE-3
COMPARISON OF ADSORPTION CAPACITIES OF DIFFERENT ACTIVATED CARBONS FOR ADSORPTION OF DIFFERENT ADSORBENT

Material	Adsorbate	Yield (%)	Ref.
Activated carbon from cotton stalk	Methylene blue	37.92	[44]
Activated carbon and metal doped TiO ₂	Geosmin	98.00	[45]
	2-MIB	97.00	[45]
Activated carbon based straw	Cephalixin	98.52	This work

Comparative study: To demonstrate the optimum adsorption capacity of AC-S, the yield of adsorption capacities (%) for cephalixin in this study were compared with those reported in the previous literature for different activated carbons prepared from waste agricultural wastes and shown in Table-3. By using activated carbon based straw as adsorbent to treat antibiotics has achieved a very high efficiency of cephalixin removal, which was found to be higher than those of other materials. In detail, Deng *et al.* [44] prepared and characterized the activated carbon from cotton stalk *via* the support by microwave and chemical activators, giving a 37.9% of methylene blue removal efficiency from aqueous solution [44]. Asghar *et al.* [45] used activated carbon and metal doped TiO₂ to remove two adsorbate targets including Geosmin and 2-MIB with high removal efficiency. Meanwhile, present study gave an evidence of the use of straw-activated carbon to enhance 98.5% of cephalixin. Therefore, it has been evaluated as fully promising material in wastewater treatment fields, in special, towards toxic antibiotics like cephalixin.

Conclusion

In this study, the effectiveness of activated carbon derived from straw to remove cephalixin molecules from aqueous solution, which was used as an efficient adsorbent is demonstrated. The material AC-S has been successfully synthesized and characterized with porosity analysis, giving a high surface area (~494 m² g⁻¹). The surface of AC-S adsorbent existed many kind of functional groups such as carboxyl, lactone, phenol and base. To introduce the strength of adsorbability, the effect of other factors were also investigated. Indeed, the impactful variables were statistically significant in evaluating the impact of an autonomous parameters, including concentration, pH solution, dosage and optimizing adsorption circumstance. Under ideal circumstance C₀ = 10 mg/L, dosage AC-S = 2.0 g/L and pH solution = 6, up to 98.52% of cephalixin removal was acquired. Boehm's result also showed an abundance of functional groups which suitable for adsorption application on the surface AC-S. Since straw, an agriculture solid waste, used in this study is locally available; the adsorption process is expected to be economically and friendly for wastewater treatment.

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CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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