Preparation of Activated Carbon from Herbal Residues and Kinetics of Cephalosporin Antibiotic Adsorption in Wastewater

Suimin Li, Qian Yang, and Yong Ye*

In order to utilize the leftover materials from traditional Chinese medicine production and to remove antibiotics from wastewater, extracted herbal residues were used as raw materials to produce activated carbon. To keep a constant quality and adsorption ability across cephalosporin antibiotic samples, all samples were divided into rhizomes, fruits, and leaves to make activated carbon by step-wise carbonization and overheated steam activation. The three kinds of carbon were mixed in the optimal ratio of 5:4:2, which had homogeneity in quality with a high adsorption capacity. Maximum adsorption of the mixture to cefalexin and cefradine were 7.1 mg/g, and the mixture removed 84% of the antibiotics from treated wastewater. Approximately 90% of the antibiotics was desorbed after ultrasonic treatment of the distilled water-carbon mixture for 10 min, and a re-adsorption capacity of 80% was maintained for next use. The adsorption process is dominated by a pseudo-second order kinetic reaction, with two active sites binding to one antibiotic molecule. The rate-limiting step is an intra-particle diffusion process. The isothermal adsorption process conforms to Langmuir and Tempkin isotherm models, showing multilayer and physical adsorption. The activated carbon from herbal residues can adsorb a low concentration of antibiotics in wastewater and be recycled after ultrasonic treatment.

Keywords: Herbal residues; Activated carbon; Adsorption; Cephalosporin antibiotics; Recycling

Contact information: Department of Pharmaceutical Engineering, School of Chemistry and Chemical Engineering, South China University of Technology, Guangzhou 510640, China;
** Corresponding author: yeyong@scut.edu.cn*

INTRODUCTION

In China, antibiotics are used for 70% of all treated clinical diseases, which is two times more than in Europe and the United States. In actuality, antibiotics are only really needed in less than 20% of these cases (Chen *et al*. 2015). The prophylactic use of antibiotics is a typical abuse of these substances; 52% of antibiotic abuse occurs when antibiotics are used prophylactically for animals, while the remaining 48% occurs within the human population (Xie *et al*. 2016). China is also the largest producer and consumer of antibiotics. Only 15% of administered antibiotics can be absorbed by the human body; the remaining 85% is discharged in their primary form by metabolism (Qiu *et al*. 2016). They enter the environment directly through sewage, aquaculture, and factories. Because it is difficult for antibiotics to biodegrade, buildup of antibiotic concentrations in the environment becomes a great threat to ecological security (Chen *et al*. 2012). It is currently difficult to remove these antibiotics from wastewater; therefore, it is necessary to find an effective way to treat wastewater contaminated with antibiotics.

Activated carbon has good adsorption properties because of its large surface area and multiple-pore structure, and it is used widely as an adsorbent (Titirici *et al*. 2015). The main raw materials that can be used to make activated carbon are wood and coal, although recent efforts have focused on agricultural by-products such as straw and fruit shells because of the limited availability of wood and coal (Reza *et al*. 2014). However, activated carbon is not usually effective for adsorbing antibiotics from wastewater because activated carbon has large pores and a low adsorption capacity.

Chinese medicinal production occupies an important position in the pharmaceutical industry because most Chinese medicines come from plant herbs, which are extracted using water and ethanol, with the residual material usually being abandoned, incinerated, or dumped, resulting in wasted resources and environmental pollution (Wu *et al*. 2015). The waste residues from the herbs can be used to make activated carbon; however, because of the variety of plant roots, stems, leaves, fruits, and other residual components in the mixture, it is very difficult to guarantee a consistent quality of activated carbon. This in turn makes it difficult to achieve a standard product based on a general process (Dubey *et al*. 2009). Additionally, the use of zinc chloride and phosphoric acid in the current process for making activated carbon can cause further pollution (Chen *et al*. 2013; Takdastan *et al*. 2016).

To utilize the leftover materials from traditional Chinese medicine production and to remove antibiotics from wastewater, a new preparation technique was studied to make uniform-activated carbon from Chinese herb residues with a steam activation method. The adsorption to cephalosporin (a major antibiotic in China) and the recyclability of activated carbon created using this technique were evaluated in this research.

EXPERIMENTAL

Materials

The herbal residues were collected from the Traditional Chinese Medicine Company in Guangzhou, China, including *Radix bupleuri*, astragalus roots, *Radix liquiritiae*, *Fructus aurantii*, *Fructus gardeniae*, *Evodia rutaecarpa*, *Herba plantaginis*, lobelia, and *Cacumen platycladi*. Wastewater containing cephalosporin antibiotics (pH 6.5) was collected from the Guangzhou Antibiotics Production Company (Guangzhou, China). Cephalexin and cefradine, which served as the controls in this study, were provided by Guangdong Xinfeng Pharmacy Company (Huizhou, China). Other reagents were of analytical purity and purchased from Guangzhou Qianhui Company (Guangzhou, China).

Methods

Preparation of activated carbon

The herbal residues were divided into three classes: rhizomes, fruits, and leaves. Activated carbon was made *via* overheated steam activation with some modification (Hata *et al*. 2016). One kilogram of herbal residues was heated to 200 °C for 1 h in a carbonization furnace (Shanghai Scientific Apparatus Company, Shanghai, China), then heated up to 400 °C for 2 h and then 600 °C for 2 h. The residues were then subsequently passed through 1 kg of 500 °C steam for 1 h. Three kinds of charcoal of varying masses were obtained: rhizome charcoal (436 g), leaf charcoal (278 g), and fruit charcoal (365 g).

Determination of iodine adsorption value

The iodine adsorption value was used as the index to evaluate the adsorption capacity of activated carbon produced in this study (Tang *et al*. 2016). A 0.5-g sample was wetted with 10 mL of 10% hydrochloride aqueous solution, heated to a boil for 30 s, and then cooled to room temperature before adding 50 mL of 0.1 M iodine solution. This sample was immediately oscillated for 15 min. The filtrate was measured at a wavelength of 288 nm by UV3300 UV-Vis spectrometer (Shanghai Meipuda Instrument Co., Ltd, Shanghai, China), and the concentration of residual iodine solution could be obtained from the standard curve of absorbance *vs.* concentration. The iodine adsorption value was calculated using Eq. 1,

$$\text{Iodine adsorption value (mg/g)} = \frac{50 \times (C_0 - C_1) \times 253.81}{W_0} \quad (1)$$

where C_0 and C_1 represent the initial concentration (M) and the remaining concentration of iodine solution (M), respectively, and W_0 is the weight (g) of the activated carbon sample.

Surface area and pore size distribution

The BET surface area and pore size distribution of samples were estimated using the standard nitrogen adsorption isotherm measured by an Autosorb-iQ Analyzer (Quantachrome Instruments, FL, USA). The samples were heated to 150 °C and evacuated for 12 h, and then nitrogen adsorption isotherms were determined at 77 K to calculate the surface area using the Brunauer-Emmett-Teller (BET) equation (Li *et al*. 2015). The total volume of samples was calculated using a t-plot based on a partial pressure P/P_0= 0.995. The pore size distribution was analyzed using the Barrett-Joyner-Halenda (BJH) model (Hadi *et al*. 2016).

Spectroscope analysis

IR spectra were measured using a Nicolet 380 FI-IR spectrograph (Nicolet Apparatus Company, USA) with KBr tablets from 4000 to 400 cm^{-1} with a resolution of 2 cm^{-1}. Element analysis was determined on Elemental analyzer Vario EL III (ELEMENTAR Company, Switzerland).

Adsorption and desorption tests of antibiotics

Four samples of waste water containing 20 μg/mL or 100 μg/mL of the control antibiotics, cefalexin or cefradin, were collected for the tests. Each sample was divided into 8 groups with 25 mL each, and separately analyzed at the following times: 1 min, 5 min, 10 min, 20 min, 40 min, 60 min, 90 min, and 120 min. A total of 0.3 g of the activated carbon was added to 25 mL of the waste water at room temperature with oscillation. The concentration of antibiotics in the filtrate was measured using an HP 1100 HPLC (Agilent Technologies Co., Ltd.; CA, USA) under the following operating conditions (Rahim *et al*. 2015): Column: Hypersil ODS (250×4.6 mm, 5 μm); Flow Phase: methanol / 0.1 M pH 6.8 phosphate buffer (80 / 20); Injection Volume: 20 μL; Flow Rate: 1 mL/min; Temperature: 25 °C; Wavelength: 260 nm. The adsorption value at "*t*" time interval was calculated using Eq. 2,

$$q_t \text{ (mg/g)} = (C_0 - C_t) \times \frac{0.025}{W} \quad (2)$$

where C_0 and C_t represent the initial concentration (μg/mL) and the remaining concentration of antibiotics at "*t*" time interval (μg/mL), respectively, and *W* is the weight (g) of activated carbon.

The antibiotics were separated from the activated carbon for recycling using an ultrasonic apparatus (Kunshan Ultrasonic Apparatus Company, Jiangsu, China). A total of 0.3 g of activated carbon was dispersed in 50 mL of water (pH 8.0) and treated with 100-W ultrasonic waves at room temperature for 5 to 30 min. The concentration of antibiotics in the water was determined using the same method as the adsorption test.

Isothermal model experiment

A total of 25 mL of cefalexin and cefradin at concentrations of 5, 10, 20, 30, 40, 60, 80, and 100 μg/mL was individually mixed with 0.3 g of the activated carbon and then oscillated at room temperature for 1 h. The adsorption capacity in equilibrium (q_e) was calculated using Eq. 3,

$$q_e \text{ (mg/g)} = (C_0 - C_e) \times \frac{0.025}{W} \quad (3)$$

where C_0 and C_e represent the initial concentration (μg/mL) and the concentration of antibiotics in equilibrium (μg/mL), respectively, and *W* is the weight (g) of activated carbon.

RESULTS AND DISCUSSION

Preparation and Formula of the Activated Carbon

While activated carbon can be made in various ways, zinc chloride and phosphoric acid are commonly used as catalysts to make activated carbon at lower temperatures, producing waste chemicals in the process (Chen *et al*. 2013; Takdastan *et al*. 2016). The carbon requires activation after carbonization, and potassium hydroxide is generally used as the activating agent (Mendoza-Carrasco *et al*. 2016). Potassium hydroxide, however, causes damage to the active adsorbate and makes recycling this carbon impossible. Because of this, no chemicals were used in the present experiments; carbonization was completed by gradual heating, and activation was achieved using overheated steam.

Rhizomes, fruits, and leaves all have different plant tissues with varying textures, so the activated carbons from them also have different pore sizes, as listed in Table 1. The activated carbon from leaves had more macropores and fewer micropores than activated carbon from fruits and rhizomes, which led to activated carbon from leaves having weaker adsorption capacity.

Table 1. Surface Area and Pore Size of Activated Carbons from Herbal Residue

Source of Activated Carbon	Surface Area (m^2/g)	Pore Volume (cm^3/g)	Microporosity Ratio (%)	Yield (%)
Rhizomes	895	0.76	93.4	43.6
Fruits	1054	0.95	67.3	36.5
Leaves	1287	1.24	35.6	27.8
Mixture	1023	0.89	82.4	36.6

To produce products with better adsorption capacity and more homogeneous quality, a ratio of the mixture with the three kinds of activated carbon was optimized by uniform design (Li *et al*. 2014). Three factors and five levels were arranged in the form of U5(5^3), as listed in Table 2. Iodine adsorption value was the index used to analyze the best combination. The best ratio of rhizomes to fruits to leaves was 5:4:2, which was used as the recipe for the adsorption tests with antibiotics.

The mixture with the 5:4:2 ratio showed a good microporosity ratio (Table 1) and was made with five replications; the iodine adsorption values were 1132, 1076, 1094, 1128, and 1203 mg/g, respectively, showing homogeneity in adsorption capacity.

Table 2. Arrangement of Factors and Levels by Uniform Design

Run	Factor A Rhizome (g)	Factor B Fruits (g)	Factor C Leaves (g)	Iodine adsorption value (mg/g)
1	1	3	1	463
2	3	1	3	662
3	4	2	5	785
4	5	4	2	1023
5	2	5	4	574

Nitrogen adsorption and desorption isotherms are illustrated in Fig. 1. There was a quick increase in the adsorption volume up to P/P_0 of 0.2 and a slow increase thereafter for all the activated carbon, indicating that they belong to Type I adsorption isotherm curves, which are attributed to microporous and monolayer adsorption. But there was a bigger hysteresis loop for the leaves, and a smaller hysteresis loop for the fruits, indicating they had irregular pores with mesoporous adsorption. The mixture had no hysteresis loop but gradually increasing adsorption volume suggestive of uniform pores in it. No deviation of desorption from the adsorption isotherm is also indicative of cylindrical pores without slit type or bottle neck type of pores in it.

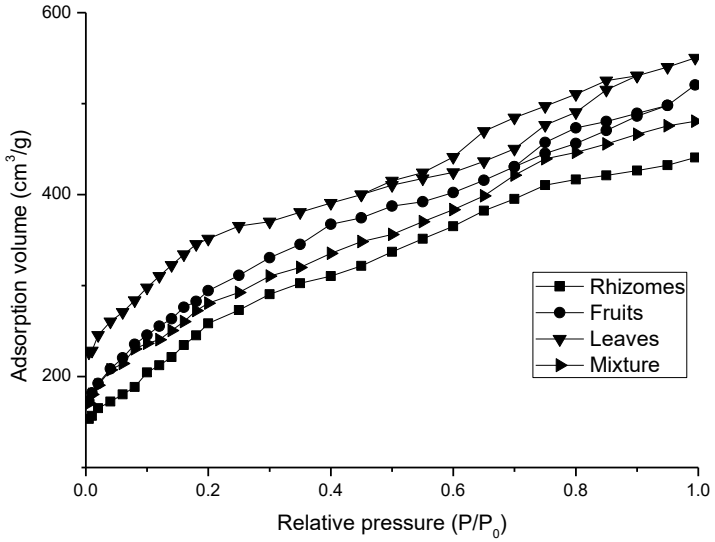

Fig. 1. Nitrogen adsorption/desorption isotherms at 77 K on the activated carbons

Elemental composition was slightly different among the activated carbons (Table 3). Leaves carbon had more nitrogen element than the others, and rhizome had more oxygen element than the others. The mixture both had the higher level of nitrogen and oxygen, which contribute to its adsorption ability. IR spectra showed a peak at 3,440 cm^{-1} which is attributed to the hydroxyl groups, and a peak at 1,610 cm^{-1} ascribed to olefin groups of aromatic ring with corresponding C-H stretching vibration peaks at 2,920 cm^{-1} and 2,860 cm^{-1}, the peak at 1,386 cm^{-1} ascribed to amide groups, the peak at 1,110 cm^{-1} ascribed to phenolic hydroxyl groups, and the other peaks at 620 cm^{-1} and 710 cm^{-1} ascribed to C-OH stretching vibration (Fig. 2). This suggests that both oxygen and nitrogen groups play an important role in the adsorption properties.

Table 3. Elemental Composition of Activated Carbons from Herbal Residue

Source of Activated Carbon	C (%)	H (%)	O (%)	N (%)
Rhizomes	45.3	5.4	48.3	0.2
Fruits	47.7	6.0	45.4	0.5
Leaves	49.2	6.5	38.4	0.6
Mixture	46.8	5.7	47.3	0.4

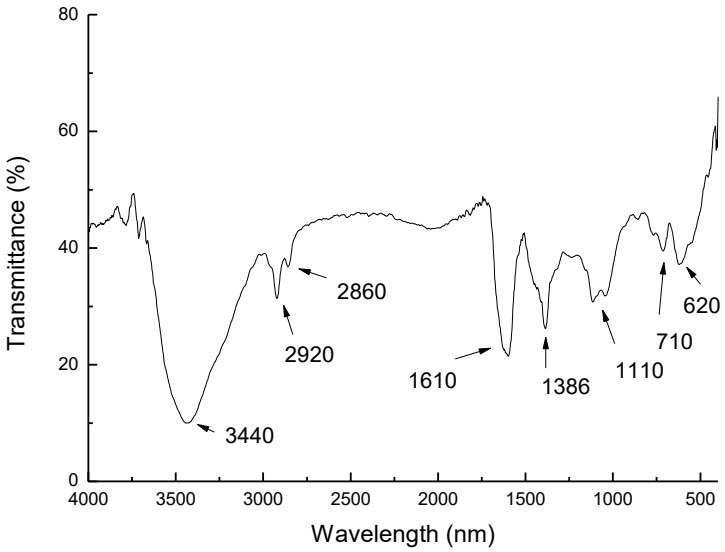

Fig. 2. IR spectra of the mixture of activated carbons from residue of Chinese medicines

Adsorption Properties of Cephalosporin Antibiotics with Activated Carbon

The adsorption curves of the activated carbon for cephalexin- and cefradine-tainted wastewater at different times are shown in Fig. 3. There was no difference seen between the two kinds of antibiotics at the same concentration. The adsorption quickly stabilized at approximately 20 μg/mL of antibiotics due to the surface adsorption of the activated carbon initially at 5 min (Foo and Hameed 2011). Adsorption slowly increased from 5 to 20 min at 100 μg/mL for both antibiotic concentrations, indicating that the antibiotics were diffusing into the inner space of the activated carbon. After 20 min, the adsorption stabilized, indicating that the process reached equilibrium. The maximum adsorption capacities of cefalexin and cefradine using the activated carbon were both

observed to have the same value, 7.1 mg/g, and the activated carbon was seen to remove up to 84% of the antibiotics from the treated wastewater.

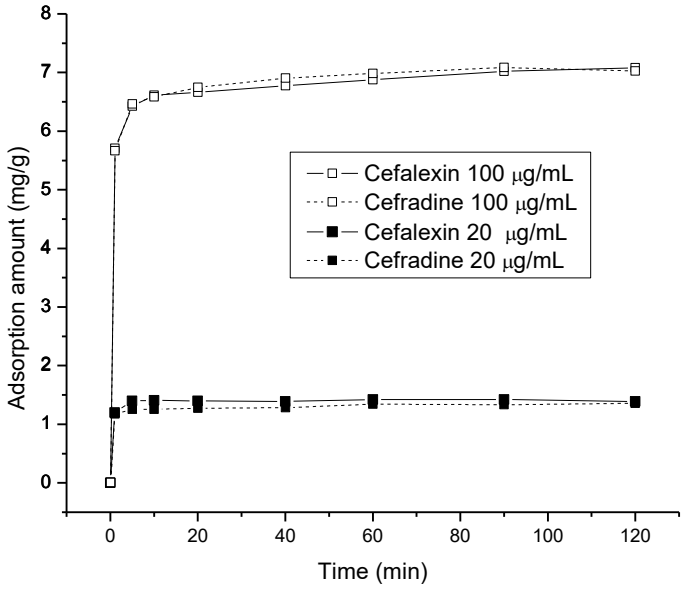

Fig. 3. Adsorption effects of activated carbon on cephalexin and cefradine

The activated carbon was treated ultrasonically for recycling at pH 8. The desorption of antibiotics and re-adsorption capacity of activated carbon were determined again. The results showed that a maximum of 90% of the antibiotics was separated from the activated carbon after 10 min of ultrasonic treatment, and the adsorption capacity of the recycled activated carbon still maintained an antibiotic adsorption capacity of 80% for a second use. Acidity has effects on adsorption and desorption of cephalexin and cefradine. They are in nonionic form in the wastewater with pH 6.5, which is advantageous for adsorption, but they exist in ionic form in alkaline condition (pH 8.0) which is better for desorption. Therefore, different pHs should be used in adsorption and desorption.

Simulation of Adsorption Kinetics

Using a kinetic model analysis of the adsorption process, the interaction between the adsorbed molecules and the active sites on the surface of the adsorbent can be estimated (Liu *et al*. 2015). If one active site binds to one molecule, the pseudo-first order kinetic equation can be described according to Eq. 4,

$$\ln (q_e - q_t) = \ln (q_e) - K_1 t \quad (4)$$

where q_e and q_t are the adsorption capacities for the antibiotics at equilibrium and at time t, respectively, t is the specific time, and K_1 is the first-order adsorption rate constant.

Alternatively, the pseudo-second order kinetic equation can be described according to Eq. 5,

$$\frac{t}{q_t} = \frac{1}{K_2 q_e^2} + \frac{t}{q_e} \quad (5)$$

where q_e and q_t are the adsorption capacities for the antibiotics at equilibrium and at time t, respectively, t is the specific time, and K_2 is the second-order adsorption rate constant.

In the above equations, K_1 and K_2 can be calculated assuming a linear relationship between $\ln(q_e - q_t)$ and t in Eq. 4 and between t/q_t and t in Eq. 5.

The parameters above were obtained based on adsorption to cephalexin and cefradine at a concentration of 100 μg/mL. The results are listed in Table 4. The coefficient of determination (R^2) was higher for the pseudo-second order kinetic model, and the calculated adsorption capacity ($q_{e\ cal}$) was similar to empirical data. This shows that the pseudo-second order kinetic model is a close fit for simulating the interaction between activated carbon and cephalexin and cefradine.

Table 4. Parameters in Pseudo-First/Pseudo-Second Order Kinetic Model

Antibiotics	Linear equation ($Y = b + mX$)	$q_{e,cal}$ (mg/g)	K	R^2
Cephalexin	Y = -0.11977 - 0.02747X	0.887	0.02747 (K_1)	0.8631
Cefradine	Y = -0.34134 - 0.02199X	0.711	0.02199 (K_1)	0.8508
Cephalexin	Y = 0.1426 + 0.14125X	7.079	0.1399 (K_2)	0.9996
Cefradine	Y = 0.09771 + 0.14158X	7.063	0.2052 (K_2)	0.9998

Rate-Limiting Step in the Adsorption Process

The adsorption process can be divided into three steps in sequence: film diffusion (adsorbate from a solution adheres to the surface of adsorbent), intraparticle diffusion (diffusion of adsorbate into the inner pores of adsorbent), and then binding of the absorbent to active sites (Nethaji and Sivasamy 2011). Because the last step is very short, it can be ignored in the analysis of the rate-limiting step.

The film diffusion process can be described according to Eq. 6,

$$\ln\left(1-\frac{q_t}{q_e}\right) = -K_3 t + A \quad (6)$$

where K_3 is the film diffusion constant, A is the equilibrium constant, q_e and q_t are the adsorption capacities for the antibiotics at equilibrium and at time t, respectively, and t is the specific time.

The intraparticle diffusion process is fit to Eq. 7,

$$q_t = K_4 t^{1/2} + C \quad (7)$$

where K_4 is the intraparticle diffusion constant, q_t is the adsorption capacity for the antibiotics at time t, and C is the equilibrium constant.

K_3 and K_4 can be calculated using the linear relationship between $\ln(1 - q_t/q_e)$ and t in Eq. 6 and between q_t and $t^{1/2}$ in Eq. 7.

The parameters were calculated using experimental data and are listed in Table 5. Even though the processes of film diffusion and intraparticle diffusion both affect the adsorption rate, intraparticle diffusion is the dominant rate-limiting step because it was seen to have a better correlation and a higher rate constant.

Table 5. Constants in the Film Diffusion and Intraparticle Diffusion Equations

Antibiotics	Film Diffusion			Intraparticle Diffusion		
	K_3	A	R^2	K_4	C	R^2
Cephalexin	0.02747	-2.07621	0.8631	0.06844	6.3278	0.9518
Cefradine	0.02199	-2.29862	0.8515	0.06647	6.3886	0.9013

Isothermal Adsorption Model

The isothermal adsorption model is used to describe the distribution of adsorbate in the liquid phase and adsorbent (Fu *et al*. 2016). The Langmuir, Freundlich, and Tempkin isothermal equations are expressed as Eqs. 8, 9, and 10, respectively,

$$\frac{C_e}{q_e} = \frac{1}{q_m K_L} + \frac{1}{q_m} C_e \quad (8)$$

where q_m is the maximum adsorption (mg/g), K_L is the Langmuir adsorption rate constant (mL/μg), and q_e is the adsorption capacity for the antibiotics at equilibrium.

$$\ln q_e = \ln K_F + \frac{1}{\mathrm{n}} \ln C_e \quad (9)$$

where q_e is the adsorption capacity for the antibiotics at equilibrium and K_F is Freundlich constant ((mg/g) (μg/mL)$^{1/n}$), n is a constant of intensity without unit.

$$q_e = B \ln K_T + B \ln C_e \quad (10)$$

where q_e is the adsorption capacity for the antibiotics at equilibrium, B is the Tempkin constant without unit, and K_T is the equilibrium binding constant (μg/mL). The constants were calculated assuming a linear relationship between q_e and C_e at different concentrations of antibiotics, and are listed in Table 6.

Table 6. Constants in Different Isothermal Adsorption Models

Antibiotics	Langmuir Model			Freundlich Model			Tempkin Model		
	K_L	q_m	R^2	K_F	N	R^2	B	K_T	R^2
Cephalexin	0.700×10^{-4}	7.992	0.980	0.040	0.447	0.924	7.907	0.230	0.991
Cefradine	8.842×10^{-7}	7.984	0.957	0.116	0.455	0.889	10.690	0.347	0.988

The Langmuir and Tempkin models both were considered fits because of their higher correlation coefficients. The maximum adsorption capacity reaches about 8.0 mg/g, and is also predicted by the Langmuir model. K_L was smaller, indicating that the monolayer adsorption capacity was lower and that multilayer adsorption plays an important role in the antibiotics adsorption. The value of "*n*" was less than 1, suggesting a weaker intermolecular force between adsorbent and adsorbate, and that physical adsorption was dominant in the adsorption process.

CONCLUSIONS

1. Rhizomes, fruits, and leaves separated from the herbal residues after extraction can be used to produce activated carbons with different surface areas and pore sizes. The optimal ratio of rhizomes, fruits, and leaves is 5:4:2, leading to higher iodine adsorption value and greater homogeneity.
2. The maximum adsorption of activated carbon for cephalexin and cefradine both is 7.1 mg/g, and activated carbon removes 84% of these two antibiotics from wastewater. Approximately 90% of these antibiotics can be desorbed with the re-adsorption capacity remaining at 80% after ultrasonic treatment.
3. The adsorption process of activated carbon to cephalexin and cefradine is dominated by a pseudo-second order kinetic adsorption reaction with two active sites binding to one antibiotic molecule. The rate-limiting step is intraparticle diffusion.
4. The isothermal adsorption process of activated carbon to cephalexin and cefradine conforms to Langmuir and Tempkin isotherm models, and shows multilayer and physical adsorption.
5. The activated carbon from herbal residues can adsorb the low concentration of cephalexin and cefradine in wastewater, and can be recycled after ultrasonic treatment in water. This technique is practical for solving herbal residues accumulation and reducing antibiotic water pollution.
6. The activated carbon will be modified with functional groups to improve its specific adsorption on other antibiotics in the next research.

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