

Full Length Research Paper

Adsorption isotherms of celebrex as non-steroidal anti-inflammation on single-walled carbon nanotubes

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The objective of this work is to study the adsorption behavior of celebrex as non-steroidal anti-inflammation on single-walled carbon nanotubes (SWCNTs). As the function of temperature and initial concentration of adsorbate were observed through adsorption isotherms, the amount of celebrex adsorbed on carbon nanotube surface increased with the concentration at constant temperature, and decreased with the increase of temperature at constant concentration. The experimental results obtained at the temperatures of 295, 300 and 305 K showed that the experimental data were properly represented by the Langmuir, Freundlich and Temkin isotherm models.

Key words: Adsorption, celebrex, non-steroidal, carbon nanotubes.

INTRODUCTION

The use of low cost absorbents, such as carbonic substances, agricultural product and wastes of product has been investigated (Nguyen and Do, 2001). In recent years, agricultural production of the separating metals from water is widely studied. These include peat soil (Hoys and McKay, 2000), wood (Poots et al., 1978), shell of pine tree (Al-Asheh and Duvnjak 1997), banana skin (Low et al., 2000), rice bran (Marshall and Johns 1996), soy and skins of cottonseed (Wafwoyo et al., 1999), peanut skin (Cimino et al., 2000), orange skin, vegetable fertilizer (Azab and Peterson, 1989) and leaves. Most of these works showed that natural products can be a good absorbent for organic acids. Since their discovery by Iijima in 1991, carbon nanotubes (multi-walled carbon nanotubes (MWCNTs)) compounds are highly developed in chemical and physical dimension (Iijima et al., 2001). The ways of synthesis of this compound, developed quickly (Rao et al., 2001). MWCNTs can adsorb so many atoms and molecules on their surface, such as adsorption of metallic elements like lithium (Bendiab et al., 2001), potassium (Clay et al., 2002), rubidium (Rao et al., 1997), cesium (Wadhawan et al., 2001) and non metallic, such as hydrogen (Cao et al., 2001), oxygen (Yang et al., 2001), nitrogen (Zhu et al., 2000) and methanol (Talapatra and Migone, 2002). Adsorption characteristic of MWCNTs is breather for adsorption of gases, such as hydrogen and other gases (Dillon and Heben, 2001). All the compounds on the surface of

MWCNTs, adsorbed two main covalent bonds and non covalent bonds (Bahr and Tour 1952; Basiuk et al., 2004). Adsorption takes places on the surface and on the carbon walls, while non covalent adsorption which is the kind of physically adsorption takes place on the MWCNTs walls. One of the characteristics of non covalent bonds adsorption on MWCNTs is that the structure of MWCNTs does not change after this adsorption and separation of the adsorption (Chen et al., 2001; Lehmann et al., 2004). All compounds on the surface of MWCNTs are adsorbed by covalence and non covalence bonds. Basically, essential fatty acid exists in liquid oil, and it has many double bonds, which one of the most important is omega-6 that is found in corn oil and canola oil, and it gives them specific characteristic. Indeed, fat in these oils are the best type of fat, because essential fatty acid do not only increase the blood cholesterol, but cause the decrease of blood cholesterol (Cunnane et al., 1993). Features of celebrex can slow down the formation of cancer clone (Denis et al., 1999), prevent breast cancer (Thompson et al., 1996), blood pressure regulation (Berry and Hirsch, 1986) reduced resistance to insulin in diabetic patients (Cunnane et al., 1993) and the body immunological resistance to antigens. NSAIDs reduce inflammation and relieve fever and pain by blocking enzymes and protein made by the body. NSAIDs, such as ibuprofen and naproxen block a protein (called prostaglandin) that makes heavy menstrual bleeding worse. Aspirin does not

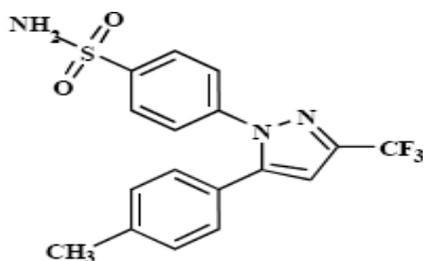


Figure 1. Chemical structure of celebrex.

block this protein. NSAIDs relieve pain and fever. They also reduce swelling and inflammation caused by an injury or a disease, such as arthritis. Some NSAIDs, such as ibuprofen and naproxen, help ease cramping and reduce blood loss from heavy menstrual bleeding. NSAIDs work well to relieve pain, decrease fever and reduce swelling and inflammation caused by an injury or disease (Gøtzsche, 2007). Some NSAIDs help reduce heavy menstrual bleeding (Bohn, 2002; Dukitt and Collins, 2008). NSAIDs can help relieve the pain of kidney stones.

Celebrex is chemically designated as 4-[5-(4-methylphenyl)-3-(trifluoromethyl)-1H-pyrazol-1-yl] benzene sulfonamide and is a diaryl-substituted pyrazole. The empirical formula is $C_{17}H_{14}F_3N_3O_2S$, and the molecular weight is 381.38; the chemical structure is as shown in Figure 1 (Teichman, 2004; Chan, 2007).

In this research, we are going to study celebrex as non-steroidal anti-inflammation on single walled carbon nanotube. We are going to understand how much of this drug can be adsorbed by carbon nanotube. We also want to find out if we can affect the inflammable molecules by putting celebrex on carbon nanotube without damaging the safe molecules.

MATERIALS AND METHODS

Celebrex with purity of 88% was purchased from Merck Co., Germany. Single-walled carbon nanotubes (SWCNTs) are produced by outer diameter of 10 to 20 nm, surface space of 250 of 280 m^2/g and high purity of 9%, and were purchased from Aldrich.

Fourier transforms infrared analysis

Fourier transform infrared (FTIR) were used in the analysis of the chemical surface groups of these adsorbents. FTIR analysis was performed using a Nexus 670 FTIR spectrometer (Thermo Nicolet, Madison) equipped with a KBr beam splitter (KBr, FTIR grade). Spectra were acquired in the 4000 to 400 cm^{-1} wave number with 4 cm^{-1} resolution. The background spectrum of KBr was also recorded at the same conditions.

Adsorption experiments

A stock solution of about 100 mg/L celebrex was prepared. The

range of celebrex concentration used is from 2 to 30 mg/L. Equilibrium adsorption experiments were performed using 40 ml screw-capped glass centrifuge tubes as batch reactor systems. Each tube containing 0.05 g SWCNTs was filled with 25 ml celebrex solution of different concentrations. All tubes were immediately sealed with PTFE-lined caps and were then mechanically shaken for 24 h in a thermostated rotary shaker at temperature of 295 ± 1 K, except for the adsorption experiments, in which temperatures of 300 and 305 K were adjusted. After equilibration, all tubes were placed vertically for 4 h at the same temperature to ensure complete sedimentation of SWCNTs from the bulk solutions. By using spectrophotometer tool adsorption rate, celebrex was obtained.

Modeling of the adsorption isotherms

Equilibrium study on adsorption provides information on the capacity of the adsorbent. An adsorption isotherm is characterized by certain constant values, which express the surface properties and affinity of the adsorbent and can also be used to compare the adsorptive capacities of the adsorbent for different pollutants. Equilibrium data can be analyzed using commonly known adsorption systems. Several mathematical models can be used to describe experimental data of adsorption isotherms (Uddin et al., 2007). The Freundlich, Langmuir and Temkin models are employed to analyse the adsorption that occurred in the experiment.

Langmuir model

The Langmuir model (Zeldowitsch., 1934; Langmuir., 1916) assumes uniform energies of adsorption onto the surface and no transmigration of adsorbate in the plane of the surface. The Langmuir equation may be written as:

$$\frac{c_e}{q_e} = \frac{1}{q_m b} + \frac{1}{q_m} c_e \quad (1)$$

where q_e is the amount of solute adsorbed per unit weight of adsorbent (mg/g), C_e the equilibrium concentration (mg/L), q_m is the monolayer adsorption capacity (mg/g) and b is the constant related to the free energy of adsorption. The Langmuir model considers several assumptions: the adsorption is localized, all the active sites on the surface have similar energies, none interaction between adsorbed molecules exists, and the limiting reaction step is the surface reaction as in the heterogeneous catalytic reaction (Fierro et al., 2008).

Freundlich model

The Freundlich model is an empirical equation based on sorption on heterogeneous surface through a multilayer adsorption mechanism (Freundlich, 1907). It is given as:

$$q_e = k_f c_e^{1/n} \quad (2)$$

where q_e is the amount of solute adsorbed per unit weight of adsorbent (mg/g), C_e is the equilibrium concentration (mol/L), k_f is the constant indicative of the relative adsorption capacity of the adsorbent ($mg/g(mg/L)^n$) and $1/n$ is the constant, indicative of the intensity of the adsorption. The linearized form of the Freundlich equation is:

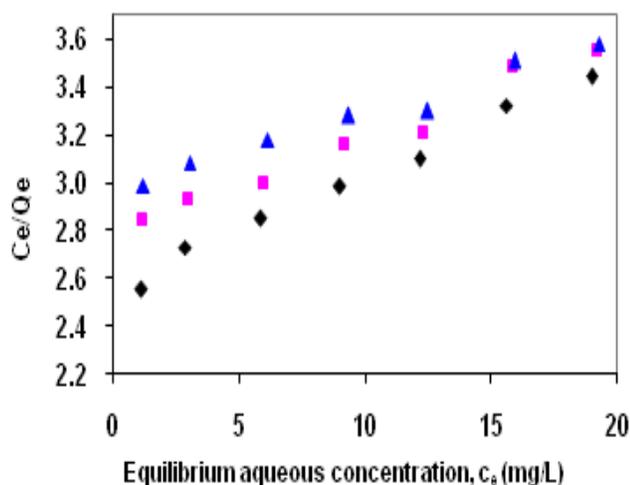


Figure 2. Langmuir isotherm of celebrex on SWCNT: ♦, 295 K ($R^2 = 0.9885$); ■, 300 K ($R^2 = 0.9238$); ▲, 305 K ($R^2 = 0.8811$).

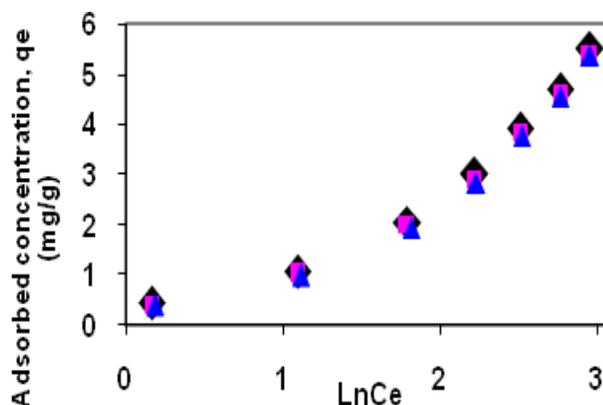


Figure 3. Temkin isotherm of celebrex on SWCNT: ♦, 295 K ($R^2 = 0.9257$); ■, 300 K ($R^2 = 0.9223$); ▲, 305 K ($R^2 = 0.9078$).

$$\ln q_e = \ln k_f + \frac{1}{n} \ln c_e \quad (3)$$

The value of k_f and n can be calculated by plotting $\ln q_e$ versus $\ln c_e$.

Temkin model

Temkin (Fierro et al., 2008; Temkin and Pyzhev 1940) suggested that, because of the existence of adsorbent-adsorbate interactions, the heat of adsorption should decrease linearly with the surface coverage. The Temkin isotherm equation assumes that the adsorption is characterized by a uniform distribution of the binding energies, up to some maximum binding energy. The corresponding adsorption isotherm can thus be adjusted by the following equation:

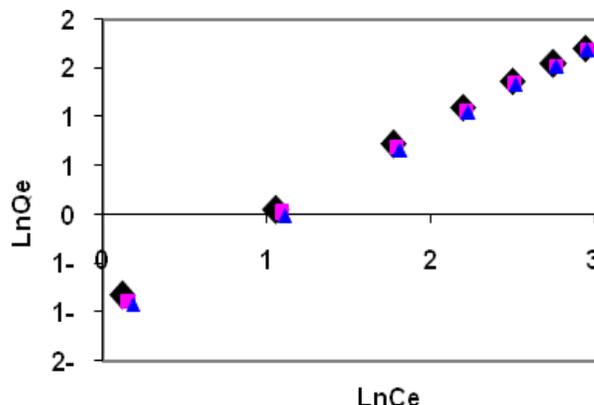


Figure 4. Freundlich isotherm of celebrex on SWCNT: ♦, 295 K ($R^2 = 0.9993$); ■, 300 K ($R^2 = 0.9988$); ▲, 305 K ($R^2 = 0.9990$).

$$q_e = \frac{RT}{b} \ln(AC_e)$$

$$q_e = B \ln A + B \ln c_e \quad (4)$$

where B is related to the heat of adsorption (L/g) and A is the dimensionless Temkin isotherm constant. The Temkin parameters (B and A) can be determined from the linear plots of q_e and $\ln c_e$.

RESULTS AND DISCUSSION

Adsorption isotherms

The Langmuir, Temkin and Freundlich isotherms of the adsorption process of celebrex on SWCNTs are as shown in Figures 2 to 4. It was observed that the experimental data were well represented by Langmuir, Freundlich and Temkin models. The values of the constants of the isotherms of Langmuir, q and b , and of Freundlich, k and n , and of Temkin, B , A and b , are as shown in Table 1. The results of Figures 2 to 4 show that in order to adsorb celebrex on carbon nanotube in the temperature range of 295 to 305 K, the Freundlich model is followed because they have more R^2 (Table 1).

Conclusion

In the light of the findings of this work, the following main conclusions can be stressed. The amount of celebrex adsorbed in the SWCNTs surface increased with concentration at constant temperature and decreased with the increase of temperature at constant concentration. The Freundlich isotherm model describes the sorption data better than the Langmuir and Temkin models.

Table 1. The values of the constants of the isotherms of Langmuir, Freundlich and Temkin on CNT.

T/K	Langmuir			Freundlich			Temkin			
	b	q	R ²	n	k (L.g ⁻¹)	R ²	A (L.mg ⁻¹)	B	b (j.mol ⁻¹)	R ²
295	0.0153	25.172	0.9885	1.095	0.3848	0.9993	0.817	1.778	13.785	0.9256
300	0.0136	32.658	0.9238	1.063	0.3849	0.9988	0.792	1.786	13.972	0.9223
305	0.0074	3.010	0.8811	1.040	0.3541	0.9990	0.769	1.778	14.065	0.9078

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