RESEARCH ARTICLE

Thermodynamic Study of Adsorption of Tamoxifen Drug on Activated Nano-Coal Particles prepared from Stem of Eucalyptus and its Diagnosis by different Techniques

Ahmed Taha Yaseen, Liqa'a Hussein Alwan

Department of Chemistry, College of Education, Samarra University, Iraq

Received: 28th June, 2020; Revised: 15th July, 2020; Accepted: 19th August, 2020; Available Online: 25th September, 2020

ABSTRACT

The research included preparing nano-activated charcoal from the stem of the eucalyptus trees. The prepared nano-coal particles were diagnosed using various techniques, such as, transmission electron microscopy (TEM), and energy-dispersive X-ray (EDX) technology.

The research included a spectroscopic study of the adsorption of tamoxifen on the prepared nano-coal, and the results showed that the appropriate concentration is 2.5×10^{-5} molar, with an appropriate weight of the adsorbent material which is 0.4-gram, the time of equilibrium is 40 minutes, and the percentage of adsorption efficiency increases with passing time, the adsorption process is dependent to a second-degree equation, according to the correlation coefficient (R^2), which gave the value of 0.9999. Thermodynamic functions were calculated for the adsorption process at different temperatures (17.5, 27.5, 37.5, and 47.5ºC), and it was found that the adsorption process is a heat emission process (ΔH negative), the adsorption is physical because it is less than 40 kJ/mol, and the free energy is negative (ΔG), and entropy adsorption was negative (ΔS), meaning less random.

Keywords: Activated nano-coal, Adsorption, Stem of eucalyptus, Tamoxifen drug.

International Journal of Drug Delivery Technology (2020); DOI: 10.25258/ijddt.10.3.3

How to cite this article: Yaseen AT, Alwan LH. Thermodynamic study of adsorption of tamoxifen drug on activated nano-coal particles prepared from stem of eucalyptus and its diagnosis by different techniques. International Journal of Drug Delivery Technology. 2020;10(3):328-331.

Source of support: Nil.

Conflict of interest: None

INTRODUCTION

Activated charcoal is a black powder characterized by its high carbon content, odorless and tasteless, non-toxic, and highly porous, and it belongs to a large family of charcoal materials that do not have a specific chemical structure.[1,2] It is one of the most used adsorbents as it has a large surface area, where the average surface area of activated charcoal is 200 to 800 m^2/g. Research has shown that 50 grams of coal possess a surface equal to 13.935 m^2, as this distance is possessed with a myriad of binding sites, which makes it have a high adsorption capacity.[3] Activated charcoal was used for medicinal purposes in the past. The Egyptian manuscripts indicated that the ancient Egyptians used activated charcoal to remove odors from rotting wounds before 1,500 BC, and that the Indian manuscripts pointed to the use of sand and activated charcoal as filters to purify drinking water before 450 BC.[4] The use of activated charcoal was first described by the scientist (Lowitz) in 1791 AD, as its use has been limited until it described the advantages of using it in research for the years 1963–65. From that time, the use of activated charcoal has increased in the United States and other countries to treat a number of poisoning cases, especially children.[5] Activated charcoal was used to sterilize water when performing surgical operations. Activated charcoal was used in World War I to protect from toxic gases for protective masks and over time, the uses of activated charcoal increased globally.[6]

In 2010, Alobaidy[7] prepared activated charcoal from the stems of Iraqi corn, and studied a number of its properties, such as, the acidic function, ash, and moisture content, and adsorption of blue methylene tincture. The study concluded that prepared charcoal has similar characteristics to commercial activated industrial charcoal.

G. Yousefi and his group[8] performed a comparison in some studies conducted to determine the adsorption properties of amitriptyline (AMT) on activated charcoal and sodium polystyrene sulfonate (SPS). The results showed high effectiveness of the activated charcoal prepared in the adsorption of these drugs.

AIM AND OBJECTIVE

- Study of adsorption of tamoxifen by activated nano-coal made from environmentally friendly and inexpensive materials.

- Diagnosis of adsorbent material prepared by several techniques (TEM and EDX).

MATERIALS AND METHODS

Equipment

Visible spectrum device-ultraviolet, Japanese-made vis spectrophotometer-UV type. FTIR spectrophotometer. Drying oven. Burning oven. X-ray diffraction device 6000X-ray diffractometer. Atommic force microscopy. Scanning electron micron. Sensitive balance.

Materials

In this study, the stem of the eucalyptus and lemon juice was used in addition to the ion-free water and tamoxifen.

Preparation of Activated Carbon from Stems of Eucalyptus Trees

Preparation of Raw Material

The stems of the lemon were taken and cut into very soft small pieces, then washed several times to clean it from suspended soil. Then, they were washed with ion-free water several times in order to get rid of the suspended impurities and make sure of its purity, then put in a drying oven at a temperature 100ºC for 3 hours for drying and to get rid of moisture from stem.

Charring Process

- The charring process was performed by taking 100 grams of sliced and dried stems and placing them in a burning dish inside the burning oven at a temperature of 550ºC for 1-hour.
- The coal is cooled, then crushed well, and placed in a sealed box, away from moisture.

Chemical Activation

- 20 grams of non-activated charcoal were accurately weighed.
- The activation procedure by adding 15 mL of lemon juice to the charcoal powder, and then mixing in the beaker completely.
- The mixture was placed inside an iron capsule that was closed by both ends, prepared manually for two hours at a temperature of 700ºC.
- Cool the resulting activated charcoal, then wash it with distilled water free of ions, several times to get rid of impurities (lemon juice residue), and get a neutral function by measuring the filtrate by a pH meter device.
- The prepared activated charcoal was dried at 120ºC for 3 hours.

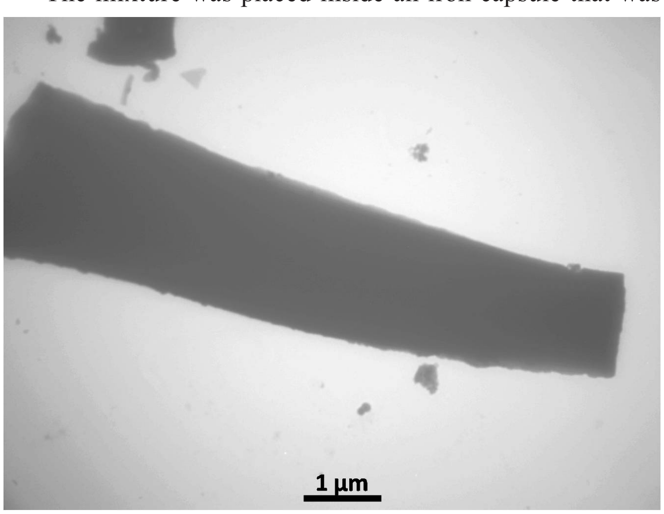

Figure 1: Carbon nanoparticles under TEM

RESULTS AND DISCUSSION

Diagnosis of prepared Activated Charcoal Surface

TEM Diagnosis

The TEM is one of the microscopic imaging devices, which is characterized by its high magnification ability and very strong, because it is possible to study the sample surface and the internal structure of the sample very accurately. Through our observation of the image, as in Figure 1, with a zooming strength 1-µm, the prepared nano-carbon particles are within the nanoscale, and its granules are a rod-like form, which makes it have a high surface area and large pores.

Energy-Dispersive X-Ray (EDX)

This spectroscopy is one of the analytical techniques that are used to analyze the elements in order to know the chemical properties of the samples as it studies the interference between the sample and the source of the X-ray excitation depending on the basic principle that each element has a distinct atomic structure that grants X-rays that describe the atomic structure of the elements in order to be identified distinctively from each other. Valenzuela-Muniz AM *et al.*[9] used this technique to find out the constituent elements of the sample of the prepared activated charcoal.

As it was evident from Figure 2 that the highest coal content is a carbon (83%), followed by oxygen (17%), which may be

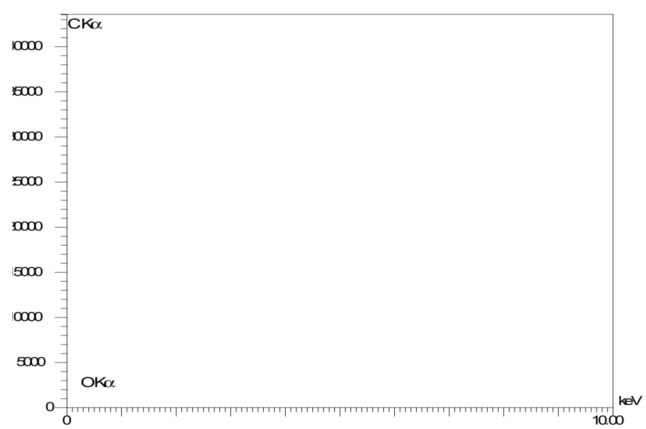

Figure 2: Nanocarbon particles under EDX

Table 1: Proportions of components that make up the sample

Element	*Weight%*	*Atomic%*
C	78.57	83
O	21.43	17
Totals	100	100

due to the moisture in the prepared activated charcoal. Table 1 shows the proportions of the constituent elements of the sample of the activated charcoal prepared.

Spectral Study

The maximum wavelength (λ_{max}) was determined by preparing the tamoxifen solution, under study, for the purpose of following the drug spectral to take advantage of the drug's ability to show clear absorptivity in the visible-ultraviolet area, and the maximum wavelength of the drug was 275 nm, where a calibration curve was built for different concentrations, as shown in the Figure 3, at a range of concentrations 1×10^{-5} to 5×10^{-5} molar, using the spectral method to give a linear relationship and confirm its compliance with the law of Bear-Lambert, in addition to studying the optimum conditions and the adsorption motility.

Determination of Equilibrium Time

In order to determine the time required for the adsorption system to reach equilibrium, the change in the concentration of tamoxifen was monitored spectrally over time and the follow-up was done using a fixed concentration of tamoxifen, which is 0.000025 molar, and constant weight of activated charcoal 0.4-gram at the greatest wavelength of the drug (275 nm) at room temperature (25ºC), and the obtained results are shown in Table 2.

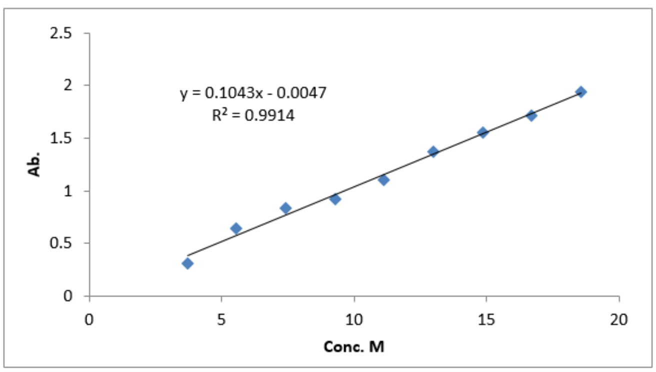

Figure 3: Titration curve for different tamoxifen concentrations

It is noted from the above results that the absorbance value was 0.922, and then decreased significantly during the first 10 minutes, i.e., adsorption of tamoxifen by activated charcoal by 82%. This high efficiency during this short period of time is due to the availability of a large number of unoccupied adsorption sites. This corresponds to the dynamic theory of adsorption, where adsorption is rapid at first and then slow.[10]

It is noted in the proven results that the adsorption efficiency has continued to increase with time by a small amount until the equilibrium state is reached at time 40 minutes, as the adsorption efficiency at this time is 88.67%. This is the highest value of adsorption efficiency and the stability of this value from 40 to 50 minutes indicates, however, the equilibrium time starts after 40 minutes of the adsorption process, and at this time the adsorption speed is equal to the extortion process, after which an increase in absorbance is observed with a decrease in the adsorption efficiency due to the repulsion process between the drug particles from the activated charcoal, and its return to the solution.

Calculation of Thermodynamic Functions for Adsorption Process

Through the study of thermodynamic functions, it is possible to identify the nature of the studied system and the nature of the powers controlling it. The value of the equilibrium constant (K_{eq}) at different temperatures represents the most important variables that we depend on it for calculating thermodynamic functions, where the K_{eq} values were calculated using the following formula:

$$K_{aq} = \frac{Xeq}{a - Xeq}$$

Where, a = initial concentration of the adsorbent substance (mg/L), X_{eq} = concentration of adsorbent at

Table 2: Time required for the absorption system

Absorption%	C_{Abs} (mg/g)	C_e (mg/L)	Absorption	Time (min)
-	-	-	0.922	0
82.5	1.915	1.617	0.164	10
84.01	1.948	1.483	0.15	20
85.47	1.982	1.348	0.136	30
88.67	2.056	1.051	0.105	40
88.67	2.056	1.051	0.105	50
80.92	1.877	1.77	0.18	60

Table 3: Calculation of Thermodynamic Functions for Adsorption Process

S°Δ (J/mole.K)	G°Δ ($J.mole^{-1}$)	H°Δ ($J.mole^{-1}$)	LnK_{eq}	K_{eq}	1/T (K^{-1})	T (K)
-39.616	-4,199.81	-15,714.29	1.738	5.689	0.00344	290.65
-39.978	-3,694.41	-15,714.29	1.478	4.387	0.00332	300.65
-40.191	-3,241.34	-15,714.29	1.255	3.508	0.00321	310.65
-38.864	-3,252.37	-15,714.29	1.22	3.071	0.00311	320.65

equilibrium, and a - X_{eq} = the remaining concentration of the adsorbent at equilibrium.

Results, as shown in Table 3, show a decrease in the equilibrium constant (K_{eq}) with increased temperatures due to a decrease in the adsorbent concentration of the drug, offset by the remaining concentration with increasing temperature. Just as the values of K_{eq} were calculated by the ratio between the amount of adsorbed material to the residue in the solution, and we note the inverse proportion between the temperature and the equilibrium constant. As for the values of ΔG^o, they were calculated by the following equation:

$$\Delta G^o = -RT\ln K_{eq}$$

From the values of ΔG^o and ΔH^o that are calculated by the Vant Hoff curve, which we get by drawing $\ln K_{eq}$ with the reciprocal of the temperature, we calculate the values of ΔS^o as follows:

$$S^o = (\Delta H^o - \Delta G^o)/\Delta T$$

It is worth noting that this study was conducted at the optimum conditions for adsorption, using the best concentration of tamoxifen (0.000025-molar), the best weight of the adsorbent substance (0.4-gram), at equilibrium time (40 minutes), and proven thermodynamic values No. 3.

The negative value of the enthalpy (ΔH^o) indicates that the adsorption of tamoxifen on the surface of the prepared activated charcoal is an exothermic process as evidence that the physical adsorption means that the powers responsible for the occurrence of the adsorption process are weak, and the negative values of the change in free energy (ΔG^o) pointing to automatically adsorption occurrence, and the low values of the change in free energy confirm the nature of the heat-emission adsorption process, Also, the change in negative entropy (ΔS^o) values indicates that the occurrence of the adsorption process reduces the randomness of the system under study because the molecules that suffer adsorption become restricted due to their attachment to the adsorbent surface, i.e., it will lose their freedom compared to the state it was before the adsorption, and this is consistent with previous studies.[11]

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