

Adsorption of Tartrazine on Medical Activated Charcoal Tablets under Controlled Conditions

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Abstract

Two types of commercial medical tablets of activated charcoal formulations (AC1 and AC2) were used as adsorbents for tartrazine. The adsorption studies were performed at controlled conditions of pH, and temperature (gastrointestinal-like conditions). It was found that pH plays a major role in the adsorption process. At pH 1.5 and 37°C the effect of different parameters affecting dye removal (salinity, adsorbent dose, initial dye concentration and stirring rate) were examined. The effects of some dietary additives like mono- and disaccharides, artificial sweeteners and glycine on the adsorption efficiency were assayed. The maximum adsorption of tartrazine on activated carbon tablets was observed at highly acidic media. The removal efficiency appears to decrease with increasing temperature and salinity indicating an exothermic process. Equilibrium adsorption isotherms for the removal of tartrazine from aqueous solution using activated charcoal tablets have been investigated. Langmuir and Freundlich's models were applied to the data related to adsorption isotherms. According to Langmuir's model data, the observed maximum adsorption capacities (q_m) were 272.85 and 456.83 mg g^{-1} at 37°C for AC1 and AC2, respectively. Medical activated charcoal tablets appear as a very prospective adsorbent for the removal of tartrazine from aqueous solution.

Keywords: Medical formulations; Activated charcoal; Tartrazine; Adsorption; Isotherm

Introduction

Tartrazine, (E102 or FD&C Yellow 5) is one of the most commonly used food additives. It is found in the following food stuffs: soft drinks, instant puddings, flavored chips, custard powder, soups, sauces, ice cream, candy, chewing gum, jam, jelly, marmalade, mustard, yogurt and many other convenience foods. Other non-nutritional products like vitamins, antacids, medicinal capsules also can contain tartrazine. For the humans, tartrazine is considered a highly toxic material and can act as catalyst in hyperactivity [1] and other behavioral problems [2]. It may also cause asthma, eczema, thyroid cancer and lupus [3,4]. Tartrazine appears to cause the most allergic reactions of all the azo dyes. Other harmful reactions can include migraine, blurred vision and itching [5-7]. Tartrazine sensitivity is mainly manifested by urticaria. It possesses high water solubility, which maximizes its harmful effects. The adsorption of tartrazine has been studied by several adsorbents like hen feathers [8], bottom ash and de-oiled soya [9], polystyrene anion exchange resins [10], multi walled carbon nanotubes [11] and activated carbon [12].

The most common industrial adsorbent is activated carbon. It is the oldest adsorbent known and is usually prepared from using one of the two basic activation methods; physical and chemical [13]. Adsorption on activated charcoal (AC) is widely employed for removal of colors and organic pollutants due to its extended surface area, microporous structure, high adsorption capacity and high degree of surface reactivity [14,15].

Activated charcoal, due to its strong adsorption properties can be effective in binding with some harmful substances in the stomach and intestine. In case of poisoning, activated charcoal can absorb or hold the toxin to its surface, and prevent its absorption in the digestive tract. It can be also used for the treatment of drug overdose as well. However, the activated charcoal that is used in the case of poisoning usually comes in powder form or just suspended in a liquid. Charcoal tablets are also used for digestive ailments, especially for indigestion, diarrhea and flatulence. Activated charcoal can absorb excess gas in the stomach and intestine and thereby, alleviate bloating, flatulence and

heartburn. Activated charcoal can interact with certain medications or affect their absorption in the intestinal tract. Therefore, it is better not to take medications one or two hours before and after taking activated charcoal tablets [16,17].

The present study aimed to examine the ability of oral medical formulations of activated charcoal to remove toxic tartrazine food coloring dye by adsorption under stomach-like conditions. The study has been carried out under different variables, as pH, temperature, adsorbent dose and dye concentration. The effect of salinity and some additives usually present in stomach as mono- and disaccharides, saccharine and aspartame, and some surfactants were examined at strong acidic solutions at 37°C.

Experimental

Reagents and solutions

Medical carbon tablets: Eucarbon[®] (AC1) and Neo Carbotrina[®] (AC2) were obtained from SEDICO Pharmaceutical Company (6 October City, Egypt) and The Arab Drug Company ADCO (El Amiriya, Egypt), respectively. Five tablets of each formulation of the compositions shown in Table 1, were crushed and homogenized to a uniform size (≤ 0.05 mm) and kept in a closed glass bottle until used.

Tartrazine (Figure 1), trisodium-5-hydroxy-1-(4-sulfonatophenyl)-4-(4-sulfonatophenylazo)-H-pyrazole-3-carboxylate is an azo dye (CI Number=19140, EEC Number=E-102) with molecular formula

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Activated Carbon	Component	Amount/tablet (mg)	% w/w
AC1	Senna leaf	105	29.09
	Rhubarb extract	25	6.92
	Activated vegetable charcoal	180	49.86
	Sulphur (sublimated)	50	13.85
	Peppermint piperitae oil	0.5	0.14
AC2	Fennel oil	0.5	0.14
	Belladonna	10	2.77
	Charcoal	100	27.77
	Di iodohydroxyquinoline	125	34.72
	Phthalyl Sulphathiazole	125	34.72

Table 1: Composition of commercial activated carbon tablets.

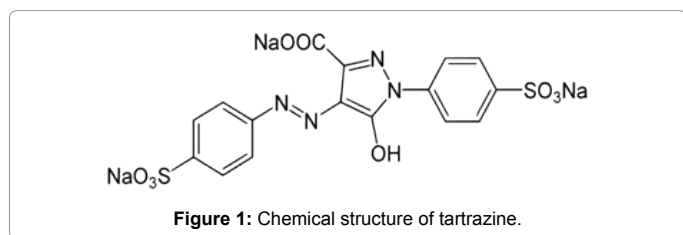


Figure 1: Chemical structure of tartrazine.

$C_{16}H_9N_4Na_3O_9S_2$ and molecular weight 534.4 was obtained from Fluka. All other reagents were of A.R. grade.

Apparatus

Absorbance measurements were performed a double-beam JascoV-530 (UV-Vis) spectrophotometer (Japan). The solution pH has been measured with a pH-meter model HI 8014, HANNA Instruments (Italy).

Adsorption studies

A 500 mg L⁻¹ stock solution was prepared by dissolving the required amount of dye in deionised water. Working solutions of the desired concentrations were obtained by successive dilution. Dye concentrations were analyzed before and after adsorption using a double-beam spectrophotometer at 428 nm versus deionized water as blank. After centrifugation at 5000 rpm, the concentrations of tartrazine in supernatant were measured and average values of two replicates were taken for each determination.

Batch adsorption experiments were carried out at temperature (37°C ± 1). A known weight of adsorbent material was added to 50 ml of the dye solution with an initial concentration of 50 to 300 mg L⁻¹. The contents were stirred thoroughly using a magnetic stirrer with a speed of 100 rpm after adding the required dose of adsorbents (4-20 g L⁻¹) for a specific period of contact time (5-120 min). The pH of the test solution was adjusted to the required value by adding either 1M HCl or 1M NaOH solution. After equilibrium, the final concentration (C_e) was determined and the percentage removal of dye was calculated using the following relationship:

$$\% \text{ Dye Removal} = \frac{C_0 - C_e}{C_0} \times 100 \quad (1)$$

Where C_0 and C_e are the initial and final (at equilibrium) concentrations of dye (mg L⁻¹), respectively

Results and Discussion

Effect of initial pH

The effect of solution acidity on tartrazine sorption by medical activated carbon was studied in the initial pH range of 1.5-11.0 and is

shown in Figure 2. The adsorption studies were carried out at 37°C. A typical pH versus percentage removal of the dye showed a remarkable decrease in percentage removal of dye at higher pH values. The decrease in the uptake of dye above pH 6.0 and thereafter no change in the adsorbed amount were observed. Maximum uptake of the dye (43.4 and 56.2%) was achieved at pH 1.5 for AC1 and AC2, respectively. Therefore, pH 1.5 was selected as an optimum value for performing subsequent studies. This pH value is also the approximate pH of the human stomach. The result obtained clearly indicates increase of protonation due to neutralization of negative charge at the surface of the adsorbents, which facilitates diffusion and provides more active surface of the adsorbents, resulting thereby greater adsorption at their surfaces. The pH of solution controls the electrostatic interactions between the adsorbent and the adsorbate. A decrease in the percentage removal with increase in pH may be due to deprotonation, which retards the electrostatic forces between sorbent and sorbate that leads to reduced sorption capacity [18]. The effect of pH on the adsorption capacity of AC is in agreement with the previously reported results for adsorption of tartrazine from aqueous solutions by hen feathers [8] and some waste materials [9].

Effect of contact time

The effect of contact time between AC and tartrazine solution was studied for AC1 and AC2 using a constant concentration (50 mg/ml) of dye solution at 37°C. The adsorption of tartrazine dye onto AC has been investigated as a function of time in the range of 5-120 min at constant stirring rate of 100 rpm. Figure 3 shows percentage removal of the dye with contact time. As seen in Figure 3, a higher removal percentage of tartrazine dye is obtained at the beginning of the adsorption. Quantitative adsorption of dye from solution was determined within about 90 min. Therefore, 90 min stirring time was found to be appropriate for maximum adsorption and was used in all subsequent measurements.

Effect of initial dye concentration

The effect of initial dye concentration in the range of 50 to 300 mg L⁻¹ tartrazine on adsorption efficiency onto AC1 and AC2 was investigated and is shown in Figure 4. It is observed from the figure that the percentage tartrazine removal decreased with the increase in initial concentration of tartrazine. Though the percent adsorption

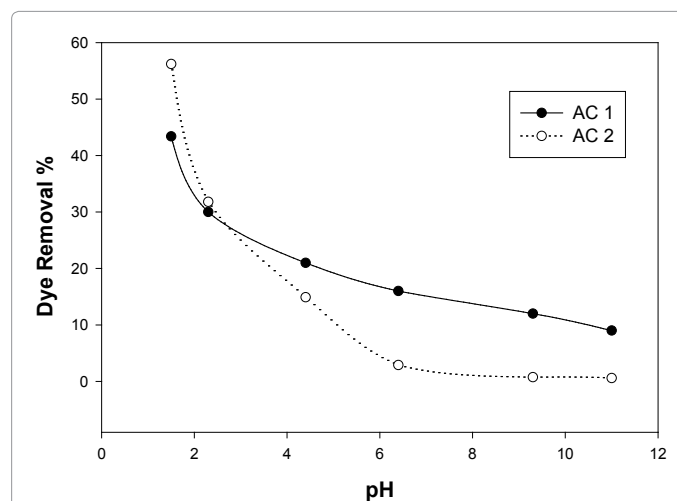


Figure 2: Effect of pH on the percent removal of tartrazine at equilibrium onto AC1 and AC2 at 37°C, C_0 100 mg L⁻¹ and AC dose 4 g L⁻¹.

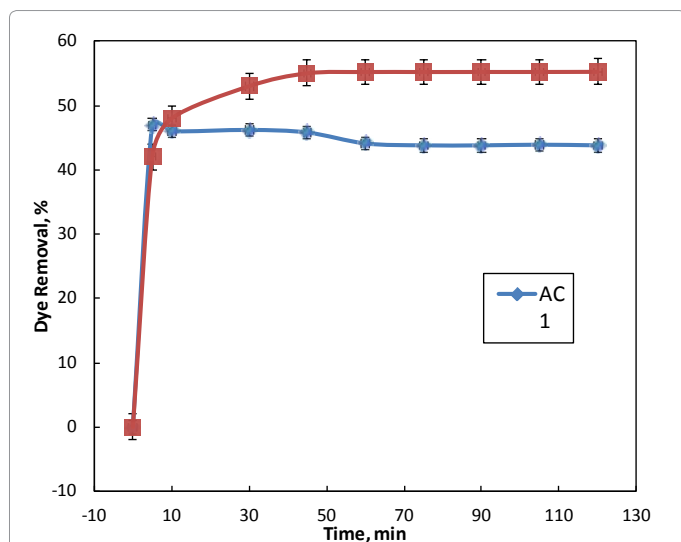


Figure 3: Effect of contact time on the percent removal of tartrazine onto AC1 and AC2 at pH 1.5, temperature 37°C, C_0 100 mg L⁻¹ and AC dose 4 g L⁻¹.

decreased with increase in initial dye concentration, the actual amount of dye adsorbed per unit mass of adsorbent increased with increase in dye concentration in test solution. The unit adsorption for AC1 was increased from 10.85 mgg⁻¹ to 35.70 mgg⁻¹ as the tartrazine concentration in the test solution was increased from 50 mgL⁻¹ to 300 mgL⁻¹. Similarly, unit adsorption for AC2 was increased from 14.05 mgg⁻¹ to 50.1 mgg⁻¹ as the dye concentration was increased from 50 mgL⁻¹ to 300 mgL⁻¹.

The initial dye concentration provides the necessary driving force to overcome the resistance to the mass transfer of tartrazine between aqueous phase and the solid phase. The increase in initial dye concentration results also an increase in the interaction between dye molecules and AC surface. Therefore, the increase of the initial concentration of tartrazine enhances the adsorption uptake of the dye [19].

Effect of activated carbon dose

The influence of adsorbent dosage on equilibrium uptake was shown in Table 2. The adsorbent dosages were taken between 2-10 gL⁻¹ at initial dye concentration of 50 mgL⁻¹. With the increasing of the activated carbon dose, the adsorption capacity increased. Increasing adsorbent dosage can be attributed to increased adsorbent surface area and the availability of more adsorption sites. The values of equilibrium concentration of dye q_e decreased with increasing the adsorbent dosage. This factor explaining this result is that adsorption sites remain unsaturated during the adsorption reaction whereas the number of sites available for adsorption site increases by increasing the adsorbent dose [20].

Effect of stirring rate

The effect of stirring rate on the dye adsorption at the adsorbent dosage of 0.10 g/25 ml, initial dye concentration of 50 mgL⁻¹, pH 1.5 was examined at equilibrium (60 min). The data obtained indicate that the adsorption capacity (q_e) decreased as the agitation speed increased from 100-400 rpm (Figure 5a). For an rpm of the 100, the sorption of the dye on AC has its maximum value. It is due the attractive force between dye and adsorption site increased to 100 rpm, but in higher

stirring rate, the dye molecules do not have enough time for contact with sorbent active sites [21].

Effect of salinity

The effect of salinity on the adsorption of tartrazine on AC was examined by adding different amounts (2-10 g L⁻¹) of NaCl to a constant concentration of tartrazine (100 mg L⁻¹). The experiments were performed at 37°C and pH 1.5. As shown in Figure 5b, the increase of salt concentration caused a remarkable decrease of adsorption efficiency for the two activated carbon sample used. Ionic strength is one of the key factors affecting the electrical double layer (EDL) structure of a hydrated particulate. The increase in ionic strength leads to a decrease in EDL thickness and an increase in the amount of indifferent ions approaching the AC surface. Thus, the results obtained can be attributed in part to increased competition between tartrazine and Na⁺ ions for surface sites with increasing the ionic strength [22].

Sorption modeling

To describe the equilibrium distribution of tartrazine between the AC surface and the aqueous solution phase, two different sorption models, Langmuir and Freundlich have been used. These models were also used to calculate the loading capacity of the two forms of AC examined in this study.

Langmuir isotherm

The most widely used isotherm equation [23] for modeling of the sorption equilibrium data is the Langmuir isotherm. This model assumes the uniform energies of adsorption onto the surface without any transmigration of adsorbate in the plane of the surface. Langmuir

Activated Charcoal Dose (g L ⁻¹)	AC1		AC2	
	% Removal	q_e (mg g ⁻¹)	% Removal	q_e (mg g ⁻¹)
4	31.84	7.95	37.00	9.25
8	52.20	6.53	56.70	7.09
12	63.00	5.25	66.20	5.52
16	67.00	4.19	70.40	4.40
20	67.51	3.37	71.80	3.59

Table 2: Effect of activated carbon dose on percent removal at equilibrium.

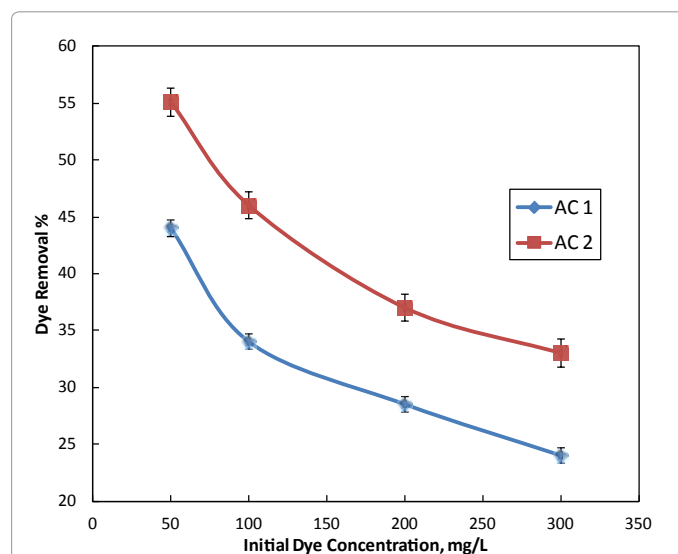
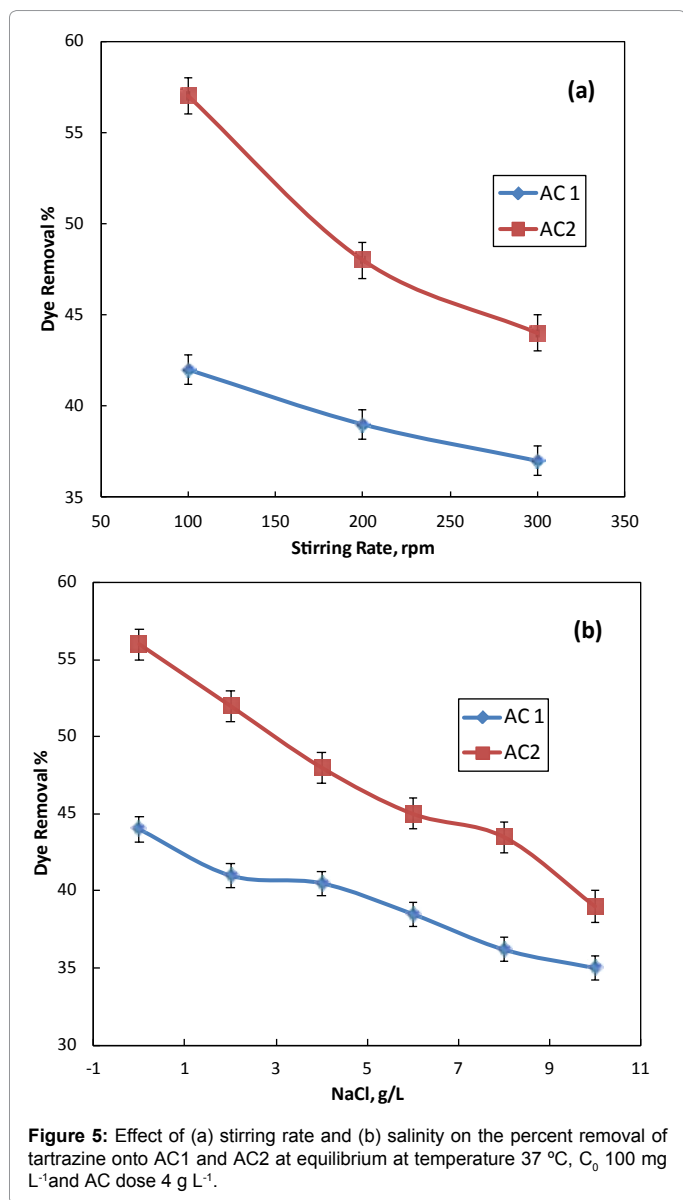


Figure 4: Effect of initial dye concentration on percent removal at equilibrium at temperature 37°C and AC dose 4 g L⁻¹.



sorption is a model based on the physical hypothesis that there are no interaction between adsorbed molecules and the adsorption energy over the entire coverage surface. This model imposes that a particular site of the adsorbent is occupied by an adsorbate molecule; no further adsorption takes place at that site, i.e. forming a monolayer of adsorbed species. The linear form of Langmuir isotherm equation is given by the equation [24]:

$$\frac{C_e}{q_e} = \frac{1}{q_m K_L} + \frac{C_e}{q_m} \quad (2)$$

where, C_e is the equilibrium concentration of the dye (mg/L), q_e is the amount of dye adsorbed per unit mass of AC (mg g⁻¹), q_m and K_L are Langmuir constants related to adsorption capacity and rate of adsorption, respectively. The Langmuir constants can be evaluated from the slope and the intercept of linear equation (Table 3). The correlation coefficients of Langmuir isotherms, R^2 are 0.9501 and 0.9606 for AC1 and AC2, respectively. The essential characteristics of the Langmuir isotherm can be expressed in terms of a dimensionless constant separation factor R_L that is given in Equation. 3: The value of

R_L indicates the type of the isotherm to be either favorable ($0 < R_L < 1$), unfavorable ($R_L > 1$), linear ($R_L = 1$) or irreversible ($R_L = 0$). The values of R_L were found to be < 1 for the two adsorbents (Table 3) suggesting the isotherm to be favorable at the concentrations studied [25].

Freundlich isotherm

This model considers a heterogeneous adsorption surface that has unequal available sites with different energies of adsorption [26] and can be represented by:

$$\ln q_e = \ln K_f + \frac{1}{n} \ln C_e \quad (3)$$

where, K_f (mg/g) and n are Freundlich constants giving an indication of how favorable the adsorption process. K_f can be defined as the adsorption or distribution coefficient and represents the quantity of dye adsorbed onto the fibers for a unit equilibrium concentration. The slope of $1/n$ ranging between 0 and 1 is a measure of adsorption intensity or surface heterogeneity, becoming more heterogeneous as its value gets closer to zero [27].

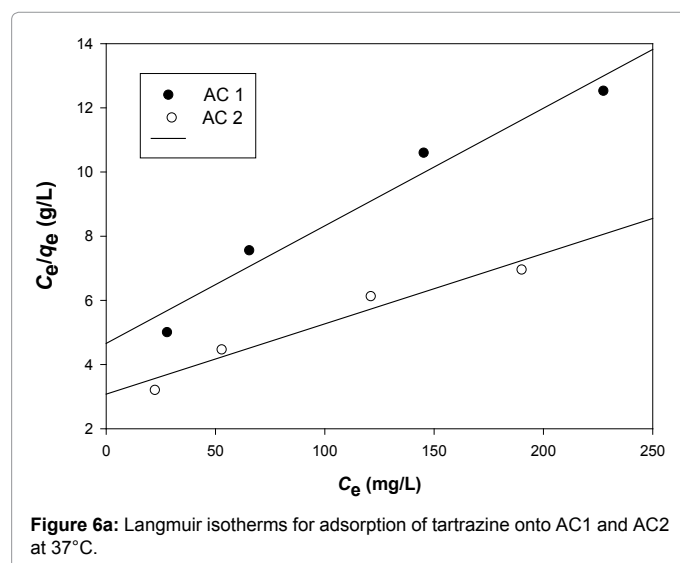
To compare the Langmuir and Freundlich isotherm models, the experimental data have been statistically processed by linear regression. The regression equations of $y=ax+b$ type and the obtained values of the correlation coefficient, R^2 , are given in Table 3. It can be seen (Table 3) that the obtained data fit better to the Freundlich model than the Langmuir model (higher values for R^2), so the formation of more than one molecular layer of tartrazine on the surface of activated carbon appears to be achieved in the case of AC1 and AC2. The Langmuir and Freundlich isotherm plots are shown in Figures 6 a and b, respectively.

Effect of temperature and thermodynamic studies

The temperatures used in this study were 27, 37 and 47°C. As shown in Table 4, the adsorption capacity of the activated carbon decreased with increase in the temperature of the system. Activated carbon usually contains polar functional groups that can be involved in

Adsorbent	Langmuir Constants			Freundlich Constants		
	q_m (mg/g)	K_L (L/mg)	$R^2 R_L$	$K_f n$	R^2	
AC1	272.85	0.00071	0.9501	0.81-0.96	0.97	1.583
AC2	456.83	0.00079	0.9606	0.83-0.97	0.83	1.768

Table 3: Langmuir and Freundlich adsorption constants for adsorption of tartrazine on AC1 and AC2.



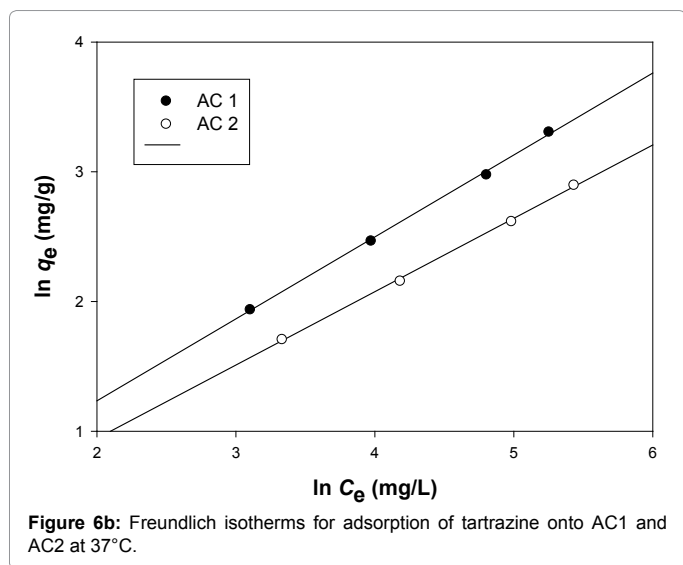


Figure 6b: Freundlich isotherms for adsorption of tartrazine onto AC1 and AC2 at 37°C.

Temperature (°C)	Tartrazine Removal %	
	AC1	AC2
27	47.40	60.0
37	39.25	57.7
47	30.26	55.8

Table 4: Effect of temperature on percent removal of tartrazine onto AC1 and AC2.

chemical bonding and are responsible for dye adsorption. The decrease in the extent of equilibrium adsorption of the dye with increasing the temperature indicated that a low temperature favored dye removal by adsorption and the adsorption of tartrazine onto AC was controlled by an exothermic process. The decrease of adsorption capacity with increasing temperature may be due to a decrease in the chemical potential of the adsorbate. It is known that decreasing sorption capacity with increasing temperature is mainly due to the weakening of sorptive forces between the active sites on the AC and anionic dye species, and also between adjacent dye molecules on the sorbed phase [28]. For a conventional mechanism of physisorption system, increase in temperature usually increases the rate of approach to equilibrium, but decreases the equilibrium capacity [29].

Thermodynamic parameters such as change in free energy ΔG° (kJ/mol), enthalpy ΔH° (kJ/mol) and entropy ΔS° (J/K/mol) were determined. ΔG° was calculated from the following equation [30]:

$$\Delta G^\circ = -RT \ln K_L \quad (4)$$

where: K_L is the Langmuir constant; T is absolute temperature and R is the gas constant (8.314 J/mol K). The apparent ΔH° and ΔS° were calculated from adsorption data at different temperatures using the Van't Hoff equation [31]:

$$\ln K_L = \frac{\Delta S}{R} - \frac{\Delta H}{RT} \quad (5)$$

where: K_L is the Langmuir constant and T is the solution temperature (K). The magnitude of ΔH° and ΔS° was calculated from the slope and intercept from the plot of $\ln K_L$ vs $1/T$ (Figure 7). The calculated thermodynamic parameters are given in Table 5. The values of ΔH° as calculated from Eq. (5) were -20.55 and -6.97 kJ/mol for AC1 and AC2, respectively. It is known that physical adsorption and chemisorptions can be classified, to a certain extent, by the magnitude of the enthalpy change. It is accepted that bonding strengths of <84 kJ/mol are

typically those of physical adsorption type bonds. Chemisorption bond strengths can range from 40 to 120 kJ/mol [32]. Based on this, the adsorption of tartrazine on activated carbon appears to be a physical adsorption process. Moreover it is expected that adsorption processes from liquid phase are exothermic due to the heat released after bond formation between solute and adsorbent [33]. The negative values of ΔS° indicate the decrease of randomness at the solid/liquid interface during the adsorption process. The adsorption of tartrazine was non-spontaneous for AC1 and spontaneous for AC2 at the temperatures under investigation as indicated from the positive and negative values of free energy (ΔG°) for the two adsorbents, respectively.

Effect of nutrient additives

It is likely that some effective nutrients may affect the adsorption efficiency of tartrazine on AC. Thus, the present part of this study was performed to develop the effect of some dietary components on the adsorption efficiency of tartrazine on AC. Different additives were added to the original dye solution in a fixed concentration of 2 gL⁻¹ and the results were compared to the tartrazine solution (Table 6). The data reveal the effect of the adsorption composition (Table 1). Table 7 shows the comparison of the proposed method to removal of tartrazine

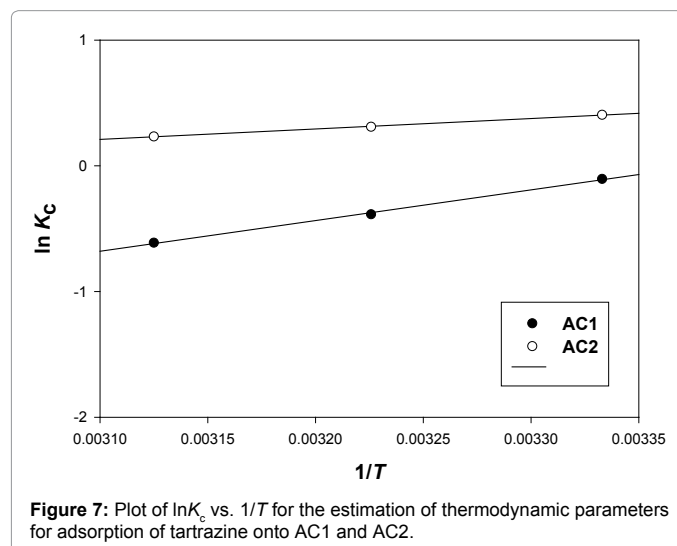


Figure 7: Plot of $\ln K_c$ vs. $1/T$ for the estimation of thermodynamic parameters for adsorption of tartrazine onto AC1 and AC2.

AC	Temperature (°C)	ΔG°	ΔH°	ΔS°
AC1	27	262.48	-20.55	-69.44
	37	1004.87		
	47	1647.33		
AC2	27	-1023.18	-6.97	-19.86
	37	-809.53		
	47	-627.27		

Table 5: Thermodynamic parameters of adsorption of tartrazine onto AC1 and AC2.

Nutrient added*	Effect on dye removal %	
	AC1	AC2
Glucose	53.92	65.12
Succarose	44.50	69.17
Glycine	55.63	63.15
Lactose	53.38	64.66
Saccharine	43.60	66.90
Aspartame	50.30	72.20

Table 6: Effect of nutrients added on the percent removal of tartrazine onto AC1 and AC2.

Determination technique	Adsorbent	Optimum conditions	Ref.
FT-IR, SEM	Saw dust	70 min, pH 3	[35]
X-ray diffraction	Bottom Ash and De-Oiled Soya	3–4 h, pH 2	[9]
Spectrophotometry	Hen feathers	pH 2	[8]
Spectrophotometry FT-IR analysis	Polystyrene anion exchangers	20 min, 50°C	[10]
IR, BET, SEM, TGA	Melamine-formaldehyde-Tartaric acid resin	15-30°C	[36]
Spectrophotometry	Polyaniline	pH range of 2-5	[37]
Spectrophotometry	Electrocoagulation	-----	[38]
Spectrophotometry	Titanium dioxide	pH 11	[39]
Spectrophotometry	Medical tablets of activated charcoal	pH 1.5 and 37°C	This work

Table 7: Comparison of the proposed method to removal of Tartrazine with those reported previously.

with those reported previously. It shows that the proposed method is the first one in terms of the use of medical products and are commonly used to removal of tartrazine by high rate and efficiency.

Conclusion

The present study was carried out to examine the ability of commercial activated carbon tablets to adsorb artificial coloring agent tartrazine from aqueous solutions. Activated carbon tablets appeared to be a promising adsorbent for the removal of tartrazine from aqueous solutions under gastrointestinal-like conditions. The optimum pH for removal process was around 1.5. The extent of dye removal increased with decreased initial concentration of the dye and also increased with increased contact time and AC dose. Adsorption of tartrazine onto the two types of AC occurred in a short time and reached equilibrium in about 90 min for the two types of tablets. AC2 had a higher adsorption capacity than AC1 under the same conditions. The results obtained are well fitted in the linear forms of Freundlich more than Langmuir adsorption isotherms for either AC1 or AC2. The adsorption process was found to be exothermic. The adsorption process is exothermic and non-spontaneous for AC1 and spontaneous for AC2. The values of entropy change suggested the probability of favorable adsorption. Medical activated charcoal tablets appear as a very prospective adsorbent for the removal of tartrazine from aqueous solution.

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