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Adsorption of Sunset Yellow (E110) Food Dye onto Granular Activated Carbon: Thermodynamic and Kinetic Evaluation

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Abstract | This study investigated the adsorption behavior of the artificial food dye Sunset Yellow (E110) using granular activated carbon as an adsorbent material. Sunset Yellow is a complex, high-molecular weight chemical compound containing several active functional groups. The adsorption characteristics of the dye were evaluated through spectrophotometric analysis using UV-Visible spectroscopy to establish a standard calibration curve and determine the remaining dye concentration after adsorption. Adsorption capacity and removal efficiency were subsequently calculated. Several factors affecting the adsorption process were examined, including the initial dye concentration, temperature range (298-318 K), adsorbent dosage, contact time and solvent type. Thermodynamic parameters of the adsorption process were determined using equilibrium constant values and the Van't Hoff equation. The calculated positive values of enthalpy change (ΔH°) and distribution coefficient (Kd) indicated that the adsorption process was endothermic in nature. Since the ΔH° values did not exceed 40 kJ/mol, the adsorption was identified as predominantly physical adsorption. The negative Gibbs free energy (ΔG°) values obtained from equilibrium constant calculations demonstrated that the adsorption process was spontaneous at lower temperatures, with spontaneity increasing as temperature increased while maintaining a constant initial dye concentration. However, increasing the initial dye concentration at constant temperature reduced the spontaneity of adsorption. In contrast, calculations based on Kd values produced positive ΔG° values, indicating non-spontaneous adsorption behavior under those conditions. Positive entropy values (ΔS and ΔS°) suggested increased molecular organization of dye molecules at equilibrium during the adsorption process. The sticking probability (S*) was calculated using the modified Arrhenius equation and the obtained values fell within the preferred adsorption range (0<S*<1), confirming the predominance of physical adsorption on the adsorbent surface. Furthermore, the apparent activation energy values indicated that the adsorption process was diffusion-controlled and favored lower temperatures within the experimental range investigated in this study.

Key Words Artificial Food Dyes, The Sticking Probability, Adsorption

INTRODUCTION

Sunset Yellow (E110) is a yellow azo dye synthesized from coal tar and possesses the following characteristics: it belongs to the monoazo dye group and exists in the form of reddish-orange solid granules. It is considered one of the harmful synthetic dyes because it is widely used in many food industries and carbonated beverages, which has led researchers to investigate the use of inexpensive and naturally available materials for its removal through adsorption [1].

The attractive force between the liquid substance adsorbed onto the surface of a solid material and the energy required to return that substance back into the solution is measured by the heat of adsorption (enthalpy, ΔH). This parameter helps determine the nature of the adsorption process, whether it is physical or chemical adsorption [2]. The heat of adsorption can be calculated using the Van't Hoff equation, as shown in Equation 1:

$$\ln Kc = \ln K_o - \frac{\Delta H}{RT} \quad (1)$$

Where:

- ΔH represents the heat of adsorption (enthalpy)
- Kc represents the adsorption equilibrium constant
- K_0 represents a constant value

- T represents the absolute temperature
- R represents the universal gas constant (8.314 J/mol·K)

Plotting the relationship between ln(Kc) and (1/T) produces a linear relationship with a straight-line slope equal to (-ΔH/R). From this relationship, the value of the heat of adsorption can be calculated. The adsorption equilibrium constant (Kc) can be determined by dividing the concentration of the adsorbed substance (Cads) by the remaining equilibrium concentration (Ce) of the non-adsorbed substance, as shown in the following equation [3]:

$$Kc = \frac{C_{ads.}}{Ce} \quad (2)$$

$$C_{ads.} = C_i - Ce \quad (3)$$

Where:

- Cads represents the concentration of the adsorbed substance (mg/L)
- Ce represents the concentration of the non-adsorbed substance at equilibrium (mg/L)
- Ci represents the initial concentration of the adsorbate (mg/L)

The values of the thermodynamic functions can be calculated using the following equations:

$$\Delta G° = -RT \ln Kc \quad (4)$$

$$\Delta G° = \Delta H - T\Delta S° \quad (5)$$

$$\Delta S° = \frac{(\Delta H - \Delta G°)}{T} \quad (6)$$

$$\Delta S = \Delta H/T \quad (7)$$

The study of the distribution coefficient (Kd), the sticking probability of adsorbate ions on the adsorbent surface (S*) and the apparent activation energy (Ea) is important for identifying the nature of the adsorption process, whether it is physical adsorption, chemical adsorption or a combination of both. This can be determined through thermodynamic analysis of the adsorption process using the modified Arrhenius equation [4].

$$S^* = (1 - \theta) \exp - Ea-/RT \quad (8)$$

$$\theta = 1 - \frac{Ce}{Ci} \quad (9)$$

$$\ln (1-\theta) = \ln S^* - Ea-/RT \quad (10)$$

Plotting the relationship between ln(1-θ) and 1/T gives a linear relationship in which the intercept represents ln(S*) and the slope represents (-Ea/R). The sticking probability (S*) is defined as the rate of adsorption for each molecular collision with the surface [5].

When the value of S* is greater than 1 (S*>1), this indicates that the ions do not adhere to the solid surface. If S* = 0, adsorption tends toward infinity on the surface, indicating chemical adsorption. When S* = 1, the adsorption process represents a combination of physical and chemical adsorption. However, when 0<S*<1, adsorption is considered favorable, indicating preferred adhesion of the adsorbate onto the adsorbent surface, with physical adsorption being the dominant mechanism [6-9].

Experimental Section

The practical aspect plays a fundamental role in all applied sciences because of its accuracy, reliability and importance in obtaining precise results, as well as in the preparation of chemical materials and the use of laboratory equipment. Therefore, this study focused extensively on the experimental procedures, which can be explained through the following sections:

Chemicals and Materials

The primary chemicals and solvents used in this study were obtained from BDH Chemicals and Fluka Chemicals. The materials used included:

- **Activated Carbon (Granular):** Granular activated carbon was used in the form of solid granules. The material was heated in an electric oven at 100°C for 3 hours and then stored in a dry, tightly sealed container
- Hydrochloric Acid (HCl)
- Sodium Hydroxide (NaOH)
- Absolute Ethanol
- Distilled Water
- Sunset Yellow Food Dye (E110)

The synthetic food dye Sunset Yellow was purchased from markets in Mosul in tightly sealed bottles manufactured by internationally recognized companies, including Ajanta Chemical Industries, India (Table 1).

Laboratory Instruments Used

Electrothermal Melting Point Apparatus-9300: Used for measuring the melting point of the dye.

pH Meter-JENWAY 3510

The instrument was calibrated using standard buffer solutions (pH 7 and pH 9). The acidity function (pH) of the synthetic food dye solution was measured at the optimum temperature before the adsorption process.

Table 1: Chemical Name, Molecular Formula and Selected Physical Properties of Food Dye E110

Property	Description
Dye Symbol	E110
Chemical Family	Monoazo
Common Name	Sunset Yellow
Scientific Name	Disodium; 6-hydroxy-5-[(4-sulfonato phenyl)diazenyl] naphthalene-2-sulfonate
Chemical Formula	C16H10N2O7S2Na2
Molecular Weight	452.37 g/mol
pH Range	8-Jun
Melting Point	300 °C
Λmax	480 nm
Molar Absorptivity (ε)	19707 L/mol·cm
Optimum Adsorption Conditions	Ci = 45.237 g/L, Temp. = 45°C, pH = 8.11

Shaker with Water Bath-Julabo SW 23
Used to control and maintain the temperature during the adsorption process.

Drying Oven-Memmert
Used for drying the food dye material under investigation.

T92+UV Spectrophotometric PG Instruments
Used for measuring ultraviolet and visible spectral absorption in order to determine the λmax values of the synthetic food dye.

CECIL Spectrophotometric 1000S
Used for measuring the absorption spectra of the dye using distilled water as solvent and blank solution with 1 cm optical cells.

Preparation of Solutions

- A 1.0 M sodium hydroxide (NaOH) solution was prepared by dissolving 0.4 g of NaOH in 100 mL of distilled water
- A 0.1 M hydrochloric acid (HCl) solution was prepared by dilution of concentrated HCl (11.63 M) according to the dilution law to obtain 100 mL of solution
- A standard solution of Sunset Yellow dye (E110) was prepared by dissolving 1 g of the dye in one liter of distilled water. Additional lower concentrations within the range of approximately (2×10^{-5} M to 10×10^{-5} M) were prepared using the dilution law

The change in the amount of adsorbed dye with time was monitored using Lambert-Beer's law to construct a calibration curve at λmax by plotting absorbance versus concentration in order to determine the molar absorptivity coefficient from the slope of the relationship according to the following equation:

$$A = \varepsilon 0000 LCA = \varepsilon L C A = \varepsilon LC$$
$$\varepsilon\varepsilon\varepsilon$$
$$ccc$$
$$lll$$

Where:

- A represents the absorbance of the synthetic food dye at various concentrations
- ε represents the molar absorptivity coefficient (L/mol·cm)
- C represents the concentration of the synthetic food dye (M)
- L represents the optical path length of the absorption cell (1 cm)

Maximum Wavelength and Calibration Curve of E110 Dye
The maximum wavelength (λmax) of the synthetic food dye was determined by recording the absorption spectrum of the dye solution at constant concentration using the T92+ UV Spectrophotometric PG Instruments device. The maximum wavelength was identified at the highest absorbance value.

For a proper adsorption study, it is essential to use an appropriate analytical method to determine the remaining concentration of the adsorbate after adsorption onto the adsorbent surface. Since the adsorbate in this study was a colored compound, the spectrophotometric method in the ultraviolet-visible region (200-800 nm) was considered the most suitable analytical technique.

The concentration of the synthetic food dye before and after adsorption was determined using the CECIL Spectrophotometric 1000S UV-Visible spectrophotometer. The obtained results were expressed in mg/L using the previously prepared standard calibration curve.

The amount of adsorbed dye was expressed in terms of adsorption capacity (qe) and adsorption efficiency (%) through estimation of the remaining concentration (Ce) and adsorbed amount (Cads), according to the following equations.

RESULTS AND DISCUSSION

This study investigated the potential effects of synthetic food dyes on human health through the application of chemical principles [10]. Numerous previous studies and scientific reports [11,12] have demonstrated considerable interest in developing effective methods for reducing or eliminating the harmful effects of synthetic food dyes on both human health and the environment. Understanding the behavior and impact of these synthetic dyes requires extensive and detailed investigations by researchers.

Table 2: Effect of Adsorbent Weight on the Adsorption Efficiency and Adsorption Capacity of Synthetic Food Dye E110 at Natural pH

gm	Ce mg/L	qe mg/gm	Percentage
0.01	5.881	15.832	34.9979
0.05	4.5784	4.469	49.39541
0.07	3.961	3.633143	56.21947
0.1	3.176	2.9357	64.89599
0.5	1.804	0.72434	80.06057

Table 3: Effect of Contact Time on the Adsorption Efficiency and Adsorption Capacity of Synthetic Food Dye E110 on 0.5 g of Granular Activated Carbon at Natural pH

t min.	Ce mg/L	qe mg/gm	Percentage
5	3.2544	0.57926	64.02785
15	2.5097	0.65373	72.25931
25	2.0389	0.70081	77.46325
35	1.8036	0.72434	80.06411
45	1.3725	0.76745	84.82923
60	1.1762	0.78708	86.99901
70	1.0194	0.80276	88.73218
80	1.0194	0.80276	88.73218

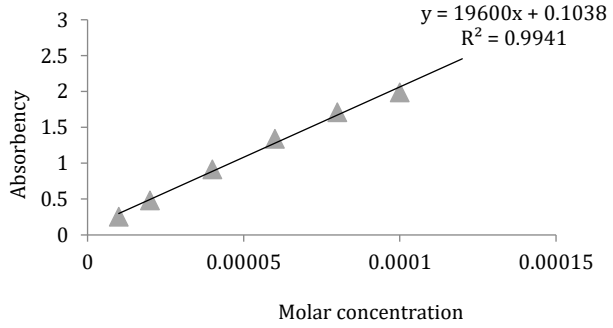

Figure 1: Calibration Curve of the Synthetic Food Dye E110

Determination of the Calibration Curve of Food Dye E110

The calibration curve of the synthetic food dye E110 was determined at its maximum wavelength (λmax), as illustrated in Figure 1, by preparing dye solutions of different concentrations and measuring their absorbance values. The relationship between absorbance intensity and molar concentration of the food dye was then plotted. The resulting relationship was expected to follow Lambert-Beer's law according to Equation (11).

From the calibration curve, straight-line relationships were observed over well-defined concentration ranges with correlation coefficient (R^2) values close to unity. In addition, the molar absorptivity coefficient values were satisfactory, indicating that this spectrophotometric method can be reliably used for determining the concentration of the synthetic food dye investigated in this study.

Since the primary objective of this research was to conduct a thermodynamic study of the adsorption process, it was necessary to determine suitable concentrations of the synthetic food dye solutions and the appropriate amount of adsorbent material (granular activated carbon) until adsorption equilibrium was achieved. This allowed evaluation of the influence of various factors on the adsorption process.

Study of the Influencing Factors

Effect of Adsorbent Weight: It was observed that increasing the weight of the adsorbent material resulted in an increase in adsorption efficiency (%) and a decrease in adsorption capacity (qe). This behavior can be attributed to the increase in the number of available active sites for adsorption on the adsorbent surface, which enhanced the adsorption of food dye molecules. Consequently, the percentage of adsorption increased, while the ratio of milligrams of adsorbed material to milligrams of adsorbent decreased, as shown in the Table 2.

For the present study, an adsorbent amount of 0.5 g was selected because it provided an appropriate level of dye removal while still retaining a measurable portion of the dye in solution, thereby satisfying the requirements of the study and enabling accurate thermodynamic calculations.

Effect of Contact Time

To determine the type, nature and behavior of the adsorption system and to evaluate the adsorption ability of the dye molecules from solution onto the solid adsorbent surface, the effect of contact time between the synthetic food dye molecules and the surface of granular activated carbon was investigated. This factor plays a significant role in the adsorption process.

The study was conducted by determining the adsorption rate and identifying the optimum contact time required for maintaining the adsorbent material in contact with the dye solution until equilibrium was reached, while all other experimental parameters were kept constant. The obtained results are presented in the Table 3.

Since the adsorption rate reaches its maximum during the initial minutes of the adsorption process, it was observed that the highest amount of synthetic food dye was transferred onto the surface of granular activated carbon at approximately 70 minutes. After this period, the adsorption percentage gradually became constant until equilibrium was achieved.

Table 4: Effect of Solvent Composition on the Adsorption Efficiency and Adsorption Capacity of Synthetic Food Dye on 0.5 g of Granular Activated Carbon at Natural pH and Optimum Temperature

ايثانول : ماء	Ce mg/L	qe mg/gm	%
90-10	3.3329	0.57145	63.16179
80-20	2.9408	0.61066	67.49563
60-40	2.744	0.63034	69.67084
40-60	2.3526	0.66948	73.99695
20-80	0.4313	0.86161	95.23288

This behavior can be explained by the fact that, at the beginning of contact between the synthetic food dye molecules and the adsorbent surface, a large number of active adsorption sites are available, allowing rapid diffusion and adsorption of dye molecules during the early stages of contact. As time progresses, the number of available active sites decreases, resulting in competition among the remaining non-adsorbed dye molecules as well as interactions with the already adsorbed molecules. Consequently, repulsive forces increase between the molecules, causing some adsorbed molecules to return to the solution until adsorption equilibrium is reached, where the number of adsorbed molecules equals the number of desorbed molecules.

Effect of Solvent on Adsorption

An appropriate organic solvent, ethanol, was selected and mixed with water in order to investigate the effect of solvent composition on the adsorption process. The adsorption efficiency and adsorption capacity of the synthetic food dye on the adsorbent surface were studied using ethanol-water solvent mixtures with ethanol volume percentages ranging from 10% to 90% under optimum experimental conditions, as shown in the Table 4.

From Table 4, it was observed that the adsorption efficiency and adsorption capacity of dye E110 increased with increasing ethanol content, corresponding to a decrease in the dielectric constant of the solvent mixture. It is well known that water possesses a higher dielectric constant (approximately 80) compared to ethanol (approximately 65). Therefore, mixing water with ethanol produces solvent systems with dielectric constants lower than that of pure water.

An increase in the dielectric constant of the solvent enhances the tendency of dissolved dye molecules to migrate toward the adsorbent surface rather than participating in solute-solute and solvent-solute molecular interactions. Consequently, the adsorption efficiency increases with increasing dielectric constant of the solvent system [13].

Effect of Initial Dye Concentration

The effect of five different initial concentrations of the food dye was investigated within the range of $(2\text{-}10)\times10^{-5}$ M over the temperature range used in this study while keeping all other experimental variables constant. A fixed volume of the food dye solution was shaken at a constant speed of 100 rpm using the same amount of adsorbent material for 70 minutes. After filtration, the remaining concentration of the dye in the solution was determined spectrophotometrically following the adsorption process. The obtained results are presented in the Table 5.

From Table 5, it was observed that increasing the initial concentration of the synthetic food dye at constant temperature resulted in a decrease in both adsorption capacity and adsorption efficiency. This behavior can be explained by the fact that increasing dye concentration increases the number of dye molecules available for adsorption while the number of active adsorption sites on the surface of granular activated carbon remains constant.

As the number of dye molecules increases, repulsive interactions among the molecules become stronger due to competition for the available active sites on the adsorbent surface. Furthermore, increasing concentration enhances molecular interactions between dye molecules as well as interactions between the dye and the solvent because the molecules become closer to each other, leading to the formation of intermolecular hydrogen bonds and Van der Waals forces. In addition, a large number of dye molecules remain in solution after equilibrium is reached, resulting in lower adsorption capacity and adsorption efficiency.

The ionization process of the synthetic food dye also decreases with increasing concentration, thereby reducing the number of ions transferred to the solid surface, which consequently decreases adsorption efficiency and adsorption capacity [14].

It was also observed that the adsorption percentage reached its highest value at the lowest dye concentration (2×10^{-5} M) and gradually decreased with increasing concentration. This indicates that adsorption is more efficient in dilute dye solutions. At higher concentrations, the number of dye molecules transferred to the adsorbent surface increases, leading to stronger competition among molecules for active adsorption sites. As a result, repulsive forces increase and some molecules return to the solution, causing a reduction in adsorption efficiency [15-17].

Effect of Temperature

The effect of temperature on the adsorption efficiency of the investigated synthetic food dye onto the adsorbent surface was studied while keeping all other experimental conditions constant. The obtained results are presented in the Table 6.

It was observed that the adsorption capacity and adsorption efficiency increased with increasing temperature at constant dye concentration, indicating that temperature acts as a positive factor in the

Table 5: Effect of Initial Concentration on the Adsorption of Synthetic Food Dye E110 onto Granular Activated Carbon

T k◦	Ci mg/L	$C_{ads.}$ mg/L	qe mg/gm	%
298	9.0474	7.4751	0.74751	82.62153
	18.0948	14.702	1.4702	81.2498
	27.1422	21.967	2.1967	80.93301
	36.1896	28.3076	2.83076	78.22026
	45.237	33.3673	3.33673	73.76108
303	9.0474	7.7857	0.77857	86.05456
	18.0948	14.947	1.4947	82.60384
	27.1422	22.0422	2.20422	81.21007
	36.1896	28.9846	2.89846	80.09097
	45.237	34.1258	3.41258	75.43781
308	9.0474	7.8582	0.78582	86.85589
	18.0948	15.1634	1.51634	83.79977
	27.1422	22.1421	2.21421	81.57813
	36.1896	29.0632	2.90632	80.30815
	45.237	34.3481	3.43481	75.92922
313	9.0474	7.8712	0.78712	86.99958
	18.0948	15.3803	1.53803	84.99845
	27.1422	22.5543	2.25543	83.0968
	36.1896	29.2639	2.92639	80.86273
	45.237	34.7153	3.47153	76.74094
318	9.0474	7.9319	0.79319	87.67049
	18.0948	15.6503	1.56503	86.49059
	27.1422	23.1177	2.31177	85.17254
	36.1896	30.1905	3.01905	83.42314
	45.237	35.9914	3.59914	79.56186

Table 6: Remaining Concentration, Adsorption Capacity and Adsorption Efficiency of the Investigated Synthetic Food Dye Using Granular Activated Carbon as an Adsorbent at Different Temperatures and Optimum Dye Concentration under Natural pH Conditions

Ci mg/L	T k◦	Ce mg/L	qe mg/gm	Kc	%
9.0474	298	1.5723	0.74751	4.754245	82.62153
	303	1.2617	0.77857	6.170801	86.05456
	308	1.1892	0.78582	6.607972	86.85589
	313	1.1762	0.78712	6.692059	86.99958
	318	1.1155	0.79319	7.110623	87.67049
18.0948	298	3.3928	1.4702	4.3329	81.2498
	303	3.1478	1.4947	4.748396	82.60384
	308	2.9314	1.51634	5.17275	83.79977
	313	2.7145	1.53803	5.665979	84.99845
	318	2.4445	1.56503	6.40225	86.49059
27.1422	298	5.1752	2.1967	4.244667	80.93301
	303	5.1	2.20422	4.322	81.21007
	308	5.0001	2.21421	4.428331	81.57813
	313	4.5879	2.25543	4.91604	83.0968
	318	4.0245	2.31177	5.744242	85.17254
36.1896	298	7.882	2.83076	3.591423	78.22026
	303	7.205	2.89846	4.022845	80.09097
	308	7.1264	2.90632	4.078244	80.30815
	313	6.9257	2.92639	4.225407	80.86273
	318	5.9991	3.01905	5.032505	83.42314
45.237	298	11.8697	3.33673	2.811133	73.76108
	303	11.1112	3.41258	3.071297	75.43781
	308	10.8889	3.43481	3.154414	75.92922
	313	10.5217	3.47153	3.2994	76.74094
	318	9.2456	3.59914	3.892814	79.56186

adsorption process on the solid surface [18,19]. This behavior can be attributed to the increase in the kinetic energy of dye molecules with increasing temperature, which facilitates their transfer from the solution phase to the solid adsorbent surface.

This effect is particularly important in the thermodynamic study of adsorption enthalpy and contributes significantly to enhanced adsorption when the interactions between the adsorbed molecules and the adsorbent surface are not purely physical or are associated with relatively strong physical interactions.

The equilibrium constant (Kc) values also increased with increasing temperature at constant concentration, confirming the enhancement of adsorption efficiency of the synthetic food dye onto the adsorbent surface. This indicates that the adsorption system is endothermic in nature. Increasing temperature enhances the interaction between dye molecules and the adsorbent

surface and promotes the sorption process, thereby increasing the adsorption equilibrium constant.

On the other hand, increasing the concentration of the synthetic food dye at constant temperature resulted in a decrease in the equilibrium constant values and consequently reduced adsorption efficiency. This behavior is attributed to the increased molecular interactions among dye molecules, which possess relatively large molecular structures. As dye concentration increases, the molecules become closer to each other, resulting in reduced molecular mobility and lower energy absorption by the adsorption system [20].

From the values presented in Table 10, the following observations were made:

- The sticking probability values (S*) of the food dye on the surface of granular activated carbon were greater than zero and less than one, which represents the preferred range according to the modified Arrhenius equation (Equation 8) [6]. The obtained S* values were very close to zero and far from unity. These values indicate that the adsorption of the synthetic food dye remained within the preferred range (0<S*<1), confirming the predominance of physical adsorption. However, in dilute solutions, the values approached the region associated with chemical adsorption behavior. As the dye concentration increased, the S* values also increased, indicating a return toward complete dominance of physical adsorption.
- The S* values increased with increasing dye concentration and remained within the preferred range for physical adsorption. Values very close to zero indicate strong adhesion of the synthetic food dye molecules onto the surface of granular activated carbon, confirming the occurrence of physical sorption of dye molecules. On the other hand, values approaching but remaining below unity suggest lower physical penetration of the dye molecules into the internal pores of the adsorbent material
- In both dilute and concentrated solutions, the apparent activation energy (AAE) values for adsorption of the synthetic food dye onto granular activated carbon were negative. The magnitude of the negative values decreased with increasing concentration. This indicates that lower temperatures are more favorable for dye removal in dilute solutions, whereas higher concentrations favor relatively higher temperatures for adsorption. This behavior can be attributed to the weaker intermolecular interactions in dilute solutions, where dye molecules remain farther apart compared with concentrated solutions. In addition, greater diffusion of dye molecules into the internal pores of granular activated carbon occurs in dilute solutions. When the S* values are extremely close to zero, spontaneous physical adsorption (physisorption) also becomes more favorabl

The low apparent activation energy values further indicate that the adsorption of the synthetic food dye onto granular activated carbon is primarily a diffusion-controlled process. Furthermore, the same trend confirms a decrease in physical sorption with increasing dye concentration in solution [31,32].

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