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Study the Adsorption Behavior of Food Colorant Dye Indigo Carmine and Loratadine Drug in Aqueous Solution

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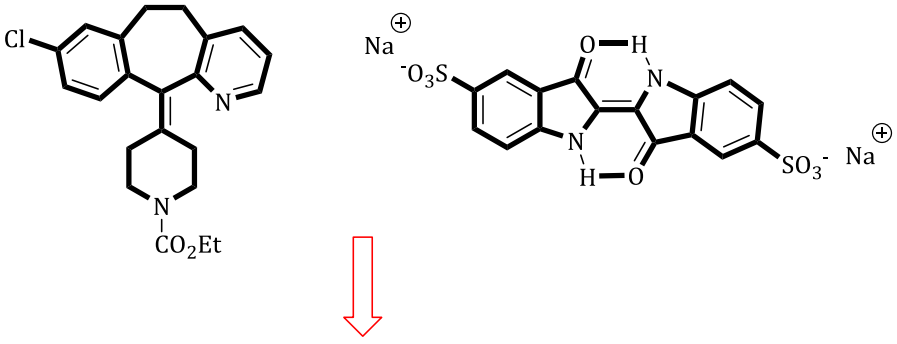
ABSTRACT

The interaction between foods additives and drugs can influence the effectiveness of drug therapy, and also on the adverse reactions assessments of numerous drugs. Food colorants dyes are chemical compounds which have been added to various types of foods. In this work, the colorant food dye indigo carmine (IC), as adsorbate, and loratadine drug, as adsorbent, was studied through the adsorption interaction behavior. The adsorption process was carried out at various concentrations of dyes, various dosages of drug, and different temperature (288-318K). The Langmuir and Freundlich adsorption isotherm were used to explain the equilibrium data, with Freundlich giving the best fit to the data. The adsorption kinetics follows pseudo-second order kinetics and the adsorption process were spontaneous, exothermic, and negative value of entropy demonstrates reductions in the disorder at the solid-solution interface after IC adsorption on loratadine.

GRAPHICAL ABSTRACT



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Introduction

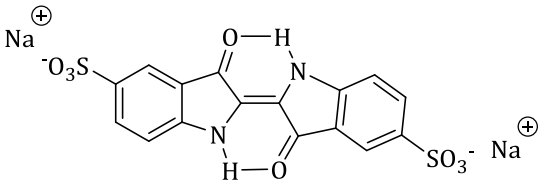
Food dyes are chemical or biological substances that are added to foods to reduce manufacturing costs and change the color of the food to attract consumers [1]. Food dyes are widely used as colorants in a variety of foods, including beverages, juices, and sweets [2].

Food coloring dyes can be either natural or artificial. The production of natural food colors is handled without using chemicals from a variety of sources such as seeds, fruits, vegetables, insects, and microorganisms [3]. Synthetic food dyes are water-soluble chemical substances that are manufactured in a factory which can be used in foods without further processing. Due to safety considerations, its original synthesis from coal tar and crude oil may have been controversial for some times [4]. These dyes have no nutritional value and may be toxic, causing many diseases from an allergic to antagonism to cancer [5]. Synthetic dyes are poorly biodegradable, harmful to the environment, cause genetic mutations and metabolic disorders, have carcinogenic effects, and bioaccumulate, particularly when extremely consumed [6]. Consuming too many food dyes in children has been associated with several adverse reactions such as allergies, hyperactivity, and difficulty concentrating [7]. The health effects of these colors are insignificant because they are identified as safe under proper production technologies and normal utilization.

Although each country controls the type and concentration of dyes permitted in food, various studies have revealed the use of inadequate dyes at levels exceeding the maximum allowed, causing serious risks to consumers [8].

Indigo carmine (IC), Disodium [2(2′) *E*]-3,3′-dioxo-1,1′,3,3′-tetrahydro[2,2′-biindolylidene]-5,5′-disulfonate, is an organic sodium salt with two SO_3H groups attached to the indigo molecule (Scheme 1) [9].

This dye is highly soluble in water, helps make coloring simple, and provides a blue color. Indigo carmine was used in foods and drinks as a chemically synthesized coloring agent. It's most widely used as an additive in capsules and tablets [10].



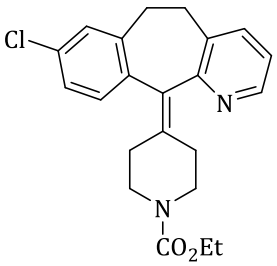
Scheme 1: Chemical structure of Indigo carmine

Despite its numerous applications, Indigo carmine causes numerous problems in human people, including skin irritation and permanent retinal damage. It has also been related to acute haemodynamic effects and respiratory illnesses including difficulty in breathing and chest tightness [11].

In the other hand, compounds used as drugs are typically organic compounds which have influences on the entire body. In general, the effects of these drugs in an overdose may be a heightened level of the therapeutic properties seen with regular use [12].

Loratadine, 4-(8-Chloro-5,6-dihydro-11*H*-benzo(5,6)cyclohepta(1,2-b)pyridin-11-ylidene)-1-piperidinecarboxylic acid ethyl ester, is a kind of antihistamine medicine to treat allergy symptoms. It is often used to treat skin irritation rash and hives, as well as nasal congestion, runny nose, itchy, and watery eyes [13].

Loratadine is a drug with a rate-limiting step in its absorption due to its dissolution or solubility. Due to be practically insoluble in water, it has a low variable oral absorption (Scheme 2) [14].



Scheme 2: Chemical structure of Loratadine

To date, no work has been carried to illustrate the risks of interactions between food dye and insoluble or slightly soluble drugs. This interaction occurs for the adsorption of food dyes on the surface of these drugs, which may inhibit the drugs' effectiveness in treating diseases even as increasing the biocompatibility of these dyes inside the body.

Considering the health hazards of these chemicals to humans, the purpose of this work is to investigate the interaction of food dyes and drugs through the adsorption process. Different adsorption isotherm will be used to illustrate the equilibrium in process like Langmuir and Freundlich models. Feasibility of the adsorption process will be evaluated by calculating kinetics and thermodynamics parameters of the process.

Materials and Methods

Indigo carmine was supplied by Hopkin and Williams Ltd. Loratadine was supplied by State Company for the manufacture of medicines and medical supplies /Samarra.

A stock solution of indigo carmine was obtained by precisely dissolving 1g of the dye in distilled water in a 1 L volumetric flask. Various concentrations (10–800mg/L) were prepared by diluting the respective dye stock solution as per requirement for the calibration curve (Figure 1) and adsorption processes were then analyzed using a Shimadzu double-beam UV–Vis spectrophotometer, the absorbance was measured at the wavelength 610 nm, as displayed in Figure 2.

Batch adsorption experiments have been conducted to study how well the system is affected by various parameters, including IC concentration, Loratadine amount, contact time, and temperature. The adsorbed amount of indigo carmine on the loratadine drug was determined by using the following equation;

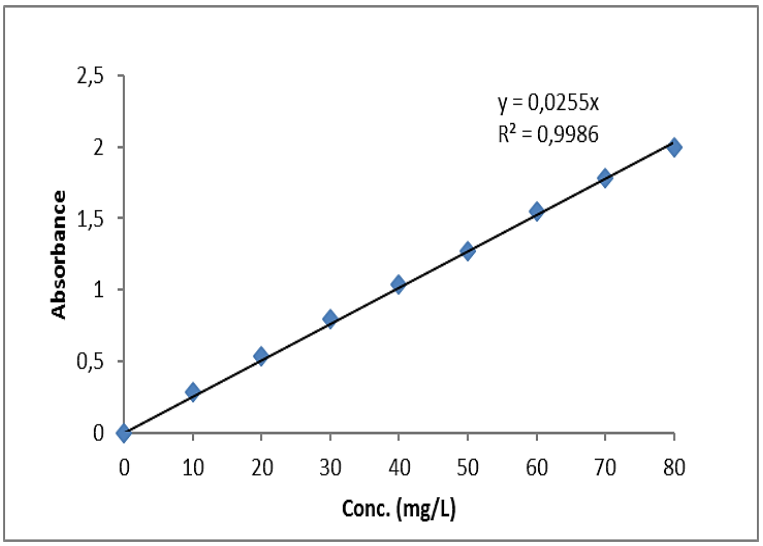


Figure 1: Calibration curve of indigo carmine

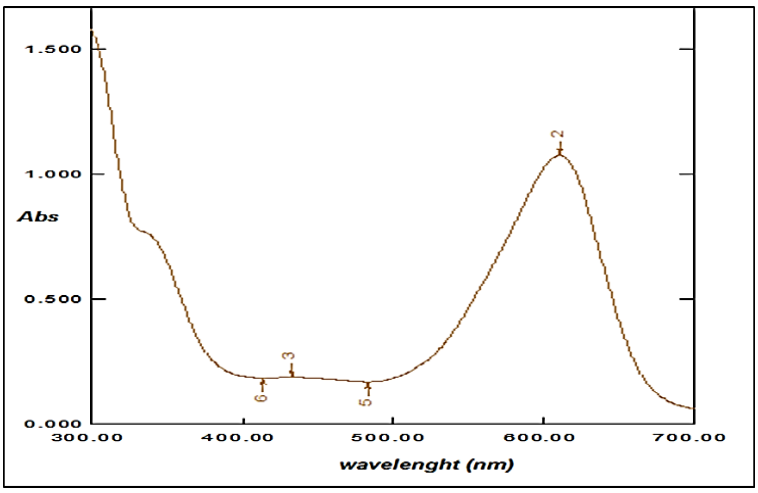


Figure 2: The UV-Visible spectra of indigo carmine

$$q_e = \frac{(C_0 - C_e)\,V}{w} \qquad (1)$$

Where C_0 and C_e are the concentration of indigo carmine (mg/L) at initial and equilibrium phase, respectively, V represent the volume of solution in liter, and w is the weight of the loratadine drug (g). The effect of adsorbent amount was studied for 40 mg/L IC solution with differing amounts of adsorbent (0.05, 0.1, 0.15, 0.2, and 0.25) g/50 mL of dye. For all experimental tests, the temperature was held to 298 K and the thermostatic shaker bath's agitation speed was set to 200 rpm. The effect of contact time on the IC adsorption process was evaluated by addition of 0.15g of loratadine in 50 mL of IC solution (40 mg/l) in 250 mL flasks. Every 10 minutes, 5 mL of sample was withdrawn and the concentration was calculated until the equilibrium is attained. In the temperature range of 288-318 K, the impact of temperature on the quantity adsorbed was investigated.

Results and Discussion

Effect of adsorbent dosage

Adsorbent dosage is a crucial factor since it significantly affects an adsorbent's capacity at a specific initial concentration of adsorbate. To find out the effect of drug dosage on the adsorption of IC, a series of adsorption experiments with different adsorbent doses were carried out from 0.05g/50 ml to 0.25g/50 mL at indigo carmine initial concentration of 40 mg L^{-1} and at constant contact time.

As depicted in Figure 3, the adsorbed IC amount enhanced with drug dosage enhancement and reaches a maximum value using 0.15 mg of loratadine.

As drug dosage was increased, further dye was adsorbed until equilibrium was achieved, and in practice, the dye adsorption remained almost constant, and then decreases with the increasing the dosage. This has occurred due to the competition for adsorption between adsorbent molecules and the splitting effect of the gradient in concentration between dye molecules and adsorbent concentration [15]. All subsequent experiments affirmed the use of this adsorbent dosage.

Effect of initial concentration and contact time

At various time periods and with different initial dye concentrations from 10 to 80 mg L^{-1}, the influence of contact time on dye adsorption was evaluated range, as depicted in Figure 4. It was found that the adsorption process was fast throughout the first contact time and afterwards slowed down. The time required to achieve equilibrium for IC adsorption on the surface of loratadine was 60 minutes. This may be caused by the fact that there are initially many adsorption sites available, but as time passes on, there is a reduction of the adsorption sites [16].

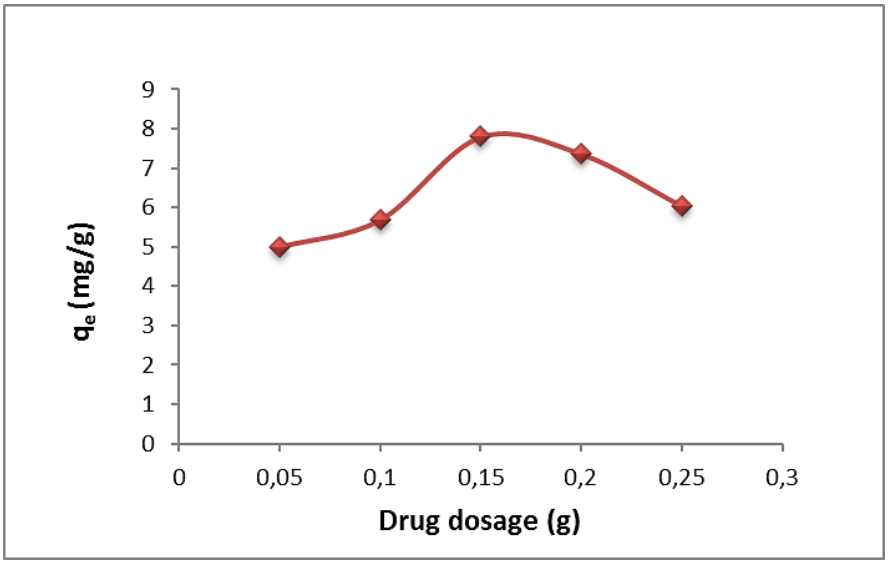


Figure 3: Effect of drug dosage on quantity adsorbed of indigo carmine

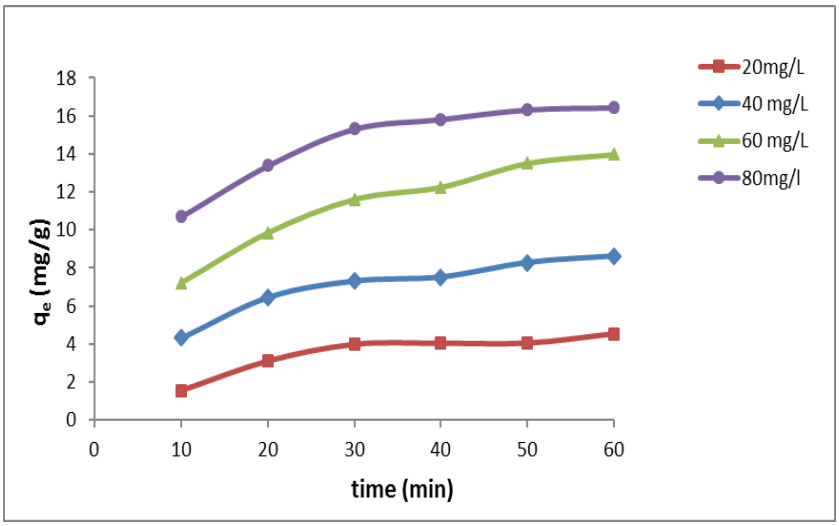


Figure 4: Effect of IC initial concentration on adsorption process

Adsorption isotherm

The adsorption isotherms help to understand the relationship between the amounts of dye adsorbed on the adsorbent at equilibrium. Figure 5 displays the relation between quantity adsorbed and concentration of dye at equilibrium which show a small slope in the beginning followed by a sharp rise indicating that the curve is the S-shape curve. This particular isotherm indicates that the surface's affinity for the adsorptive is low at low concentrations and increases at higher concentrations [17].

To establish an adsorption isotherm, the adsorption data were fit to the Langmuir and Freundlich models. The Langmuir adsorption isotherm assumes a monolayer, uniform adsorption site, and limited adsorption location, there may be a saturation level at which no additional adsorption occurs. Likewise, it supposes that molecules adsorbing on nearby sites will not interact with one another. A surface with a finite number of identical sites and monolayer coverage, the Langmuir equation is established and given as follow [18]:

$$\frac{C_e}{q_e} = \frac{1}{K_L . Q°} + \frac{C_e}{Q°} \qquad (2)$$

In which, Q° (mg/g) and K_L (L/mg) represent the Langmuir constant. Q° is the maximum adsorption capacity and K_L is about energy adsorption constant. By plotting C_e/q_e linearly against C_e, the slope and interception were used to get the Langmuir constants Q° and K_L, respectively. Figure 6 represents applying Langmuir isotherm equation on experimental results of adsorption of IC dye on loratadine as an adsorbent.

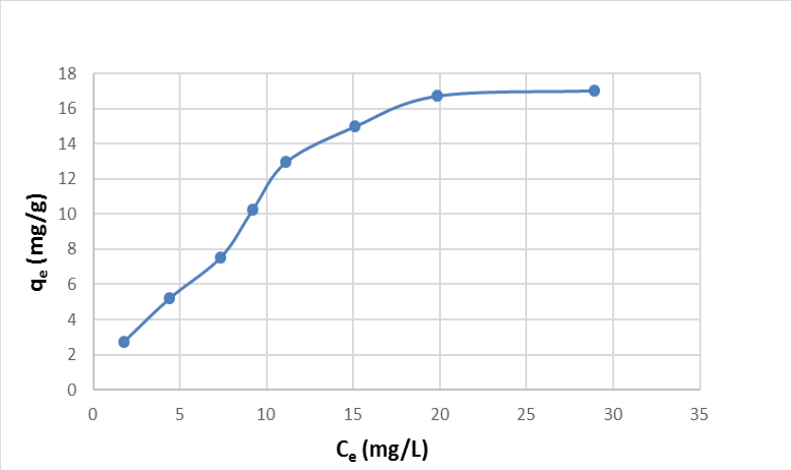


Figure 5: Adsorption isotherm of indigo carmine on loratadine at 298K

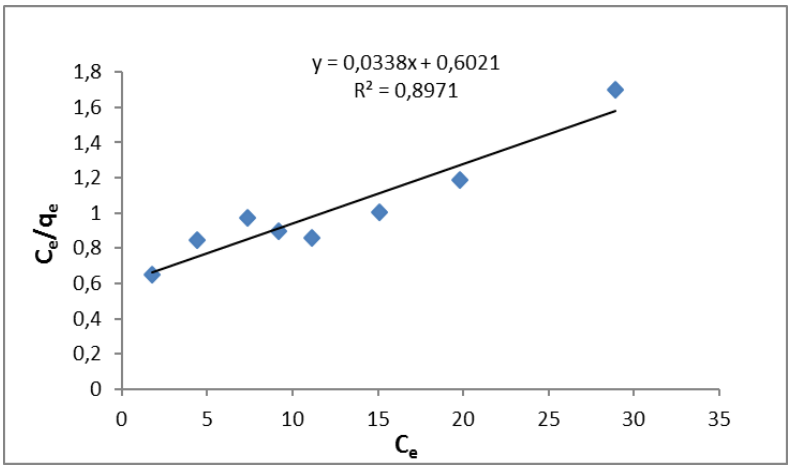


Figure 6: Linear form of Langmuir isotherm of adsorption of IC

Freundlich isotherm 19 model is derived from Langmuir isotherm and assumes a heterogeneous surface of adsorption capacity and it is suitable for multilayer process and is given by:

$$\ln q_e = \ln K_f + \frac{1}{n} lnC_e \qquad (3)$$

The dimensionless constant n is related with the effectiveness of adsorption, and K_f is the Freundlich constant (mg/L), which corresponds to capacity of the adsorption. The n value reflects the degree of nonlinearity between adsorption process and concentration of dye. The values of K_f and n were determined, respectively, from the intercept and slope of the fitted line of $\ln q_e$ against $\ln C_e$, as depicted in Figure 7.

From the results presented in Table 1, it is obvious that the R^2 (regression coefficients) value of Freundlich isotherm was greater than that estimated from Langmuir. This indicated that the Freundlich model fit well with the data of IC dye adsorption onto loratadine drug. Positive cooperative interaction and a heterogeneous nature of the adsorption were indicated from n values which was greater than unity [20]. In addition, the value of maximum monolayer capacity (Q^0) estimated from Langmuir equation was decrease with rise in temperature indicating increasing adsorption capacity at a low temperature.

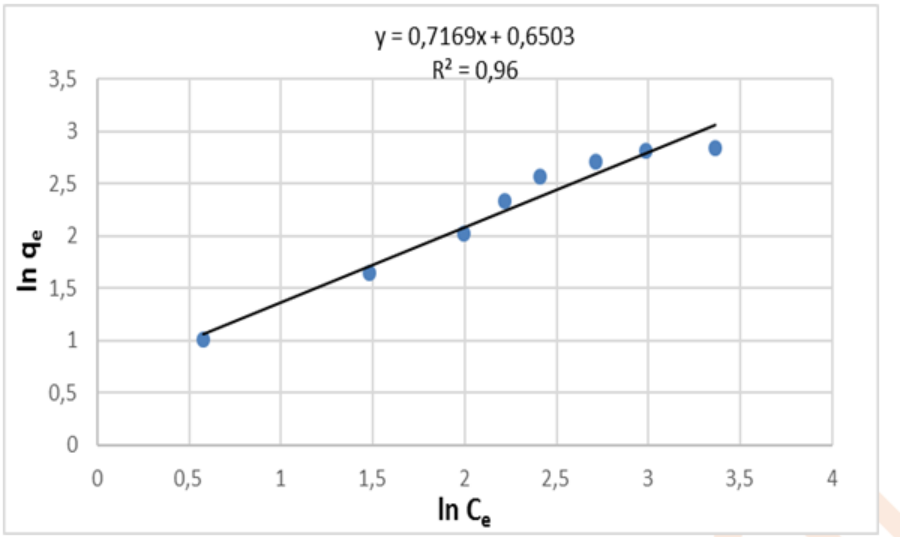


Figure 7: Linear form of Freundlich isotherm of adsorption of IC

Table 1: Kinetics parameters of adsorption of IC dye on loratadine drug

models	Langmuir equation			Freundlich equation		
Temperature	R^2	Q^0 (mg/g)	K_L(Lmg^{-1})	R^2	K_f (Lmg^{-1})	n
288	0. 8450	34.24	0.0526	0.9713	2.130	1.387
298	0.8971	29.58	0.0561	0.960	1.916	1.394
308	0.9434	26.178	0.0661	0.9577	2.029	1.477
318	0.9735	21.142	0.0572	0.9488	1.644	1.582

Kinetics of adsorption

To clarify the mechanisms of IC adsorption on drug surfaces, several kinetics approaches were employed. The adsorption kinetic behavior of IC (40 mg/l) onto drug at four temperatures (288-318 K) has been studied using the Lagergren-first-order equation and the pseudo-second-order equation. Depend on the values of the linear regression correlation coefficient (R^2); the best-fit model was adopted. The pseudo-first order kinetic model has the following linear equation form 21:

$$\log(q_e - q_t) = log q_e - (k_1/2.303)\, t \qquad (4)$$

In which, q_e is the amount of IC adsorbed on the surface of the drug at equilibrium, q_t is the IC amount adsorbed on the surface of the drug at any time, and k_1 is the rate constant of the pseudo-first order kinetics (min^{-1}). The intercept and slope of plots of log (q_e - q_t) verses t often used to estimate the q_e and k_1, respectively. The outcome of such a plot is depicted in Figure 8.

The linear form of the pseudo-second order kinetic model (22) is given as follows:

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \left(\frac{1}{q_e}\right) t \qquad (5)$$

Where k_2 is the pseudo-second order kinetics' rate constant (g.mg^{-1}.min^{-1}). To determine k_2 and q_e, the intercept and slope of plots of t/qt against t, was achieved as illustrated in Figure 9.

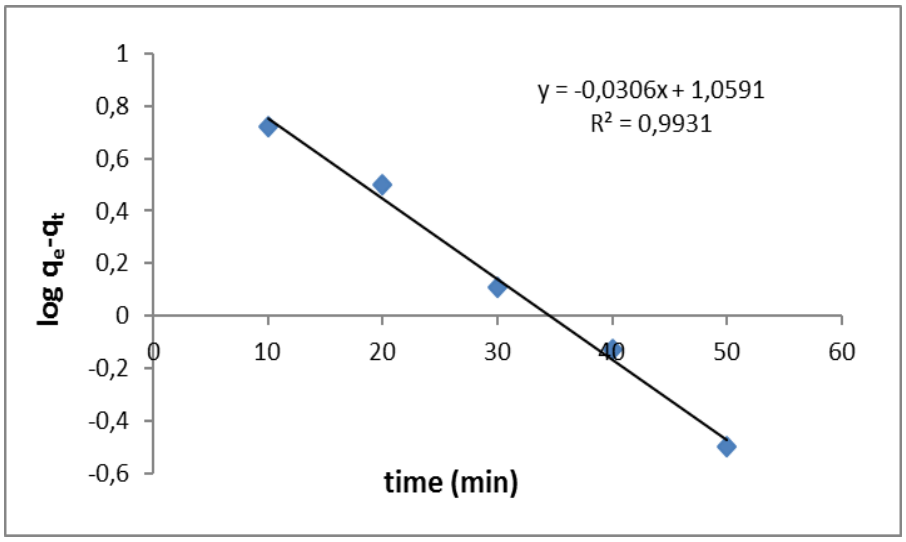


Figure 8: Plot of pseudo first order kinetics for adsorption of IC dye at 298K

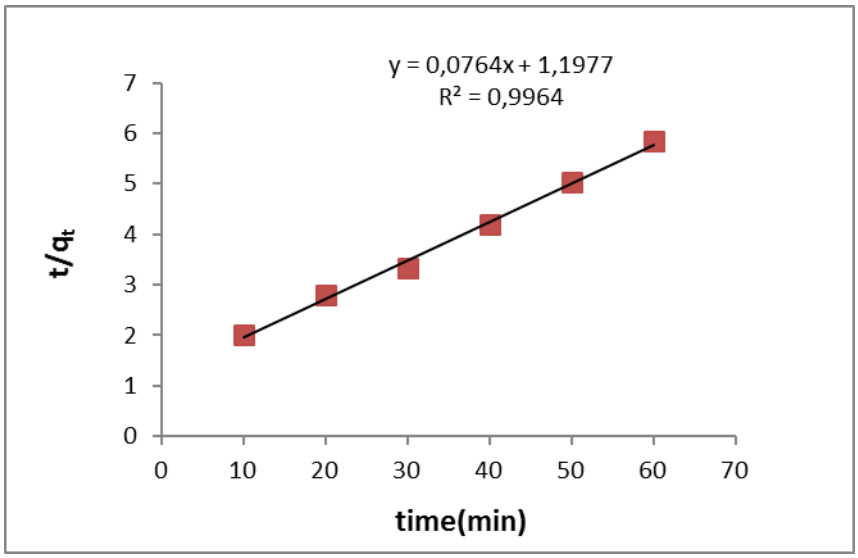


Figure 9: Plot of pseudo second order kinetics for adsorption of IC dye at 298 K

The correlation coefficients (R^2) of the two equations are closely as indicated in Table 2; however, the value of R^2 for the pseudo-second order equation is higher. This predicted that the pseudo second order was a better kinetic model compared to pseudo-first order model to express the kinetics of the process.

Table 2: Kinetics parameters of adsorption of IC dye on loratadine drug

Kinetics equations	Pseudo First order			Pseudo Second order		
Temperature	R^2	k_1 (min^{-1})	q_e (mg/g)	R^2	k_2(g mg^{-1} min^{-1})	q_e (mg/g)
288	0.9710	0.0502	6.362	0.9965	0.0100	12.195
298	0.9931	0.0704	11.457	0.9964	0.0048	13.089
308	0.8851	0.0677	15.463	0.9929	0.0021	15.060
318	0.9076	0.0891	18.105	0.9972	0.0036	12.987

Thermodynamic of the Process

To investigate the thermodynamic properties (free energy change (ΔG°), enthalpy change (ΔH°), and entropy change (ΔS°) associated with the interaction between IC dye and drug, the adsorption experiments were carried out at four temperature ranges (288- 318 K).

The following equations (6) and (7) have been used to calculate the thermodynamic properties [23]:

$$\Delta G° = -RT \ln K_L \quad (6)$$

$$\ln K_c = \frac{\Delta S°}{R} - \frac{\Delta H°}{RT} \quad (7)$$

Where the value of the universal gas constant R in joule is (8.314 J/mol K), T is the temperature in kelvin, and K_c is the equilibrium constant (L/g). The values of (ΔH°) and (ΔS°) were calculated from the slope and intercept of the van't Hoff plots of ln(K_c) against 1/T. Figure 10 demonstrates plotting of this relation, while the thermodynamic parameters obtained at various temperatures are represented in Table 3. The feasibility and spontaneous adsorption nature are verified by the negative values of ΔG°. The finding that the negative value of ΔG° decreases as temperature increases implies that lower temperatures are more favorable for the adsorption of IC on lotatadine.

As can be seen, the adsorption reaction was exothermic process because the value of (ΔH°) was

negative (-15.217 kJ/mol). Furthermore, practically the value of (ΔH°) is less than 40 kJ/mol. This provided evidence that the IC adsorption onto loratadine was a physisorption mechanism [24].

The negative value of entropy (ΔS°) (-40.632 J/K mol), shows how the irregularity in the adsorption system's solid/liquid interface decreased and that the adsorption capacity lowered as a consequence of the temperature rise [25].

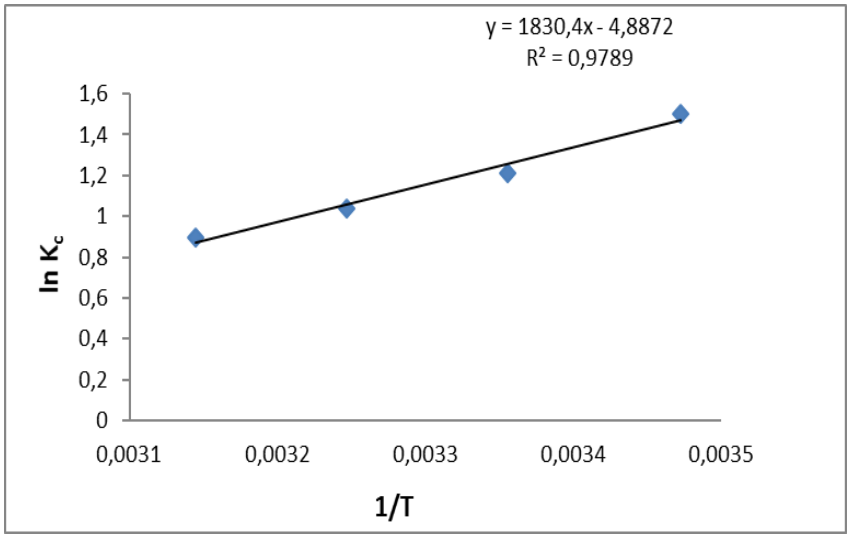


Figure 10: Plots of Van't Hoff for IC adsorption onto loratadine

Table 3: Thermodynamic data for the IC adsorption onto loratadine drug

Temp.(K)	ΔG° (kJ/mol)	ΔH°(kJ/mol)	ΔS° (J/K.mol)
288	-3.598	-15.217	-40.632
298	-2.993		
308	-2.665		
318	-2.367		

Conclusion

It is crucial to take precautions before clinical instances since interactions between drugs and food colorant dyes are widespread throughout the world. The interaction between indigo carmine dye as a food additives and drug loratadine was proven by the adsorption of dye on the surface of drug. The Freundlich isotherm model was proven to be the most efficient model to illustrate the adsorption isotherm of IC dye onto loratadine drug. The kinetics of IC adsorption on the drug loratadine can be modeled by the pseudo-second order equation, because it closely matched the experimental results. The spontaneous and exoothermic nature of the IC adsorption onto loratadine was demonstrated by thermodynamic study. The negative ΔH◦ value, which is -15.217 kJ/mol, indicates that the IC adsorption on loratadine is physisorption.

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Authors' contributions

All authors contributed to data analysis, drafting, and revising of the paper and agreed to be responsible for all the aspects of this work.

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