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Kinetic and Thermodynamic Study for Removal of Carmosine Dyes(E122) Using $CuFe_2O_4$ NP

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ABSTRACT

The optimum conditions for removal (adsorption) of carmosine dye on the $CuFe_2O_4$ NP surface at pH 4, 50°C, an adsorbent dosage of 0.1 g, a dye concentration of 5 mg/L and a contact period of 30 minutes were done. The maximum efficiency of dye removal demonstrates about 99 % at 50°C and pH equal to 4 that due to the acidic natural of this dye. Based on kinetic model, the order of adsorption of dye on $CuFe_2O_4$ NP surface was found to be Pseudo-Second Order. Furthermore, as a result of the thermodynamic analysis, negative values of Gibbs Free Energy change and positive values of enthalpy change indicated that the adsorption method took place spontaneously with an endothermically. The enthalpy change value is 51.35308 kJ mol^{-1} that proved the adsorption kind is chemical adsorption. The activation energy of adsorption for this dye on the $CuFe_2O_4$ NP surface is obtained as a negative value because of the reaction occurs in multi-steps, which depending on the found two various charges on $CuFe_2O_4$ NP surface : Cu^{2+} and Fe^{3+}. The suggested binding mechanism of dye on $CuFe_2O_4$ NP surface was performed.

Keywords: Carmosine dye (E122), Removal treatment, Adsorption kinetics, Thermodynamics, $CuFe_2O_4$ NP.

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Introduction

Dyes are thought to be an important source of organic molecule pollution in wastewater[1]. They have a major impact on the toxicity and appearance of wastewater. Organic pollutants include aliphatic compounds that contain polycyclic aromatic hydrocarbons, dichlorodifluoromethane, alkenes, alkynes, and tetramethyl ammonium ions. The benzene rings found in these neutral, nonpolar chemical compounds are a significant cause of environmental problems. Dye is used as a coloring agent in many industries, such as leather, textiles, paper, plastics, food, medicine, and cosmetics. Dyes can be categorized into many groups based on their chemical composition, application method, use, or kind of chromophore. [2]. In order to prepare and serve food that is plentiful, delicious, and nourishing, food additives play a critical role in meeting the demands of the expanding population[3]. Synthetic natural food additives, as well as flavoring or coloring agents, can be utilized[4]. Colorings give food the appealing appearance that consumers seek[5]. Numerous industries, including textiles, leather, paper, food, pharmaceuticals, and cosmetics, use azo dyes. Azo groups (–N=N) attached to aromatic rings in their chemical structures are what define azo dyes. Various industries utilize carmoisine as a food coloring agent. Carmoisine Sky, which is a synthetic red food dye made from dye colorings, is distinct from Azorubine, Food Red 3Azorubin S, brillantcarmoisine O, and Acid Red 14. Usually, sodium salt is included. The powder ranges from red to maroon. It has been applied in situations where food needs to be heat-treated after fermentation. contains E122 carmoisine, which is present in foods like pudding[6].It is used as a red pigment when solubility in water .The chemical structure of disodium 4-hydroxy-3-(4-sulfonat-1-naphthylazo) naphthalene-1sulphonate (chemical formula $C_{20}H_{12}N_2Na_2O_7S_2$)[7]. The dye's capacity to produce and reductively cleave aromatic amines—which can cause cancer and build up through food chains to interact with bodily fluids including saliva and stomach secretions—determines whether it can be classified as a factor that is detrimental to humans[8]. The potential use of copper ferrite nanoparticles ($CuFe_2O_4$ NPs) in water purification has drawn the attention of numerous researchers in recent years. The $CuFe_2O_4$ NPs are an inexpensive magnetic material that is stable in a different conditions[9]. This work

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purposes to study the removal of carmosine dye on $CuFe_2O_4$ NP surface at optimum conditions. The binding mechanism of dye on $CuFe_2O_4$ NP surface shall suggested.

EXPERIMENTAL

Chemical materials

Without any further processing, all chemicals are bought from Fluka, BDH, and Sigma-Aldrich. $CuFe_2O_4$, which is supplied by Fluka, is utilized as a catalyst at 99.9% purity. As a local enterprise in Mumbai, India, the carmoisine (E122) dye is supplied as food coloring. With the formula structure $C_{20}H_{12}N_2$ $Na_2O_7S_2$, this dye is considered a monoazo dye. Its IUPAC name is a salt of 2-(4-sulpho-1 naphthylazo)-1-naphthol-4-sulfonic acid, and its MWt is 502.42 g/mol., the chemical structure describes in Figure 1.

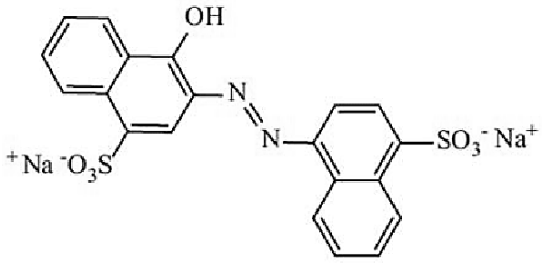

Figure 1. Chemical structure of carmoisine (E122) dye.

Removal of Carmosine Dye using $CuFe_2O_4$ NP

For adsorption kinetics studies, adsorption via $CuFe_2O_4$was achieved whilst varying adsorption-related parameters to define the influences of adsorption time, temperature, adsorbent dose, and pH. A glass beaker containing 0.1 g of the adsorbent (100 mL) was mixed with 100 mL of the 5 ppm carmosine dye solution at 25^{o}C via magnetic stirring. At time intervals of 0,5,10,15, 20,25 and 30 min, centrifugation was used to separate the solution from the adsorbent (at 4000 rpm). The concentration of the remaining dye was measured at a λ_{max} of 515 nm after determining the absorbance through a spectrophotometer that detects UV light. The removal efficiency (E%) can be calculated depending on Eq. (1), the adsorption capacity at equilibrium, q_e, (mg/g) and at a different time q_t (mg/g), can be measured via the use of Eqs. (2), and (3), respectively[10,11].

$$E(\%) = \left(\frac{C_o - C_e}{C_o}\right) \times 100 \qquad (1)$$

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$$q_e = \frac{(C_o - C_e) \times v}{w} \quad (2)$$

$$q_t = \frac{C_O - C_t}{w} \times V \quad (3)$$

Where: V is the volume of the dye solution (L), *w* is the mass of the adsorbent nanoparticles in (g), C_o is the initial dye concentration(mg/L), C_t is the residue dye concentration (mg/L) at time t, and C_e is the equilibrium dye concentration after adsorption (mg/L).

Result and Discussion

Effect of contact time.

The contact time refers simply to the amount of time the adsorbent is in contact with the dye solution. This effect was investigated for (5 ppm) carmosine dye for ranged contact times (10, 20, and 30 min), at pH of dye solution equal to 4. The amount of carmosine dye that was adsorbed on the $CuFe_2O_4$surface, was found to increase with increasing contact time when keeping all other parameters constant as shown in Figure 2. This a significant elevates in the adsorption efficiency of carmosine dye at 84.22 % with contact times that reflecting its effective collections between dye molecules and the $CuFe_2O_4$ nanoparticles surface[9].

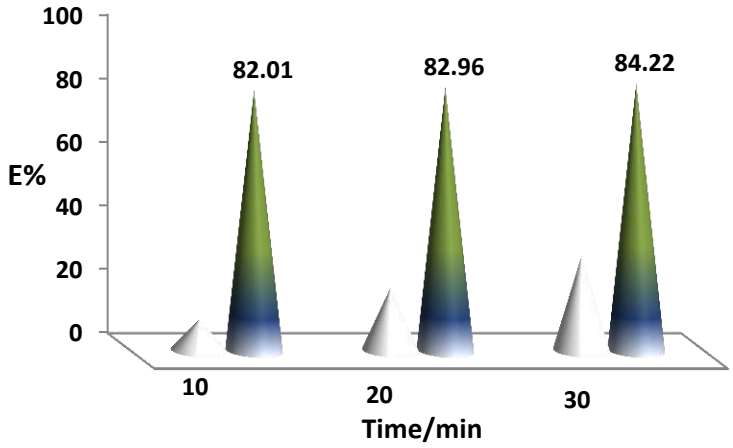

Figure 2 .The relation between removal efficiency of Carmosine dye and contact time using $CuFe_2O_4$ NP.

Kinetic of Dye Removal in the Presence of $CuFe_2O_4$ NP

The contact time can be defined as the amount of time the adsorbent is in contact with the dye solution. The influence on carmosine dye (5 ppm) was calculated for different times (10, 20, 30,

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40, 50 and 60 min), with solutions fixed at pH 4. There are many of kinetic study models available that one can use to define adsorption kinetics; previous studies have used pseudo-first order and pseudo-second order [12-14] kinetics to this end. In this study, these kinetics were modeled as per Eq. (4) and Eq. (5), respectively and shown in Figure 3(a,b).

$$\ln(q_e - q_t) = \ln q_e - k_1 t \qquad (4)$$

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

where k_1 (min^{-1}) and k_2 (g $mg^{-1}min^{-1}$) are the rate constants for the first-order and the pseudo-second-order models, respectively. q_e and q_t in ($mg.g^{-1}$) are the quantities adsorbed at equilibrium and an arbitrary time t (min)[15,16].

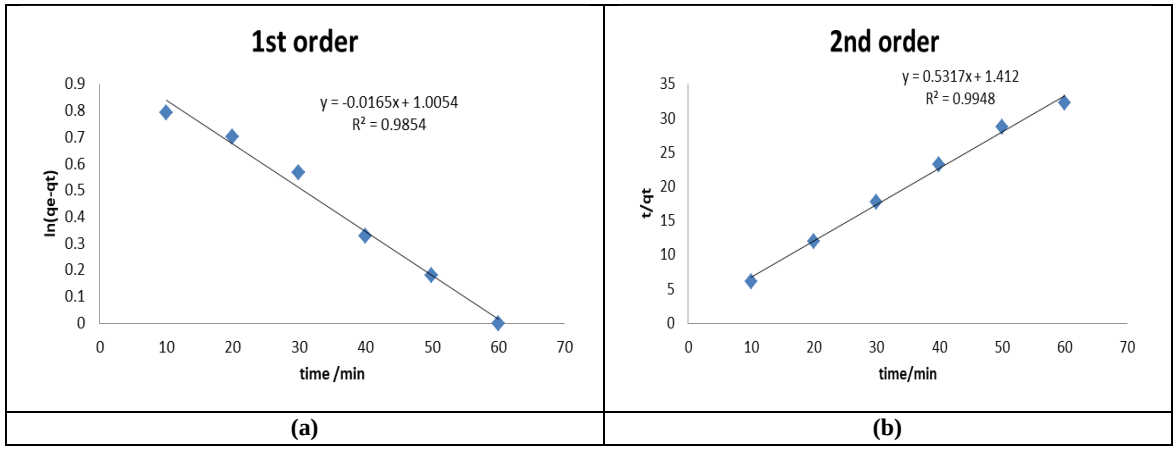

Figure 3. (a) First-order model and (b) second-order model for adsorption of Carmosine dye on the surface of $CuFe_2O_4$ NP.

TABLE 1. The first- and second-order reaction parameters for carmoisine dye on the surface of $CuFe_2O_4$ NP.

Pseudo-First-Order			Pseudo-Second-Order		
k_1 (1/min)	q_e (mg/g)	R^2	k_2 (g/mg.min)	q_e (mg/g)	R^2
0.0165	1.0054	0.9854	0.5317	1.8876	0.9948

Based on Figure 3(a,b), the calculating of first- and second-order reaction constants for the adsorbance of Eosin yellow dye on the surface of the $CuFe_2O_4$ was done. The results of these studies are listed in Table 1. The values of the correlation coefficient, R^2, for the pseudo-second-

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order model characterized the maximum to investigational adsorption data for the carmoisine dye on the $CuFe_2O_4$ NP surface. Furthermore, the calculated q_e values for the pseudo-second order model[17] were found to be very close to those found for the experimental data, as compared with those calculated for the pseudo-first order model.

Effect of Initial pH on Removal of Carmoisine dye using $CuFe_2O_4$ NP

The effect of pH was studied by changing this parameter (pH 4, 5, 6, 7 and 8) with a fixed concentration of dye in each instance (5 mg/L). Fig.4 shows that an acidic medium at pH 4 offers the optimal removal efficiency of the dye, because carmosine dye is itself naturally acidic. The maximum removal efficiency for carmosine dye using as adsorbent 93.8 % at pH=4. Furthermore, an increase in pH of the solution leads to a decline in the removal efficiency according to the following pH values: 4 > 5 > 6 > 7 > 8 [18].This behavior indicates that the surface charge of $CuFe_2O_4$ becomes negative in basic medium, which leads to an increase in the electrostatic repulsion of Carmoisine dye, which is itself anionic.

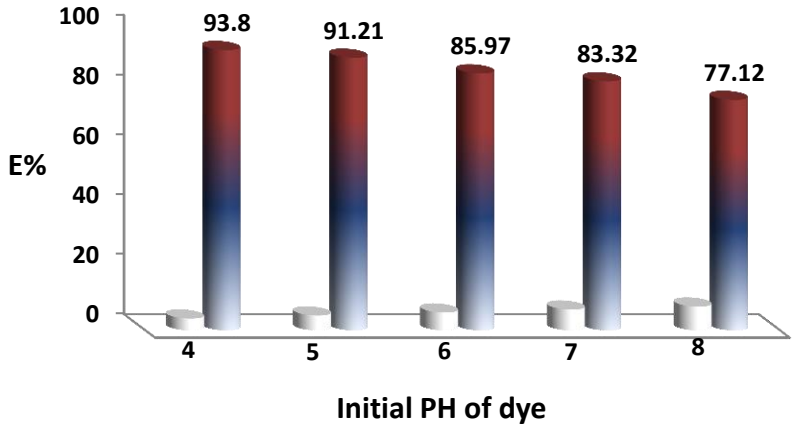

Figure 4. The influence the pH of the Carmosine dye solution on the removal efficiency of $CuFe_2O_4$ surface.

The Influence of Temperature on Dye Removal

Temperature is a vital parameter in adsorption methods. For any route it is important to find to what temperature an effect on the efficiency of adsorbent. Studies are showed at different temperatures were 10, 20, 30, 40 and 50 ^oC by keeping all other parameters constant [19].The experiments were succeeded to conclude the thermodynamic parameters of the adsorption

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processes at different temperatures, as appearance in table 2. According to the Van't Hoff plot in Figure 5, the interaction between dye and adsorption surface was found to be endothermic, with a ΔH^o of 51.35308 kJ/mol, this result is proved the adsorption kind is chemical adsorption[20,21]. The ΔS^o can be found from the intercept, which is 0.201016 kJ/mol.K, showing that the adsorbed units were less random [22].

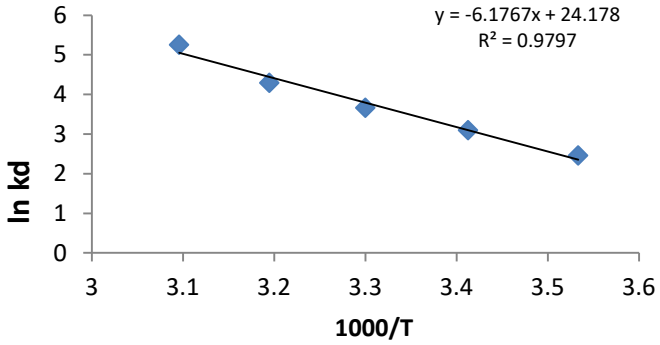

Figure 5. The Van't Hoff equation of adsorption Carmosine dye.

The activation energy, E_a, is dependent on the magnitude of $\Delta H^{o,}$ as per the following Eq.6[23].

$$E_a = \Delta H^o + RT \qquad (6)$$

Where R is the universal gas constant and T is the absolute temperature.

The negative values for all ranged of studied temperatures, that may be attitude to happened of adsorption process on $CuFe_2O_4$ surface with multi-step because of this spinel contains two different charge on its surface Cu(II) and Fe(III) that given various of potential points for binding.

The Gibbs free energy, ΔG^o, can be calculated using Eq. 7 depending on adsorption constant (kd) [23,24].

$$\Delta G^o = -RTlnk_d \qquad (7)$$

The negative $\Delta G°$ values show that the adsorption of carmosine dye onto $CuFe_2O_4$ is spontaneous under experimental conditions, as presented in Figure 6.

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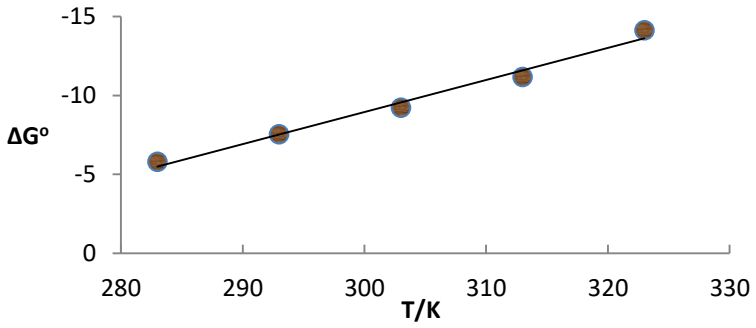

Figure 6. Relation of Gibb's free energy change (ΔG^o) for the adsorption Carmosine dye versus temperature .

Table 2: Thermodynamic parameters determined for Carmosine dye adsorption onto $CuFe_2O_4$ NP. at different temperatures.

T(K)	Δ S^o (kJ/mol.K)	Δ G^o (kJ/mol)	ΔH^o (kJ/mol)	E_a (kJ/mol)
283	0.2010	-5.7830	51.35308	-49.0002218
288	0.2010	-7.5265		-48.9170818
293	0.2010	-9.2046		-48.8339418
298	0.2010	-11.1549		-48.7508018
323	0.2010	-14.1063		-48.6676618

Because of the reaction is endothermic the efficiency increases with increasing the temperature[25], so the efficiency is found to be 99.479 at 50 ^oC and pH=4, as shown in Fig 7.

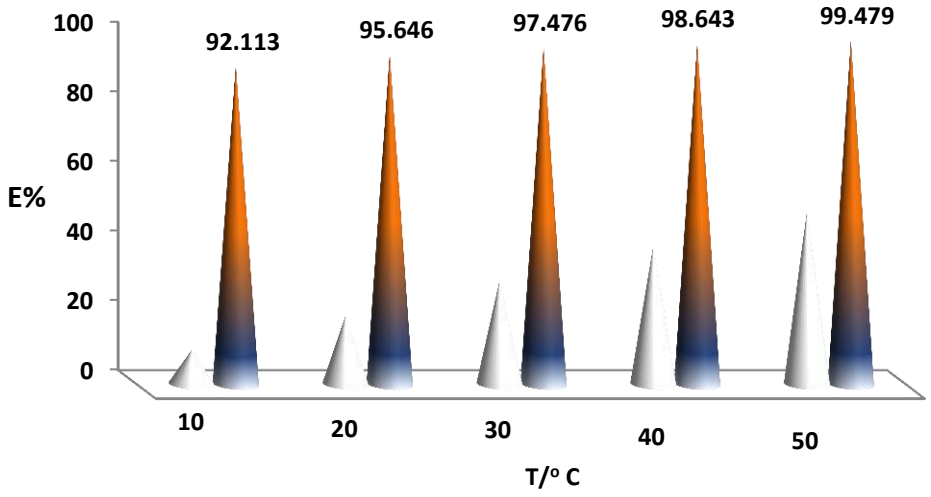

Figure 7. The efficiency relation with different temperature of $CuFe_2O_4$ NP.

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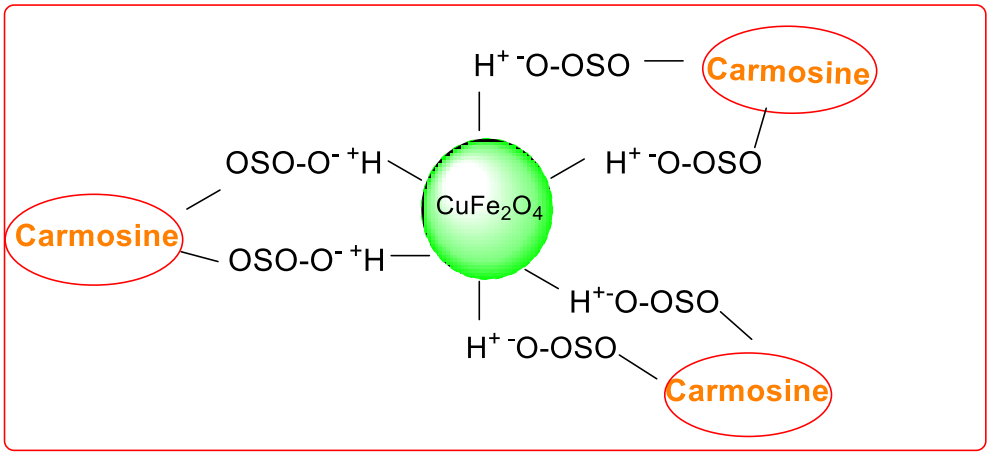
Mechanism of dye binding on catalyst

Electrostatic interactions between the negatively charged dye (-OSOO-) and the positively charged (H^+) on $CuFe_2O_4$ NP surface in an acidic media, as showing in schematic 1, the adsorption mechanism illustrates the surface interactions between carmosine molecules and functional groups on $CuFe_2O_4$ NP.



Scheme 1. The surface interactions between Carmosine dye and functional groups on $CuFe_2O_4$ NP at pH= 4.

Conclusion

The Carmosine dye is successfully removal by $CuFe_2O_4$ NP. In terms of adsorption thermodynamics, ΔH^o, ΔS^o and ΔG^o values were calculated for $CuFe_2O_4$ NP. According to these results, the adsorption process occurs endothermic, low random and spontaneously. The adsorption type for this removal process demonstrates chemical adsorption at pH 4 and 50 ^oC. The kinetic model was studied and obtained as pseudo second-order depending on dye and $CuFe_2O_4$ NP surface.

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Author contributions The manuscript was prepared with contributions from all authors. All authors have approved the final version of the manuscript.

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Data availability No datasets were generated or analyzed during the current study.

Declarations

Ethical approval Not applicable.

Competing interests The authors declare no competing interests.

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