The Sorption of Erythrosine E127 Dye onto Commercial Kaolin Thermokinetic Studies

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Abstracts: In this study, a highly efficient and economical method was used to remove E127 dye using commercial kaolin (CK) as the adsorbent. The optimal pH for adsorption was found to be 4.7, and the maximum surface concentration of the dye was 53.8 $mg.g^{-1}$ when 125 mg of adsorbent was used. The Freundlich isotherm was used to describe the adsorption process, which suggested that the active sites in the adsorption process were randomly selected. The adsorption capacity (q_{max}), enthalpy (ΔH), and entropy (ΔS) were calculated to be 74.6 $mg.g^{-1}$, -12.3 $KJ.mol^{-1}$, and 75.0 $J.mol^{-1}$, respectively. Both thermodynamic and kinetic analyses indicated that the adsorption process was spontaneous, involved physical adsorption, and followed the pseudo-second-order (PSO) model. The method was effective in extracting the E127 dye up to four times.

Keywords: Erythrosine Dye (E127), Kaolin, Thermodynamic, Kinetics, Isotherm, And Adsorption Processes.

1. INTRODUCTION

Synthetic dyes are used to show the aesthetic appearance of canned foods; despite their toxic effects, they have attracted great attention due to their pollution of the environment, especially wastewater [1-3]. In the food process, a large amount of industrial dyes are consumed. Annually, 10-15% of these dyes are discharged into wastewater [4-6]. Among these dyes, erythrosine (E127) is a pink-red (cherry-colored) and soluble in water. E127 dye is widely used in various fields, such as canned food, especially canned meat, medicine, cosmetics, wool, and nylon [7, 8]. E127 dye threatens human health when consumed in large quantities, which increases hyperthyroidism as a result of varying levels of thyroid hormone [9, 10]. 0.1 mg per kg of body weight per day is the acceptable daily intake (ADI) of E127 dye, based on the WHO, FDA, and EU Scientific Committee for Food (SCF) [11]. Previous studies reported that 60–200 mg of E127 dye was given for 14 days as an oral clinical dose, and its effect was weak on humans[12]. Several studies reported different methods for removing artificial dyes from aqueous solutions, such as oxidation, electrochemical methods, precipitation, coagulation, and membrane separation. However, these methods are expensive, and some of them are not effective in removing dyes [13, 14] Among all these methods, adsorption has been shown to be the most effective method in removing industrial dyes and the least expensive [1, 13]. Many adsorbents are used for the adsorption of industrial dyes from aqueous solutions. Many factors affect the selection of adsorbents, including adsorption capacity, efficiency, cost, type of pollutants, toxicity, easy recovery, and reusability [15, 16]. Activated carbon (AC) is one of the most common adsorbents used in the adsorption of artificial dyes due to its high efficiency and simplicity in the sorption of artificial dyes from aqueous solutions [1, 5, 17, 18]. Among these adsorbents, clays, silicates, and aluminosilicate minerals are the main components of clay, which includes less than 2 µm of soil, rock sediment, and water [15, 19]. Generally, the clays have a high efficiency of adsorption due to the ions charged on the surface and a large surface area (800 $m^2.g^{-1}$) [15, 20, 21]. Kaolinite, montmorillonite, and bentonite are used as clay-based adsorbents. Kaolinite is usually used to remove dyes in aqueous solutions due to its high stability, large specific area, and reduced expansion [15, 22, 23]. Various experimental parameters are used to study adsorption, such as adsorbent dose, pH, contact time, ionic strength, temperature, and dye concentration [9, 24]. A multi-parameter study will be conducted to determine the best experimental parameters for the adsorption of E120 dye onto CK. In the previous studies mentioned, a single parameter was studied with fixed values for other parameters [9, 25]. In this project, we have developed a simple method for analysis of E127 dye from aqueous solutions using CK treatment using Adsorption isotherm data, thermodynamic parameters, kinetic models (Pseudo first order PFO, and Pseudo second order PSO), and

adsorption models (Langmuir, Freundlich, and Temkin) at different temperatures. Furthermore, the study determined the experimental parameters of the adsorption of E120 dye onto CK, including adsorbent dose, pH, contact time, ionic strength, and temperature.

2. MATERIEL AND METHOD

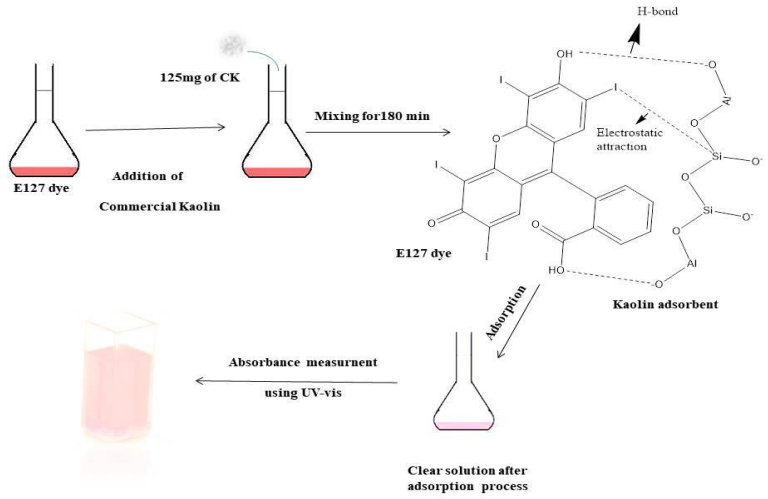
2.1 Materials and Apparatus

All materials used in this study were analytical grade reagents and used as received without any further purification: sodium Hydroxide from Loba Cheime, filter paper from Whattman, Hydrochloric acid from Loba Cheime, Sodium chloride from Loba Chemie. The dye sample of erythrosine was obtained from sigma-Aldrich; commercial kaolin was purchased from sigma-Aldrich. A double beam UV-vis spectrophotometer (SP-3000 nano, Optima, Japan) was used to produce absorption spectra, and an Ezdo pH meter-pp-201 was used to measure pH. (Taiwan). This experiment made use of a water bath shaker from Daihan Scientific in Korea.

2.2 Adsorption Procedure

Adsorption parameters were studied in batches to evaluate the efficacy of adsorption procedures between E127 dye and CK at different conditions such as contact time, adsorbed mass, pH, ionic strength, and temperature. A 0.2-2.25 µM ranged concentration of E127 dye was prepared in a 100-mL volumetric flask.

A 1 µM concentration of E127 dye was added to 100 mL, followed by 125mg of CK in the E127 solution as previously described. The pH 4.7 was adjusted by adding HCl and shaking at 25 °C for 30 min. 5 mL of the as-prepared solution were taken out, centrifuged for 5 min at 5000 rpm, and then filtered through whattman filter paper No. 2. The concentration remaining was measured at 525nm. Scheme 1 shows the general steps for the proposed methodology.



Scheme 1 shows the general steps for the proposed methodology, where step I shows the DM-µ-SPE method while step II shows the synthesis of the fluorescent N-FCDs for the determination of E 120 via a dynamic quenching mechanism.

The equations formula (1) and (2) were used to calculate the surface concentration q_e ($mg.g^{-1}$), and percentage removal (% removal) [26, 27].

$$qe = \frac{(Ci - Ce)}{m} X\ V \quad \text{..... (1)}$$

$$\% \, Removal = \frac{(Ci - Ce)}{Ci} X \, 100 \quad \ldots\ldots\ldots (2)$$

Where, $Ci, Ce, m, and \ V$ are initial, equilibrium concentration (µM), mass of CK (mg), and volume of aqueous solution (mL), respectively.

2.3 Kinetic Models Procedure

The kinetic models (PFO and PSO) were studied at E127 dye concentration (1µM), 125 mg CK adsorbed, pH 4.7, and at 25 ◦C. The remaining concentration of E127 was measured. The surface concentration q_t (mg.g^{-1}) at different times was calculated by equation formula (3)

$$qt = \frac{(Ci - Ct)}{m} X \, V \quad \ldots\ldots\ldots (3)$$

Where C_t is the concentration (µM) of the liquid-phase at different times in the aqueous solution.

3. RESULTS AND DISCUSSIONS

3.1 Spectroscopic Maximum Excitation Wavelength of E127 Dye

The maximum excitation wavelength (λ_{Ex}) was observed at 525 nm using spectrophotometric spectra, as shown in Figure 1. The correlation coefficients, R^2, limit of detection (LOD), and limit of quantification (LOQ), were 0.9982, 0.16, and 0.52, respectively.

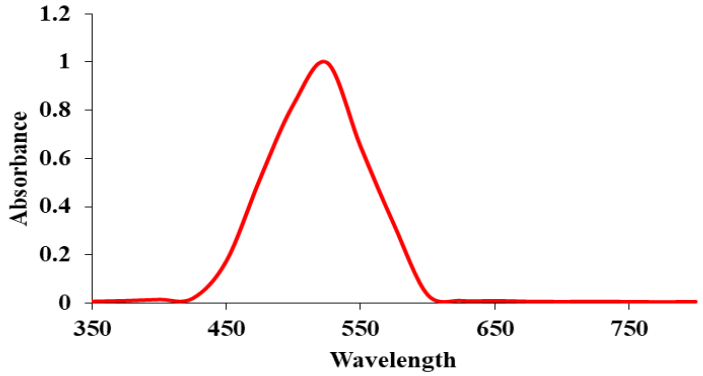


Figure 1. Absorption spectrum of E127 dye

3.1.2. Effect of Time, Ph, Adsorbed Mass, Ionic Strength, and Temperature

The contact time was studied to determine the equilibrium time and to find the efficacy of the adsorption of E127 dye onto CK. Figure 2a shows the q_e increasing with time at 180 min, then becoming almost constant. E127 dye absorption onto CK at 180 min was 80%. So, the suitable contact time for the adsorption process is 180 min. Adsorbent mass was studied due to its effect on the adsorption process and its effect on the removal of dye from aqueous solutions. Figure 2b shows the effect of CK weight on the removal of E127 dye. The qe value increased with increasing the weight of CK to 125 mg, and then the values of qe were at equilibrium between 150 – 500 mg. Therefore, the optimum mass for the adsorption process was 125mg. Figure 2c shows the qe values were quite reduced from 53.8 mg.g^{-1} at pH 4.7 to 6.7 mg.g^{-1} at pH 12, as a result of the similarity of the negative charge between the CK and E127 dyes (electrostatic repulsion).[9, 28] The pH of 4.7 was chosen as the optimum parameter for pH solution. The previous literature reported similar results for adsorption of E127 dye onto CK,[7] de-oiled mustard,[29] and de-oiled soya[9]. Figure 2d shows a study of the effect of ionic strength at concentrations

ranging from 1000 to 5000 µM on the absorption of E127 dye at 180 min contact time and the addition of 125 mg of CK. The adsorption of CK is considerably increased when the NaCl salt is added to the dye solution. At 1000 µM NaCl solution, qe has increased from 9.9 to 54.7mg.g^{-1} at 5000 µM. The increased qe with an increased concentration of NaCl salt is due to the migration of the NaCl salt ions to the KC surface.[30] The adsorption of E127 dye on CK was studied at temperatures of 25, 30, 40, and 50 ^{o}C. As shown in Figure 2e, the qe of E127 dye slightly decreased as temperatures increased, which indicates that the sorption process of E127 dye is not related to the temperature and that the active site of CK is damaged at higher temperatures[31].

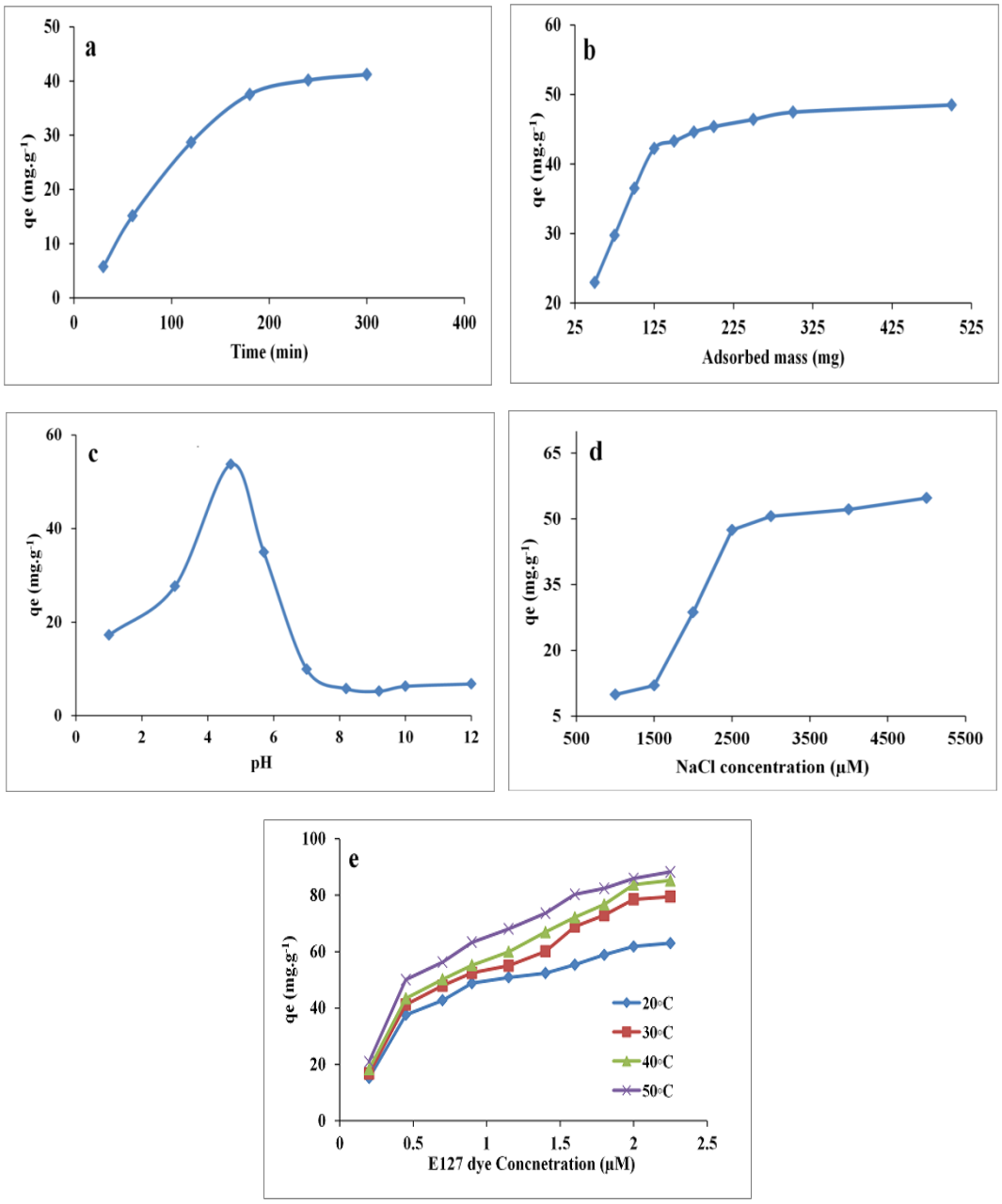


Figure 2. Impact of contact time (a), mass of KC (b), pH (c), ionic strength (d), concentration, and temperature (e) on the surface concentration of E127 dye onto CK.

3.2. Adsorption Isotherm

To determine the ability of CK to absorb E127 from aqueous solutions the Langmuir, Temkin and Freundlich models were used. The Langmuir theory describes an adsorption process that occurs at homogeneous sites of the adsorbent; no further adsorption can occur once the dye molecules bind to a site [32]. According to the equation formula (3)

$$\frac{Ce}{qe} = \frac{1}{klqm} + \frac{ce}{qm} \quad \text{..... (3)}$$

Where kl, qm, qe and Ce are the Langmuir constant (μM^{-1}), maximum surface concentration ($mg.g^{-1}$), adsorption capacity ($mg.g^{-1}$), and equilibrium concentration (μM), respectively. $\frac{1}{qm}$ and $\frac{1}{klqm}$ are found from the slope and intercept, respectively, of a straight line plot of $\frac{Ce}{qe}$ vs. ce. The Freundlich model describes the adsorption process for interactions between multilayer and heterogeneous surfaces.[32, 33] According to the equation formula (4)

$$\ln qe = \ln kf + \frac{1}{n}\ln Ce \quad \text{..... (4)}$$

Where kf and n are sorption capacity and sorption intensity, respectively. n and kf are found from the slope and intercept, respectively, of a straight line plot of $\ln qe$ vs. $ln\ Ce$. The Temkin model assumes that the heat of adsorption has an inverse relationship with increasing the coverage of the adsorbent surface while ignoring the high and low concentrations. It also features uniform power distribution for maximum bonding.[32] According to the equation formula (5)

$$qe = B ln\ A + B\ lnCe \quad \text{..... (5)}$$

Where B and A are heat adsorption ($J.mol^{-1}$) and Temkin constants ($L.g^{-1}$). Table 1 show that Freundlich model is the best for adsorption of E127 dye and CK, indicating that the adsorption is heterogeneous. Table 1 also shows the R^2 values for both the Langmuir and Temkin models. The values indicate that the two models are less suitable. Table 1 shows the n values in the Freundlich E127 absorption model (1.1 to 3.2). Both values of A and B in Temkin model showed that the reaction resulting from adsorption of E127 and CK is a physical sorption.

Table 1 List the isotherm parameters of the Langmuir, Freundlich, and Temkin models.

Langmuir				Freundlich			Temkin		
T.(K)	R^2	$qmax$	kl	R^2	kf	n	R^2	A	B
298	0.9875	74.6	2.6	0.9903	49.9	2.6	0.9824	8.9	22.6
303	0.9647	100.0	1.9	0.9294	59.8	1.9	0.9389	4.9	36.4
313	0.9727	105.3	1.9	0.9755	64.8	1.1	0.9767	5.5	37.3
323	0.9898	103.1	0.4	0.9373	74.1	3.2	0.9499	18.2	25.4

3.3. Adsorption Thermodynamic Parameters and Kinetic Models

Enthalpy, entropy and free energy were used to determine the adsorption of E127 dye from CK through equation formula (6) and (7).

$$\ln kd = \frac{\Delta S}{R} - \frac{\Delta H}{RT} \quad \text{..... (6)}$$

$$\Delta G = \Delta H - T\Delta S \quad \text{..... (7)}$$

Where the ΔH enthalpy (KJ.mol^{-1}), R gas constant is (8.3145 J.mol^{-1}.K^{-1}), ΔS entropy (J.K^{-1}.mol^{-1}), and kd distribution coefficient. The values of ΔH and ΔS were found to be -12.3 and 75.0, respectively, as represented in Table 2. The adsorption process of the E127 dye onto CK was not spontaneous, and exothermic due to the positive value of ΔG, and the negative value of ΔH. The adsorption of E127 dye onto CK was highly random during the solid-liquid interface due to the positive value of the ΔS. The values of (ΔG) decrease with increasing the temperature, which indicates that the adsorption of E127 with CK was endothermic [34].

Table 2 Thermodynamic parameters for the adsorption of E127 dye onto CK

ΔH	ΔS	ΔG (kJ.mol^{-1})			
		298 K	303 K	313 K	323K
-12.3	75.0	-22.3	-22.8	-23.5	-24.2

The PFO and PSO were used to examine kinetic models to assess E127 dye adsorption in CK. Table 3 shows that the $qe\ exp.$ value (mg.g^{-1}) is not close to the $qe\ cal.$value (mg,g^{-1}), the R^2= 0.9171 indicates that the PFO does not correspond to the sorption of E127 dye in the KC. The experimental value of qe is consistent with the calculated " qe " and R^2= 0.9891 indicates that PSO corresponds to E127 dye adsorption in the CK

Table 3 Kinetic parameters for Pseudo-first-order PFO and pseudo-second-order PSO models

E127 conc. (μM)	PFO			PSO		
1.0	k_1 (min^{-1})	$qe\ cal.$ (mg.g^{-1})	R^2	k_2 (g.mg^{-1}.min^{-1})	$qe\ cal.$ (mg.g^{-1})	R^2
	0.09	2.4	0.9171	27.8	0.5	0.9891

3.4. Reusability of Kaolin to Remove E127 Dye

The reuse of the CK to extract E127 was examined in an aqueous solution. Figure 3 shows the values of reuse of the CK (98.9%, 91.1%, 82.3% and 69.2%), respectively, showing that the CK can extract the E127 dye after four cycles.

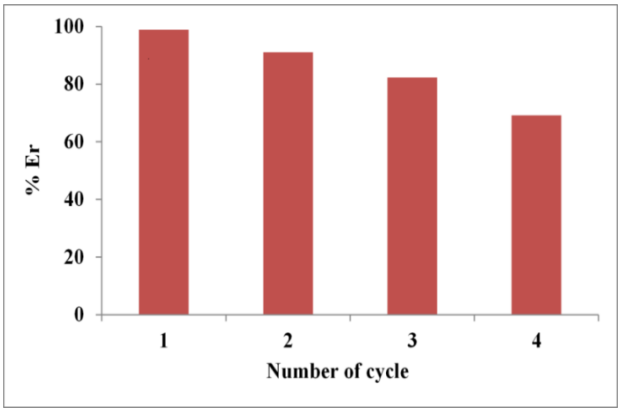


Figure 3. Reuse to extraction of E127 dye using CK.

3.5. The E127 Dye Adsorption Capacity of the CK Adsorbents was Compared to Some Previous Reports.

Table 4 was used to compare the q_{max} of the CK adsorbents in this study to the effects of E127 dye on CK adsorbent in previous reports. The results showed that the q_{max} varied depending on the experimental conditions

used. The adsorption capacity of the E127 dye in this study also varied compared to other CK adsorbents. Table 4 demonstrates that kaolin has notably high adsorption efficiency for E127 when compared to all of the adsorbents mentioned in the literature, with the exception of activated carbon (AC) binary systems and commercial activated carbon (CAC).

Table 4 The adsorption capacity (q_{max}) an isotherm models of various adsorbents reported in the literature for the adsorption of E127 dye.

Adsorbent	q_{max} ($mg.g^{-1}$)	Isotherm model	Ref.
AC single system	65.0	Langmuir	[35]
AC binary system	90.0	Langmuir	[35]
Fe_3O_4@C-nanodots	6.0	Langmuir	[36]
Commercial activated carbon	89.3	Langmuir	[9]
Hen feathers	15.4	Langmuir	[1]
Chitosan/ chitin/hydrogel SiO_2 hybrid	71.3	Langmuir	[37]
Bentonite clay	1.1	Langmuir	[38]
Commercial kaolin	74.6	Langmuir	This study

CONCLUSION

This study investigated the sorption of E127 dye onto CK and found that the PSO kinetic model provided the best fit to the experimental data. The temperature variations were used to determine the values of ΔH, ΔS, and ΔG. The negative value of ΔH suggests that the adsorption process of E127 dye onto CK is endothermic, which is characteristic of a physical adsorption process. The positive value of ΔS indicates that there are some structural changes at the solid-liquid interface. Given the outcomes and variables that were acquired, therefore, it can be inferred that CK can be an effective and affordable adsorbent for removing E127 dye from aqueous solutions.

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CONFLICT OF INTEREST

The authors declare no conflict of interest

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