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Article

# Thermodynamic and kinetic study of the adsorption of organic pollutants using environmentally friendly materials

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## Abstract

An AFM and BET were used to diagnose Flint surface, and the results were displayed. where it was determined that the specific surface area of Flint is 6.2697 $m^2$ $g^{-1}$. The biggest peak that may be obtained by a certain amount of Flint surface, with granules and an average diameter of 63.84 nm, is 31.0535 nm in height. The aqueous solution of Erythrosine was removed using the adsorption capabilities of Flint. The range of maximal dye adsorption was found to be 10.19-43.77%. At temperatures of (298,308,318,328)K, the Freundlich model was utilized since Temkin model could only be partially applied, Langmuir model was not appropriate, and the Freundlich model was the only one that worked. Reason being, out of the three models, it is the one that will be most useful in the real world. Physical adsorption is another option. Based on the results, the pseudo-second-order model, One of the three adsorption process kinetic models that were taken into consideration, was significantly applicable. Through the application of thermodynamic functions based on the enthalpy value, it was determined that the process was exothermic, with a negative value of -19.9471 kJ/mol. Similarly, the value of entropy was negative, which was -2.7962 J/mole.K. The process was shown to be exothermic adsorption, spontaneous, and occurring without randomization when Gibbs free energy was calculated.

**Keywords:** Removal; Pollution; Flint; Erythrosine; BET Surface Area.

## 1.Introduction

The term "pollutant" refers to any material—liquid, solid, or gaseous—that is introduced to the environment. Due to its pervasiveness in society, pollution ranks high among environmental and human concerns [1]. Disposal of many types of trash, including organic, petrochemical, petroleum, and industrial materials, is a common cause of this pollution [2].

Potential threats to human health include drinking water contaminated with organic dyes. Many worry that industrial dyes pose a threat to water quality. Contamination of the environment is still an issue, and it becomes much worse when the toxins are resistant and persistent. The xanthene family of synthetic colours is a subset of this category [3].

Clay is a major contributor to soil contamination because of the large amounts of toxins it can absorb. In organic pollution, clay particles often contain cations of innocuous elements like sodium and calcium, which can easily swap with toxic heavy metals like lead, cadmium, and mercury. Clay may be able to bind a significant number of organic pollutants due to its large surface area and capacity for a wide range of molecular interactions outside of ion exchange [4].

Flint is a soft, white mineral that has numerous uses. Porcelain makes extensive use of flint, which is abundant in Nigeria. Extensive study has demonstrated its efficacy in eliminating harmful metals from wastewater. It was found to be effective in removing nitrate, methylene blue, hexavalent chromium, and Congo Red from water-based solutions [5].

Synthetic dye from the xanthene family, is used as a food colorant [6]. The food sector, which includes the manufacturing of chocolate, biscuits, meat, candies, and chewing gum, as well as the pharmaceutical and cosmetics industries, make extensive use of Erythrosine (E127) as a colorant. [7]. With a solubility of 70g/L in water and excellent stability in both basic and neutral solutions, Erythrosine is a versatile drug. Fig.1 shows the chemical structure of synthetic E127 [8].

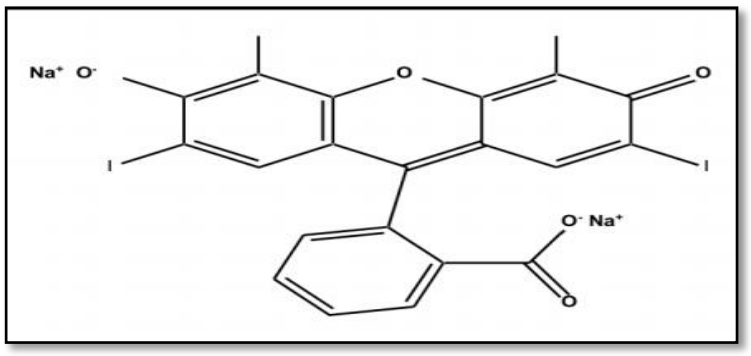


**Figure 1. Chemical structure of Erythrosine [8].**

## 2.Materials and method

### 2.1. Preparation of E127 stock solution

A standard solution was prepared by dissolving (1g) of E127 dye in de-ionized distilled water in a beaker. A 1000 ml volumetric flask was then used to dilute this solution. A solution of E127 was prepared with a concentration range of (5.0 - 50.0)ppm.

### 2.2. Preparation of Flint

An enough quantity was measured and transferred to a glass beaker to wash Flint clay. To further eliminate any surface impurities, the clay was rinsed ten to twelve times using de-ionized distilled water. Six hours were spent in a lab oven at 100 °C to dry the surface. Grind the dry surface with an electric grinder. With a granular size of (75µm), the Flint was passed through a molecular filter. Thereafter, it was preserved for the examination by placing it in a plastic container.

### 2.3. Diagnostics of the flint surface using Atomic Force Microscopy

Atomic force microscope (AFM) It is a high-resolution microscope that can produce two- and three-dimensional images of surfaces and analyze samples at a thousand times higher resolution than optical microscopes, which can only diagnose objects that are a few nanometers in size [9].
Fig2 shows the results of using AFM instrument to take two- and three-dimensional pictures of the surface topography of Flint. Investigating a sample that had been crushed using a sieve with a granular size of (75µm) or less yielded the following results:
As shown in Fig. 2, the highest height that a specific amount of Flint surface can reach is (31.0535)nm.

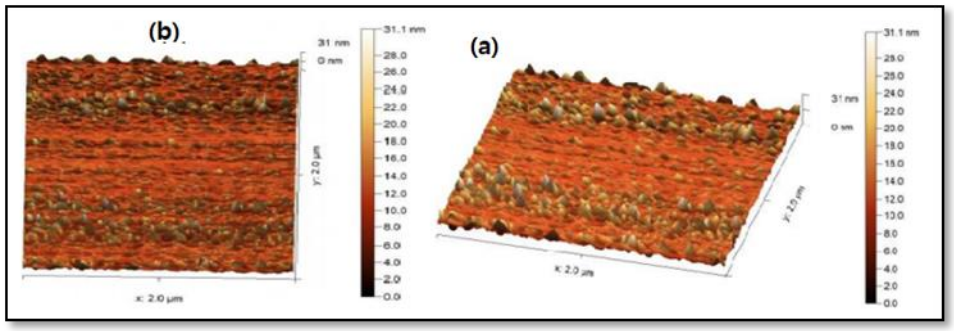


**Figure 2. (a) 3D and (b) 2D image of Flint under AFM**

According to the results of the particle analysis, the granules' distribution varied, and their diameters ranged from 57.35 nm to 76.32 nm, with an average of 63.84 nm. These findings are presented in Fig.3 and Scheme 1. In accordance with the chart's subsequent analysis. Many of the granules measured less than 70 nm in diameter, while bigger granules were sometimes present.

Many of the granules measured less than 70 nm in diameter, while bigger granules were sometimes present.

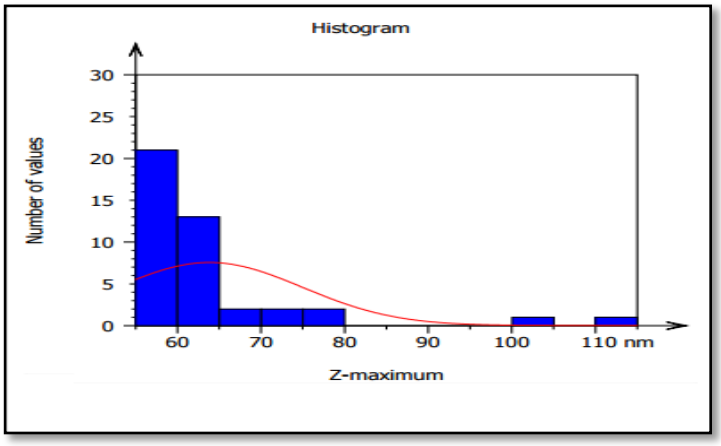
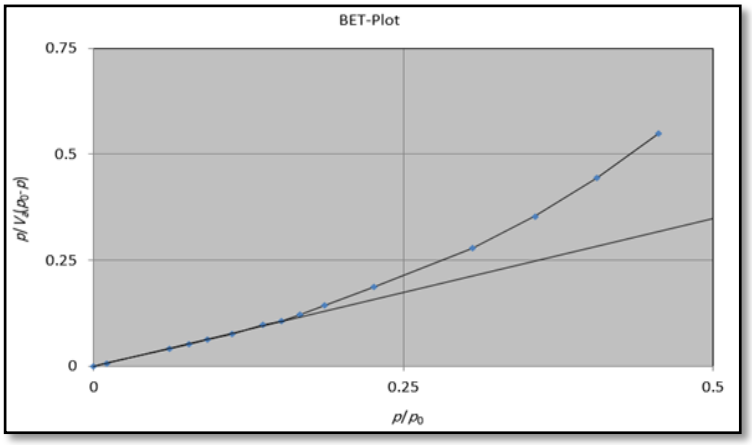



**Figure 3. Analysis of data of Flint surface particles under AFM**

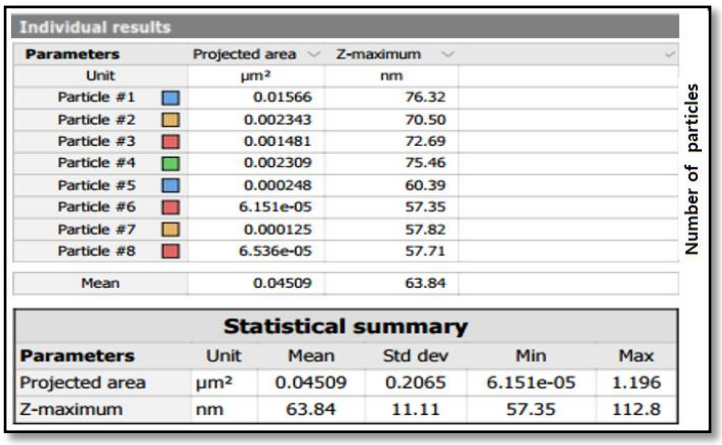



**Scheme1. The distribution of granules on Flint based on granule diameter.**

## 2.4.BET Surface Area of Flint

The specific surface area ($S_{BET}$) for flint is determined by the $N_2$ adsorption and desorption isotherm at (77.00) K. The pore structure and BET adsorption desorption curve are shown in Fig.4. Based on the IUPAC classification system, it is shown that the adsorption-desorption isotherm curve belongs to Type IV, and Flint has a mesoporous pore shape. With a specific surface area of (6.2697 $m^2$ $g^-$

[1]), an average pore width of (7.7947 nm), and a total pore volume of (0.0122 $cm^3$ $g^{-1}$). [10]



Individual results

| Parameters | Projected area | Z-maximum |
|---|---|---|
| Unit | µm² | nm |
| Particle #1 | 0.01566 | 76.32 |
| Particle #2 | 0.002343 | 70.50 |
| Particle #3 | 0.001481 | 72.69 |
| Particle #4 | 0.002309 | 75.46 |
| Particle #5 | 0.000248 | 60.39 |
| Particle #6 | 6.151e-05 | 57.35 |
| Particle #7 | 0.000125 | 57.82 |
| Particle #8 | 6.536e-05 | 57.71 |
| Mean | 0.04509 | 63.84 |

Number of particles

Statistical summary

| Parameters | Unit | Mean | Std dev | Min | Max |
|---|---|---|---|---|---|
| Projected area | µm² | 0.04509 | 0.2065 | 6.151e-05 | 1.196 |
| Z-maximum | nm | 63.84 | 11.11 | 57.35 | 112.8 |

**Figure 4. BET plot of Flint adsorption**

## 2.5. Determination of the maximum wavelength (λmax) for E127

The λmax was found by using a standard solution of E127. A visible and ultraviolet spectrophotometer was used to measure it within the range of (190-800 nm). The greatest wavelength at which E127 absorbs light is (526 nm) nanometers. We utilized a quartz cell that was 1 cm thick for the measurement, Fig.5 shows E127 dye's absorption spectrum.

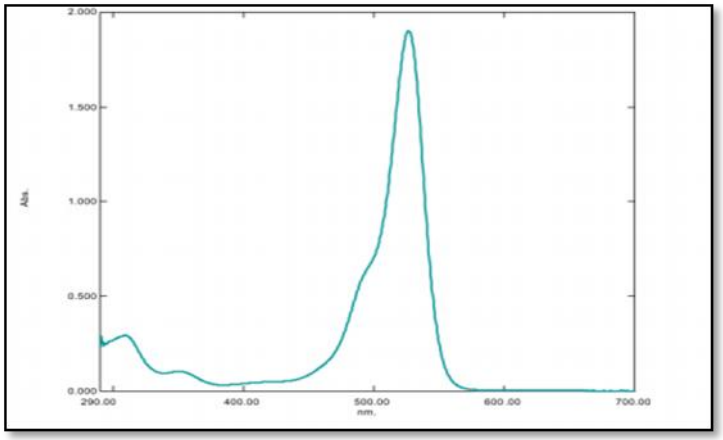



**Figure 5. UV-visible absorption spectrum of E127**

## 3.Results and Discussion

### 3.1. Impact of the adsorbent weight (Flint) on the capacity for adsorption

To find out how much weight the Flint should have to remove E127 dye via adsorption on its surface, we investigated a range of Flint weights from 0.05 to 0.7g at 298K with a primary dye concentration of 50mg/L. Fig.6 shows that the adsorbent surfaces were more successful in removing the dye from its water-based solution when the number of active sites generated Adsorption for E127 was raised. A steady increase in percentage of removal (%R) was seen as Flint weight increased by (0.05 to 0.9)g, or (10.19 to 43.77)%. All of the adsorption sites at Flint surface were saturated, which led to the percentage of elimination to remain constant at the optimal weight of (0.6)g. of Flint, which was (43.77)% [11].

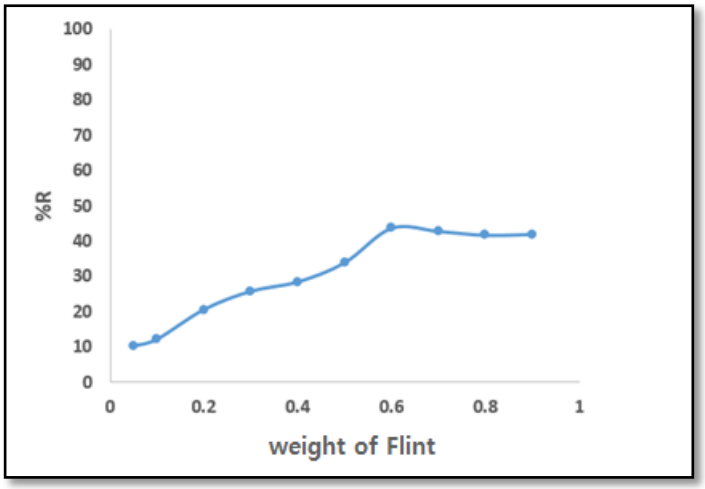



**Figure 6. Flint's weight and its impact on the adsorption of E127**

### 3.2. Determine the equilibrium time.

At initial dye concentrations of (50mg/L) and at different temperatures (298,308,318,328)K, The impacts of Flint's equilibrium time with the dye E127 aqueous solution for granule size (≤75µm) and Flint's ideal weight (0.6g) were investigated. Fig.7 shows the linear relationship between the passage of time and the adsorption process on Flint. According to the results, E127 dye adsorption process on Flint has an equilibrium duration of (40 min).

**Figure 7. Effect of equilibrium time on the amount of E127 that was adsorbed on Flint**

### 3.3. Adsorption Kinetics Models studies

Applying pseudo-first-order (PFO), pseudo-second-order (PSO), and Elovich kinetic models, the adsorptive behavior of E127 on Flint was examined. Here is the linear form of PFO that has been stated. [12]

$$\ln(qe - qt) = \ln qe - K1t \_\_\_\_\_1$$

qt in unite (mg/g) is capacity for adsorption at time, qe in unite (mg/g) is capacity for adsorption at equilibrium, and K1 is the rate constant for adsorption in unite (min-1). To determine K1, we look at the linear plot of in (qe-qt) against time (t), as shown in Fig.8a.

To find Pseudo first order constants, follows formula (2). [13]

$$\frac{t}{qt} = \frac{1}{K2qe^2} + \frac{1}{qe}\, t \_\_\_\_\_2$$

$K_2$ in unite (g mg-1. min -1) is (PSO) process's adsorption rate constant. The plot of t/qt vs. time (t) is linear, and K2 is calculated from the intercept of this curve Fig.8b.

The (Elovich) constants are determined using the constants ∘ and β as shown in formula (3). [14]

$$qt = \frac{1}{\beta}\ln(\beta\alpha) + \frac{1}{\beta}\ln t \_\_\_\_\_3$$

The desorption process constant is denoted by ∝ and is measured in units of (mg.g-1.tim-1), while the initial adsorption rate constant is denoted by β and is measured in units of g.mg-1. In addition, the values of are computed using the slope and intercept shown in Fig.8c. [15]

The strong ($R^2$) values and converging of the theoretical and actual (qe) values make PSO and Elovich models usable; this is demonstrated by the results in Table1 and an examination of the R2 values for the three Kinetic models. Real (qe) values didn't match theoretical values, and there were not enough (R2) values to use PFO Kinetic models. [16,17]

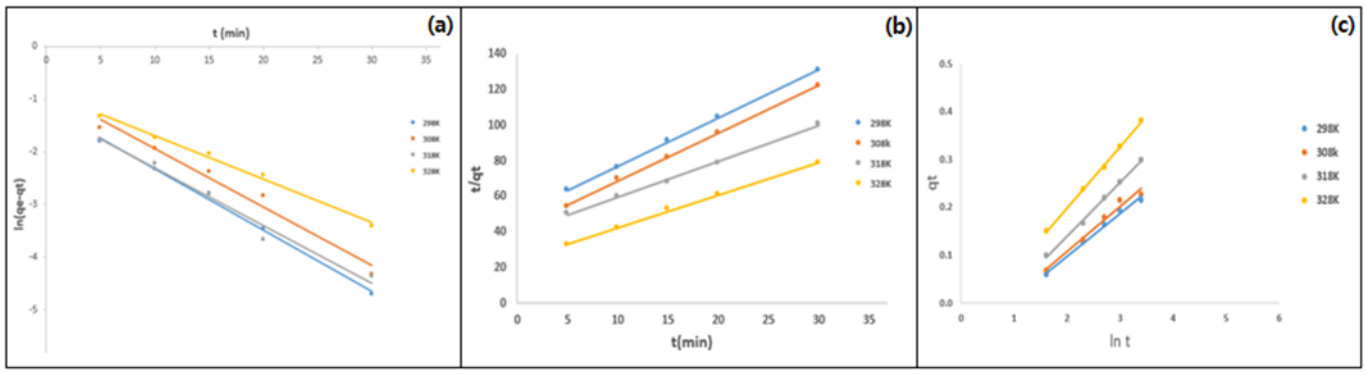



**Figure 8. (a) PFO, (b)PSO, (c)Elovich kinetic models for the adsorption of E127 on Flint at different temperatures.**

**Table (1) Adsorbent Flint surface's kinetic constant values at various temperatures**

| T(K) | Pseudo first order | | | Pseudo second order | | | Elovich | | |
|---|---|---|---|---|---|---|---|---|---|
| | K1 (min-1) | qe (mg/g) | $R^2$ | K2 (g.mg-1.min-1) | qe (mg/g) | $R^2$ | β (g.mg-1) | ∝ (mg.g-1.min-1) | $R^2$ |
| 298 | 0.1163 | 0.313 | 0.997 | 0.1496 | 0.3667 | 0.9997 | 11.2107 | 0.0363 | 0.9913 |
| 308 | 0.1091 | 0.2955 | 0.9785 | 0.1716 | 0.3726 | 0.9989 | 10.6496 | 0.0398 | 0.9786 |
| 318 | 0.1105 | 0.4318 | 0.976 | 0.1021 | 0.4981 | 0.9977 | 8.8967 | 0.0527 | 0.9978 |
| 328 | 0.0823 | 0.4162 | 0.9923 | 0.1376 | 0.5469 | 0.9987 | 7.9064 | 0.0821 | 0.9987 |

## 3.4. Adsorption isotherms

We used a range of temperatures (298, 308, 318, and 328) K, various concentrations of E127 (5-60 mg/L), the optimal adsorbent surface weight of Flint (0.6 g), particle size (≤75µm), and an equilibrium time (40 min) to analyze the adsorption isotherms. Adsorption isotherms of E127 dye on Flint at various temperatures are shown in Fig.9 as a function of both qt(mg/g) and (Ce (mg/L).

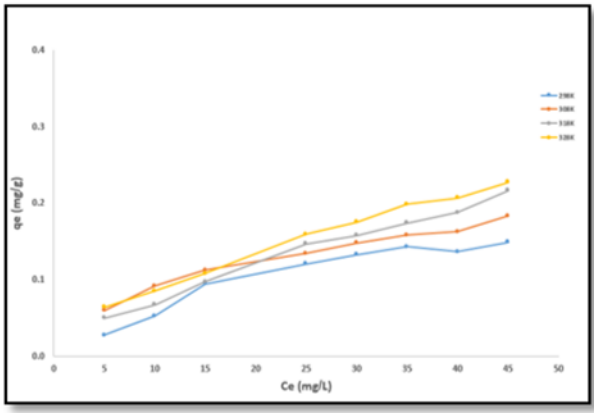



**Figure 9. Isotherm for adsorption of E127 on Flint**

In accordance with Freundlich's adsorption principles, the dye's adsorption curve on Flint type (L3) is depicted in Fig.9. In accordance with Giles' classification, this demonstrates that various forces are applied during the adsorption process on the adsorbent surface. The more area the adsorbent surface covers, the more material is adsorbed on the surface [18]. Because adsorption takes place horizontally on the adsorbent surface, the adsorbent will occupy a bigger part of the surface. Thus, occupied areas on the surface with limited adsorption potential and a small amount of adsorption will occur. [19]

### 3.5. Adsorption Isotherm Models

After analyzing the acquired data using three adsorption process isotherm models, The findings for the Temkin, Freundlich, and Langmuir model constants as well as correlation coefficients are shown in Table 2. Fig10 shows the adsorption isotherm models. Through the application of equation (4), Freundlich model constants $K_f$ in unite (mg/g) and (n) were determined. [20]

$$\log(qe) = \log(Kf) + \frac{1}{n}\log Ce \quad \text{\_\_\_\_\_4}$$

By applying formula (5), we were able to determine $K_L$ in unit (L/mg) which represents Langmuir constants and qemax in unite (mg/g). [21]

$$\frac{Ce}{qe} = \frac{Ce}{qe\ max} + \frac{1}{qe\max Kl} \quad \text{\_\_\_\_\_5}$$

Applying formula (6), BT (J/mol) and Temkin constants in unit (L/mg) were taken into consideration. [22]

$$qe = BT * lnKT + BT * lnCe \quad \text{\_\_\_\_\_6}$$

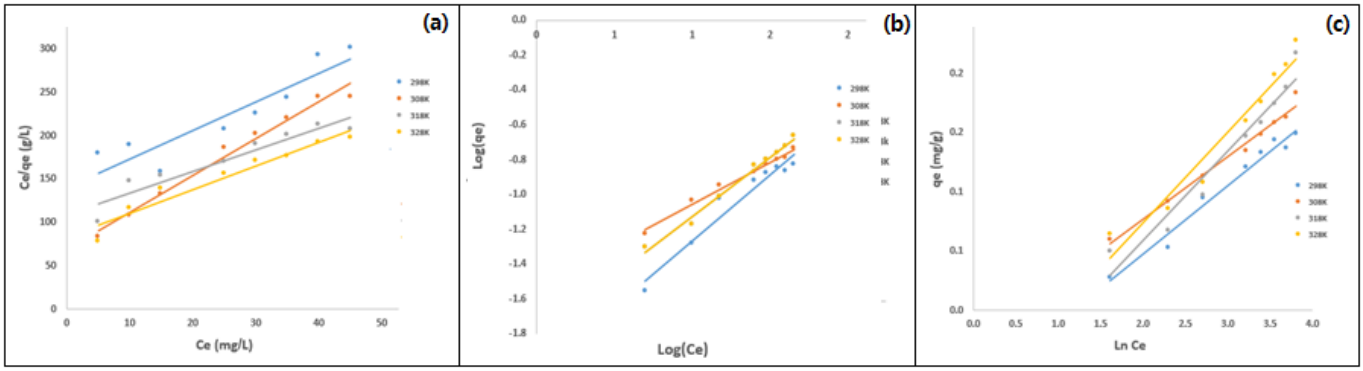



**Figure 10. (a)Langmuir, (b)Freundlich, (c)Temkin isotherms for adsorption of E127 on Flint at different temperatures.**

| T(K) | Langmuir | | | Freundlich | | | Temkin | | |
|---|---|---|---|---|---|---|---|---|---|
| | KL (L/mg) | qemax (mg/g) | $R^2$ | Kf (mg/g) ) | n | $R^2$ | KT (L/mg) | BT (J/mol) | $R^2$ |
| 298 | 0.0234 | 0.3035 | 0.8454 | 0.0093 | 1.3137 | 0.9482 | 0.5763 | 7.5354 | 0.976 |
| 308 | 0.0618 | 0.2346 | 0.9821 | 0.0291 | 2.0859 | 0.9883 | 0.3564 | 7.3207 | 0.9822 |
| 318 | 0.0227 | 0.4021 | 0.9037 | 0.0155 | 1.4641 | 0.9898 | 0.3068 | 5.9072 | 0.9447 |
| 328 | 0.0323 | 0.3684 | 0.9347 | 0.0228 | 1.6697 | 0.9901 | 0.2951 | 4.0026 | 0.9482 |

**Table (2) The adsorbent surface Flint's isotherm constant values at various temperatures.**

The adsorption process in this experiment may be well described by Freundlich's isotherm equation, as shown in Table 2, which also demonstrates that the values of (n) must be between 1 and 10. This demonstrates that physical forces control the adsorption process. Freundlich constant (Kf) for the adsorbent surface Flint decreases with increasing temperature, lending credence to the idea that the adsorption process is exothermic. There were several different adsorption energy locations on the heterogeneous surfaces where the adsorption process occurred [23]. Langmuir model does not fit, as shown in Table 2, due to the relatively low linear correlation coefficient at the same test temperatures [24]. Similarly, Temkin's model has limited applicability because the surface correlation coefficient is always less than 0.9. [25]

### 3.6. Thermodynamic study for adsorption of E127 on Flint

By calculating thermodynamic constants like Gibbs free energy , Entropy and Enthalpy ($\Delta G°$, $\Delta S°$,$\Delta H°$), the impact of temperature on the adsorption process's behavior could be shown [26]. A look at Table 4. reveals the thermodynamic function values. E127 adsorption process's equilibrium constant on Flint was estimated by using Equation (7) to calculate the values of the thermodynamic functions at each temperature, which were necessary for the adsorption of dye on the adsorbent surface [27].

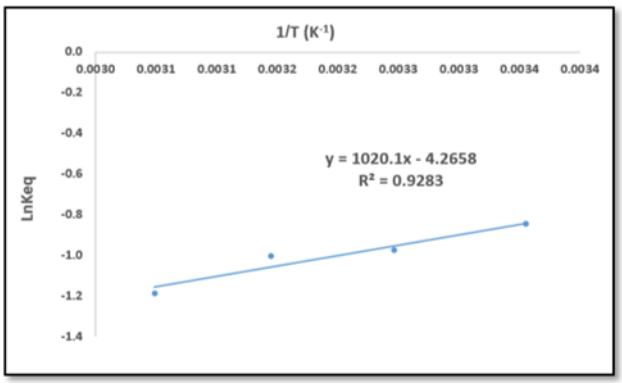



**Figure 11. Vant Hof's equation for adsorption E127 dye on Flint**

$$Keq = \frac{qe * W(g)}{Ce * V\ (l)} \quad \text{_____7}$$

Plotting Fig. 10 between lnKeq and 1/T from Table 3 will yield the equations for ΔH° and ΔS°. The slope is denoted by (- ΔH°/R) and the intercept by (ΔS°/R) [28].

$$\ln Keq = \frac{-\Delta H°}{RT} + \frac{\Delta S°}{R} \quad \text{_____8}$$

**Table (3) Thermodynamic values of E127 dye on the Flint at different temperatures.**

| Adsorbent | T (K) | ΔG° KJ/mole | ΔH° KJ/mole | ΔS° J/mole.k |
|---|---|---|---|---|
| Flint | 298 K | -6.953 | -19.9471 | -2.7962 |
| | 308 K | -4.3169 | | |
| | 318 K | -5.5058 | | |
| | 328 K | -3.9068 | | |

To calculate the change in Gibbs free energy, utilizes Equation (9) [29].

$$\Delta G° = -RTInKeq \quad \text{_____9}$$

**Table (4) values of the adsorption E127 dye thermodynamic equilibrium constants at various temperatures and on Flint.**

| Adsorbent | T (K) | $1/T\ (K^{-1})$ | $K_{eq}$ | Ln $K_{eq}$ |
|---|---|---|---|---|
| Flint | 298 K | 0.0034 | 0.2792 | -1.2758 |
| | 308 K | 0.0032 | 0.3874 | -0.9483 |
| | 318 K | 0.0031 | 0.3664 | -1.004 |
| | 328 K | 0.003 | 0.4257 | -0.854 |

The enthalpy values were lower than 40 KJ/mol, confirming physical adsorption, and the fact that the adsorption process was exothermic shows that it occurred on

Flint [30]. The regularity, rather than randomness, of the surface dye adsorption process is demonstrated by the negative entropy values. It is not a spontaneous adsorption process for E127 dye on Flint because the Gibbs free energy values are positive [31].

## Conclusions

The experiment showed that Flint absorbed Erythrosine dye with a removal rate of 43.77%. According to the results of the used kinetic constants, (PSO) model has the best match. matching Freundlich isotherm as an additional condition. The lack of spontaneity in the adsorption process is shown by the negative values of ΔG°, which, along with the negative values of ΔS° and ΔH°, suggests that the adsorption process is becoming more regular, is exothermic, and physical.

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