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Adsorption of Sudan III Dye onto Commercial Bentonite: Optimal Conditions, Isotherm Modeling, and Kinetic Analysis

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Abstract

In this research, a cost-effective and highly efficient technique was employed to removal of Sudan III dye using a commercially available bentonite (CB). The greatest concentration of dye on the surface was measured to be 25.03 mg.g-1 when 50 mg of adsorbent was used, and it was found that 1.0 was the most acceptable pH for the adsorption process. The Langmuir isotherm, which defined the adsorption process, showed that the active sites in charge of adsorption were chosen at random. The results of the calculations showed that the enthalpy (ΔH), entropy (ΔS), and adsorption capacity (qmax) were, respectively, 107.3 mg.g-1, 19.7 KJ.mol-1, and 54.7J.mol-1. According to the pseudo-second-order (PSO) model, both thermodynamic and kinetic analyses showed that the adsorption process was not spontaneous but rather involved physical adsorption.

Keywords: *adsorption processes, thermodynamics, kinetics, isotherm, Sudan III dye, and commercial bentonite (CB).*

1. Introduction

Recently, artificial colorings have received great attention due to their frequent use in canned foods, even though they are harmful to the environment, especially with regard to water pollution (Gupta, Mittal, Kurup, & Mittal, 2006; Kadirvelu et al., 2003; C. Li, Zhong, Wang, Xue, & Zhang, 2015). A significant amount of industrial colors are used in the food processing process, and annually, 10-15% of these dyes are released into wastewater (Garg, Gupta, Yadav, & Kumar, 2003; Jain & Sikarwar, 2009; Lee, Choi, Thiruvenkatachari, Shim, & Moon, 2006). Sudan III, a fat-soluble dye, belongs to the diazo lysochrome dye family and bears structural similarities to azobenzene. It’s used in acrylic emulsions, waxes, and hydrocarbon products, It is distinguished by its red hue and solubility in methanol (Mittal, Mittal, Kurup, & Singh, 2006; UYSAL & ARAL, 1998). Sudan dyes have been found in foods like pizza, noodle soup, fish sauce, chili powder, and Worcestershire sauce (Abu-Alrub, Amer, & Alkahtani, 2014; Pardo, Yusà, León, & Pastor, 2009). Excessive use of Sudan III dye offers a concern to human health since it can cause high levels of thyroid hormone, increasing the possibility of developing hyperthyroidism (Al-Degs, Abu-El-Halawa, & Abu-Alrub, 2012; Bernstein, Haugen, & Frey, 1975). The International Agency for Research on Cancer (IARC) categorized Sudan

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dyes as Group 3 carcinogens (Abu-Alrub et al., 2014; Ikhazuangbe P.M.O., 2017; Papillomaviruses, 2011). Several methods were used to separate Sudan III dye from aqueous media, such as precipitation, electrochemical, oxidation, and coagulation. Still, these methods tend to be expensive, and some of them show limited efficiency in totally eradicating dyes (Salleh, Mahmoud, Karim, & Idris, 2011; Yahyaei, Mousavi, Parvini, & Mohebi, 2016). Adsorption has emerged as the most efficient and cost-effective way for removing industrial dyes (Gupta et al., 2006; Yahyaei et al., 2016). Several adsorbents are used to assist in the adsorption of artificial dyes from aqueous media (Shahadat & Isamil, 2018; Unuabonah & Taubert, 2014). Activated carbon (AC) is generally recognized as a routinely used adsorbent in the process of removing synthetic colors from water solutions. It is preferred for its excellent efficiency and simplicity in trapping synthetic dyes during the sorption process (Gupta, Ali, & Saini, 2007; Gupta et al., 2006; Jain & Sikarwar, 2009; Rozada, Calvo, Garcıa, Martın-Villacorta, & Otero, 2003). Clay is one of the principal adsorbents used for removing dyes from aqueous media, due to the silicates and aluminosilicate minerals found in soil, rock sediment, and water particles smaller than 2 µm (Krishna Gopal Bhattacharyya & Gupta, 2008; Shahadat & Isamil, 2018). Clays have excellent adsorption effectiveness due to the presence of charged ions and their large surface area (about 800 $m2.g^{-1}$). This combination of elements contributes to their excellent adsorption capacity (Adeyemo, Adeoye, & Bello, 2017; Murray, 2002; Shahadat & Isamil, 2018). Bentonite, kaolinite, and montmorillonite are widely used as clay-based adsorbents in a variety of applications. Bentonite is used to remove dyes from aqueous media due to its stability and specific surface area (Mustapha et al., 2019; Shahadat & Isamil, 2018; Uddin, 2017). Adsorption processes are investigated using a variety of experimental parameters, including adsorbent masses, pH, time, concentration of salt, temperature, and initial dye concentration. These characteristics are studied to get insights into the adsorption behavior and maximize the efficiency of the operation (Al-Degs et al., 2012; Brereton, 2003). A detailed multi-parameter investigation will be carried out to find the ideal experimental settings for Sudan III dye adsorption on CB (carbon black). Unlike earlier research that concentrated on a single parameter while leaving other parameters constant, this study will analyze the combined influence of numerous elements to discover the most effective circumstances for the adsorption process (Al-Degs et al., 2012; Alkahtania, Abu-Alrubb, & Mahmoudc, 2017). In this project, we have developed a simple method for CB treatment-based analysis of Sudan III dye in methanol solvent. The analysis makes use of pseudo-first-order PFO and pseudo-second-order PSO models, as well as the adsortion isotherm. The investigation also looked at the experimental parameters for the adsorption of Sudan III dye onto CB, such as temperature, concentration of NaCl, time, pH, and CB dose.

2. Experiment

2.1 Materials and instruments

In this study, the chemical materials used were of analytical grade. (NaOH) sodium hydroxide, HCl hydrochloric acid, and NaCl sodium chloride were obtained from Loba Chemi. Filter paper from Whatman. The Sudan III dye and commercial bentonite were procured from Loba Chemi and Sigma-Aldrich, respectively. Absorption spectra were obtained using a double-beam spectrophotometer (Cinitra 1010, GBC). pH values were taken using an Ezdo pH meter (pp-201) from Taiwan.

2.2 Sorption procedure

The adsorption process between Sudan III dye and CB adsorbent was investigated under various conditions, including time, CB mass, pH, concentration of NaCl, and temperature. A concentration range of 0.1–0.9 mM of Sudan III dye was prepared in a 50-mL volumetric flask. For the experimental procedure, 50 mL of the Sudan III dye solution with a concentration of 0.5 mM was combined with 50 mg of CB. The pH was modified to 1.0 using HCl, and the solution was agitated at 25 °C for 120 min. Subsequently, A volume of 10 mL was extract out of the prepared solution, centrifuged at 10000 rpm for 10 minutes, and then filtered through filter paper grade 2. Then, at 515 nm, the remaining concentration was measured.

The surface concentration q_e (mg.g-1) was determined using equation (1) (Bauer, Jacques, & Kalt, 2001; Mahnashi, Abu-Alrub, Amer, & Alqarni, 2021).

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

Where Ci, Ce, m,and V are the initial and equilibrium Sudan III dye concentration (mM), weight of CB (mg), and volume of methanol solution (mL), respectively.

2.3 Kinetic models procedure

The kinetic models, namely Pseudo First Order (PFO) and Pseudo Second Order (PSO), were examined under specific conditions, including a concentration of 0.5 µM for Sudan III dye, 50 mg of CB adsorbent, a pH of 1.0, and a temperature of 25 °C. Equation formula (2) was used to determine the qe at various time (t) intervals based on the measured amount of Sudan III dye remaining in the sample.

$$qe = \frac{(Ci - Cf)}{m} X V \quad .... (2)$$

The concentration (C_f) in mM of the liquid phase at different time points in the aqueous solution is represented by the variable C_f.

3. Results and discussions

3.1 Absorption spectrum, limit of detection (LOD), and limit of quantitative (LOQ) of Sudan III dye

The spectrophotometric spectra depicted in Fig. 1 revealed that the highest excitation wavelength (λ_{Ex}) observed was 515 nm. The correlation coefficient (R^2) was determined to be 0.9929, while the limit of detection (LOD) and limit of quantification (LOQ) were calculated as 0.004mM and 0.014 mM, respectively.

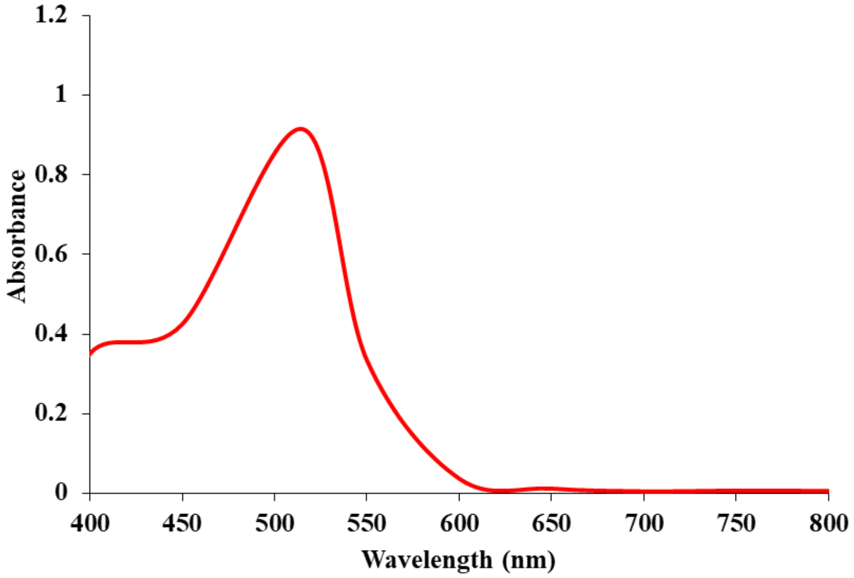

Fig.1 Absorption spectrum of Sudan III dye

Table 1 provides a summary of the analytical performances of previous approaches and the newly proposed method for detecting Sudan III. In comparison to the prior methods, the proposed technique exhibits enhanced sensitivity and a reduced detection limit.

Table.1 Comparison of LOD in different methods

Analyte	Method	LOD (mM)	Ref.
Sudan III dye	Biosensor	0.073	(Y. Li, Wang, Bai, & Wang, 2017)
	SERS	0.012	(Wu et al., 2018)
	HPLC	0.016	(Daood & Biacs, 2005)
	Spectrophotometer	0.004	This work

3.2. The effects of contact time, pH, CB mass, NaCl concentration, and temperature on the sorption process were investigated.

The study investigated the contact time to determine the time required to reach equilibrium and evaluate the effectiveness of Sudan III dye adsorption onto CB. Fig.2a illustrates that q_e (adsorbed amount) increases with time until it reaches a plateau at 120 minutes. At this time, the adsorption of Sudan III dye onto CB was measured to be 0.40. Therefore, 120 minutes was chosen as the optimal contact time for the adsorption process. The effect of adsorbent mass on the sorption technique and the qe of Sudan III dye in methanol solution were also investigated. Figure 2b shows how CB mass affects the Sudan III dye extract. The qe value increased as the CB mass increased up to 50mg, then stabilized at 300 and 500mg. Consequently, the ideal mass for the sorption process was found to be 50 mg (Daood & Biacs, 2005). Fig.2c indicates a significant reduction in q_e values from 0.48 $mg.g^{-1}$ to 0.32 at pH 1.0 to pH 12, respectively. This can be attributed to the similarity in negative charge between CB and Sudan III dyes, resulting in electrostatic repulsion. (Al-Degs et al., 2012; Shakir, Elkafrawy, Ghoneimy, Beheir, & Refaat, 2010).The pH value of 1.0 was identified as the optimal parameter for the solution's pH. Similar results have been shown in previous studies of the adsorption of the Sudan III dye formula on activated carbon. (Abu-Alrub et al., 2014). Fig. 2d presents a study investigating the impact of ionic strength within the concentration range of 500 to 3000 mM on the absorption of Sudan III dye. The study was conducted at a contact time of 120 minutes with the addition of 50 mg of CB. The results indicate that the adsorption of CB significantly increases when NaCl salt is introduced to the dye solution. At a 500 µM NaCl solution, the q_e value decreases from 0.43 - 0.26 $mg.g^{-1}$ at 3000 mM. The migration of Cl ions to the surface of Sudan III dye is responsible for the drop in qe that occurs when the concentration of NaCl salt increases (Issa & Al-Degs, 2009). The study examined the adsorption of Sudan III dye on CB at temperatures of 25, 30, and 40 °C. As depicted in Fig. 2e, the qe of Sudan III dye exhibited a minor decrease with increasing temperatures. This suggests that the sorption process of Sudan III dye is not influenced by temperature and that higher temperatures may potentially damage the active sites of CB (Krishna G Bhattacharyya, SenGupta, & Sarma, 2014).

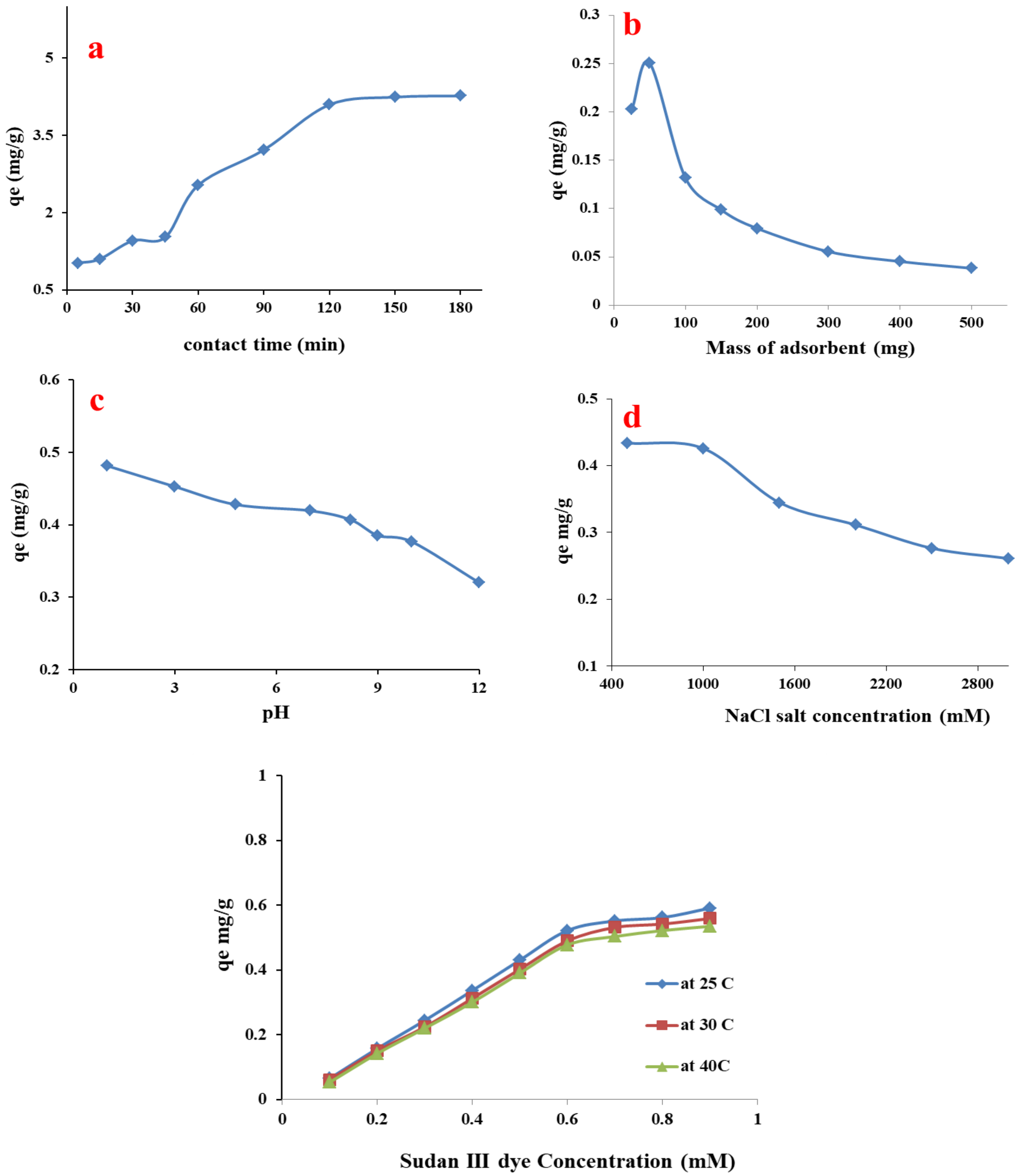

Fig. 2 The surface concentration of Sudan III dye onto CB was investigated in relation to various factors: contact time (a), mass of CB (b), pH (c), NaCl concentration (d), and temperature (e).

3.3. Adsorption isotherm

The Langmuir, Temkin, and Freundlich models were employed to assess the adsorption capacity of CB for absorbing Sudan III dye from methanol solutions. The Langmuir model is based on the assumption that adsorption takes place at homogeneous sites on the adsorbent, and once the dye molecules bind to a site, no further adsorption can occur (Inyinbor, Adekola, & Olatunji, 2016). According to the equation formula (3)

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

Where kl, qm, qe and Ce are the Langmuir constant (mM^{-1}), maximum surface concentration ($mg.g^{-1}$), adsorption capacity ($mg.g^{-1}$), and equilibrium concentration (mM), 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 characterizes the adsorption process as interactions between multilayer and heterogeneous surfaces (Inyinbor et al., 2016; Patil, Renukdas, & Patel, 2011) According to the equation formula (4)

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

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 decreases as the coverage of the adsorbent surface increases, disregarding high and low concentrations. It also incorporates a uniform power distribution to maximize bonding (Inyinbor et al., 2016) According to the equation formula (5)

$$qe = B\ln A + B \ln Ce \ldots\ldots\ldots\ldots\ldots. (5)$$

Table 2 demonstrates that the Langmuir model is the most effective in describing the adsorption of Sudan III dye and CB, suggesting a uniform adsorption process. Additionally, the R^2 values for both the Freundlich and Temkin models, as presented in Table 2, indicate that these models are less appropriate for describing the adsorption phenomenon. In the Freundlich Sudan III absorption model, Table 2 displays n values ranging from 0.90 to 1.2. Furthermore, the values of A and B in the Temkin model suggest that the adsorption of Sudan III dye and CB is a physical sorption reaction.

Table 2 presents the isotherm parameters for 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	107.3	1.6	0.9608	1.3	1.0	0.9622	10.4	0.26
303	0.9647	90.8	2.7	0.9714	1.8	1.2	0.9389	10.2	0.25
313	0.9727	100.1	2.1	0.9702	1.4	0.9	0.9767	9.9	0.24

3.4. Adsorption thermodynamic parameters and kinetic models

The adsorption of Sudan III dye from CB was analyzed using equations incorporating enthalpy, entropy, and free energy.

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

$$\Delta G = \Delta H - T\Delta S \ldots\ldots\ldots\ldots\ldots\ldots\ldots...\ldots (7)$$

The enthalpy (ΔH) with a value of 19.7 $KJ.mol^{-1}$, the gas constant (R) of 8.3145 $J.mol^{-1}.K^{-1}$, the entropy (ΔS) of 54.7 $J.K^{-1}.mol^{-1}$, and the distribution coefficient (kd) were utilized to assess the adsorption of Sudan III dye onto CB. Table 3 displays the obtained values of ΔH and ΔS. The adsorption process of Sudan III dye onto CB was determined to be spontaneous and exothermic, as indicated by the positive values of ΔG and ΔH. The adsorption at the solid-liquid interface exhibited a high degree of randomness, attributed to the positive ΔS value. Additionally, the values of ΔG decreased as the temperature increased, indicating that the adsorption of Sudan III dye with CB was endothermic (Sah et al., 2022).

Table 3 presents the thermodynamic parameters related to the adsorption of Sudan III dye onto CB

ΔH	ΔS	ΔG ($kJ.mol^{-1}$)		
		298 K	303 K	313 K
19.7	54.7	3.4	3.2	2.3

The adsorption of Sudan III dye onto CB was investigated using PFO (pseudo-first-order) and PSO (pseudo-second-order) kinetic models. Table 4 demonstrates that the experimental value of qe ($mg.g^{-1}$) is not in close agreement with the calculated qe cal. value ($mg.g^{-1}$) in the case of PFO, as indicated by an R^2 value of 0.8202. This suggests that the PFO model does not accurately represent the sorption process of Sudan III dye in CB. On the other hand, the experimental value of q_e aligns well with the calculated qe value in the case of PSO, as evidenced by an R^2 value of 0.9957. This indicates that the PSO model is a suitable representation of the adsorption of Sudan III dye in CB.

Table 4 presents the kinetic parameters for the PFO and PSO models

Sudan III dye conc. (mM)	PFO			PSO		
	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
0.5	0.03	57.22	0.8202	99.51	74.63	0.9957

3.4. The Sudan III dye adsorption capability of CB adsorbents was compared to earlier studies.

Table 5 was utilized to compare the q_{max} of the CB in this study to the effects of Sudan III dye on CB adsorbent in previous reports. The results showed that the q_{max} varied depending on the experimental conditions used. The qmax of the Sudan III dye in this study also varied compared to other CB. Table 5 demonstrates that bentonite has notably high adsorption efficiency for Sudan III dye when compared to all of the adsorbents mentioned in the literature.

Table 5 Comparison of the qmax of Sudan III dye with previous studies

Adsorbent	q_{max} ($mg.g^{-1}$)	Isotherm model	Ref.
grafted silylated bentonite	95.0	Langmuir	(Saeed et al., 2020)
AC binary system	7.8	Langmuir	(Abu-Alrub et al., 2014)
chlorotrimethylsilane, (OTCS)	76.92	Langmuir	(Saeed et al., 2020)
diphenyldichlorosilane, (TPCS)	97.08	Langmuir	(Saeed et al., 2020)
n-octyltrichlorosilane, (ODTCS)	76.92	Langmuir	(Saeed et al., 2020)
Commercial bentonite CB	107.3	Langmuir	This study

4. Conclusion

The Sudan III dye adsorption onto CB was examined in this study, and it was found that the PSO kinetic model fit the experimental data the best. The values of ΔH, ΔS, and ΔG were determined using the temperature variations. The exothermic effect of the Sudan III dye adsorption process on CB is indicated by the positive value of ΔH, which points to a physical adsorption mechanism. It may be concluded that CB can function as an efficient and economical adsorbent for the removal of Sudan III dye from aqueous solutions based on the variables and results obtained.

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Conflict of interest

The authors declare no conflict of interest

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