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Snail Shell (*Rostellariella*) as a Low Cost Adsorbent for Safranine Dye Removal from Aqueous Solution

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Abstract

In this paper, snail shell powder was used an adsorbent for safranine dye due to its low cost, high efficiency and high adsorption capacity. Experiments were conducted at a temperature of 298 K to find out the effect of pH, concentration, weight, ionic strength and equilibrium time. Also, the best conditions for the adsorption of Safranin dye were implemented at a weight of 0.0200 g of snail shell powder. The removal ratio was 96.09 % at a concentration of 9mg/L and 20 minutes as adsorption time at a temperature of 298 K. The objective of the study to analyze the equilibrium isotherms. The data collected from the experiments were analyzed by the three models of adsorption: Langmuir, Temkin, and Freundlich. The data were suitable for Freundlich isotherm. The calculated thermodynamic information of the process shows that the removal process occurs through an active exchange of random molecules.

Keywords: Adsorption Isotherms, Freundlich, Langmuir, Safranine Dye, Snail Shell and Temken.

Introduction

Clean and healthy water is one of the serious and important issues for human existence[1]. However, we note that agricultural water and wastewater impede the provision of easy water to surrounding communities and ecosystems. About eighty% of the population in the world faces actual problems related to fresh water materials and water protection under the infection of the available water assets through commercial and urban activities, so various methods have been applied to treat industrial wastewater such as advanced oxidation processes (AOPs), reverse osmosis, sedimentation, ion exchange, adsorption, ozone, filtration with coagulation process[2]. However, most of these processes require very high operational and capital costs[3]. Dyes are basic compounds and are commonly used in many production functions such as papermaking, plastics, and cosmetics, and about 15% annually of dye production is dumped into major water sources as pollution[4, 5]. Dyes are called toxic organic substances of low biodegradability and play the main function in a few environmental problem as aesthetic pollution, eutrophication and perturbations of the aquatic gadget[6] .Some aromatic organic dyes are extremely hard-to-degrade and have toxic natures as exemplified by the cationic dye safranine [7] . In addition, the presence of dyes as pollutants within aquatic assets can reduce the large amount of water responsible for mutagenic results and carcinogens that may also afflict humans and the surrounding natural world[8] . The snail shell has got the same primary construction as different Mollusk shells. It includes 3 layers, which bestow it enough energy to withstand the external conditions

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and shocks that can be uncovered to it [9] . So the current research offers low fee, lively and effective, had that would be hired in the remedy the waters of lakes and river. Snail shell powder has been extensively utilized in removal a few dyes like CBB-G250 [10], and azure B [11]. The aim for this work is to study removing the active Safranine dye using adsorption method on snail shell powder.

Materials and Methods

Adsorbate (Safranine Dye):

Safranine (SF) is a cationic dye widely used in industrial sectors [12, 13]. Prepare a safranine dye solution with a concentration of 100 ppm by dissolving 0.1 g of safranine in 1000 (mL) of distilled water. The properties of safranine dye shown in Table.1. Its chemical composition is shown in Fig 1

Table 1. Properties of safranine dye.[14]

IUPAC Name	3,7-diamino-2,8-dimethyl-5-phenylphenazinium Chloride
Formula	$C_{20}H_{19}ClN_4$
Source	HIMEDIA
Aqueous solubility	Soluble
Molecular Weight g/mol	350.84
color	Red
λ max	520 nm

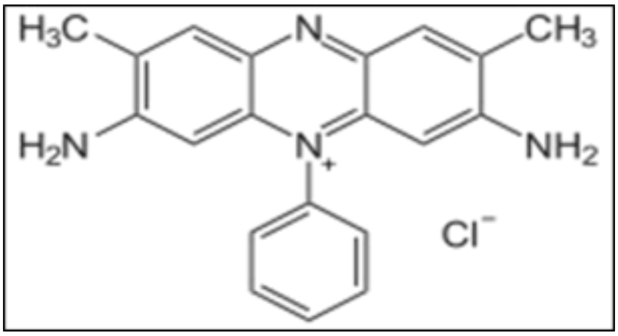

Figure 1. The chemical structure of safranine [15]

Adsorbent Surface (Snail Shell)

Snail shell was turned into amassed from its supply (seaside of Euphrates River)River .It was washed cautiously by means of the tap water and distil water to take away the contaminations form its surface. Then it was dried in the oven at 100 °C for twenty-four hours then, the dried materials was ground to obtain fine particles through an electric grinder into finer particles. The ground powder was sifted using a laboratory sieve whose size is 75 microns and stored in a sealed package for use for this research [16].Chemical analysis, as shown in Table 2.

Table 2. Chemical analysis of snail shell

NO	Molecular structure	(X-Ray) Analyzer %
1	CaO	52.70
2	SiO_2	2.40
3	Al_2O_3	0.68
4	Fe_2O_3	0.44
5	MgO	1.5
6	SO_3	0.28
	Other organic compounds	42

Batch Adsorption Experiments:-

All experiments were carried out at a temperature 298K. Safranine dye solution diluted 9 ppm containing 0.0200 g of snail shell powder (absorbent) put in a thermostat water bath shaker, 140 rpm. The filtered solution containing the unabsorbed dye was analyzed by a UV-Visible Double beam spectrophotometer -1800 at the wavelength equal to λ_{max}. The effect of contact time, adsorbent dose, ionic effect, pH, and temperature were studied. The expression $\frac{X}{m}$ defined as the amount of adsorbate in (mg) present in a given mass of adsorbent (g).

$$Re\% = \frac{(C_\circ - Ce)}{C_\circ} \times 100 \ \ldots\ldots..1$$

$$Q_e = \frac{(C_\circ - Ce)V}{m} \ \ldots\ldots\ldots\ldots2$$

Where:-

RE%: is the percentage of removal, $q_e = \frac{X}{m}$ (mg/g) is the amount of adsorbate adsorbed per unit mass of adsorbent surface, V: Volume of the solution (L), C_0: initial safranine concentration (mg/L), C_e: equilibrium equilibrium dye concentration in solution, m: mass of the adsorbent surface [17].

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Results and Discussion

Characteristics of Surface Adsorbent

The surface (snail shell) was diagnosed Using FT-IR, AFM and SEM techniques, as shown in Figs 2,3 and 4, respectively.

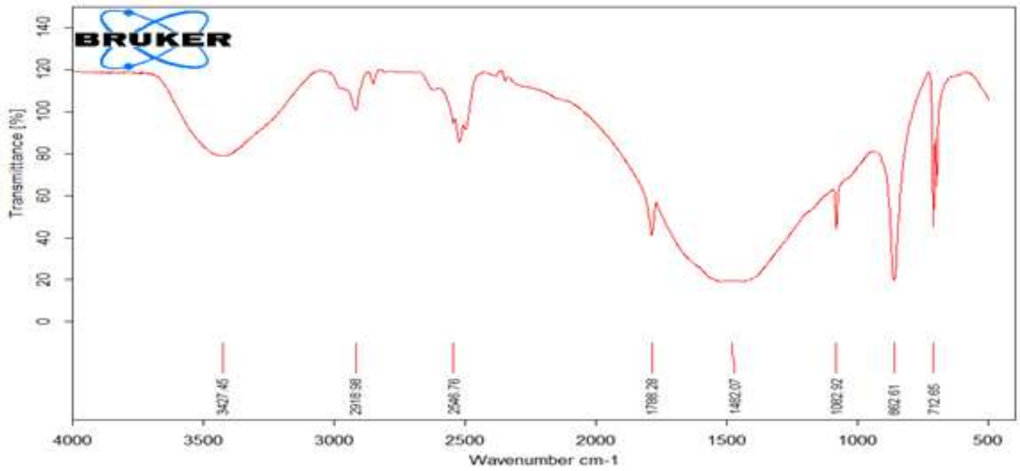

Figure 2. FT-IR analysis for adsorbent surface (snail shell powder)

From the results, the FT-IR spectrum (Fig 2) of the surface of the snail shell powder with a pointed fang that shows presence of the main absorption bands where we notice the absorption band at 3427.45 cm^{-1}, which results from the vibrational frequency of the hydroxyl groups, which belongs to $Fe(OH)_3$, $Al(OH)_3$ group, and the absorption band is at 712.65 cm^{-1} , which belongs to the bond in the Si-O group. The spectrum also showed an absorption band at 699.69 cm^{-1}, which returns to the absorption Fe-O group. The spectrum also showed an absorption band at 862.61cm^{-1}, which represents the absorption of the group Al-O, these chemical groups represent the effective sites on which the adsorption process occurs[18].

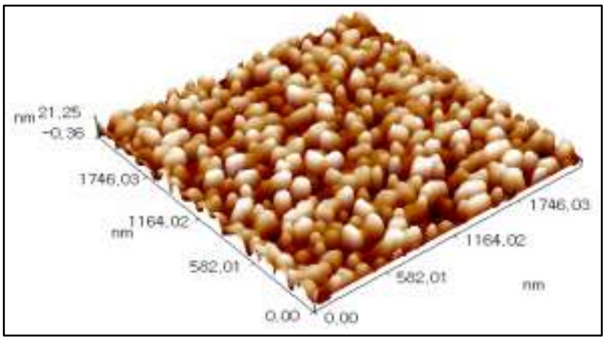

Figure 3. AFM analysis for adsorbent surface (snail shell powder)

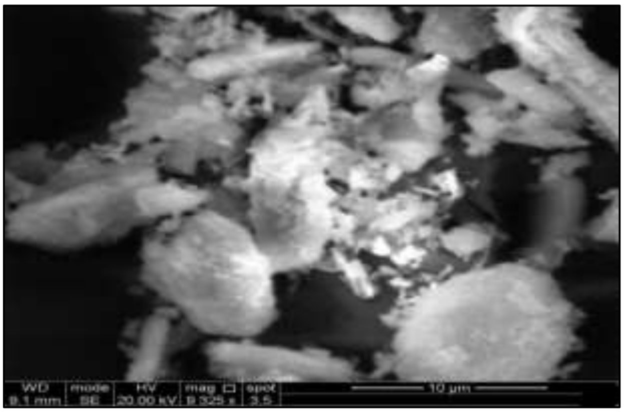

Figure 4. SEM analysis for adsorbent surface (Snail Shell powder)

AFM is the important and common technology which acts as an instrument for magnification, measurement diagnosis. It's employed in the realm of nanotechnology for example way the topography of surfaces. We notice from Fig 3, a three dimensional image, the surface snail shell during which the layers or particles are distributed, the surface snail shell appears irregular, interspersed with gaps.

The scanning electron microscope (SEM) was also used, and the surface of the snail shell powder was diagnosed, where Fig 4 shows spherical shapes formed from well-correlated clusters in terms of size and shape.

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Contact Time:

The contact time of safraninre dye was studied on snail shell at a temperature of 298 K[19]. The experiments were conducted at different times 5-120 minutes, at a concentration of 9 ppm of safranin.0.0200 grams of snail shell. The results shown in Fig 3 showed that the best equilibration time is 20 min, where it gave the best removal percentage of safranin dye (96.09%). So time 20min. was used for all other experiments.

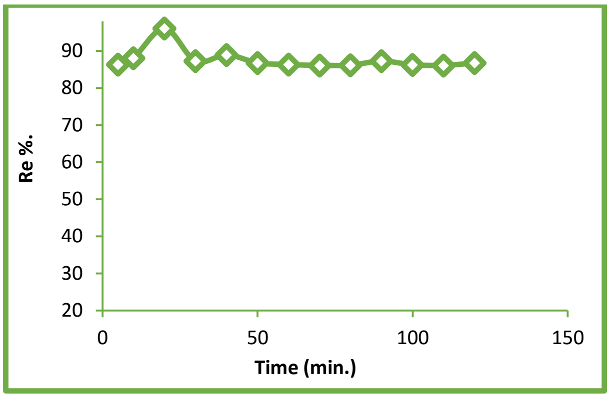

Figure 5. Effect of Contact time on adsorption of safranin dye on the adsorbent surface (snail shell)

Adsorbent Dose:

In general, the safranine dye removal percentage increases with the increase of the adsorbent dosage. Therefore, a group of different weights of the adsorbent material 0.005-0.09 g was tested. The results we obtained indicate that the elimination rate increases as the weight of the adsorbent surface Snail shell increases because of the surface area of the increasing absorbent material. So the removal percentage reached constant value, which is the saturation of the active sites of snail shell, and therefore 0.0200 g was chosen as the best weight for the adsorbent.

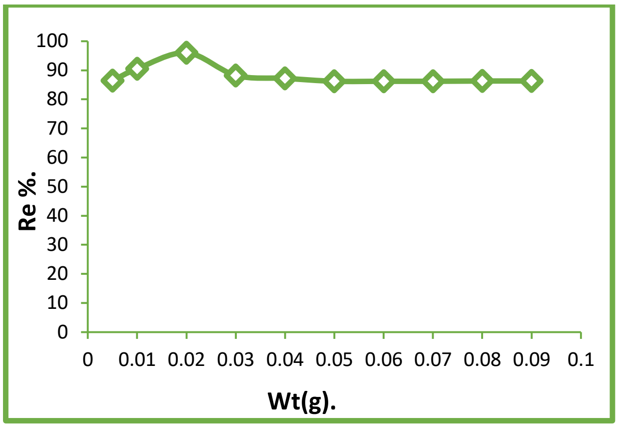

Figure 6. Effect of adsorbent dose on adsorption of safranin dye on the adsorbent surface (snail shell)

pH value:

pH effects on adsorption process[19]. Therefore, safranine solutions were prepared at different concentrations and pH 2-12 using 0.5 N HCl and 0.5 N NaOH. Fig 7 shows that the best removal percentage occurred at pH 8[20]. The result shows that the lowest value of removal in pH 2, that the ratio of elimination increased with increases of the pH until it reached pH 8 [21].After pH 8, there was no noticeable increase, but there was decrease in the ratio of elimination. Therefore, acid function 8 was selected, approved, and fixed in other experiments.

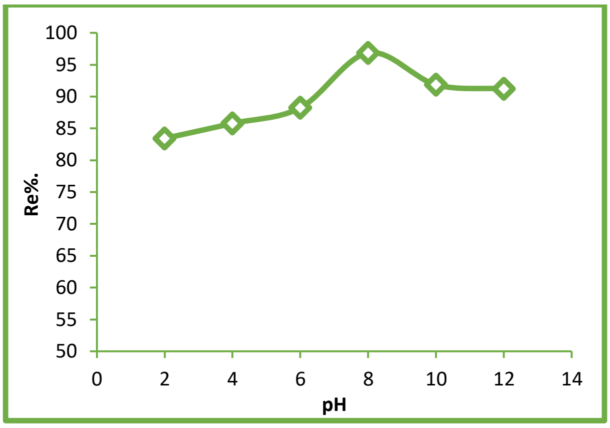

Figure 7. Effect of pH value on adsorption of safranin dye on the adsorbent surface (snail shell)

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Ionic Strength:-

Many concentrations of sodium chloride, potassium chloride, magnesium chloride and calcium chloride 0.0200-0.0700 M were taken to study the effects of the ionic strength on the ratio of the removal efficiency. Fig 8 shows impact the ionic strength on the removal efficiency ratio. The effect of the concentration of these salts on the dye indicates that the increase in the salt concentration corresponds to a decrease in the percentage of removal.

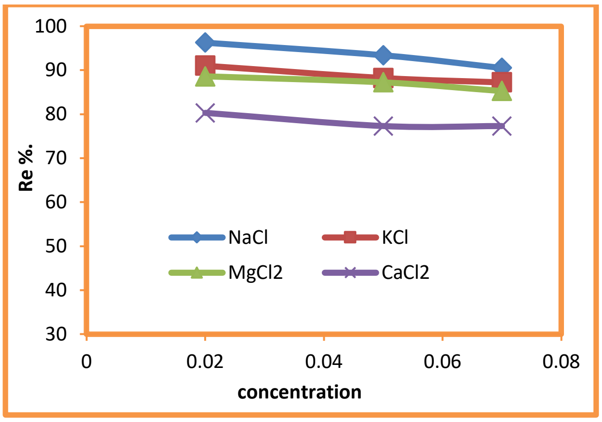

Figure 8. Effect of Ionic strength on adsorption of safranin dye on the adsorbent surface (snail shell)

Temperature

In this study, the thermodynamic functions ΔG^0, ΔH^0 and ΔS^0 were calculated for the adsorption of Safranine dye on Snail Shell in a range of temperatures 298-338 K using Eqs 3-6[22].

$$Keq = \frac{q_e * m}{Ce * V} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots 3$$

$$\Delta G^0 = -RT \ln Keq \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots 4$$

$$\Delta G^0 = \Delta H^0 - T\,\Delta S^0 \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots 5$$

$$\Delta S^0 = \Delta H^0 - \Delta G^0 / T \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots 6$$

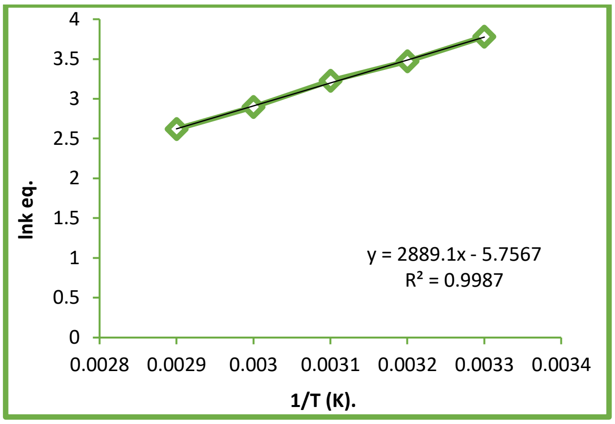

Figure 9. Effect of temperature on adsorption of safranin dye on the adsorbent surface at pH=8 (snail shell).

Table 3. The thermodynamic functions ΔGº, ΔHº, ΔSº for the adsorption of Safranine dye on the adsorbent surface (Snail Shell) at range of temperatures (298-338)K

Adsorption system	Temperature (K)	ΔGº (kJ. mol^{-1})	ΔHº (KJ. mol^{-1})	ΔSº (J.$mol^{-1}.K$)
	298	-9.3654		-0.0491
	308	-8.8884		-0.04912
SF	318	-8.528	-24.0199	-0.04871
	328	-7.9071		-0.04912
	338	-7.3661		-0.04927

The negative value of ΔGº means that the adsorption of dyes is spontaneous under these conditions. And the negative value of ΔHº meaning that the process is exothermic, this indicates only the adsorption process occurs and that the adsorbed molecules spread on the surface slows down their diffusion velocity. This leads to a decrease in the mutual reaction between the surface and the adsorbent molecule, and with the increase in temperature, it will separate the bonds between them, the negative signs of the ΔS^0 scores indicate the decrease in randomness (randomness of adsorbed molecules with adsorption) [23].

Adsorption Isotherm Study:

The adsorption mechanism and properties of safranine dye on the snail shell were further studied by obtaining an adsorption isotherm. At temperature ranges from 298 to 338 K .0200g from the adsorbing surface, 9 mg/L of the safranine dye, equilibrium time 20min, this figure proves that the

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adsorption amplitude increases with increasing the concentration of equilibrium for safranin. The isotherms obtained from the experiments were identical to S-curve in form at Giles' discretion [24].

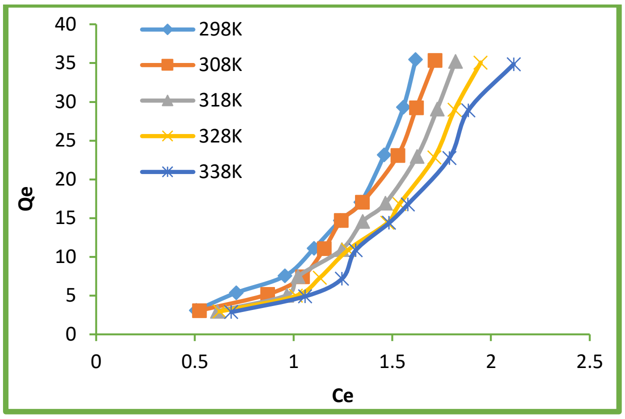

Figure 10. The adsorption isotherms of safranine dye at pH=8 using 0.02g from snail shell powder are shown at different Temperature.

Langmuir Isotherm Model:

This model is one of the most important isotherms, especially in the description and study of monolayer adsorption, as it assumes uniformity of energy on the surface of the adsorbent material. The Langmuir model is defined in the form of the following equations:

$$Q_e = \frac{a\, b\, C_e}{1+b\, C_e} \text{.................... } 7$$

Where the :- a and b are the Langmuir constants (L/mg)[22].

$$R_L = \frac{1}{(1+b\, Co)} \text{.................... } 8$$

The value of R_L indicates the category of the isotherm to be either unfavorable [25], linear [25] Irreversible [1]or favorable{ $0 < R_L < 1$} [26] .

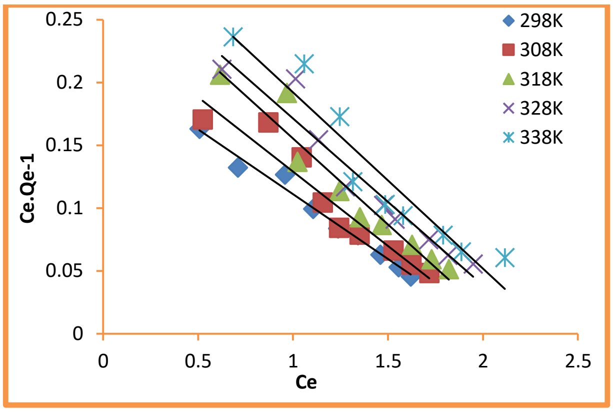

Figure 11. The Langmuir isotherms model for adsorption safranine dye over the studied Temperatures.

Freundlich Isotherm Model:

The Isotherm Freundlich equation Eq 9 was applied to the experimental data for safranine dye. The values of the Freundlich (n and K_f) constants were calculated from the slope and straight line segment obtained from drawing the relationship between logqe and logCe, respectively[27].

$$\log(Qe) = \log(K_f) + \frac{1}{n}\log(Ce) \text{ ....9}$$

where: K_f L/mg and n are Freundlich constants [28].

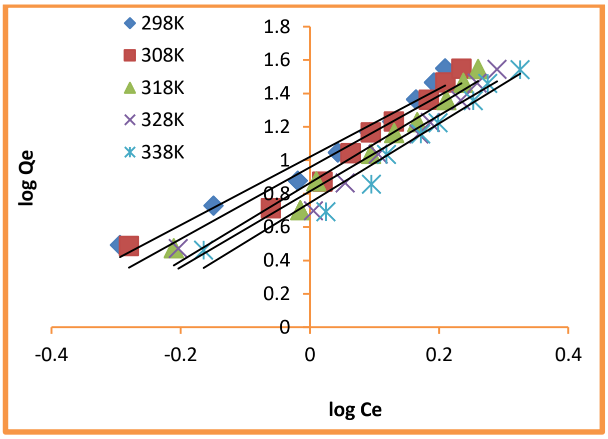

Figure 12. The Freundlich isotherms model for adsorption safranine dye over the studied temperatures.

Temkin Isotherm Model:-

The application of the Temkin isotherm was tested on the experimental data of (safranin - snail shell) adsorption system Eq 10, and from drawing the relationship between qe vs. lnCe, and from the slope and section of the obtained straight lines, the values of the constants B and A_t were calculated, respectively[29].

Q_e= B lnA_t + B lnC_e ………………...10

where:- A_t , B is a Temkin constant[28].

Then the isotherm constants (a, b, RL) for Langmuir and (n, Kf) were calculated for the Freundlich model, as well as the Temkin model (B, AT) with linear correlation coefficients as shown in Table 4.

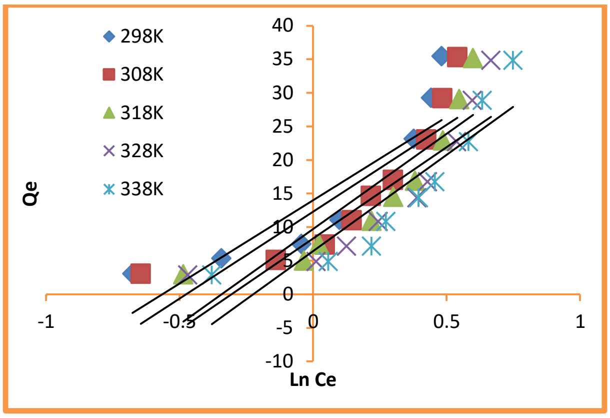

Figure 13.The Temkin isotherms model for adsorption safranine dye over the studied temperatures.

Table4. The values of the Langmuir, Freundlich, and Temkin parameters of adsorption isotherms At (298-338) K

	Temperature (K)	**Isotherm**									
		Langmuir				Freundlich			Temkin		
		- a (mg/g)	**- b (mg/L)**	R^2	**- RL**	K_f	**n**	R^2	**B**	A_T	R^2
SF	298	9.6899	0.4815	0.9823	0.2717	10.4737	0.4905	0.9617	24.785	2.6387	0.7556
	308	8.4317	0.4791	0.9186	0.2971	9.0677	0.4667	0.9404	25.91	2.5091	0.7458
	318	7.3099	0.4680	0.9343	0.2638	7.3146	0.4272	0.9635	28.69	2.2723	0.7847
	328	7.5358	0.4365	0.9351	0.2620	6.5132	0.4402	0.9537	27.144	2.1174	0.7672
	338	7.1326	0.4216	0.9035	0.3108	5.5628	0.4212	0.9601	28.687	1.8598	0.8026

The values of the constant (a mg/g) in the Langmuir equation, which represents a constant associated with the maximum adsorption capacity, where the adsorption capacity is better the higher the value of the constant. As for the value of the constant (b mg/L), it is related to the adsorption energy.

The values of the constant K_f in the Freundlich equation are an approximate indicator of the adsorption capacity. As the value of n in the Freundlich equation, the greater it is, the more preferred it is in adsorption, because it is related to the method of attachment of the dye molecules on the adsorbent surface[28].

Conclusion

An experimental study showed that snail shell powder could be a good choice for safranine dye removal. The safranine dye was removed well, where the percentage was 96.09%, through the stability of each of the following: Adsorbent dose 0.0200 g, safranine concentration safranine concentration 9 ppm, contact time 20 minutes and pH 8 at a temperature of 25 °C, Therefore the snail shell can be used as an effective adsorbent due to its high adsorption capacity, as well as its low cost.

Functions of thermodynamics showed the adsorption for safranine dye pigment using surface snail shell it's a spontaneously isothermal reaction.

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Authors' Declaration

- Conflicts of Interest: None.
- We hereby confirm that all the Figures and Tables in the manuscript are ours. Furthermore, any Figures and images, that are not ours, have been included with the necessary permission for re-publication, which is attached to the manuscript.
- Ethical Clearance: The project was approved by the local ethical committee in University of Kerbala.

Authors' Contribution Statement

F. B. Z. wrote a part of the manuscript and collected the samples. M. A. wrote another part of manuscript and interpretation the data. E.T.K. analyzed all Results. All Authors read the manuscript carefully and approved the final version.

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صدفة الحلزون (ذو الناب المدبب) كممتز منخفض التكلفة لإزالة صبغة السفرانين من محاليلها المائية

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الخلاصة

في هذا البحث ، تم استخدام مسحوق قشرة الحلزون كمادة مازة لصبغة السفرانين بسبب تكلفته المنخفضة وكفاءته العالية وكذلك قدرته العالية على الامتصاص. أجريت التجارب في درجة حرارة 298 كلفن لمعرفة تأثير الأس الهيدروجيني ،الوزن ،القوة الأيونية، زمن التوازن ودرجة الحرارة. كما تم تنفيذ أفضل الظروف لامتزاز صبغة السفرانين بوزن 0.0200 جم من مسحوق قشرة الحلزون. كانت نسبة الإزالة 96.09٪ بتركيز 9.0000 ملجم / لتر و 20 دقيقة كوقت اتزان عند درجة الحرارة 298 كلفن. الهدف من الدراسة هو تحليل ايزوثيرم متساوي درجة حرارة . تم تحليل البيانات التي تم جمعها من التجارب وفق نماذج الامتزاز الثلاثة Langmuir :و Temkinو .Freundlich كانت البيانات مناسبة لتساوي درجة حرارة .Freundlich توضح المعلومات الديناميكية الحرارية المحسوبة للعملية أن عملية الإزالة تحدث من خلال التبادل النشط للجزيئات العشوائية.

الكلمات المفتاحية: ايزوثيرمات الامتزاز, فريندلش, لانكماير, صبغة السفرانين, صدفة الحلزون, تكمن.