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Iraqi Porcelanite Rocks for Efficient Removal of Safranin Dye from Aqueous Solution

Ali Hussein Chafat [1] ***Muneer A. Al-Da'amy*** [1] ***Eman Talib Kareem*** [2*]

[1]Department of Chemistry, College of Education for pure science, University of Kerbala, Iraq.
[2]Department of Chemistry, College of Science, University of Kerbala, Iraq.
*Corresponding author: iman.talib@uokerbala.edu.iq
E-mail addresses: ali.chafat@s.uokerbala.edu.iq, moneer.a@uokerbala.edu.iq

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Abstract:

This research includes a study of the ability of Iraqi porcelanite rocks powder to remove the basic Safranine dye from its aqueous process by adsorption. The experiments were carried out at 298Kelvin in order to determine the effect of the starting concentration for Safranin dye, mixing time, pH, and the effect of ionic Strength. The good conditions were perfect for safranine dye adsorption was performed when0.0200g from that adsorbed particles and the removal max percentage was found be 96.86% at 9 mg/L , 20 minutes adsorption time and at PH=8 and in 298 K. The isothermal equilibrum stoichiometric adsorption confirmed, the process data were examined by Langmuir, Freundlich and Temkin adsorption equations at different temperatures (298-338)K.The results of the adsorption were good suited for Freundlich and Langmuir Isotherm more than Temkin. Thermodynamic data found for adsorption processes ΔG^0, ΔH^0, ΔS^0 indicates this is the removal step is through the adsorption operations which is spontaneity, exothermic and indicates a decrease in the randomness of that adsorbed dye molecules. Isotherms obtained from the experiments were identical to (S-curve) in form at Giles' discretion.

Keywords: Freundlich Isotherm, Langmuir Isotherm, Porcelanite Rocks, Safranin dye, Temkin Isotherm

Introduction:

The water of rivers and lakes is polluted as a result of the use of large quantities of dyes in multiple and different industries, where this pollution can be considered one of the earliest things dangerous to society and the environment [1]. For a long time and over wide periods of time, dyes of all kinds organic and inorganic have been used in a variety of applications and fields biological medical applications, printing, cosmetics, anti-corrosion coatings and leather, where dyes were classified according to what was mentioned above into safe, dangerous for humans and harmful [2]. Because of many uses, there is usually a lot of precipitation of colored residues in the waters of lakes and rivers. There is the toxic effect, which prevents or forms a barrier to the passage or access of sunlight to the organisms that live in the water, where a dark layer is formed on the surface of the water. These negatives and effects are a stumbling block to a very important process in the ecosystem which is photosynthesis [3, 4].These dyes contain on complex molecular aromatics structures that which make them more stable and difficult to degrade [5].Usually dye compounds have a complex structure with aromatic rings in the backbone, which adds a lot of stability and makes them difficult to degrade. According to previous studies, most organic dyes contain an azo group Egchromophores. In addition; it also contains minerals such as compounds, anthraquinones thiazines...etc. in order to increase its reactivity and solubility [6] .there is the ability to remove dyes from wastewater by using many methods, methods and techniques such as sedimentation, adsorption, oxidation by ozone, coagulation, flocculation, exchange ion and Biological methods. In this study, we will use the adsorption method because of its advantages, which are low cost, easy to implement, and environmentally friendly. The adsorption method using activated carbon as an adsorbent can be considered one of the most active types of surface adsorbents, but it has a cost [7,8]. The purpose for this study which is to study removal the active Safranine dye using adsorption method on Iraqi Porcelanite powder.

Materials and Methods:

Safranin Dye

All of the compounds we employ in our research are of global origin, and no purifying methods are applied to them. In this part of the experiment we prepared a solution of Safranin dye by dissolving 0.0200g of Safranin dye in 10 ml of water and good continuous shaking, and then we got a clear solution until the volume is complete 200 ml to get a solution of the dye that we are studying. Using a concentration of safranin dye 100 mg/L, these are the properties of safranin dye shown in Table.1. Its chemical composition is shown in Fig. 1

Table 1. Specifications of Safranine dye [9]

Iupac Name	3,7di amino 2,8Di methyl5phenyl Phenazinium Chloride
Molecular formula	$C_{20}H_{19}ClN_4$
Molecular Weight	350.85g/mol
λ max	530nm
Generic Name,	(C.I)basic Red2

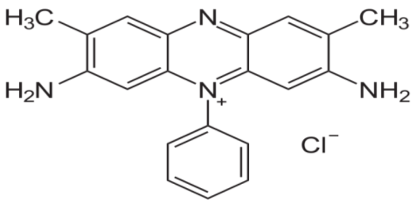

Figure 1. Structural formula ForSafranin Dye [10]

The adsorbent material (Iraqi porcelanite Rocks)

Natural Iraqi Porcelanite rocks were obtained from the ministry for industry and minerals, the General Company for geological survey and mining. These rocks are sprayed and washed using distilled water in order to remove unwanted substances, impurities and get rid of water-soluble substances from salts and others.The material is placed in a thermal oven at a temperature of 110 °C in order to drying the sample after washing. Then, the dried materials ground to obtain fine particles through an electric grinder into finer particles. The powder, which was ground, is sift using a laboratory sieve whose size is (75 microns) and stored in a sealed package for use for this research [11]. Chemical Analysis, as reported in Table. 2.

Table 2. Chemical analysis of Iraqi Porcelanite rocks

NO	Molecular structure	(X-Ray) Analyzer %
1	SiO_2	74.0300
2	CaO	5.6203
3	MgO	3.6501
4	Al_2O_3	3.6500
5	P_2O_5	1.1602
6	Fe_2O_3	0.9700
7	TiO_2	0. 1710
8	K_2O	0.1700
9	Na_2O	0.0607
10	Cr_2O_3	0.0201
11	SO_3	0.0091
12	MnO	0.0040
13	L.O.I	11.2001
	Sum	100.76

The information of the (FT-IR) spectrum in Fig. 2, showed the presence the main absorption bands, as we notice that the bands are between (3547-3466 cm^{-1}) resulting from the stretching vibrating for Hydroxyl groups. The absorption band in (1183 cm^{-1}) belongs to the bond at(Si-O)group. This spectrum like that showed an absorption band in (455 cm^{-1}), which represents the absorption of the Fe-O group. The spectrum showed an absorption band at (796 cm^{-1}), which represents that absorption for the Al-O group, that these chemical groups represent effective sites on which the adsorption process occurs due to their negative charge, which gives the ability of these groups to form chemical bonds or physical bonds because they contain an electron pair or electrostatic charge can participate in the formation of a physical bond or a chemical bond [12]

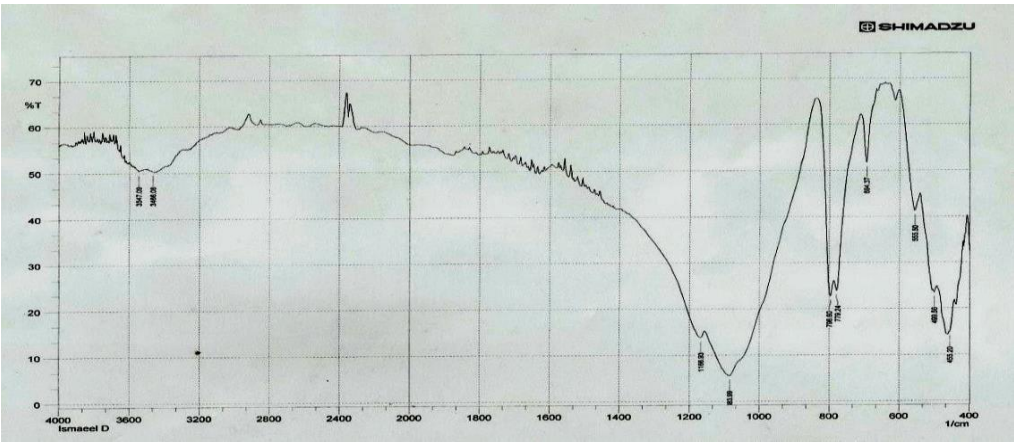

Figure 2. The FT-IR spectrum of Iraqi porcelanite Rocks.

The chemical analyzes(x-rd) of the rocks revealed that they consist of a mixture of quartz, and opal-CT, which is known to be an irregular overlap between the phases of Crestolite and tridamite-alpha, as well as the presence of some clay phases kaolin, alias and Smectite that are intertwined with silica [13]. Silica is also found in these rocks, with a high percentage of up to 74.0300%. Calcium oxide represents the main part of the secondary compounds accompanying the composition of these rocks, as the general rate of it is 5.6200%. There are other compounds, but in lower rates, they are titanium dioxide TiO_2, magnesium oxide, MgO and oxide Aluminum AL_2O_3 and some other Oxides [14].The Fig. 3, demonstrates of X-RD spectrum of the rocks.

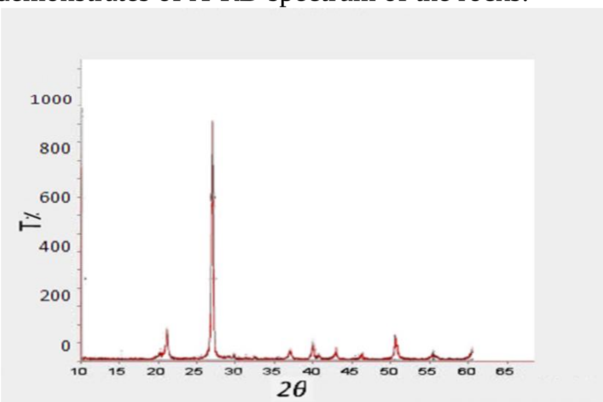

Figure 3. The X-RD spectrum of Iraqi porcelanite Rocks

The surface formation for Iraqi porcelain rocks were photographed using Scanning Electron Microscopy (SEM). Fig. 4, showed the porous nature of the surface and the wide surface area

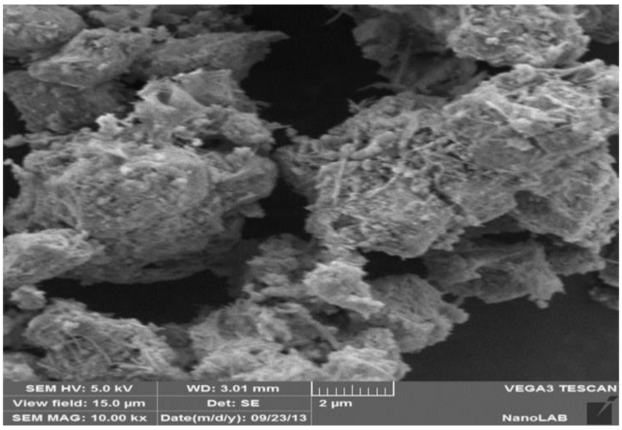

Figure 4. The SEM image of Iraqi porcelanite Rocks.

AFM is the important and common technologies as an instrument for magnification, measurement diagnosis, It's employed in the realm of nanotechnology for example way the topography of surfaces is viewed using Nano measurements it's known mapped.We notice from Fig. 5, a three dimensional image the surface Iraqi porcelanite Rocks during which the layers or particles are distributed appears irregular, interspersed with gaps.

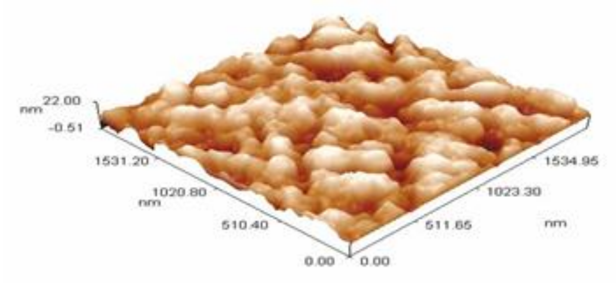

Figure 5. AFM image of the surface of Iraqi porcelanite Rocks

Experimental part for adsorption

Studies have been conducted by using 0.0200g adsorbent surface weight, 25ml safranin dye solutions 9mg/L, 150rpm through making use of water bath (thermal shaker). In each solution, the supernatant contain dye was examined by UV-Vis 1800 dual-beam digital a device with a wavelength that corresponds to λ_{max}.the effects of contact time, surface weight, ionic strength, pH and temperature were investigated.

$$\text{Removal \%} = \frac{C_0 - C_e}{C_0} \times 100 \ldots\ldots 1$$

Where

C_O, C_e Indicate the starting concentration and equilibrium concentration of residual dye mg/L straight [15].

Results and Discussion:

Contact Time Impact

By examining the relationship between contact time and the efficacy of removal Safranin using Iraqi porcelanite rocks the equilibrium time was confirmed, as seen in the Fig. 6.The end resulting is indicates time of equilibrium reached after 20min of operation time.

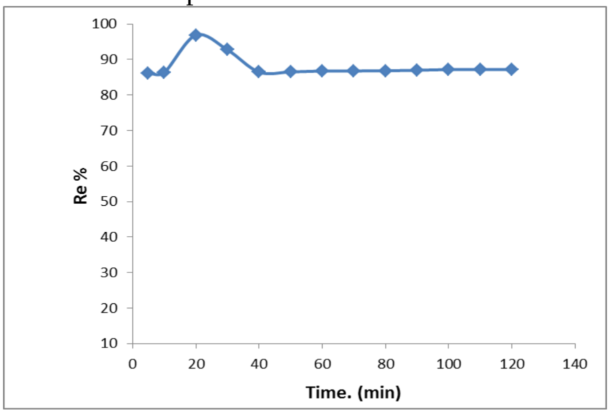

Figure 6. impact contact time in adsorption Safranin using Iraqi porcelanite Rocks at Temp. 298 K, Conc of dye 9mg/L, the adsorbent's weight 0.0200g

Impact weight of the adsorbent

The actual experiments were conducted using a range of weights ranging from (0.0050-0.0900) g, the dye concentration was initially 9 mg/L, and the temp at298K Fig. 7. The results we obtained indicate that the elimination rate is increases as the weight of the adsorbent surface porcelanite rocks 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 porcelain rocks, and therefore 0.0200 g was chosen as the best weight for the adsorbent.

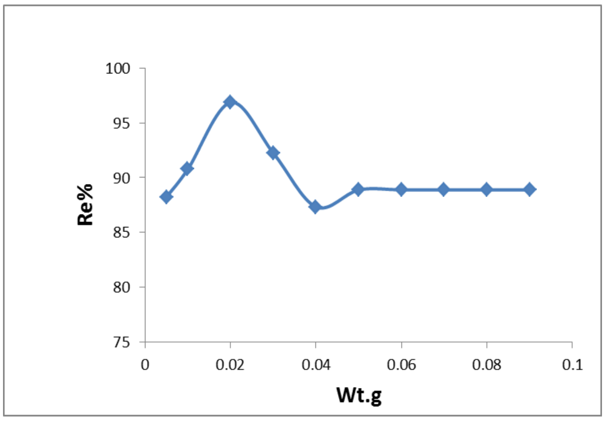

Figure 7. Impact weight of the adsorbent of Safranin using Iraqi porcelanite rocks Temp. 298K, Conc of dye 9mg/L

Impact of acid function

To study the impact of Acid function on removal rate, range of acid function values (2, 4, 6, 8, 10, and 12) were selected. 9mg/L was taken, which is the dye concentration at the start, then pH arrangement was taken as in the above range using 0.01N of HCl and NaOH. 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. 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. Fig. 8, shows this process.

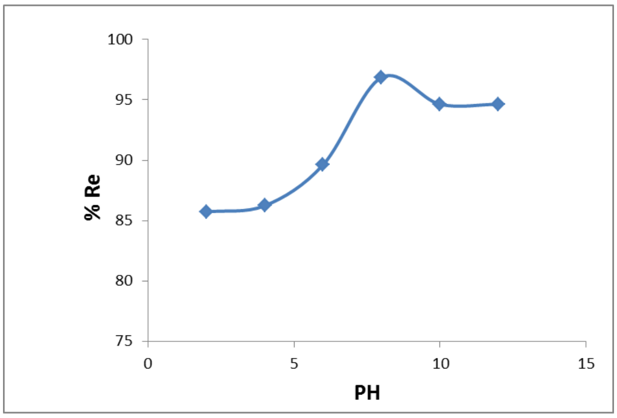

Figure 8. Impact of acid function in adsorption of Safranin using Iraqi porcelains rocks at Temp.298K, Conc dye 9mg/L, the adsorbent's weight 0.0200g

Impact of ionic strength.

Different concentrations of NaCl, KCl, $MgCl_2$ and $CaCl_2$ (0.0200-0.0700M) were chosen to research and show impact the ionic strength on the removal efficiency ratio, The effect of the concentration of these salts on the dye is, as the increase in the salt concentration corresponds to a decrease in the percentage of removal as shown in Fig. 9.

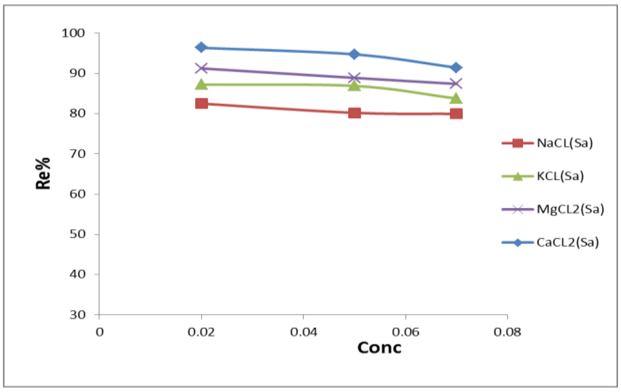

Figure 9. Impact the ionic straight in adsorption of Safranin using porcelanite rocks at Temp. 298 K, Conc dye 9mg/L, the adsorbent's weight 0.0200g

Impact the Temperature

The study of impact of change in temperature has been explained to show and understand the adsorption process's nature. Table. 3 contains information for adsorption, Free energy to Gibbs (ΔG^0), the term enthalpy (ΔH^0) and entropy (ΔS^0) which are found as Eqs. 2,3,4,and 5.[16]

$$Keq=\frac{Q_e\, m}{C_e\, V} \dots\dots\dots\ 2$$

Where Keq is the equilibrium constant for the adsorption process at each temperature, Qe (mg/g) is the amount of dye adsorbed at equilibrium (adsorbent capacity), Ce(mg/L) the concentration of safranin dye at equilibrium, V and m is the volume of the solution (L) and m the adsorbent mass (g) respectively. Fig. 10, show Vant Hoff equation for the adsorption of Safranin dye on a porcelanite surface

$$\Delta G^0 = -RT\ln Keq \dots\dots\dots\ 3$$

ΔG^0: is a Gibbs energy change (kJ/mol), R: is the ideal gas constant (8.314 J/mol. K)
T: is the absolute temperature (K)

$$\ln Keq = \left(\frac{-\Delta H^0}{RT}\right) + Conc \dots\dots\dots\ 4$$

$$\Delta S^0 = \frac{\Delta H^0 - \Delta G^0}{T} \dots\dots\dots\dots\dots\dots 5$$

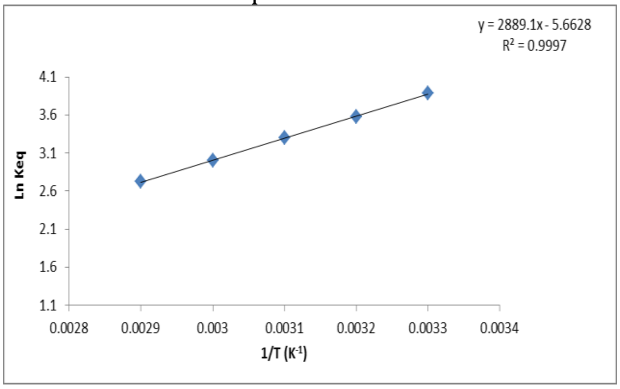

Figure 10. draw the Vant Hoff equation for the adsorption of Safranin dye on a porcelanite surface

Table 3. Function values ΔG^0, ΔH^0 and ΔS^0 in Safranin using porcelanite Rocks (298-338K)

	Tempe. Kelvin	ΔG^0 kJ/mol	ΔH^0 kJ/mol	ΔS^0 kJ/mol.K
Safranin	298.	-9.6132	-24.0199	-0.0483
	308	-9.1445		-0.0482
	318	-8.7106		-0.0481
	328	-8.1798		-0.0482
	338	-7.6472		-0.0484

ΔG^0 and ΔH^0 values that are negative imply that the adsorption of Safranin dye using surface the porcelanite Rocks has been a spontaneous exothermic process [17, 18] while the ΔS^0 scores indicate the decrease in randomness (randomness of adsorbed molecules with adsorption) [19]

Adsorption Isotherms

Figure. 11 hows isothermal Safranin dye adsorption on surface porcelanite Rocks at acid function 8, temperature (298-338)K,0.0200g from the adsorbing surface,9mg/L of the safranin dye, equilibrium time 20min,this figure proves that the 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.

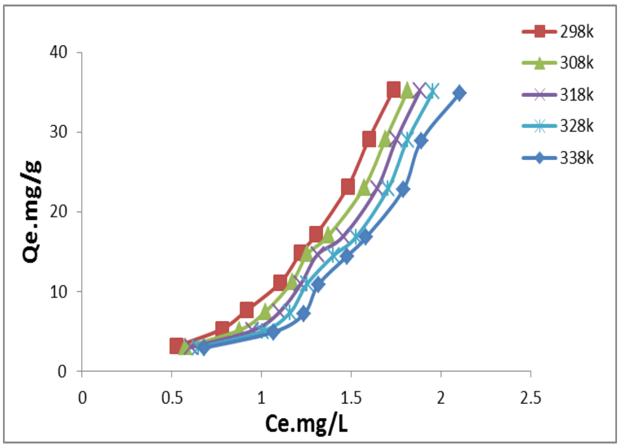

Figure 11. Isotherm adsorption for Safranin dye from aqueous solution using surface the porcelanite Rocks at different Tempe.

Langmuir Isotherm

Definition of Langmuir isotherm (Eq. 6) states the adsorption process takes place across homogeneous sites of the adsorbent.

$$Q_e = \frac{a\,b\,C_e}{1+b\,C_e} \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ 6$$

Where;

Q_e is defined as the quantity of safranin adsorbent at the time of equilibrium (mg/g) .

(a,b) are the constants of Langmuir.

$$RL = \frac{1}{1+bC_0} \ldots\ldots\ldots\ldots\ldots\ldots\ldots,\ 7$$

Where

RL Value explain that adsorption kinds is Irreversible(RL=0),preferred(0<RL<1), endothelial(RL=1)or unfavorable(RL >1) [20] .The a, b values are calculated from the slopes (1/a) and intercepts (1/ab) of linear plots of Ce /Qe versus Ce are shown in Fig. 12.

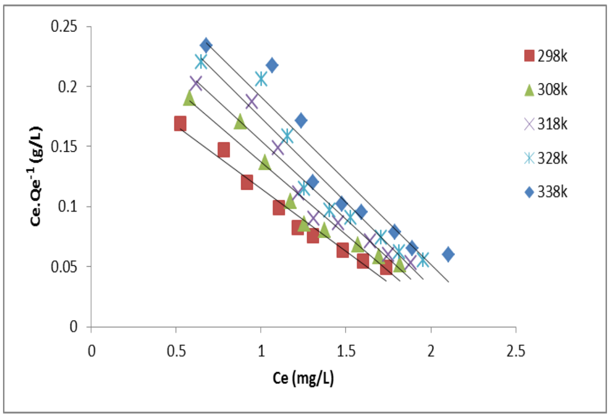

Figure 12. Langmuir isotherms for Safranin dye using surface the porcelanite Rocks at different Temp.

Freundlich Isotherm

Freundlich isotherm pattern of adsorption states that it is multi-layered adsorption across heterogeneous active sites Freundlich isothermal.

$$LogQ_e = Log\ kf + \frac{1}{n} LogC_e \ldots\ldots\ldots\ldots\ 8$$

Where k_f, n Freundlich's constants [21] . Fig. 13 show the applicability of equation well when plotting Log Qe against the values of Log Ce.

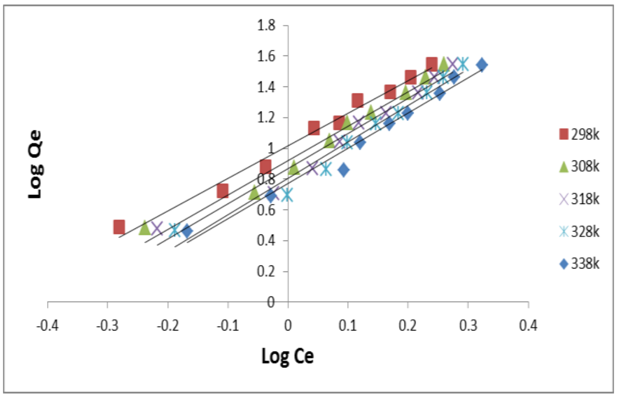

Figure 13. Freundlich isotherm Safranin dye using surface the porcelanite Rocks at different Tempe.

Temkin Isotherm

The Temkin isotherm is commonly used in the following way:

$$Q_e = \beta\ln At + \beta\ln C_e \ldots\ldots\ 9$$

Where At the binding constant for equilibrium, β connected to the adsorption heat [22], where The Temkin curves for Safranin dye adsorption are shown in Fig. 14. 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.

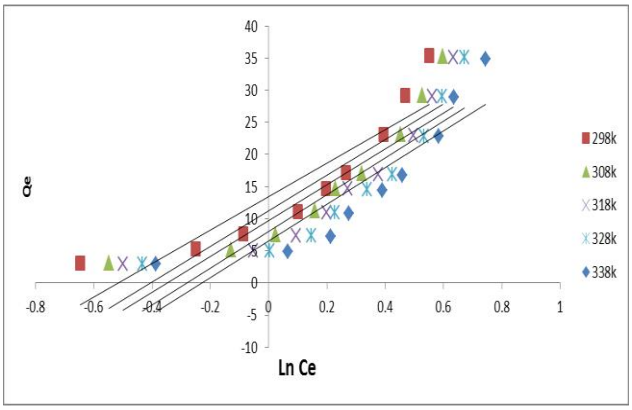

Figure 14. The Temkin isotherm for Safranin dye using surface the porcelanite Rocks at different Tempe.

Table 4. Adsorption isotherm values each of Langmuir, Freundlich, and Temkin at (298 –338) K

Temp. K		Langmuir isotherms				Freundlich isotherms			Temkin isotherms		
		a (mg/g)	b (mg/L)	R2	RL	Kf (mg/g)	n	R2	B	AT(mg/L)	R2
	298	-9.7371	-0.4727	0.9815	-0.4347	11.2591	0.5971	0.9557	14.0011	3.0579	0.6448
	308	-8.4818	-0.4649	0.9609	-0.4436	9.5499	0.5839	0.9522	14.3081	2.7636	0.6297
Safranin	318	-6.3452	-0.4929	0.9531	-0.4083	8.2871	0.5308	0.9624	15.6241	2.3579	0.6459
	328	-6.2035	-0.4657	0.9445	-0.4427	7.5111	0.5264	0.9582	15.5262	2.2536	0.6339
	338	-6.2267	-0.4401	0.9337	-0.4808	6.6743	0.5198	0.9575	15.6591	2.1049	0.6387

Conclusions:

The present research proved this porcelain rock is an adsorbent that can be used effectively to remove Safranin dye from its aqueous solution well reducing pollution the outcomes provide other benefits for waste water from industry treatment. Functions of thermodynamics showed the adsorption for Safranin dye pigment using surface porcelanite rocks it's a spontaneously isothermal reaction .The isothermal Freundlich model gave a good linear relationship with the adsorption for Safranin dye using surface the porcelanite Rocks in a time20 minutes in the range (298-338 K) and surface weight (0.0200 g).

Authors' declaration:

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

Authors' contributions statement:

A. H. Ch. 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 Authores read the manuscript carefully and approved the final version of their MS.

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صخور البورسيلانيت العراقية لازالة فعالة لصبغة السفرانين من محلولها المائي

علي حسين جفات[1] **منير عبد العالي الدعمي[1]** **ايمان طالب كريم[2]**

قسم الكيمياء، كلية التربية للعلوم الصرفة، جامعة كربلاء، كربلاء، العراق.
قسم الكيمياء، كلية العلوم، جامعة كربلاء، كربلاء، العراق.

الخلاصة:

يتضمن هذا البحث دراسة قابلية مسحوق صخور البورسلينايت العراقية على إزالة صبغة السفرانين القاعدية من محاليلها المائية عن طريق الامتزاز ، وأجريت التجارب عند 298 كلفن من أجل تحديد تأثير تركيز الابتدائي لصبغة السفرانين ، زمن التماس ، الدالة الحامضية ، وتأثير الشدة الأيونية. كانت الظروف الفضلى لامتزاز صبغة السفرين عند 0.0200 غرام من وزن السطح الماز ووجدت نسبة الازالة 96.86٪ عند تركيز 9 ملغم / لتر ، ووقت الامتزاز 20 دقيقة وعند دالة حامضية = 8 و درجة حرارة 298 كلفن . تم ايجاد ايزوثيرمات الامتزاز واختبار القيم العملية بتطبيق معادلات لانكماير وفريندليش وتيمكين عند درجات حرارة مختلفة (298-338) كلفن ، وكانت نتائج الامتزاز مطابقة لايزوثيرم فريندليش ولانكماير أكثر من ايزوثيرم تيمكين. تشير قيم الدوال الثرموديناميكية التي تم حسابها ΔG^0، ΔH^0، ΔS^0 إلى أن عملية الامتزاز تجري تلقائية وباعثة للحرارة وتشير إلى انخفاض عشوائية جزيئات الصبغة الممتزة. ايزوثيرامات الامتزازالتي تم الحصول عليها من التجارب جاءت مطابقة للشكل S حسب تصنيف جيليز .

الكلمات المفتاحية : ايزوثيرم فريندلش، ايزوثيرم لانكماير، صخور البورسلينايت، صبغة السفرانين، ايزوثيرم تيمكن .