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Adsorption of Methyl Violet Dye onto Cresson Seeds: Thermodynamic and Kinetic Studies

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The adsorption of methyl violet dye onto Cresson (*Lepidium sativum*) seeds also known as Garden cress as an adsorbent from its aqueous solution has been carried out. The effect of experimental coefficient parameters like connection time, primary concentration of adsorbate (methyl violet) and amount of adsorbent dose and temperature were evaluated to obtain the optimum condition for adsorption of methyl violet onto Cresson seeds using batch adsorption. The adsorption information were mathematically analyzed using adsorption isotherms like Langmuir, Freundlich and Dubinin-Radushkevich isotherms in order to study adsorption mechanism of methyl violet dye onto Cresson seed. The isotherms of adsorption were advanced and equilibrium data adjusted well to Freundlich isotherm model. The kinetic studies indicated that the process of adsorption followed the model of second-order kinetic. The thermodynamic parameters (ΔGº, ΔHº and ΔSº) were estimated and the obtained negative value of ΔGº indicate a spontaneous adsorption process, positive ΔHº value obtained designate about the endothermic properties of adsorption process and positive ΔSº value obtained during the adsorption indicated the increased randomness.

Keywords: Cresson seed, Methyl violet dye, Adsorption kinetics.

INTRODUCTION

Artificial dyes of great extent assortment and amount are used by various industries for colouring their eductors [1-3]. Plausible quantity of the dyes appended in the practicability to be left over semi-finished and finally find their way to water collection. Presence of dyes in a water collection position earnest cause of danger to the environment like the change in colour, nature of the water and makes it infelicitous for human exhaustion. The growing environmental problems caused by dye flowing out have driven plausible research exertions to decrease of dye wastewater. Methyl violet is in particular important because of its extensive appliance in antibacterial [4], temper inks, textiles, bacteria categorization and in paints [5,6]. The intake of methyl violet may cause excitation to the breathing apparatus and swallowing exemplary causes distress to the digestive system [7]. Furthermore, methyl violet dye is rebellious and hard to control to demean because of the existence of three aryl groups, which each one is bonded to N_2 atom that interplay with one or two CH_3 groups (Fig. 1) [8]. An extensive range of technologies had been developed for removal by artificial dyes with aqueous solutions to diminution their effect in the environment, inclusive membrane filtration processes [9], adsorption mechanics [10], clotting [11], developed processes of oxidation [12] and processes of ozonation [13]. Between the above-referred to techniques, sorption is higher quality to the different mechanisms in terms which have a low cost, elasticity, modesty of a plan, case of procedure and insensitivity to toxic environments [14]. The adsorption is one of the greatest qualified mechanisms can be valid for methyl violet dye removal [15,16] and the mercantile adsorbents as plant seeds are costing much, to examine is perform by some researchers for recognizing low-priced adsorbents [17].

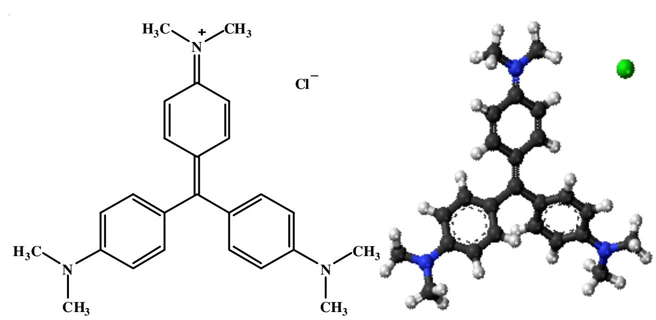

Fig. 1. Chemical and 3D structure of methyl violet dye

Various agricultural products have been considerably studied for removal of pollutants, these contain peat, wood, pine bark, peanut shells, plant seeds, wool, banana pith, compost and leaves [18]. Cresson seeds can be employed as an origin of agricultural based products. The seeds swell upon moistening and once swollen comprise of a rigid core with a cancellate swollen external layer. The gummous layer of blown seeds is a pectinous template, made up of plausible amounts of unesterified galacturonic acid with a large capaciousness of hydration [19,20]. The aim of this study is to use Cresson seeds as an adsorbent for the removal of methyl violet dye from aqueous solution and to study the effect of specific coefficients such as, initial concentration of methyl violet, adsorption efficiency, adsorbent dose and the temperature for maximum removal of methyl violet dye in its aqueous solutions and also pseudosecond-order kinetic models were used to fit the experimental data. Kinetic parameters, rate constants, equilibrium adsorption capacities, and related correlation coefficients for each kinetic model were calculated and discussed.

EXPERIMENTAL

Sorbent: Cresson seeds (*Lepidium sativum* seeds), brownish red in colour, were purchased from the local market, prepared as whole seed, the plants producing these seeds grown extensively in Iraq. The required quantity of seeds were washed with distilled water then allowed to swell for 30 min and directly used as adsorbent.

Sorbate: The commercial analytical methyl violet dye appearance green to dark-green powder (m.f.: $C_{25}H_{30}N_3Cl$) and m.w. 407.986 g/mol: λ_{max} was 590 nm, mono isotopic mass 407.212830 Da, EC Number 208-953-6, m.p.: 205 to 215 °C, soluble in water and ethanol, CAS Registry Number: 548-62-9 was used for the preparation of stock solution of 40 mg/L, by dissolving 0.01 g from methyl violet dye with 250 mL distilled water. A series of diluted solutions were prepared (30, 20, 10 mg/L) by fresh diluting of the stock solution, to compute the best λ_{max} using UV-visible spectrophotometer.

Methods: The experiments of adsorption were executed using batch equilibrium method [21]. Swollen seeds were taken in 250 mL round bottom flask containing 125 mL of methyl violet dye solution. The flasks were placed on a hot plate with magnetic stirrer (Bibby Strlintd, UK) and shaken at 150 rpm at 25 °C, Finally predetermined time intervals, adsorbent was dismissed by centrifugation with 150 rpm, the supernatant assayed using UV-visible spectrophotometer. Seeds uptake by the adsorbent at equilibrium (Q_e) was computed by eqn. 1:

$$Q_e = (C_0 - C_e)\ V/W \quad (1)$$

Q_e = amount sorbed (mg/g) of methyl violet dye, C_0 = initial concentration (mg/L) of methyl violet dye, C_e = equilibrium concentration (mg/L) and V = total volume of solution (L) and W = adsorbent used mass of Cresson seeds (g) [22,23]. Removal percentage or adsorption percentage (%R) were calculated using eqn. 2:

$$R\ (\%) = [(C_0 - C_e)/C_0] \times 100 \quad (2)$$

RESULTS AND DISCUSSION

Effect of adsorption parameters

Effect of contact time: Cresson seeds (0.5 g) mixed with 125 mL of initial concentration (7 mg/L) of methyl violet dye at 25 ºC for different time (5, 10, 20, 30, 40, 50, 60, 70 and 80 min) and centrifuged. Methyl violet concentration were determined as above and the contact time on adsorption of methyl violet is demonstrated in Fig. 2. The equilibrium was attained after shaking for 80 min, thus 70 min was accepted as optimum time for adsorption of methyl violet on Cresson seeds, additionally growth in contact time did not exhibit any increase in adsorption due to a saturation in a surface sites [24].

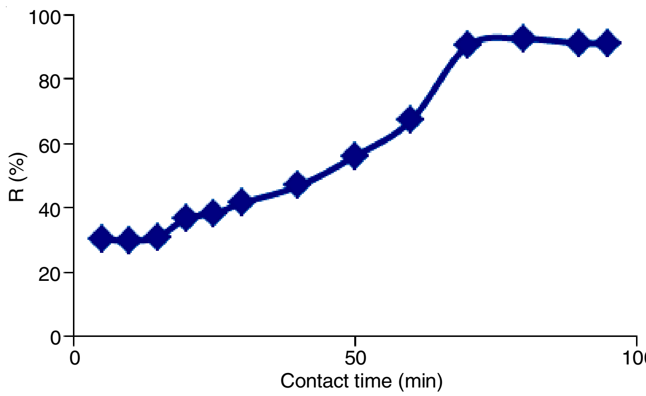

Fig. 2. Influence of contact time upon adsorption methyl violet dye onto Cresson seeds

Effect of adsorbent doses: A primary concentration methyl violet dye (40 mg/L) were used in conjunction with different amount of swollen Cresson seeds of (0.1, 0.3, 0.5, 1 and 1.2 g), the other parameters were kept constant; contact time 70 min, agitation speed 150 rpm; temperature 40 ºC. Methyl violet uptake was found to increase with increase in Cresson seeds dosage up to 1.2 g of seeds. Therefore, optimum Cresson dose was chosen as 1 g for the subsequent experiment as shown in Fig. 3.

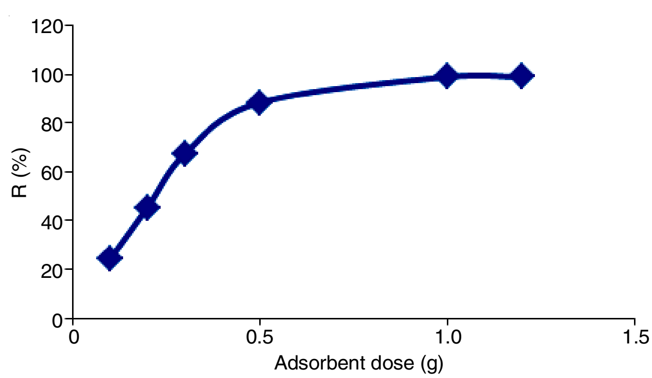

Fig. 3. Influence of adsorbent dose on adsorption methyl violet dye on to Cresson seeds

Effect of primary concentrations: Experiential evaluates for adsorption by different concentrations of methyl violet (3, 4, 7, 8, 9 and 10) mg/L are shown in Fig. 4. As the concentration of methyl violet increases, more and more sites of surface are covered at higher concentration, the capacity of adsorbent obtain consumed caused by non-availability of surface location [21].

Effect of temperature: Experiments were executed at different temperatures 25, 30, 35, 40 and 45 ºC in conjunction with the optimum other parameters, contact time 70 min, adsor-

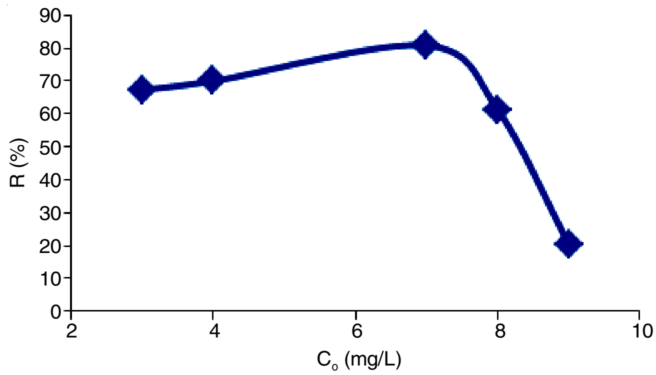

Fig. 4. Effect of primary concentration at adsorption process using methyl violet dye on to Cresson seeds

bent dose 1 g, agitation speed 150 rpm. Fig. 5 shows that at 40 °C, the maximum removal of dye can be obained, so this temperature was chosen for further experiments.

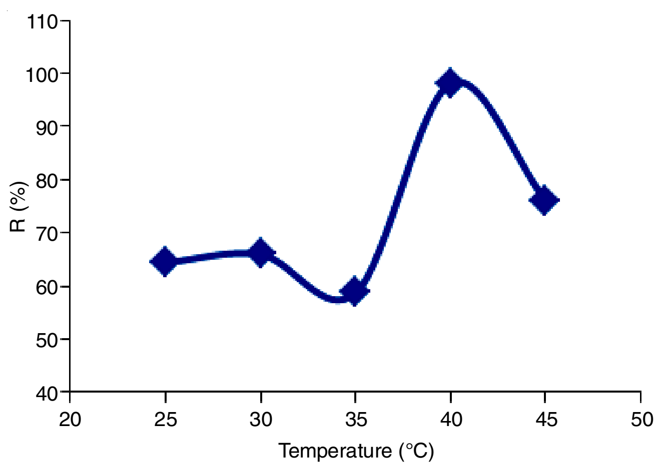

Fig. 5. Influence of temperature on adsorption of methyl violet dye onto Cresson seeds

Adsorption isotherms: To determine the adsorption capacity and potentiability for selecting the adsorbent for removal of methyl violet dye, several adsorption isotherms are required. So modelling of experimental data from adsorption processes is a very important means of predicting the mechanisms of various adsorption systems. For batch experiments executed, the most efficient state parameters chosen were; Cresson seeds dose 1g, contact time 70 min and agitation speed 150 rpm. Adsorption isotherm study was carried out by five different temperatures (25, 30, 35, 40 and 45 °C). The equilibrium values obtained are depicted in Table-1.

Langmuir adsorption isotherm: The Langmuir adsorption isotherm is valid for monolayer adsorption on to a level with a limited number of homogeneous situation [24]. It is founded on evaluation of adsorption identity, such as evenly plentiful adsorption, monolayer surface covering and no influence between adsorbed type [25,26]. In agreement with the Langmuir isotherm, the adsorption practicability can be express as:

$$\frac{C_e}{Q_e} = \frac{C_e}{Q_m} + \frac{K_L}{Q_m} \tag{3}$$

C_e (mg/L) is equilibrium concentration in solution of methyl violet dye, Q_e (mg/g) = adsorbed amount at equilibrium per unit weight, Q_m (mg/g) = the adsorption ability at the highest level denominated effective dissociation constant. The linear plot of C_e/Q_e *vs.* C_e proposed the suitability of Langmuir isotherm (figure not shown). The values of Q_m and K_L were computed by the slope and intercepts and are listed in Table-2. Langmuir constants relates to adsorption ability and rate of adsorption consecutively.

TABLE-2
LANGMUIR PARAMETERS OF ADSORPTION ISOTHERM WITH THEIR CORRELATION COEFFICIENT ON ADSORPTION OF METHYL VIOLET DYE ON CRESSON SEEDS

Temp. (K)	Q_m	K_L	R^2
298	0.275	9.676	0.963
303	0.478	0.630	0.878
308	0.479	2.762	0.831
313	1.103	0.352	0.840
318	2.109	0.167	0.795

Freundlich adsorption isotherm: Batch isotherm data fitted to the linear format with the Freundlich adsorption isotherm is commonly expressed by sequential equation [27].

$$\ln Q_e = \ln K_f + (1/n)\, C_e \tag{4}$$

where K_f and n are computed and the data are provided in Table-3. The values indicated the adsorption capacitance and adsorption strength consecutively. The results showed that n > 1, which indicates that adsorption is favourable as show in Table-4, favourable sticking of adsorbate to adsorbent [28]. The suitability of Freundlich isotherm indicating that

TABLE-1
EQUILIBRIUM PARAMETERS ON ADSORPTION OF METHYL VIOLET DYE ON TO CRESSON SEEDS

At 25 °C			At 30 °C			At 35 °C			At 40 °C			At 45 °C		
C_e	Q_e	C_e/Q_e	C_e	Q_e	C_e/Q_e	C_e	Q_e	C_e/Q_e	C_e	Q_e	C_e/Q_e	C_e	Q_e	C_e/Q_e
0. 033	0. 065	0. 505	0. 179	0. 064	0. 102	0. 027	0. 263	0. 102	0. 141	0. 052	2. 424	0. 130	0. 042	3. 076
0.062	0.075	0.744	0.224	0.062	1.468	0.170	0.116	1.468	0.131	0.047	2.787	0.1812	0.058	3.103
0.555	0.199	3.022	0.558	0.112	4.169	0.771	0.185	4.169	0.711	0.237	2.999	0.659	0.238	2.768
0.600	0.303	3.202	0.803	0.158	1.687	0.331	0.196	1.687	0.525	0.153	3.438	0.682	0.209	3.267
1.971	0.346	3.916	1.912	0.323	2.525	1.327	0.525	2.525	1.391	0.346	4.0179	1.399	0.379	3.684
2.110	0.454	4.867	1.925	0.291	4.494	1.591	0.429	4.494	1.951	0.383	5.094	1.632	0.485	3.367
1.810	0.304	5.950	2.109	0.272	2.266	1.156	0.510	2.265	1.150	0.316	3.636	1.626	0.458	3.654
2.170	0.276	7.876	3.129	0.315	9.331	3.000	0.369	9.331	2.320	0.514	4.518	0.950	0.300	3.166
1.590	0.199	7.969	1.701	0.185	2.176	1.129	0.519	2.176	2.388	0.543	4.402	2.871	0.662	4.340
3.626	0.276	14.038	2.131	0.251	3.006	0.714	0.237	3.006	0.741	0.182	3.071	1.434	0.407	3.526
3.990	0.269	14.805	4.330	0.363	7.458	2.506	0.352	7.458	1.911	0.471	4.124	2.591	0.572	4.532

TABLE-3
FREUNDLICH ISOTHERM MODEL PARAMETERS TO THE ADSORPTION OF METHYL VIOLET DYE ON CRESSON SEEDS

At 298 K		At 303 K		At 308 K		At 313 K		At 318 K	
ln C_e	ln Q_e	ln C_e	ln Q_e	ln C_e	ln Q_e	ln C_e	ln Q_e	ln C_e	ln Q_e
-3.404	-2.737	-1.720	-2.749	-3.144	-2.776	-1.958	-2.959	-3.144	-2.775
-2.891	-2.597	-1.496	-2.785	-1.772	-2.157	-2.032	-3.0533	-1.772	-2.157
-0.511	-1.162	-0.584	-2.187	-0.261	-1.688	-0.340	-1.439	-0.260	-1.688
-0.029	-1.193	-0.213	-1.845	-1.106	-1.628	-0.644	-1.879	-1.106	-1.628
-0.302	-1.063	0.648	-1.130	0.283	-0.643	0.330	-1.061	0.283	-0.643
0.747	-0.791	0.654	-1.229	0.255	-0.846	0.668	-0.959	0.255	-0.846
0.593	-1.190	0.746	-1.302	0.145	-0.673	0.139	-1.152	0.145	-0.673
0.775	-1.289	1.141	-1.156	1.203	-0.997	0.842	-0.666	1.203	0.998
0.464	-1.612	0.531	-1.686	0.121	-0.656	0.870	-0.616	0.121	-0.656
0.129	-1.356	0.757	-1.381	-0.337	-1.437	-0.299	-1.703	-0.337	-1.438
1.384	-1.311	1.312	-1.998	0.893	-1.045	0.648	-0.752	0.892	-1.044

adsorption by swollen Cresson seeds may be govern by physisorption. From the values of the regression coefficient R^2 (Table-4), Freundlich isotherm gave good and fitted better than Langmuir isotherm from the calculated results for experimental data R^2.

TABLE-4
FREUNDLICH ISOTHERM PARAMETERS OF ADSORPTION AND THEIR CORRELATION COEFFICIENT ON ADSORPTION OF METHYL VIOLET DYE ON CRESSON SEEDS

Temp. (K)	K_f	n	R^2
298	0.227	2.899	0.815
303	0.169	1.629	0.952
308	0.135	2.041	0.783
313	0.261	1.217	0.988
318	0.403	1.456	0.759

Dubinin-Radushkevich isotherm: This adsorption isotherm model was selected to calculate the quality porosity of biomass and the seeming energy of adsorption. The represented model can be expressed as follows:

$$Q_e = Q_s\, e^{-K_{ad}\varepsilon^2} \quad (5)$$

$$\ln Q_e = \ln Q_s - K_{ad}\varepsilon^2 \quad (6)$$

K_{ad} is the isotherm constant ($mol^2\ J^{-2}$), Q_s is the Dubinin-Radushkevich isotherm constant connected to level of sorbate sorption by sorbent level [29,30].

$$E\ (\text{sorption energy}) = 1/\sqrt{2K_{ad}} \quad (7)$$

$$\varepsilon = RT \ln [1 + 1/C_e] \quad (8)$$

The obvious energy (E) of adsorption from Dubinin-Radushkevich isotherm model can be calculated by using a relation given in eqn. 7 [31]. Tables 5 and 6 showed the high value of E = 5000 J mol^{-1} indicates that the adsorption process is physisorption.

TABLE-6
CALCULATED DATA OF DUBININ-RADUSHKEVICH ISOTHERM

Temp. (K)	K_{ad} ($mol^2\ J^{-2}$)	Q_s (mg/g)	E (kJ/mol)	R^2
298	2×10^{-8}	0.2272	5000	0.859
303	8×10^{-8}	0.2792	2500	0.860
308	3×10^{-8}	0.3745	4100	0.742
313	7×10^{-8}	0.4132	2700	0.921
318	6×10^{-8}	0.5091	2900	0.491

Lagergren kinetic rate equation: The equation of Lagergren kinetics has been mainly used for the adsorption of an adsorbate from an aqueous solution [32]. Lagergren is authenticated for the pseudo-first order rate equation for liquid-solid adsorption and summarized as in eqn. 9:

$$\ln (Q_e - Q_t) = \ln Q_e - K_1 t \quad (9)$$

The pseudo second-order equation as eqn. 10, which is applied to empirical data:

$$t/Q_t = 1/K_2\, Q_e^2 + t/Q_e \quad (10)$$

where Q_e and Q_t are the adsorption capacities at equilibrium and at (t) time (mg/g), respectively, K_1 (min^{-1}) is the adsorption rate constant for pseudo-first order [33,34] and K_2 = constant

TABLE-5
EXPERIMENTAL DATA OF DUBININ-RADUSHKEVICH ISOTHERM ON ADSORPTION OF METHYL VIOLET DYE ONTO CRESSON SEEDS

At 298 K		At 303 K		At 308 K		At 313 K		At 318 K	
ln Q_e	ε^2	ln Q_e	ε^2	ln Q_e	ε^2	ln Q_e	ε^2	ln Q_e	ε^2
-2.737	73170151	-2.7488	22549901	-2.776	66577894	-2.9599	29559650	-2.775	32654557
-2.5967	53252129	-2.785	18302064	-2.157	24398784	-3.0533	31437740	-2.157	24566648
-1.1617	5902756	-2.187	6693774	-1.688	4538839	-1.439	5215264	-1.688	5952397
--1.1931	3076678	-1.845	4116652	-1.628	12693844	-1.879	7692924	-1.628	5690521
-1.063	1879657	-1.130	1123131	-0.643	2068555	-1.061	1985137	-0.643	2030991
-0.791	923783	-1.229	1110768	-0.846	1559529	-0.959	1158447	-0.846	1595141
-1.190	1187618	-1.302	955793	-0.673	2547418	-1.152	2648691	-0.673	1604506
-1.289	881741	-1.156	488060	-0.997	452163	-0.666	869008	0.998	3611289
-1.612	1461358	-1.686	1356912	-0.6564	2638387	-0.616	827732	-0.656	62369
-1.356	364144	-1.381	939480	-1.437	5028452	-1.703	6936560	-1.438	1954698
-1.311	307022	-1.998	273986	-1.0435	773048	-0.752	1198358	-1.044	743912

TABLE-7
CALCULATED DATA OF LAGERGREN KINETIC RATE EQUATION ON ADSORPTION OF METHYL VIOLET DYE ON TO CRESSON SEEDS

At 298 K			At 303 K			At 308 K			At 313 K			At 318 K		
Time (min)	ln (Q_e – Q_t)	t/Q_t	Time (min)	ln (Q_e – Q_t)	t/Q_t	Time (min)	ln (Q_e – Q_t)	t/Q_t	Time (min)	ln (Q_e – Q_t)	t/Q_t	Time (min)	ln (Q_e – Q_t)	t/Q_t
5	-2.392	46.664	5	-3.668	57.654	5	3.156	68.399	5	-2.380	34.602	5	-2.553	32.272
10	1.961	17.289	10	-4.061	99.255	10	1.703	203.045	10	-2.080	89.102	10	-3.013	54.444
20	-1.598	161.079	20	-2.526	100.099	20	0.887	210.084	20	-2.782	114.187	20	-3.477	99.056
30	-2.828	215.123	30	-5.116	282.353	30	-6.812	262.467	30	-4.287	164.745	30	-3.826	142.180
40	-2.645	313.603	40	-3.487	363.636	40	-2.240	423.280	40	-3.016	212.653	40	-4.351	182.648
50	-3.252	312.769	50	-4.234	511.509	50	-4.493	478.377	50	-3.079	261.643	50	-4.509	226.143
60	-3.393	363.665	60	-4.923	571.565	60	-3.989	617.284	60	-3.244	302.877	60	-5.573	262.009
70	-3.619	407.545	70	-5.719	642.378	70	-4.034	714.286	70	-3.355	346.198	70	-5.878	304.347
80	-4.466	427.608	80	-6.223	57.654	80	-4.092	808.081	80	-3.778	364.602	80	-5.998	349.112

TABLE-8
PARAMETERS OF KINETIC MODELS TO ADSORPTION WITH METHYL VIOLET DYE ON CRESSON SEEDS

Temp. (K)	Pseudo first-order			Pseudo second-order		
	K_1	Q_e	R^2	K_2	Q_e	R^2
298	0.052	0.693	0.459	0.891	0.182	0.937
303	0.033	0.046	0.574	-4.826	0.103	0.975
308	0906	4.317	0.512	2.139	0.105	0.979
313	0.016	0.084	0.377	0.688	0.216	0.993
318	0.049	0.089	0.966	1.306	0.239	0.999

TABLE-9
THERMODYNAMIC PARAMETERS FOR ADSORPTION OF METHYL VIOLET DYE ON CRESSON SEEDS

Temp. (K)	1/T	K_{eq}	ln K_{eq}	ΔH° (J mol^{-1})	ΔG° (J mol^{-1})	ΔS° (J mol^{-1} K^{-1})
298	0.00336	1.1246	0.1174	9037.3180	-290.377	31.3009
303	0.00330	1.1696	0.1566		-393.959	31.1253
308	0.00323	1.2112	0.1916		-489.806	30.9322
313	0.00319	1.3300	0.2852		-740.921	31.2404
318	0.00314	1.4130	0.3457		-912.441	31.2885

of adsorption rate for pseudo second-order in (g/mg min) and (t) is time (min). The parameters obtained by the application of the two kinetic models are reported in Table-7.

The value of rate constant (R^2) calculated from the equations of pseudo first-order and pseudo second-order are found to be for pseudo first-order is = 0.578 and for second order is 0.827, which is more than the value of pseudo first-order equation, thus suggested that the interaction between Cresson seeds and methyl violet dye follows the pseudo-second order mechanism.

Thermodynamic analysis: The Langmuir parameter was used to determine the thermodynamic parameters. When the system temperature increases, the degree of adsorption increases, this become clear from K_{eq} values increase with increase in temperature, which means endothermic process, this verified by ΔHº positive value. The positive value of ΔS° suggested the increased randomness during the process of adsorption. The ΔGº with negative value suggests that adsorption is spontaneous. The equilibrium values obtained are depicted in Table-9.

Conclusion

This study confirmed that Cresson seeds can be used for removal of methyl violet dye in aqueous solution. The removal efficiency reaches 76 % in some cases. The adsorption process based on solution with temperature effect. The adsorption process was best fitted with Freundlich adsorption model and kinetics fitted to pseudo-second order model. According to experimental consequence, Cresson seeds is recommended as an available and safe biosorbent to the removal. Thermodynamic parameters such as ΔH°, ΔS° and ΔG° suggested that the adsorption process was endothermic physisorption and spontaneous.

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