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BIOSORPTION OF CONGO RED FROM AQUEOUS SOLUTIONS USING *CLEODENDRUM FRAGRANS LEAVES*: KINETIC AND THERMODYNAMIC STUDIES

Ravi Kumar Madugula[1*] King Pulipati[2], AytenewGetaye[3], EliasBarsenga[4] and DejenMandefro[4]

ABSTRACT

The aim of the present study is to remove Congo red (CR) dye from their aqueous solution using *Cleodendrum fragrans* leaves powder as biosorbent in a batch study. The variables covered in a batch study were contact time, solution pH, initial CR dye concentration, biosorbent dosage, average particle size of the biosorbent and temperature. The kinetic and isotherm studies of biosorption of CR dyes onto *Cleodendrum fragrans* biosorbent was investigated using various kinetic pseudo first order, pseudo-second order and isotherm models (Langmuir, Freundlich, Temkin). The experimental data of biosorption of the dye onto the *Cleodendrum fragrans* biosorbent fitted well with the Langmuir isotherm model. The isotherm reveals that the biosorption of the dye onto *Cleodendrum fragrans* biosorbent was favourable. Thermodynamic parameters such as Gibbs free energy, enthalpy change and entropy change were also estimated for the biosorption of CR dye. The thermodynamic studies indicated that the biosorption of Congo red dye onto *Cleodendrum fragrans* biosorbent was spontaneous, feasible and endothermic.

Keywords: Congo red, *Cleodendrum fragrans*, pollution control, aqueous solution, Biosorption.

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Corresponding Author: Ravi Kumar Madugula

Email: kumar4611@gmail.com

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[1,3,4] Department of Chemical Engineering, College of Engineering and Technology, Samara University, Afar, Ethiopia.
[2]Department of Chemical Engineering, Andhra University College of Engineering, Visakhapatnam, AndhraPradesh, INDIA.

1. INTRODUCTION

Fresh water is a limited resource in many parts of the world. And this will become more dearth's due to increasing in population, urbanization and climate change. The major fact for this paucity of fresh water is not only due to demand for water but also due to pollution in fresh water ecosystem. Due to the pollution created by human beings in this ecosystem, the usable water has decreased drastically and the cost of purifying the water has increased drastically. The main source of water pollution are point source and non-point source. Point sources include pipeline discharge of pollutants such as domestic sewage discharge, industrial waste effluents from factories or plants to receiving waters. It contrast, non-point pollution results from storm runoff, which transports polluting materials diffusely over land. Color in the liquid effluents can also decrease the capacity of plants in photosynthesis. Uses of different kinds of shampoos by human beings are major threat to the water and now a day they act as water pollutants **[1]**. There are more than 10,000 commercially available dyes with over 7 lakh times of dyestuff being produced annually across the world **[2].** The dye based effluent is a considerable source of non-aesthetic pollution since the presence of small amount of dye (below 1 ppm) is clearly visible. Many researchers found that colorant may cause problems in aquatic ecosystem in several ways as follows **[3].** Biosorption technique was most favorable procedure among all the physico-chemical and adsorption, flocculation combined with flotation, membrane filtration, electro kinetic coagulation, ozonation, oxidation, precipitation and Ion exchange methods **[4, 5]**

2. MATERIALS AND METHODS

2.1 Preparation of dye solution

Stock solutions of Congo red concentration 1000 mg/L was prepared by dissolving 1 g of 100% Congo red in 1000 ml of distilled water. The solution was prepared using standard flasks. The range of concentration of the prepared dye solutions varied between 20 and 200 mg/l was prepared using the stock solution of dye.

2.2. Preparation of biosorbent

The green colored *Cleodendrum fragrans leaves* used in the present study were collected from the college of Engineering, Andhra University, Visakhapatnam, India. The collected leaves were washed with deionised water several times to remove dirt particles. The washing process was continued till the wash water contains no dirt. The washed leaves were then completely dried in sunlight for 20 days. The dried leaves were then cut into small pieces and powdered using domestic mixie. In the present study the powdered materials in the range of 53–152 μm particle size were directly used as biosorbents without any pretreatment.

2.3. Chemicals

The other chemicals used in the present study were NaOH and HCl. The pH of solutions is adjusted with 0.1 N HCl and 0.1 N NaOH. The entire chemicals used were analytical reagent (AR).

2.4. Batch mode biosorption studies

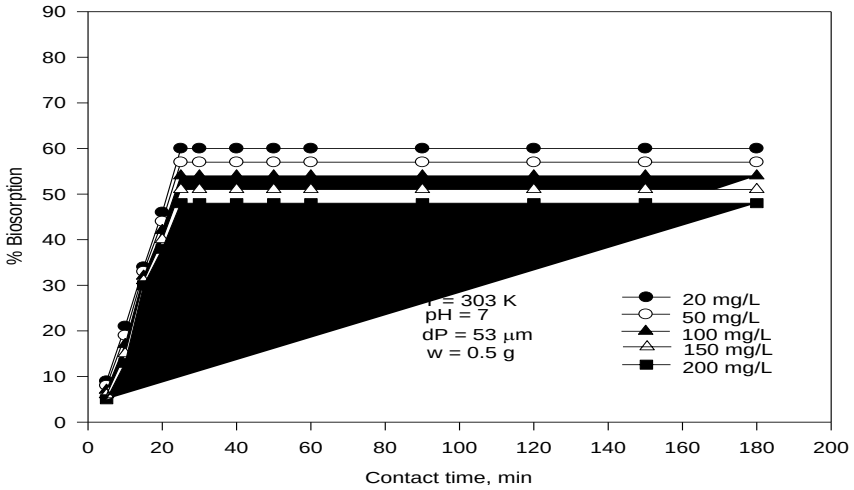
Batch biosorption equilibrium experiments were conducted in 250 ml conical flasks at a constant agitation speed (180 rpm). All the experiments were carried out at room temperature (± 30^0C). The concentrations of both the dyes before and after sorption were determined using UV- Vis spectrophotometer by monitoring the absorbance for the dye used.

3. RESULTS AND DISCUSSION

3.1. Effect of contact time

The percentage of biosorption was determined at different contact times and the results are shown in Fig1. The figure reveal that the percentage of biosorption Congo red was steeply increased with an increase in contact time from 5 to 25 min and there after reached plateu after attaining equilibrium 25 min for Congo red. Therefore, contact time of 25 min is sufficient for the removal of Congo red under the experimental conditions used in this study. The percentage of biosorption of *Cleodendrum fragrans* leaves as biosorbent for Congo red dye removal was increased from 9 to 60 %, with an increase in contact time from 5 to 25 min for an initial dye concentration of 20 mg/L. For 200 mg/L, the percentage of biosorption increased from 5 to 48 % mg/g, respectively with an increase in contact time from 5 to 25 min **[6, 7].**

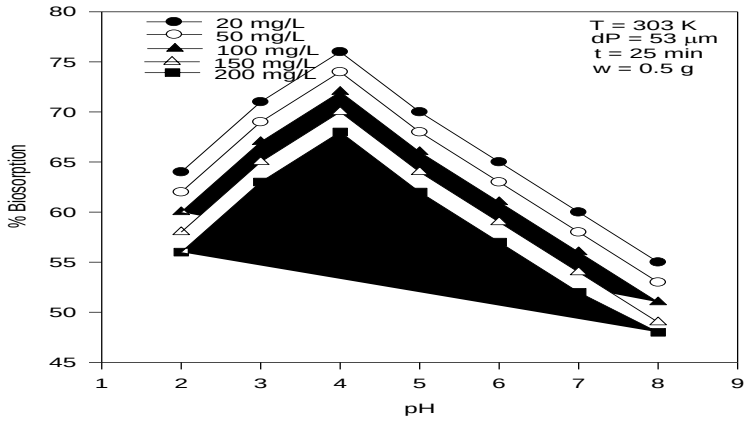
FIGURE1. EFFECT OF CONTACT TIME ON % BIOSORPTION OF CONGO RED USING *CLEODENDRUM FRANGRANS* LEAVES AS BIOSORBENT



3.2. Effect of solution pH

The solution pH is one of the important controlling parameters of the biosorption process. The effect of solution pH on % biosorption for the removal of Congo red dye was studied. From the Fig.2. It was observed that the increase in solution pH from 2 to 4 for Congo red resulted in an increase in percentage of biosorption from 64 to 76 % for Congo red for an initial concentration of 20 mg/L. For an initial concentration of 200 mg/L, the percentage of biosorption was increased from 56 to 68 % for Congo red with an increase in solution pH from 2 to 4 **[8-14].**

FIGURE2. EFFECT OF SOLUTION PH ON % BIOSORPTION OF CONGO RED USING *CLEODENDRUM FRANGRANS* LEAVES AS BIOSORBENT



3.3. Effect of initial concentration of dye

The effect of initial dye concentration on the percentage of biosorption is shown in Fig.3. It is evident from the figure that the percentage of biosorption decreased with an increase in initial concentration of dye from 20 to 200 mg/L at all temperatures. The percentage of biosorption of Congo red decreased from 72 to 44 % and 78 to 52% for Congo red with an increase in initial concentration of Congo red from 20 to 200 mg/L at the temperature of 283 K and 323K, respectively **[15,16].**

3.4. Effect of biosorbent dosage:

The result obtained is shown in Fig.4, illustrate that the percentage of biosorption was increased with an increase in biosorbent dosage. The percentage of biosorption increased from 76 to 91 % from biosorbent dosage 0.5 to 4 g for an initial concentration of Congo red 20 mg/L. For an initial concentration of Congo red 200 mg/L, the percentage of biosorption increased from 68 to 83 % from biosorbent dosage 0.5 to 4g **[17-23].**

FIGURE3. EFFECT OF INITIAL CONCENTRATION OF DYE ON % BIOSORPTION OF CONGO RED USING *CLEODENDRUM FRANGRANS* LEAVES AS BIOSORBENT

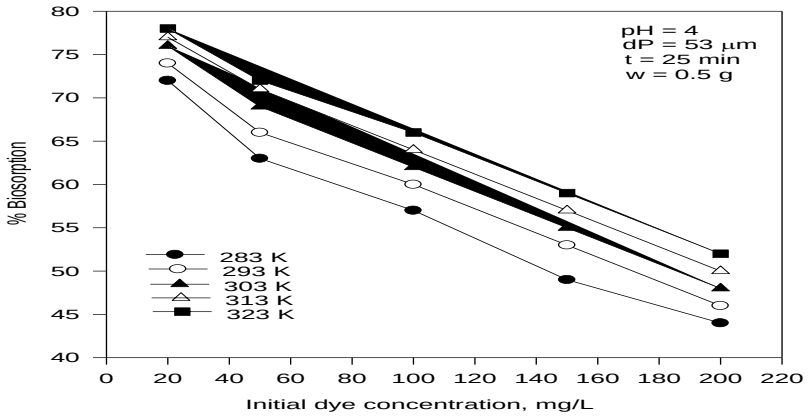
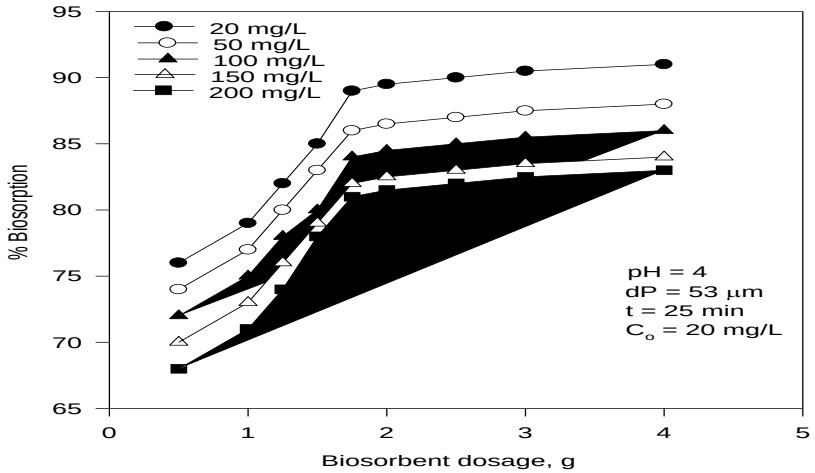


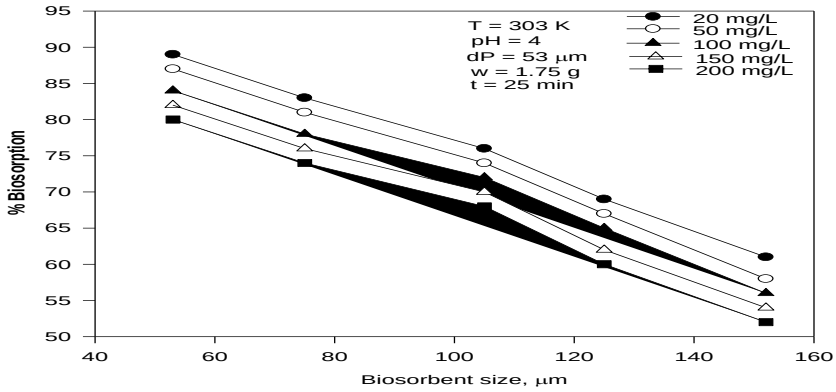
FIGURE 4. EFFECT OF BIOSORBENT DOSAGE ON % BIOSORPTION OF CONGO RED USING *CLEODENDRUM FRANGRANS* LEAVES AS BIOSORBENT



3.5. Effect of particle size of biosorbent:

The result obtained is shown in Fig.5. The result indicated that the percentage of biosorption was decreased with an increase in average particle size of biosorbent. The percentage of biosorption decreased from 89 to 61 % and 80 to 52 % with an increase in average particle size of biosorbent from 53 to 152 μm for an initial concentration of Congo red 20 mg /L and 200 mg/L, respectively **[24-30].**

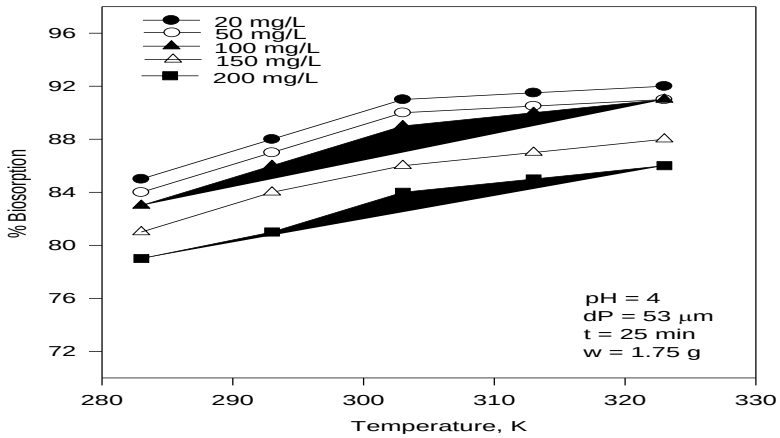
FIGURE 5. EFFECT OF AVERAGE PARTICLE SIZE ON % BIOSORPTION OF CONGO RED ONTO *CLEODENDRUM FRANGRANS LEAVES* BIOSORBENT



3.7. Effect of Temperature

The result obtained is shown in Fig.6, The figure indicated that the percentage of biosorption was increased with an increase in temperature of the solution. This suggests the endothermic nature of the biosorption process. The percentage of biosorption increased from 85 to 92 % and 79 to 86 % with an increase in solution temperature from 283 K to 323 K for an initial concentration of Congo red 20 mg /L and 200 mg/L, respectively **[31-37].**

FIGURE 6. EFFECT OF TEMPERATURE ON % BIOSORPTION OF CONGO RED ONTO *CLEODENDRUM FRANGRANS* LEAVES BIOSORBENT



4. EQUILIBRIUM STUDIES

4.1. Langmuir adsorption isotherm:

The applicability of Langmuir adsorption isotherm model was analyzed using the experimental data by plotting C_e/q_e versus C_e. Fig.7.show the Langmuir plot for the biosorption of Congo red dye at a temperature 303 K and The separation factor (R_L)

values at different initial dye concentrations for the dye was determined and shown in Fig.8. **[38-42].**

FIGURE 7. LANGUMUIR ISOTHERM FOR BIOSORPTION OF CONGO RED ONTO *CLEODENDRUM FRANGRANS* LEAVES BIOSORBENT.

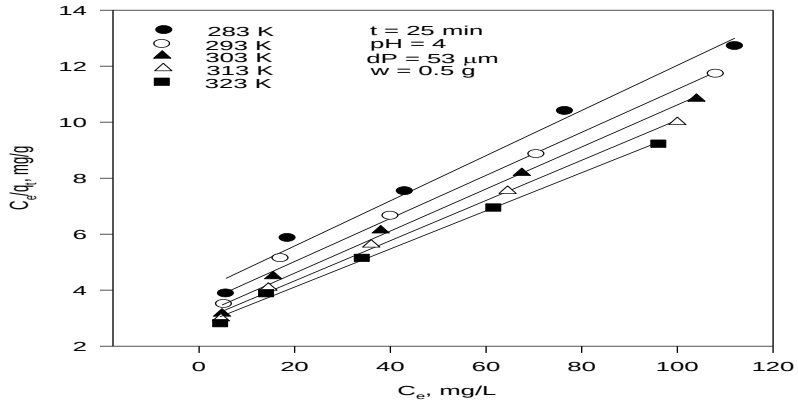
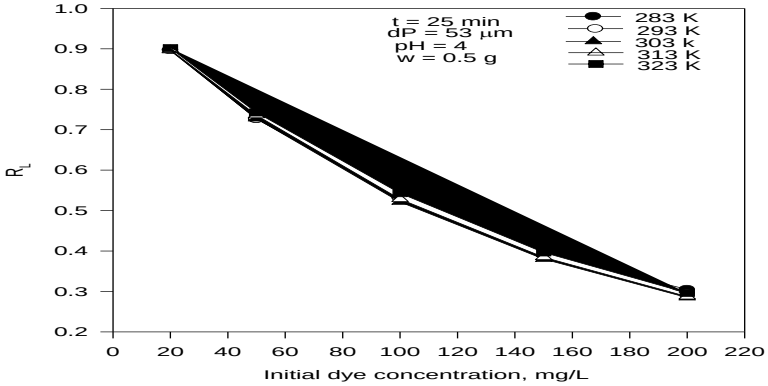


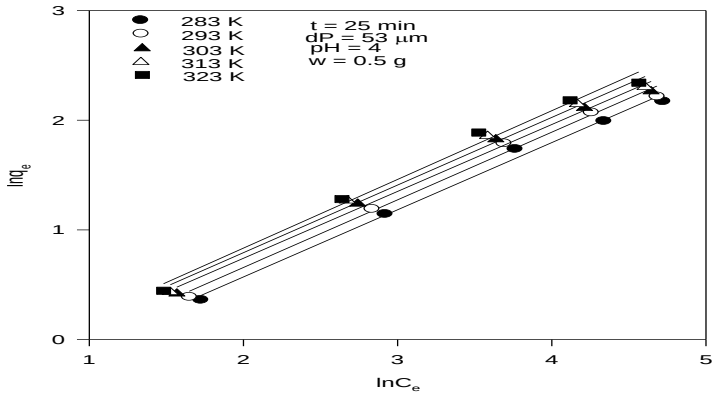
FIGURE 8. SEPARATION FACTOR FOR BIOSORPTION OF CONGO RED ONTO *CLEODENDRUM FRANGRANS* LEAVES BIOSORBENT.



4.2. Freundlich isotherm

The experimental data was tested for the fitness of Freundlich isotherm model by using linear graphical method. The biosorption data was analyzed by plotting ln q_e versus ln C_e shown in Fig.9.

FIGURE 9. FREUNDLICH ISOTHERM FOR BIOSORPTION OF CONGO RED ONTO *CLEODENDRUM FRANGRANS* LEAVES BIOSORBENT.



4.3. Temkin isotherm

Temkin isotherm studies were conducted to evaluate the biosorption potentials and to assess the variation of biosorption energies during the biosorption of Congo red dye using *Cleodendrum fragrans* leaves as biosorbent. The biosorption data was analyzed according to the linear form of Temkin model and is shown in Fig.10, for the removal of Congo red dye. The linear Temkin isotherm constants B_T and A_T, were determined from the slope and intercept of the plots of q_e versus ln C_e .

FIGURE 10. TEMKIN ISOTHERM FOR BIOSORPTION OF CONGO RED ONTO *CLEODENDRUM FRANGRANS* LEAVES BIOSORBENT.

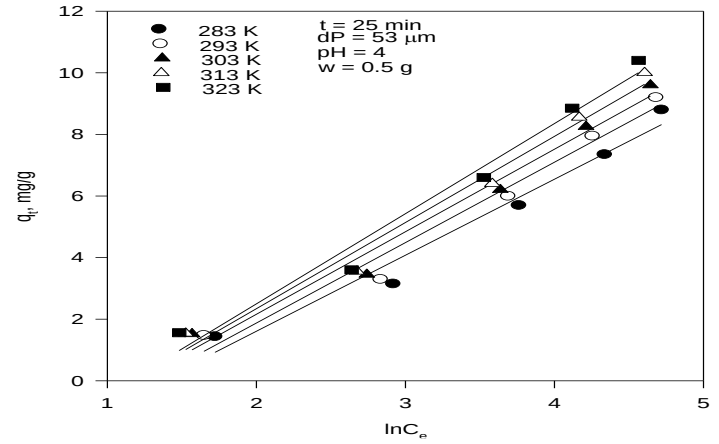


TABLE 1. BIOSORPTION ISOTHERM CONSTANTS FOR CONGO RED REMOVAL USING *CLEODENDRUM FRAGRANS* BIOSORBENT.

Temperature, T(K)	Langmuir isotherm			Freundlich isotherm			Temkin isotherm		
	q_{max}, (mg/g)	K_L, (L/mg)	R^2	n	K_f, ($mg^{1-1/n}$ $L^{1/n}$/g)	R^2	b_T, (J/mol)	A_T, (L/g)	R^2
283	12.4069	0.0203	0.9876	0.6123	0.5202	0.9937	953.15	0.2594	0.9698
293	13.0055	0.0220	0.9931	0.6170	0.5614	0.9908	932.97	0.2774	0.9742
303	**13.3191**	**0.0241**	**0.9951**	**0.6099**	**0.6194**	**0.9904**	**939.97**	**0.3026**	**0.9769**
313	13.9275	0.0248	0.9966	0.6164	0.6438	0.9891	930.25	0.3123	0.9787
323	14.6907	0.0247	0.9962	0.6263	0.6579	0.9892	917.74	0.3177	0.9766

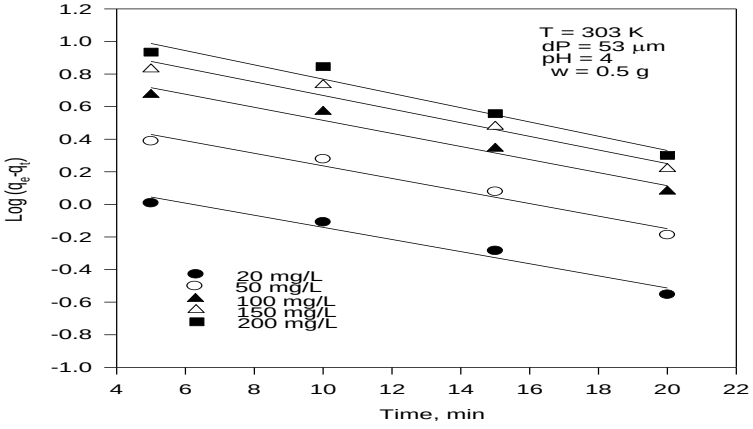
High correlation coefficient, R^2, values suggest that the biosorption process could be due to heterogeneous surface coverage. This is in good agreement with the result of Langmuir isotherm for Congo red dye.

5. KINETIC STUDIES

5.1. Pseudo- first-order kinetic model

The pseudo-first-order kinetic model was developed based on the solid adsorption capacity. The experimental results were analyzed to test the pseudo-first-order kinetic model and the linear plots of ln ($q_e - q_t$) versus t is shown in Fig.11.

FIGURE 11. PSEUDO FIRST ORDER KINETIC MODEL FOR CONGO RED BIOSORPTION ONTO *CLEODENDRUM FRANGRANS LEAVES* BIOSORBENT.



5.2. Pseudo-second-order kinetic model:

The values of equilibrium biosorption capacity and second order rate constants were determined from the slope and intercept of the linear plot of t/q_t versus t in Fig.12. The values obtained are tabulated along with the correlation coefficient (R^2) values in Table.2 for Congo red dye.

FIGURE12. PSEUDO SECOND ORDER KINETIC MODEL FOR CONGO RED BIOSORPTION ONTO *CLEODENDRUM FRANGRANS* LEAVESBIOSORBENT.

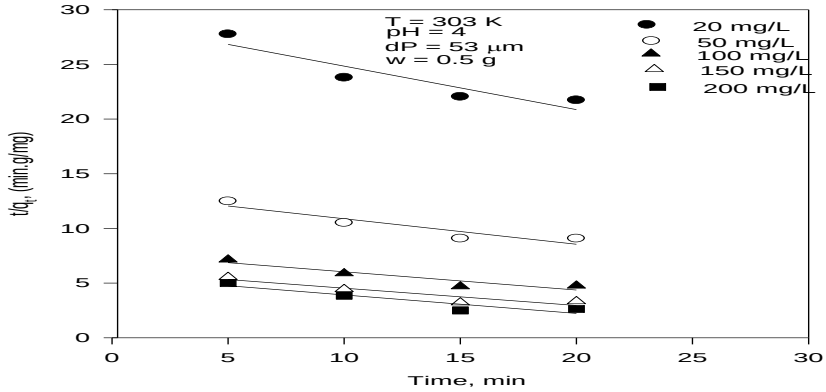


TABLE - 2.KINETIC MODEL PARAMETERS FOR CONGO RED REMOVAL USING *CLEODENDRUM FRANGRANS* BIOSORBENT.

Initial Conc, mg/l	**Lagergren-first order**			**Pseudo-second order**		
	q_e, (mg/g)	**k_1,(1/min)**	**R_1^2**	**q_e, (mg/g)**	**k_2(g/mg.min)**	**R_2^2**
20	1.2598	0.0856	0.9672	3.1338	0.0036	0.8390
50	1.8626	0.0886	0.9682	2.2497	0.0062	0.8760
100	2.5000	0.0921	0.9677	1.6270	0.0103	0.7910
150	2.9624	0.0962	0.9655	1.1724	0.0171	0.7715
200	3.3417	0.1016	0.9615	0.8325	0.0281	0.7582

6. THERMODYNAMIC STUDIES:

Thermodynamic studies provide information about the feasibility of the process and nature of biosorption process. In order to estimate the thermodynamic parameters for the biosorption of Congo red dye using *Cleodendrum frangrans* leaves as biosorbent, the experiments were conducted and data were analyzed. The values of ΔH° and ΔS° were calculated from the slope and intercept of the linear Van't Hoff plot i.e ln (q_e/c_e) vs (1/T). These plots are shown in Fig.13 for Congo red dye. The estimated

thermodynamics properties along with the correlation coefficients (R^2) are tabulated in Table-3.

FIGURE13. VAN'T HOFF RELATION FOR THE DETERMINATION OF THERMODYNAMIC PARAMETERS FOR THE BIOSORPTION OF CONGO RED ONTO *CLEODENDRUM FRANGRANS* LEAVES BIOSORBENT.

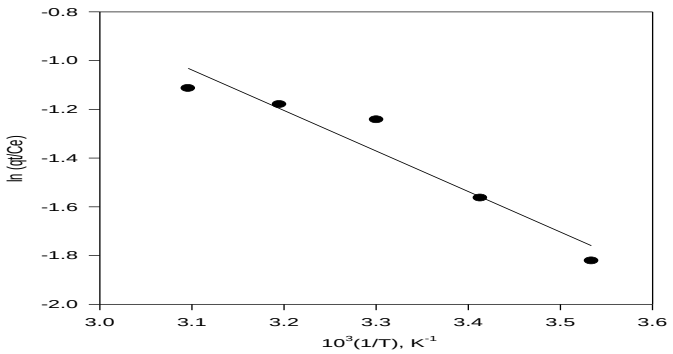


TABLE.3: THERMODYNAMIC PARAMETERS FOR THE BIOSORPTION OF CONGO RED ONTO *CLEODENDRUM FRANGRANS* LEAVES BIOSORBENT.

Temperature, K	**Congo red**		
	ΔG^0 (kJ/mol)	**ΔH^0 (J/mol)**	**ΔS^0 (J/k)**
283	–2.2269	31.8340	78.8019
293	-2.3057		
303	–2.3845		
313	–2.4633		
323	–2.5421		

7. CONCLUSION

The data obtained from the biosorption studies showed that a contact time of 25 min was sufficient for the maximum removal of Congo red dye from aqueous solution using *Cleodendrum fragrans* biosorbent. The experimental data of biosorption of the dye onto the *Cleodendrum fragrans* biosorbent fitted well with the Langmuir isotherm model. The isotherm reveals that the biosorption of the dye onto *Cleodendrum fragrans* biosorbent was favourable. The maximum removal efficiency was predicted to be 93.54% at a temperature of 303.4865 K, solution pH of 3.9991, and initial dye concentration of 19.6312 mg/L and biosorbent dosage of 1.7660g.

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