DIJBAR
www.daagu.org
Volume-2 Issue-1

STUDY ON REMOVAL OF CONGO RED DYE ONTO CLEODENDRUM FRAGRANS LEAVES: OPTIMIZATION OF PROCESS PARAMETERS USING RESPONSE SURFACE METHODOLOGY (RSM)

M.Ravi Kumar*[1], P.King[2], AytenewGetaye[1], Mohammednur Seid[1] and Nuru Ibrahim[1]

ABSTRACT

The Objective of the present study is to remove Congo red (CR) dye onto *Cleodendrum fragrans* leaves powder as biosorbent in a batch study. The effect of process variables used were contact time, solution pH, initial CR dye concentration, biosorbent dosage, average particle size of the biosorbent and temperature. The kinetic, isotherm and Thermodynamic studies of removal of CR dye onto *Cleodendrum fragrans* biosorbent were investigated. In the present study Congo red dye uptake was found to be 13.31mg/g by using *Cleodendrum fragrans* leaves as biosorbent. Response surface methodology (RSM) was adopted to study the correlations among the process variables affecting the process and to optimize the process to give high dye removal. The maximum removal efficiency was predicted to be 93.54% at a temperature of 303.48 K, solution pH of 3.99, initial dye concentration of 19.63 mg/L, biosorbent dosage of 1.76 g.

Keywords: Congo red, Cleodendrum fragrance, dye removal, Response Surface Methodology

Citation: M.Ravi Kumar, P.King, AytenewGetaye, Mohammednur Seid and Nuru Ibrahim (2020). STUDY ON REMOVAL OF CONGO RED DYE ONTO CLEODENDRUM FRAGRANS LEAVES: OPTIMIZATION OF PROCESS PARAMETERS USING RESPONSE SURFACE METHODOLOGY (RSM). Daagu International Journal of Basic & Applied Research-DIJBAR. Volume2, Issue-1, pp (29-42)

Corresponding Author:
Dr. M.Ravi Kumar

Email: kumar4611@gmail.com

Received: 24/01/2020

Accepted: 02/03/2020

Published: 29/06/2020

[1]Department of Chemical Engineering, College of Engineering and Technology, Samara University, Afar, Ethiopia.
[2]Department of Chemical Engineering, Andhra University College of Engineering, Visakhapatnam (A.P) INDIA.

1. INTRODUCTION

Water is an irreplaceable natural resource on this earth on which all life depends as a valuable natural resource, it comprises marine, estuarine, fresh water (river and lake) and ground water environments, across costal and inlands areas. Even though there is a huge water resource in this world, about 97% of water is salt water (marine) and only 3% is fresh water, and in this small fraction of fresh water most of it is locked up in polar ice caps and just 0.003% is readily available to us in the form of ground water and surface water. 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 sources 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 **[1]**. The textile industry consumes more than 107 kg of die per year of which 90% ending upon fabrics **[2].** According to the central pollution control board (CPCB), India, there are approximately a million known dye and dye intermediates out of which 5000 are produced commercial, based on their use, dyes are classified into 15 groups. They are acid dyes, azo dyes, basic dyes, direct dyes, food dyes, metal Complex, mordant dyes, whitening agent, pigment dyes, reactive dyes solvent dyes, sulphur dyes and VAT dyes. According to their chemical structure that can be classified into cationic (basic), anionic (direct, acid and reactive dyes) and nonionic dyes (disperse dyes) **[3].** Congo red is an organic compound, the sodium salt of 3,3′-([1,1′-biphenyl]-4,4′-diyl)bis(4-aminonaphthalene-1-sulfonic acid). It is an azo dye. Congo red is water-soluble, yielding a red colloidal solution; its solubility is greater in organic solvents. However, the use of Congo red has long been abandoned, primarily because of its carcinogenic properties. Congo red dye when dissolved in water has a red colour with an absorbance maximum at 497 nm. The removal of dyes from wastewater is so challenging that the government legislation required textile wastewater to be treated constantly with effect to process that can efficiently remove these dyes from wastewater during the past three decades. Biosorption technique was most favorable procedure among all the physico-chemical and adsorption, flocculation combined with flotation, membrane filtration, electrokinetic 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 mixer. 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. Response Surface Methodology

Optimization of different process parameters depending on the dye removal is highly required for the effective design and accurate control of biosorption technique. So, response surface methodology is time saving and precise alternative to conventional optimization methods. The main objective is to optimize the response surface that is shaped under the influence of process parameters.The effect of different process parameters such as temperature (X_1), solution pH (X_2), biosorbent dosage (X_3) and initial dye concentration (X_4) on Congo red dye removal onto *Cleodendrum fragrans* leaves were studied by using full factorial rotatable central composite design (CCD). Total thirty experiments, which include 16 cube point runs, 8 axial point runs and 2 centre point, 4 dummy runs were required and all of them were done in duplicate. All the experiments were conducted at contact time (t) and a constant speed of agitation 180 rpm.All independent variables were coded to five levels as X_i according to Eqn.1.

$$X_i = \frac{(X_i - X_{oi})}{\Delta X_i},\ i= 1, 2, 3,\ldots\ldots\ldots k \qquad \text{----Equ.1}$$

Where X_i is the dimensionless value of an independent variable, x_i is the real value of an independent variable, x_{0i} is the real value of the independent variable at the centre point, and Δx_i is the step change. A second degree polynomial equation (Eqn. 2) was developed to estimate the percentage of biosorption of dye at different operating conditions of the biosorption process by using STATISTICA 6.0 (Stat Soft Inc.).

$$Y=b_0+b_1X_1+b_2X_2+b_3X_3+b_4X_4+b_{11}X_1^2+b_{22}X_2^2+b_{33}X_3^2+b_{44}X_4^2+b_{12}X_1X_2+b_{13}X_1X_3+b_{14}X_1X_4+b_{23}X2X_3+b_{24}X_2X_4+ b_{34}X_3X_4 \qquad \text{----Equ.2}$$

Where , Y is the predicted response, X_1, X_2, X_3 and X_4 are independent variables: b_0 is an offset term; b_1,b_2,b_3 and b_4 are linear effects; b_{11}, b_{22}, b_{33} and b_{44} are squared effects and b_{12}, b_{13}, b_{14}, b_{23}, b_{24} and b_{34} are interaction terms.

3. RESULTS AND DISCUSSION

3.1. Effect of contact time

The effect of contact time on the biosorption process using *Cleodendrum frangrans leaves* as biosorbent was studied separately by varying initial dye concentrations (20 – 200 mg/L) at solution pH 7, temperature 303 K and constant speed of agitation 180 rpm. The percentage of biosorption and dye uptake values were determined at different contact times. The percentage of biosorption and dye uptake were steeply increased

with an increase in contact time from 5 to 25 min for Congo red and thereafter reached plateau after attaining equilibrium at 25 min for Congo red. The rapid biosorption at the early stages of contact period indicates the availability of a greater number of accessible sites on the surface of the biosorbent and also due to increase in the concentration gradient between adsorbate in solution and adsorbate on the biosorbent surface. At later stages of biosorption, the external biosorbent surface was saturated with dye molecules, thus the biosorption proceeds on interior walls of the biosorbent surface. The slower biosorption rate towards equilibrium was due to the availability of less number of available vacant sites on the biosorbent surface and also the repulsive forces between the adsorbed dye molecules on the biosorbent surface and dye molecules present in the aqueous solution.

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 and equilibrium dye uptake of *Cleodendrum frangrans leaves* biosorbent for the removal of Congo red dye was studied separately at different initial concentration of dyes (20 to 200 mg/L), temperature of 303 K and constant speed of agitation 180 rpm by varying solution pH from 2 to 8. The biosorption of Congo red was strongly pH dependent. The % biosorption and dye uptake of the biosorbent were low at solution pH 2 and increased gradually with an increase in pH from 2 to 4 for Congo red and thereafter decreased with further increase in pH from 4 to 8. The variation in the % biosorption and dye uptake with change in pH was due to the influence of solution pH on the activity of functional groups present on the biosorbent surface and chemical properties of dye molecules present in the aqueous solution.

3.3. Effect of initial concentration of dye

The effect of initial dye concentration for the biosorption of Congo red was studied at concentration range from 20 to 200 mg/L at a fixed dosage of biosorbent 0.5 g for 50 ml of solution, average particle size of 53 µm, temperature in the range 283 - 323K, solution pH 4 and constant speed of agitation 180 rpm. The percentage of biosorption was decreased and the dye uptake was increased for the dye.

3.4. Effect of biosorbent dosage

Biosorbent dosage is an important parameter in biosorption studies, because this factor determines the capacity of a biosorbent for a given initial concentration of biosorbate. The influence of biosorbent dosage on the biosorption of Congo red dye from aqueous solution was studied for initial dye concentration of 20 -200 mg/L at solution pH 4 for Congo red, temperature 303 K and constant speed of agitation 180 rpm by varying the biosorbent dosage from 0.5 to 4 g. The percentage of biosorption was increased with an increase in biosorbent dosage and the dye uptake was decreased with an increase in biosorbent dosage.

3.5. Effect of particle size of biosorbent

The effect of average particle size of the biosorbent on the biosorption of the dye, Congo red was investigated with solution having different initial dye concentrations (20 - 200 mg/L) at solution pH 4, temperature 303K and constant speed of agitation

180 rpm by varying the average particle size of biosorbent from 53 to 152 µm. The percentage of biosorption and dye uptake was decreased with an increase in average particle size of biosorbent. This could be due to the fact that the smaller particles have shortened diffusion paths and increases total surface area, and therefore, the ability to penetrate all internal pore structures of biosorbent is very high.

3.6. Effect of Temperature

The influence of temperature on the biosorption of Congo red dye was studied for initial dye concentration of 20-200 mg/L at solution pH 4, constant speed of agitation 180 rpm, and temperature of 283, 293, 303, 313, and 323 K. The percentage of biosorption and dye uptake were increased with an increase in temperature of the solution. This suggests the endothermic nature of the biosorption process.

3.7 Equilibrium studies

The equilibrium isotherm studies provide the equilibrium relationships between biosorbent and adsorbate and also give the information for the calculation of capacity of the biosorbent for an adsorbate. Equilibrium data is highly essential for the efficient design, operation and control of biosorption systems. Thus, it is highly important to establish the most appropriate equilibrium model which gives the true equilibrium data. Therefore, the isotherm results were analyzed using the Langmuir, Freundlich and Temkin models.

3.8 Kinetic Studies

The kinetics of biosorption studies describe the solute uptake rate and residence time of sorption reaction. These parameters are highly essential for efficient design of biosorption process. The kinetic data provides the necessary information required for optimizing the operating conditions in full scale batch biosorption process. The kinetic data also helps in determining the rate limiting step involved in the biosorption process. Hence, kinetic models were tested for the biosorption of Congo red dye using *Cleodendrum fragrans* leaves as biosorbent.

3.9 Thermodynamic Studie

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 fragrans* leaves leaves as biosorbent, the experiments were conducted and data were analyzed. The values of $\Delta H°$ and $\Delta S°$ were calculated from the slope and intercept of the linear Van't Hoff plot i.e $\ln (q_e/c_e)$ vs $(1/T)$. It is apparent from the results that the values of ΔG^0 were obtained at studied experimental conditions. The values of ΔG^0 indicate the spontaneous nature of the biosorption process and confirm the feasibility of the biosorption process. The decrease in values of ΔG^0 with increase in temperature shows an increase in feasibility of biosorption at higher temperature. These results are also in conformity with the results of adsorption isotherms (Langmuir isotherm).The results also revealed that the biosorption process is endothermic in nature, because ΔH^0 possess positive values. The

positive value of ΔS° indicates that the increased randomness at the solid-solute interface during biosorption process.

4. Optimization of biosorption process parameters using response surface methodology (RSM)

The response surface methodology is a collection of useful mathematical and statistical technique for analyzing the effects of several independent variables on response of interest. The main objective of RSM is to determine the optimum operational variables of the process. It is also used to evaluate the relative significance of process variables even in the presence of complex interactions. RSM offers certain advantages in adsorption process like improved percentage yield, closer confirmation of the output response to nominal and target requirements, reduced process variability and reduced developmental time and overall costs. Therefore, the aim of the present study is to conduct Congo red biosorption onto *Cleodendrum frangrans* leaves biosorbent and to investigate the combined effects of various process variables on percentage of biosorption using central composite design in response surface methodology. The variables such as temperature, solution pH, dosage of the biosorbent and initial dye concentration are all the four essential parameters of biosorption studies, significantly affected the percentage of biosorption of the dye and dye uptake of the biosorbent. Hence, these four variables were selected as independent variables to find the optimized conditions for maximum percentage of biosorption of Congo red dye onto *Cleodendrum frangrans* leaves biosorbent using response surface methodology.The influence of various process parameters such as temperature (x_1), solution pH (x_2), biosorbent dosage (x_3) and initial dye concentration (x_4) on the biosorption of Congo red onto the biosorbent was studied by using full factorial central composite design (CCD). The response was expressed as the percentage of biosorption of Congo red dye. A central composite design with 30 experiments, which includes 16 cube point runs, 8 center point runs, 2 axial point runs and 4 dummy runs, was used for the optimization of process variables for the removal of Congo red dye from aqueous solution.The Levels of different process variables used in CCD for the biosorption of Congo red dye onto *Cleodendrum frangrans* leaves biosorbent as shown in Table-1 and An Experimental design matrix along with percentage of biosorption of Congo red dye onto *Cleodendrum frangrans* leaves biosorbent as shown in Table-2.

Table-1. Levels of different process variables used in CCD for the biosorption of Congo red dye onto *Cleodendrum frangrans* leaves biosorbent.

Variable	Name	Range and levels				
		-2	-1	0	1	2
X_1	pH of aqueous solution	2	3	4	5	6
X_2	Initial concentration, C_o, mg/L	10	15	20	25	30
X_3	Biosorbent dosage, w, g	1.25	1.5	1.75	2	2.25
X_4	Temperature, T, K	283	293	303	313	323

Table-2. Experimental design matrix along with percentage of biosorption of Congo red dye onto *Cleodendrum frangrans* leaves biosorbent

Experimental Run no	x_1	x_2	x_3	x_4	% Biosorption of Congo red	
					Experimental	
					76.4800	Predicted
1	-1	-1	-1	-1	76.9800	76.4917
2	-1	-1	-1	1	77.7200	77.0375
3	-1	-1	1	-1	78.6000	77.7075
4	-1	-1	1	1	75.7200	78.5883
5	-1	1	-1	-1	76.3000	75.6875
6	-1	1	-1	1	76.3800	76.3183
7	-1	1	1	-1	77.3800	76.4483
8	-1	1	1	1	76.9800	77.4142
9	1	-1	-1	-1	77.3200	76.9708
10	1	-1	-1	1	77.6800	77.2817
11	1	-1	1	-1	78.2800	77.6917
12	1	-1	1	1	75.8800	78.3375
13	1	1	-1	-1	76.2800	75.9217
14	1	1	-1	1	76.2200	76.3175
15	1	1	1	-1	76.9000	76.1875
16	1	1	1	1	70.5200	76.9183
17	-2	0	0	0	70.4800	70.4808
18	2	0	0	0	79.3800	70.4642
19	0	-2	0	0	77.2000	79.3742
20	0	2	0	0	77.9800	77.1508
21	0	0	-2	0	79.8200	77.9642
22	0	0	2	0	79.5800	79.7808
23	0	0	0	-2	80.9200	79.5842
24	0	0	0	2	93.5000	80.8608
25	0	0	0	0	93.5000	93.5000
26	0	0	0	0	93.5000	93.5000
27	0	0	0	0	93.5000	93.5000
28	0	0	0	0	93.5000	93.5000
29	0	0	0	0	93.5000	93.5000
30	0	0	0	0		

The coefficient of determination (R^2) value provides a measure of variability in the observed response that can be explained by the experimental process variables and their interactions. The coefficient of determination (R^2) is found to be 0.996 for Congo red indicates that high correlation between the observed and predicted values. For further validation of the model, adjusted R^2 was used for confirming the model competence. The adjusted R^2 was found to be 0.99 for Congo red, which indicates that the proposed model was fitted well with the experimental data. These results indicate that the predicted R^2 values are in reasonable agreement with the adjusted R^2 values. The value of coefficient of determination indicates that 99% variation in the biosorption of Congo red is attributed to the independent variables explained by the model. The Parity plot for Congo red biosorption shown in fig1.

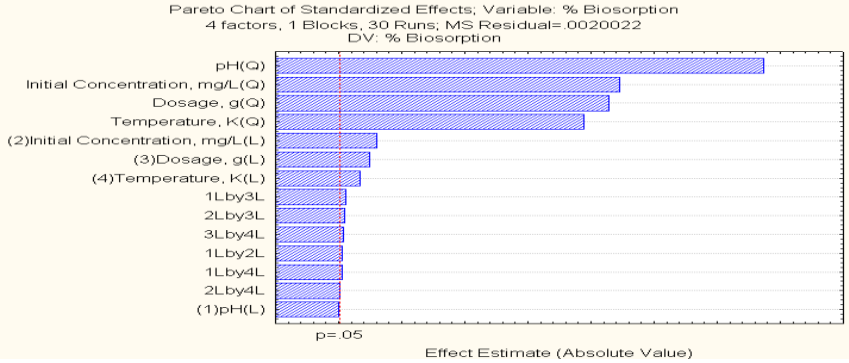


Fig.1. Parity plot for Congo red biosorption.

The significance of each parameter was evaluated by student t test and p values for Congo red, Response surface regression coefficient for removal of Congo red is given in Table.3. From these results the P values were found to be 0. 0.000064, 0.000098, 0.076887 and 0.000002. This implies that the linear effects of temperature, solution pH, dosage and initial dye concentration are highly significant for Congo red. Apart from the linear effects of the variables, the RSM also gives an insight into the quadratic and interaction effects of the process variables. It is evident from the results that the quadratic effect of all independent variables for Congo red was highly significant because their P-values were less than 0.05. As mentioned in Table.3, the magnitude of t –values give the positive or negative influence on the dependent variable. The linear terms of temperature, solution pH, biosorbent dosage and initial dye concentration have positive influence on the percentage of biosorption. The quadratic terms of temperature, solution pH, biosorbent dosage and initial dye concentration shows a negative influence on biosorption process for Congo red dye. The interaction effect was found to be significant for solution pH and biosorbent dosage for Congo red. Whereas interaction effects between remaining variables were found to be statistically insignificant because their P-values were > 0.05.

Table 3.Response surface regression coefficients for removal of Congo red

Term	Coeff.	SE coeff.	t-value	P-value
const	-3291.25	8.531229	-385.789	0.000000
x_1	48.94	0.357461	136.916	0.000000
x_2	-5.76	0.008544	-673.802	0.000000
x_3	6.06	0.071492	84.811	0.000000
x_4	-0.15	0.000342	-445.861	0.000000
x_1^2	200.25	1.460324	137.128	0.000000
x_2^2	-58.51	0.136702	-428.012	0.000000
x_3^2	20.10	0.052754	381.087	0.000000
x_4^2	-0.03	0.000085	-388.510	0.000000
x_1x_2	-0.01	0.002237	-5.475	0.000064
x_1x_3	-0.49	0.044746	-11.062	0.000000
x_1x_4	-0.01	0.001119	-5.252	0.000098
x_3x_2	-0.09	0.008949	-10.168	0.000000
x_4x_2	0.00	0.000224	1.900	0.076887
x_3x_4	0.03	0.004475	7.487	0.000002

The statistical significance of these regression model equations was evaluated by the F-test ANOVA. The significance of each variable was determined by F-values and p-values. It was observed from the Table .4. That the F statistic values for all regression were higher, which indicate that the developed models were highly significant and adequate to represent the relationship between the percentage of biosorption of the dye and process parameters. The larger fisher F- value with a low probability values (<0.05) demonstrate that the developed mathematical models were fitted well to the experimental data. From the ANOVA results, it is evident that the linear and quadratic effects were highly significant in comparison with interaction effects for the dye. However, for Congo red, solution pH and biosorbent dosage were significant Optimum values of variables obtained from regression equations for the removal of Congo red dye by *Cleodendrum fragrans* shown in Table-5.

Table 4. Analysis of variance for removal of Congo red dye

Factors	SS	Df	MS	F	P
x_1	0.000	1	0.0004	0.2	0.654794
x_2	909.027	1	909.0270	454009.1	0.000000
x_3	7.415	1	7.4148	3703.3	0.000000
x_4	398.025	1	398.0253	198791.8	0.000000
x_1^2	4.950	1	4.9504	2472.5	0.000000
x_2^2	366.795	1	366.7950	183194.0	0.000000
x_3^2	2.445	1	2.4448	1221.1	0.000000

x_4^2	302.215	1	302.2149	150939.7	0.000000
x_1x_2	0.060	1	0.0600	30.0	0.000064
x_1x_3	0.245	1	0.2450	122.4	0.000000
x_1x_4	0.055	1	0.0552	27.6	0.000098
x_3x_2	0.207	1	0.2070	103.4	0.000000
x_4x_2	0.007	1	0.0072	3.6	0.076887
x_3x_4	0.112	1	0.1122	56.1	0.000002
Error	0.030	15	0.0020		
Total SS	1445.239	29			

Table 5. Optimum values of variables obtained from regression equations for the removal of Congo red dye by *Cleodendrum fragrans*.

Parameter	Optimum value for Congo red dye
Solution pH	3.9991
Initial dye concentration, mg/l	19.6312
Biosorbent dosage, g	1.7660
Temperature, K	303.4865
% Biosorption	93.54

The response surface contour plots of the percentage of biosorption of dye versus the interactive effect of all the process parameters are shown in Figs.2 to 7 for Congo red dye.

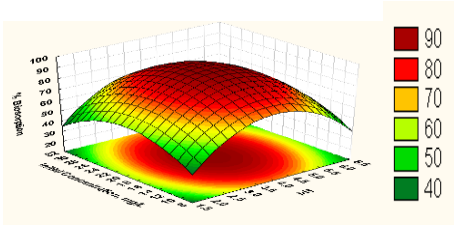


Fig.2. Response surface plot of solution pH vs initial dye concentration for the biosorption of Congored.

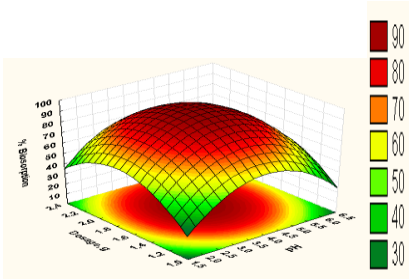


Fig.3. Response surface plot of solution pH vs Biosorbent dosage for the biosorption of Congo red.

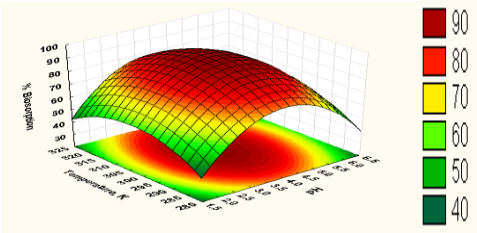


Fig.4. Response surface plot of solution pH vs temperature for the biosorption of Congo red.

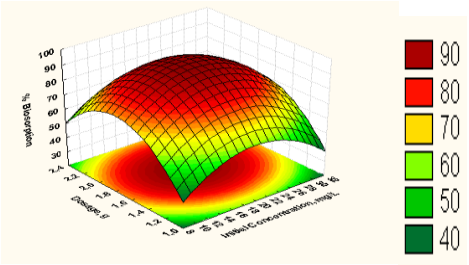


Fig.5. Response surface plot of initial dye concentration vs biosorbent dosage for the biosorption of Congo red.

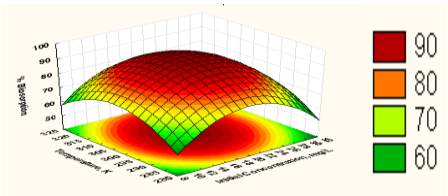


Fig.6. Response surface plot of initial dye concentration vs temperature for the biosorption of Congo red

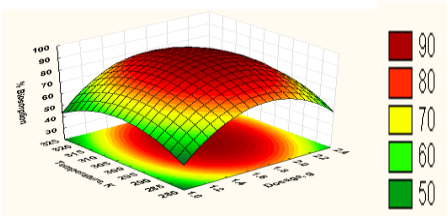


Fig.7. Response surface plot of biosorbent dosage vs temperature for the biosorption of Congo red

The maximum percentage of biosorption of level is indicated by the surface confined in the smallest curve (circular or elliptical) of the response plot. The optimal set of

conditions for maximum percentage of biosorption of Congo red is pH = 3.99, initial concentration of dyes in aqueous solution = 19.63 mg/L, biosorbent dosage = 1.76 g and temperature = 303.48 K. The extent of biosorption of Congo red at these optimum conditions was 93.54 %. The optimum values for process variables obtained by using response surface methodology were verified by conducting experiments at these optimum conditions. It was found that the maximum % biosorption of 93.54% for Congo red was obtained at these optimum parameters. These results are very nearer to that of the results of RSM studies, which proved that the statistical experimental design using RSM could be effectively used to optimize various process parameters for improving the biosorption of Congo red dye onto *Cleodendrum frangrans* leaves biosorbent.

4. CONCLUSION AND RECOMMENDATION

The optimization of experimental parameters like temperature, solution pH, biosorbent dosage and initial dye concentration were carried out using response surface methodology (RSM) with central composite design (CCD) for the removal of Congo red dye onto *Cleodendrum Frangrans* biosorbent. The optimized process variables obtained from the RSM for the biosorption of Congo red were in close agreement with the experimental data. The maximum removal efficiency was predicted to be 93.54% at a temperature of 303.48 K, solution pH of 3.99, and initial dye concentration of 19.63 mg/L and biosorbent dosage of 1.76g.

5. REFERENCES

1. Ramachandra, T.V., Ahalya, N., Rajasekara Murthy C. Aquatic ecosystems: conservation, restoration and management, Capital Publishers, New Delhi, (2005).
2. Kassim, T.L.S.M.J.N.M. Acidic and Basic Dyes Removal by Adsorption on Chemically Treated Mangrove Barks, International Journal of Applied Science and Technology, 2(3) (2012) 270-276.
3. S. Mishra, G., Tripaty, M. A critical review of the treatments for decolourization of textile effluent, Color age, 40 (1993) 35-38.
4. Sulak, M.T., Demirbas, E., Kobya, M. Removal of Astrazon Yellov.1 7GL from aqueous solution onto wheat bran J. Biores. Technol., 98 (2007) 2590-2598.
5. Xie, K., Zhao, W., He, X. Adsorption properties of nan10-cellulose hybrid. Dyes from aqueous solution, Carbo hyd . Polym., 83 (2011) 1516-1520.
6. Ho, Yuh-Shan, and Gordon McKay. "Sorption of dye from aqueous solution by peat." Chemical engineering journal, 70 (2) (1998) 115-124.
7. Kavitha, D., and C. Namasivayam. "Experimental and kinetic studies on methylene blue adsorption by coir pith carbon."Bioresource Technology, 98 (1) (2007) 14-21.
8. Tan, Boon Hai, Tjoon Tow Teng, and AK Mohd Omar. "Removal of dyes and industrial dye wastes by magnesium chloride." Water research, 34 (2) (2000) 597-601.
9. Namasivayam, C., N. Muniasamy, K. Gayatri, M. Rani, and K. Ranganathan. "Removal of dyes from aqueous solutions by cellulosic waste orange peel." Bioresource Technology, 57 (1) (1996) 37-43.

10. Garg, Vinod K., Moirangthem Amita, Rakesh Kumar, and Renuka Gupta. "Basic dye (methylene blue) removal from simulated wastewater by adsorption using Indian Rosewood sawdust: a timber industry waste." Dyes and pigments, 63 (3) (2004) 243-250.
11. Namasivayam, C., R. Radhika, and S. Suba. "Uptake of dyes by a promising locally available agricultural solid waste: coir pith." Waste management, 21 (4) (2001) 381-387.
12. Qu, Song, Fei Huang, Shaoning Yu, Gang Chen, and Jilie Kong. "Magnetic removal of dyes from aqueous solution using multi-walled carbon nanotubes filled with Fe2O3 particles."Journal of Hazardous Materials, 160 (2-3) (2008) 643-647.
13. Garg, V. K., Rakesh Kumar, and Renuka Gupta. "Removal of malachite green dye from aqueous solution by adsorption using agro-industry waste: a case study of Prosopis cineraria."Dyes and Pigments 62, no. 1 (2004): 1-10.
14. Forgacs, Esther, Tibor Cserhati, and Gyula Oros. "Removal of synthetic dyes from wastewaters: a review." Environment international, 30(7) (2004) 953-971.
15. Gong, Renmin, Yi Ding, Mei Li, Chao Yang, Huijun Liu, and Yingzhi Sun. "Utilization of powdered peanut hull as biosorbent for removal of anionic dyes from aqueous solution." Dyes and Pigments, 64 (3) (2005) 187-192.
16. Garg, Vinod K., Moirangthem Amita, Rakesh Kumar, and Renuka Gupta. "Basic dye (methylene blue) removal from simulated wastewater by adsorption using Indian Rosewood sawdust: a timber industry waste." Dyes and pigments, 63 (3) (2004) 243-250.
17. Kannan, Nagarethinam, and Mariappan Meenakshi Sundaram. "Kinetics and mechanism of removal of methylene blue by adsorption on various carbons—a comparative study." Dyes and pigments, 51 (1) (2001) 25-40.
18. Namasivayam, C., and Dyes Kavitha. "Removal of Congo Red from water by adsorption onto activated carbon prepared from coir pith, an agricultural solid waste." Dyes and pigments, 54 (1) (2002) 47-58.
19. Amin, Nevine Kamal. "Removal of reactive dye from aqueous solutions by adsorption onto activated carbons prepared from sugarcane bagasse pith." Desalination, 223(1-3) (2008) 152-161.
20. Papić, Sanja, Natalija Koprivanac, Ana Lončarić Božić, and Azra Meteš. "Removal of some reactive dyes from synthetic wastewater by combined Al (III) coagulation/carbon adsorption process." Dyes and pigments, 62(3) (2004) 291-298.
21. Lata, Hem, V. K. Garg, and R. K. Gupta. "Removal of a basic dye from aqueous solution by adsorption using Parthenium hysterophorus: an agricultural waste." Dyes and pigments, 74(3) (2007) 653-658.
22. Moussavi, Gholamreza, and Maryam Mahmoudi. "Removal of azo and anthraquinone reactive dyes from industrial wastewaters using MgO nanoparticles." Journal of Hazardous Materials, 168 (2-3) (2009) 806-812.
23. Garg, V. K., Rakesh Kumar, and Renuka Gupta. "Removal of malachite green dye from aqueous solution by adsorption using agro-industry waste: a case study of Prosopis cineraria." Dyes and Pigments, 62 (10 (2004) 1-10.
24. Robinson, T., B. Chandran, and P. Nigam. "Removal of dyes from an artificial textile dye effluent by two agricultural waste residues, corncob and barley husk." Environment International, 28 (1-2) (2002) 29-33.

25. Al-Ghouti, M. A., M. A. M. Khraisheh, S. J. Allen, and M. N. Ahmad. "The removal of dyes from textile wastewater: a study of the physical characteristics and adsorption mechanisms of diatomaceous earth." Journal of Environmental Management 69 (3) (2003) 229-238.
26. Chuah, T. G., A. Jumasiah, I. Azni, S. Katayon, and SY Thomas Choong. "Rice husk as a potentially low-cost biosorbent for heavy metal and dye removal: an overview." Desalination, 175 (3) (2005) 305-316.
27. Khraisheh, M. A. M., M. A. Al-Ghouti, S. J. Allen, and M. N. Ahmad. "Effect of OH and silanol groups in the removal of dyes from aqueous solution using diatomite." Water Research, 39 (5) (2005) 922-932.
28. Khattri, S. D., and M. K. Singh. "Removal of malachite green from dye wastewater using neem sawdust by adsorption."Journal of Hazardous Materials, 167 (1-3) (2009)1089-1094.
29. Mckay, G. B. H. S., H. S. Blair, and J. R. Gardner. "Adsorption of dyes on chitin. I. Equilibrium studies." Journal of applied polymer science, 27 (8) (1982) 3043-3057.
30. El-Geundi, Mohammad S. "Colour removal from textile effluents by adsorption techniques." Water Research, 25 (3) (1991) 271-273.
31. Aksu, Zümriye, and Sevilay Tezer. "Equilibrium and kinetic modelling of biosorption of Remazol Black B by Rhizopus arrhizus in a batch system: effect of temperature." Process Biochemistry, 36 (5) (2000) 431-439.
32. Al-Degs, Yahya S., Musa I. El-Barghouthi, Amjad H. El-Sheikh, and Gavin M. Walker. "Effect of solution pH, ionic strength, and temperature on adsorption behavior of reactive dyes on activated carbon." Dyes and pigments, 77 (1) (2008) 16-23.
33. Wang, Shaobin, Huiting Li, and Longya Xu. "Application of zeolite MCM-22 for basic dye removal from wastewater."Journal of colloid and interface science, 295 (1) (2006) 71-78.
34. Gupta, V. K., I. Ali, and Dinesh Mohan. "Equilibrium uptake and sorption dynamics for the removal of a basic dye (basic red) using low-cost adsorbents." Journal of colloid and interface science, 265 (2) (2003) 257-264.
35. Daneshvar, N., M. Ayazloo, A. R. Khataee, and M. Pourhassan. "Biological decolorization of dye solution containing Malachite Green by microalgae Cosmarium sp."Bioresource technology, 98 (6) (2007)1176-1182