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*Environmentally friendly oxidation processes*

# REMOVAL OF CONGO RED DYE BY ACTIVATED CARBON OBTAINED FROM *Prosopis juliflora*

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## ABSTRACT

Nowadays, activated carbon has been widely used to remove dye because of its good adsorption properties. Water pollution is the major outbreak for all over the world. It is caused by the polluted substances which are released from the industrial areas. This study uses activated carbon to remove textile dye. The activated carbon is prepared from the plant source *Prosopis juliflora.* The cheapest, easiest, and most environmentally friendly technique is activated carbon derived from plant sources. The chosen plant source is commonly utilised for fuel, firewood, and charcoal in rural regions since it is generally available and has excellent burning properties. Congo red dye is widely used in textile industry. Our study demonstrates the removal of Congo red dye using activated carbon which is prepared from the plant source *Prosopis juliflora.* The effective degradation was done by batch adsorption. The various parameters such as temperature, pH, dye concentration of Congo red dye were determined by using response surface methodology under optimum conditions. This study shows the effective removal of dye by low-cost adsorbent.

*Keywords*: activated carbon, *Prosopis juliflora,* Congo red, response surface methodology.

## AIMS AND BACKGROUND

Pollution is caused by natural resources and manmade resources. Our environment may deal with some contaminants and harmful substances which may cause huge impact on surrounding system. Various types of pollution occur due to several pollutants. Among that water pollution is the major outburst all over the world. It has been raised from the industrial revolution of the mid-19th century by introducing amazing

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technologies along with some water pollutants. By the middle of the 20th century, it dispels to the countries all over the world. After that, it affects the water quality, aquatic surrounding system and it also makes a mess around the human day-to-day life. The industrial sectors discharge a toxic pollutant dye substances that may mix with the water around cities and cause water pollution[1]. Still now, 80% of the wastewater is not treated well. The eco-friendly treatment of wastewater is essential to our study. Dye is a coloured substance that can come from artificial or natural sources. It has been extensively utilised in numerous industrial and field applications. Various industrial fields like paper, textile, leather, and plastics industries used some toxic dye. Among that, Congo red dye is widely used in textile industry, it is an acid-base indicator dye. Still, it was used to stain tissues for microscopic examination. This dye is known to be human carcinogen and mutagen, where it metabolised into benzidine. Removal of toxic chemical dye from the available plant source, which is environmentally friendly, and the use of activated carbon does not cause any harm the surrounding systems[3]. When Congo red dye is present in a free solution, the solvent's composition affects the colour and absorption characteristics in addition to the pH. Various techniques have been used for dye removal. Adsorption is described as a material's concentration on a solid surface. The removal of dye has been accomplished with excellent results by the adsorption method. Adsorbent activated carbon is more efficient when compared to others. Activated carbon from plant source is considered to be ease and environmentally friendly[4]. *Prosopis juliflora* (in Tamil, Seemai karuvelam) has been split up in all over the state of Tamil Nadu. It has been widely used as fuel, firewood, and timber in the rural areas. Although it is an environmental intruder, it does have some medical benefit. So that the government of Tamil Nadu has been planned to uproot the trees for the next ten years. The prototype of activated carbon from the plant source *Prosopis juliflora* is most abundant plant and it is available at low cost[5]. Since the plant is not be considered as eco-friendly, the study aimed to identify the degradation efficiency of Congo red dye by using activated carbon prepared from *Prosopis juliflora*. The study was also done under consideration of various parameters such as temperature, pH and dye concentration using Response surface methodology.

## EXPERIMENTAL

*Collection of Prosopis juliflora*. The major raw material used in this research work was *Prosopis juliflora*, a small tree of the family Fabaceae, a kind of mesquite that was obtained from different locations in Pallavaram, Tamil Nadu. Initially the plant was washed and shade dried for 5 days. Then the plant was powdered and further taken for chemical activation.

*Preparation of adsorbent*. The plant was treated with sulphuric acid for activation process. It was impregnated with 1:1 ratio and left it for 24 h. Then was oven dried at 100°C for 3 days. Then it was introduced into carbonisation process. The carbonised material was washed several times using distilled water to remove the excess sulphuric acid. The resulted activated carbon was then crushed and blended using mortar and pestle. Then it was stored for further degradation[6].

*Preparation of dye*. The Congo red dye is a type of anionic dye that is widely used in textile, rubber and plastic industry. The solutions of Congo red were prepared by dissolving in various concentrations such as 20, 40, 60, 80, and 100 mg of Congo red in 1 l of distilled water. The pH of the solution was adjusted with 0.1 N HCl or 0.1 N NaOH.

### ANALYTICAL STUDIES

The presence of various functional groups present in the adsorbent material was studied using Fourier Transform Infrared (FTIR). The FT-IR spectra of the Congo red dye before and after adsorption were carried out as a qualitative analysis to gain better insights into the surface functional groups available on the surface of the investigated adsorbent because the chemical structure of an adsorbent is of vital importance in understanding its adsorption nature.

### RESPONSE SURFACE METHODOLOGY

Response surface methodology (RSM) is a collection of mathematical and statistical techniques through which we can determine the optimum concentrations or conditions using Box-Behnken designs. RSM is a second order polynomial equation which works on the principle of relationships between diverse explanatory variables and response variables. The measure of performance of the process is called response and the variables that are used in the process are called independent variables. Experimental domain depicts the maximum and minimum limits of independent variables. Experimental design is the system created with several independent variables using matrix[7]. Box Behnken and central composite design are some of the experimental designs. Dependant variables are output variables produced by independent variables. Residual is the differences resulted between calculated and experimental value of the process under determined condition. Factors are also known as independent variables used in the process which indicate pH, temperature, dye concentration, agitation time. The system was explained by the following second order polynomial equation:

$$Y = \beta_0 + \beta_1 X_1 + \beta_2 X_2 + \beta_3 X_3 + \beta_{12} X_1 X_2 + \beta_{13} X_1 X_3,$$

where $Y$ is the response measured (dye degradation, %) and $X_1$, $X_2$, $X_3$ are the parameters. The optimum value is obtained by the regression equation, by analysing the points[8,9].

In the present work four significant parameters for Congo red (pH, temperature, activated carbon concentration and dye concentration) were chosen according to Central Composite Design. The four parameters for Congo red are the pH: 6–10, temperature: 45–85°C, dye concentration: 80–120 mg, and activated carbon concentration: 5–15 g.

Response Surface Methodology (Central Composite Design) was developed for four parameters over 30 trials using the Minitab 21 commercial software which is shown in Tables 1 and 2.

**Table 1**. Level of parameters tested for Congo red in Central Composite Design

| S. No | Parameters | Range |
|---|---|---|
| 1 | pH | 6–10 |
| 2 | Dye concentration | 80–120 mg |
| 3 | Activated carbon Concentration | 5–15 g |
| 4 | Temperature | 45–85°C |

**Table 2**. Trials given by Central Composite Design for Congo red dye

| S. No | Run order | pH | Temperature (°C) | Dye concentration (mg) | Activated carbon concentration (g) |
|---|---|---|---|---|---|
| 1 | 1 | 6 | 85 | 80 | 5 |
| 2 | 2 | 10 | 45 | 80 | 5 |
| 3 | 3 | 8 | 65 | 100 | 10 |
| 4 | 4 | 6 | 85 | 120 | 15 |
| 5 | 5 | 6 | 45 | 80 | 15 |
| 6 | 6 | 10 | 85 | 120 | 5 |
| 7 | 7 | 10 | 85 | 80 | 15 |
| 8 | 8 | 6 | 45 | 120 | 5 |
| 9 | 9 | 10 | 45 | 120 | 15 |
| 10 | 10 | 8 | 65 | 100 | 10 |
| 11 | 11 | 8 | 65 | 140 | 10 |
| 12 | 12 | 8 | 65 | 100 | 10 |
| 13 | 13 | 8 | 65 | 100 | 5 |
| 14 | 14 | 4 | 65 | 100 | 10 |
| 15 | 15 | 8 | 25 | 100 | 10 |
| 16 | 16 | 12 | 65 | 100 | 10 |
| 17 | 17 | 8 | 65 | 100 | 20 |
| 18 | 18 | 8 | 65 | 100 | 10 |
| 19 | 19 | 8 | 105 | 100 | 10 |
| 20 | 20 | 8 | 65 | 60 | 10 |
| 21 | 21 | 6 | 45 | 80 | 5 |
| 22 | 22 | 10 | 45 | 80 | 15 |
| 23 | 23 | 6 | 85 | 80 | 15 |
| 24 | 24 | 6 | 85 | 120 | 5 |
| 25 | 25 | 10 | 85 | 80 | 5 |
| 26 | 26 | 8 | 65 | 100 | 10 |
| 27 | 27 | 10 | 45 | 120 | 5 |
| 28 | 28 | 8 | 65 | 100 | 10 |
| 29 | 29 | 6 | 45 | 120 | 15 |
| 30 | 30 | 10 | 85 | 120 | 15 |

## RESULTS AND DISCUSSION

The present study shows that the activated carbon obtained from *Prosopis juliflora* has the capacity to remove the Congo red dye.

### FTIR CHARACTERISATION

Fourier-transform infrared spectroscopy (FTIR) is a technique that works by analysing the infrared spectral compounds. The FTIR analysis was made to identify the functional groups of the compounds. It was done to identify whether nitrogen, carbon, oxygen, hydrogen, sulphur group are present in the activated carbon sample. The identification of functional group is to check which functional group plays important role in dye degradation[6].

### CHARACTERISATION OF ACTIVATED CARBON USING FTIR

The qualitative analysis of activated carbon was carried out by FTIR spectrum. The FTIR spectrum of activated carbon obtained from *Prosopis juliflora* is shown in Fig. 1 and the functional groups are shown in Table 3. The spectral range is measured between 400–4000 $cm^{-1}$ and the figure illustrates that the band at 3851.95 and 3403.01 $cm^{-1}$ are due to O–H bending and OH stretching which implies the presence of alcohol group, respectively. The band observed at 2920.81 $cm^{-1}$ is attributed to the presence of aldehyde group, C–H stretching. The band at 1700.40 $cm^{-1}$ is an indication of carboxylic group, C=O stretching. The band at 1619.57 $cm^{-1}$ is due to the unsaturated ketone C=C stretching. The band formed at 1192.73–1121.89 $cm^{-1}$ is due to C–O stretching. Finally, the band at 752.51 $cm^{-1}$ is due to C–H bending while at 656.77 $cm^{-1}$ and 603.70 $cm^{-1}$ – to C–H stretching.

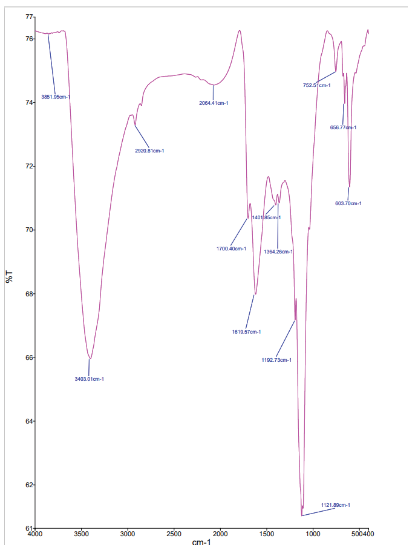


**Fig. 1**. FTIR analysis of activated carbon

**Table 3**. FTIR analysis of activated carbon

| No of peaks | Wave number ($cm^{-1}$) | Functional group |
|---|---|---|
| 1 | 3851.95 | O–H bending |
| 2 | 3403.01 | O–H stretching |
| 3 | 2920.81 | C–H stretching |
| 4 | 2064.41 | C=C stretching |
| 5 | 1700.40 | C=O stretching |
| 6 | 1619.57 | C=C stretching |
| 7 | 1401.85 | O–H bending |
| 8 | 1364.26 | C–H bending |
| 9 | 1192.73 | C–O stretching |
| 10 | 1121.89 | C–O stretching |
| 11 | 752.51 | C–H bending |
| 12 | 656.77 | C–H stretching |
| 13 | 603.70 | C–H stretching |

## FTIR CHARACTERISATION OF CONGO RED DYE BEFORE AND AFTER ADSORPTION

The FTIR spectra of Congo red before and after adsorption were analysed and there is a shift in bands observed which is illustrated in Figs 2 and 3. Tables 4 and 5 illustrate the presence of functional groups in FTIR analysis. The bands at 3330.45, 2084.00, and 1637.77 $cm^{-1}$ were shifted to 3338.12, 2107.06, and 1637.89 $cm^{-1}$, respectively. The shift in bands showed the functional groups participation in adsorption process. The bands at 3330.45 and 3338.12 $cm^{-1}$ were due to O–H stretching indicating the presence of alcohol group. The band observed at 2084.00 and 2107.06 $cm^{-1}$ was due to presence of C=C stretching. The band at 1637.77 and 1637.89 $cm^{-1}$ was indication of presence of C=C aromatic compounds.

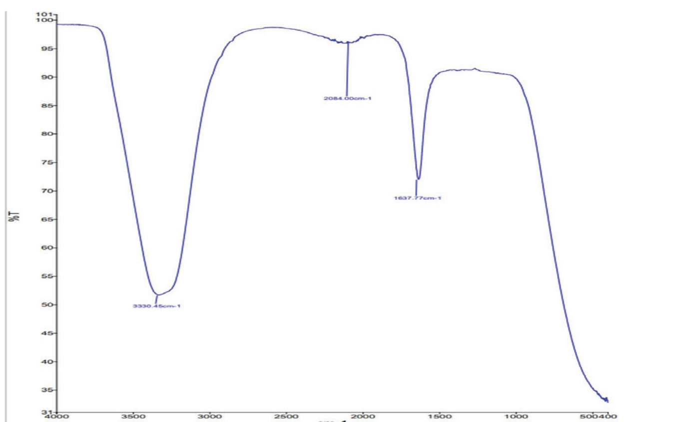


**Fig. 2**. FTIR analysis of Congo red before adsorption

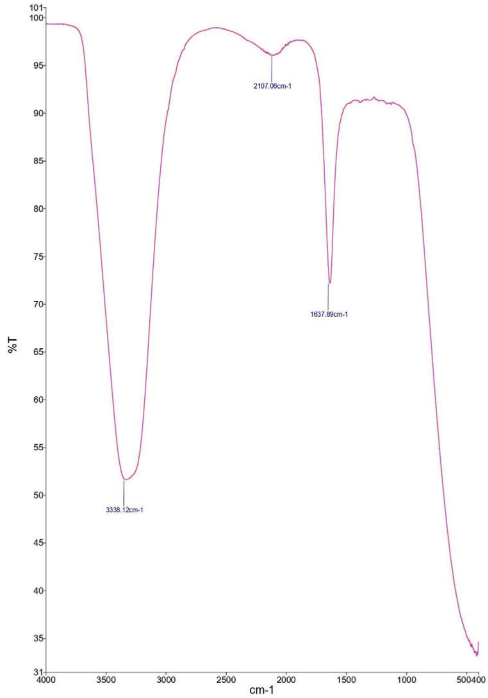


**Fig. 3**. FTIR analysis of Congo red after adsorption

**Table 4**. FTIR analysis of Congo red before adsorption

| No of peaks | Wave number ($cm^{-1}$) | Functional group |
|---|---|---|
| 1 | 3330.45 | O–H stretching |
| 2 | 2084.00 | C=C stretching |
| 3 | 1637.77 | C=C stretching |

**Table 5**. FTIR analysis of Congo red after adsorption

| No of peaks | Wave number ($cm^{-1}$) | Functional group |
|---|---|---|
| 1 | 3338.12 | O–H stretching |
| 2 | 2107.06 | C=C stretching |
| 3 | 1637.89 | C=C stretching |

**Table 6**. Central Composite Design for parameters of Congo red dye degradation

| S. No | Run order | pH | Temperature (°C) | Dye concentration (mg) | Activatedcarbon concentration (g) | Experimental value (%) (% removal) | Predicted value (%) |
|---|---|---|---|---|---|---|---|
| 1 | 1 | 6 | 85 | 80 | 5 | 68.31 | 69.0299 |
| 2 | 2 | 10 | 45 | 80 | 5 | 64.30 | 64.7399 |
| 3 | 3 | 8 | 65 | 100 | 10 | 94.50 | 92.7003 |
| 4 | 4 | 6 | 85 | 120 | 15 | 68.70 | 69.1349 |
| 5 | 5 | 6 | 45 | 80 | 15 | 68.01 | 67.9216 |
| 6 | 6 | 10 | 85 | 120 | 5 | 71.12 | 72.0833 |
| 7 | 7 | 10 | 85 | 80 | 15 | 76.20 | 76.6482 |
| 8 | 8 | 6 | 45 | 120 | 5 | 67.60 | 68.0266 |
| 9 | 9 | 10 | 45 | 120 | 15 | 77.15 | 77.3049 |
| 10 | 10 | 8 | 65 | 100 | 10 | 94.40 | 92.7003 |
| 11 | 11 | 8 | 65 | 140 | 10 | 85.67 | 84.1844 |
| 12 | 12 | 8 | 65 | 100 | 10 | 94.50 | 97.9123 |
| 13 | 13 | 8 | 65 | 100 | 5 | 64.79 | 64.1744 |
| 14 | 14 | 4 | 65 | 100 | 10 | 78.50 | 77.9811 |
| 15 | 15 | 8 | 25 | 100 | 10 | 69.34 | 68.3311 |
| 16 | 16 | 12 | 65 | 100 | 10 | 87.50 | 86.3127 |
| 17 | 17 | 8 | 65 | 100 | 20 | 73.83 | 72.7394 |
| 18 | 18 | 8 | 65 | 100 | 10 | 94.50 | 97.9123 |
| 19 | 19 | 8 | 105 | 100 | 10 | 73.48 | 72.7827 |
| 20 | 20 | 8 | 65 | 60 | 10 | 80.30 | 80.0794 |
| 21 | 21 | 6 | 45 | 80 | 5 | 64.76 | 64.7773 |
| 22 | 22 | 10 | 45 | 80 | 15 | 72.56 | 73.4207 |
| 23 | 23 | 6 | 85 | 80 | 15 | 70.39 | 70.4807 |
| 24 | 24 | 6 | 85 | 120 | 5 | 69.28 | 69.2507 |
| 25 | 25 | 10 | 85 | 80 | 5 | 70.35 | 70.0090 |
| 26 | 26 | 8 | 65 | 100 | 10 | 94.50 | 92.7873 |
| 27 | 27 | 10 | 45 | 120 | 5 | 69.45 | 70.1907 |
| 28 | 28 | 8 | 65 | 100 | 10 | 94.40 | 92.7873 |
| 29 | 29 | 6 | 45 | 120 | 15 | 68.78 | 69.9523 |
| 30 | 30 | 10 | 85 | 120 | 15 | 76.69 | 77.5040 |

The efficiency of Congo red dye degradation is investigated using Central Composite design which is shown in Table 6. The results were obtained by using the experimental design to identify the optimal conditions among the significant factors such as pH, temperature, dye concentration and activated carbon concentration[10,11].

**Table 7**. ANOVA for Congo red: Central Composite Design

| Source | DF | Adj. SS | Adj. MS | F-value | *P*-value |
|---|---|---|---|---|---|
| Model | 16 | 3162.19 | 197.64 | 53.92 | 0.000 |
| Linear | 4 | 269.17 | 67.29 | 18.36 | 0.000 |
| pH | 1 | 104.13 | 104.13 | 28.41 | 0.000 |
| Temperature ($^{o}$C) | 1 | 29.73 | 29.73 | 8.11 | 0.014 |
| Dye concentration (mg) | 1 | 25.28 | 25.28 | 6.90 | 0.021 |
| Charcoal concentration (g) | 1 | 110.04 | 110.04 | 30.02 | 0.000 |
| pH × pH | 1 | 426.08 | 426.08 | 116.24 | 0.000 |
| Temperature × temperature | 1 | 1282.83 | 1282.83 | 349.97 | 0.000 |
| Dye concentration × Dye concentration | 1 | 426.08 | 426.08 | 116.46 | 0.000 |
| Charcoal concentration × charcoal concentration | 1 | 1487.35 | 1487.35 | 405.77 | 0.000 |
| pH × temperature | 1 | 0.71 | 0.71 | 0.19 | 0.667 |
| pH × dye concentration | 1 | 4.11 | 4.11 | 1.12 | 0.309 |
| pH × charcoal concentration | 1 | 28.76 | 28.76 | 7.85 | 0.015 |
| Temperature × dye concentration | 1 | 10.26 | 10.26 | 2.80 | 0.118 |
| Temperature × charcoal concentration | 1 | 3.49 | 3.49 | 0.95 | 0.347 |
| Dye concentration × charcoal concentration | 1 | 1.94 | 1.94 | 0.53 | 0.480 |

Adj. SS – Adjusted sum of square; Adj. MS – Adjusted mean square.

The analysis of variance (ANOVA) for the degradation of Congo red dye by *Prosopis juliflora* derived activated carbon is given in Table 7. *P* value less than 0.05 indicates that the models are significant. *P* value more than 0.05 indicates that the models are insignificant.

Based on the results, the relationships between the removal percentage of dye and significant parameters are expressed by the second order polynomial equation:

$$\text{experimental value } (Y) = 186.2 + 13.86A + 2.483B + 2.087C + 5.898D - 0.9853A\times A - 0.017097B\times B - 0.009863C\times C - 0.2946D\times D + 0.0053A\times B + 0.0127A\times C + 0.1341A\times D - 0.00200B\times C - 0.00467B\times D - 0.00348C\times D,$$

where $A$ is pH, $B$ – the temperature ($^{o}$C), $C$ – the dye concentration (mg), and $D$ – the charcoal concentration (g).

From the equation above, $Y$ is the response and the coefficient of each parameter indicates the effect of parameter on dye removal.

**Table 8**. Model summary

| S | $R^2$ | $R^2$ (adj.) | $R^2$ (pred.) |
|---|---|---|---|
| 1.91456 | 98.52 | 96.69 | 94.92 |

Table 8 illustrates the model summary in which the $R^2$ value is the measure of variability in the observed response values can be explained by experimental factors. The $R^2$ value is 98.52 and the adjusted $R^2$ value is 96.69 for the removal of dye using activated carbon.

## EFFECTS OF PROCESS PARAMETERS

The graphical, three-dimensional representation is called a contour plot. The plots show the interaction between the given parameters and the optimum condition for degradation. The contour plot of the second order polynomial equation is done by the interaction of two variables within the experimental ranges[12].

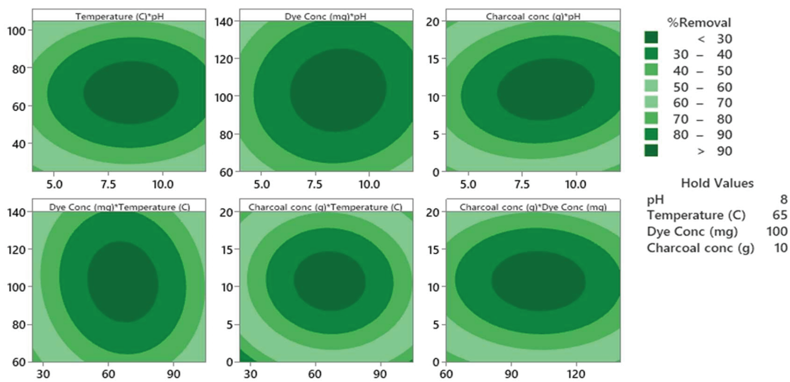


**Fig. 4**. Contour plots for Congo red removal showing the interaction between parameters

Figure 4 shows the contour plots of interaction between the various process parameters in dye removal. The interaction at pH 7.5 to 8 and temperature 60–65°C also showed significant removal of dye. At pH 8 and at dye concentration of 100 mg the interaction was reported significant. Also, the interaction of activated carbon concentration of 10–15 g and pH 8, dye concentration of 80–100 mg and temperature 60–65°C, activated carbon concentration of 10 g and temperature 65°C, activated carbon concentration of 10 g and dye concentration of 80–100 mg all showed a significant degradation of Congo Red.

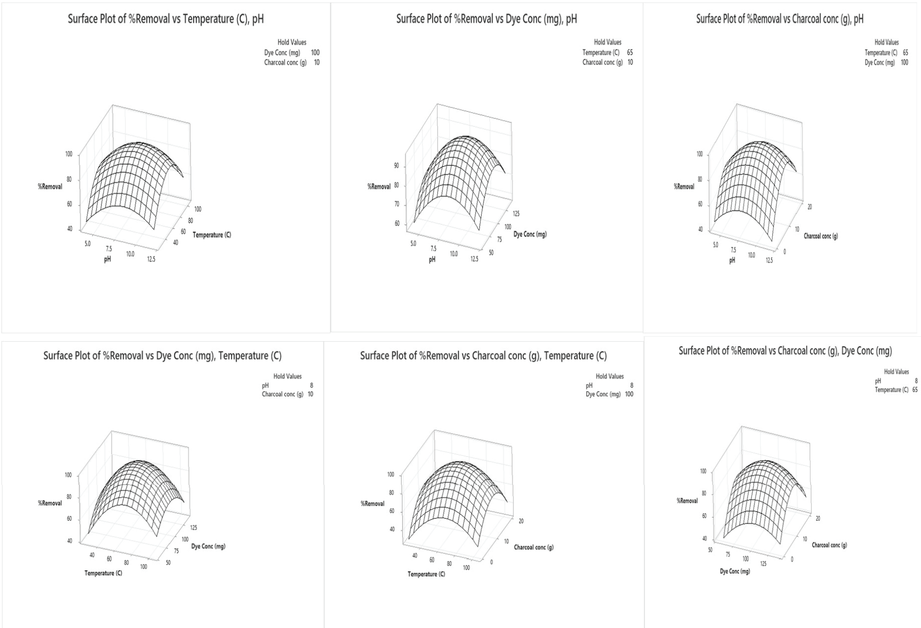


**Fig. 5**. Interactive effects of various parameters on dye removal

The surface plot indicates the interaction between the parameters of degradation process as shown in Fig. 5. The curved surface of the plot demonstrates the significant interaction between two parameters. From the figure it is inferred that the increase of pH at level of 8 shows significant increase in dye removal after that there is a decline in degradation efficiency. The optimum pH for degradation is between 7 and 9. The percentage removal also increased with increase in temperature between 60 to 65°C. The activated carbon concentration in the range of 10 to 15 g and dye concentration at 100 mg/l also showed significant dye removal. When limited activated carbon is added the degradation efficiency is low since the active site on the surface where bonded with increased amount of dye.

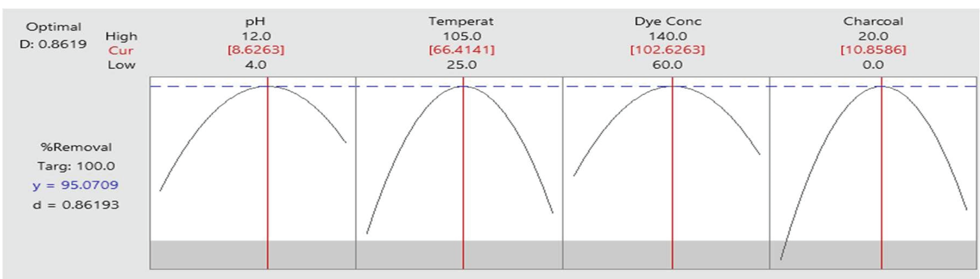


**Fig. 6**. Optimal prediction for Congo red dye degradation

From Response Surface Methodology, we can confirm the significant value for degradation of Congo red dye using *Prosopis juliflora* derived activated carbon is at pH 8.5, temperature of 65°C, dye concentration of 100 mg, and activated carbon concentration – 10 g. The optimal predicted value (Y) is 95.07 (Fig. 6).

## CONCLUSIONS

The study results demonstrated that the use of activated carbon obtained from *Prosopis juliflora* showed higher efficiency of 94.5% degradation of Congo red dye. The significant value for Congo Red dye degradation is at pH 8, temperature 65°C, dye concentration 100 mg, and activated carbon concentration – 10 g. The results reveal that the activated carbon obtained from *Prosopis juliflora* can be considered as a potential adsorbent of dyes. So, the above study implies that the plant-derived activated carbon provides cheaper and eco-friendly degradation of textile dyes.

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