

# Removal of Congo Red by Carbonized Low-Cost Adsorbents: Process Parameter Optimization Using a Taguchi Experimental Design

Ghanshyam Barman, Arvind Kumar,\* and Prateek Khare

Department of Chemical Engineering, National Institute of Technology, Rourkela Orissa 769 008, India

**ABSTRACT:** The Taguchi method was applied as an experimental design to establish the optimum conditions for the removal of congo red (CR) in batch experiments. The chosen experimental factors and their ranges were: pH (2 to 10), temperature [(40 to 60) °C], contact time [(5 to 500) min], and concentration of CR [(10 to 500) mg·L<sup>-1</sup>]. The orthogonal array L<sub>25</sub> and “the larger, the better” response were selected to determine optimum removal conditions onto commercial and low-cost adsorbents. The adsorbents were also characterized for their valuable adsorbable properties such as point of zero charge and carbon specific surface area. The influence of the different factors on the adsorption of CR from solution is explained in terms of electrostatic interactions by considering the dye species and the character of the surface. The exhausted low-cost adsorbents along with the sorbed CR can be used as such or as fire briquettes to recover their energy value, and resulting bottom ash may be used to make fire bricks.

## 1. INTRODUCTION

Congo red (CR) is one of the major “direct” dyes used in the dyeing industries in India.<sup>1</sup> Textile industries consume considerable amounts of water and use a variety and large quantity of chemicals throughout different process stages in their manufacturing processes and hence produce large amounts of wastewater.<sup>2</sup> Dye contamination in wastewater poses severe environmental problems.<sup>3</sup> Apart from many other reasons, dye-stuffs receive significant attention from researchers who are interested in the development of treatment processes at their optimum removal conditions for textile wastewater because of its strong color which affects the aesthetic nature of water.<sup>1,4</sup> There are several conventional treatment methods reported in literature for treating dye-containing wastewaters.<sup>3</sup>

Among various treatment technologies, adsorption onto activated carbon and low-cost adsorbents has been proved to be one of the effective and reliable physicochemical treatment methodologies for dye removal.<sup>5</sup>

Activated commercial carbon (ACC) has been widely employed for the treatment of industrial effluents because of its high capacity for adsorption of pollutants.<sup>6</sup> Despite its prolific use in water and wastewater industries, ACC remains an expensive material. As a consequence, many investigators have studied the feasibility of using low-cost adsorbents derived from different sources (agricultural wastes, etc.) for the removal of various dyes and pollutants from wastewaters and to compare their adsorption capacity for the adsorption of dyes under identical experimental conditions.<sup>1,7</sup>

The purpose of the present study is, therefore, to study the process of adsorption of CR onto powdered and granular AC and indigenously prepared low-cost adsorbents such as bagasse fly ash (BFA), rice husk ash (RHA), coconut shell carbon (CSC), fruit juice extract (FJE), BAEL (*Aegle marmelos*), and spent tea granules (STG) and to research the possibility of using them as low-cost adsorbents for the removal of dyes in general and CR in particular. This investigation also examines the influence of various process parameters for their optimal performance such

as the initial concentration of CR ( $C_0$ ), contact time ( $t$ ), temperature ( $T$ ), and initial pH of dye solution on CR adsorption onto powdered and granular AC and their comparison with the adsorption capacity of various prepared adsorbents using the Taguchi statistical method.

## 2. EXPERIMENTAL SECTION

**2.1. Adsorbents and Their Preparation.** BAEL, CSC, STG, BFA (ash from biofuel producer), and fruit juice pulp (carbon) were obtained locally in the vicinity of Rourkela, Orissa, washed with hot water (70 °C), and dried. The commercial grade PAC was obtained from HiMedia Research Laboratory, Mumbai, and the GAC was obtained from SD Fine Chemicals, Ltd. (Mumbai, India), and these were used as procured. RHA was obtained from Vandana Power Ltd. (Bilaspur, Chhattisgarh).

Powder and granular activated carbon (AC) were used without chemical activation. The chemical activation of BFA, RHA and STG was carried out by treating all three of these with 0.01 M HCL solution and dried at 105 °C for 12 h. To remove the residual acid that was left in BFA, RHA and STG were soaked in 3 % NaHCO<sub>3</sub>. Finally the adsorbents were dried at 105 °C for 12 h and cooled in the desiccators for further use. BAEL, CSC, and FJE were obtained from the local market of Rourkela city. They were shredded using a ball mill and sieved for obtaining the particular granular size. A similar procedure was followed for chemical activation to BAEL, CSC, and FJE as used for BFA, RHA, and STG. The materials were dried and calcined in a muffle furnace starting from room temperature to 500 °C (time period of 4.5 h) and then washed with double-distilled water (DDW) two or three time until the ash contents were removed. To obtain end use adsorbents, the procedure was followed as for BFA, RHA, and STG.

**Received:** June 5, 2011

**Accepted:** September 29, 2011

**Published:** October 20, 2011

Table 1. Experimental Results for CR Uptake onto Various Adsorbents and Their Corresponding S/N Ratio

no.	pH	t	T	C <sub>o</sub>	q <sub>e,PAC</sub>	SNR	q <sub>e,GAC</sub>	SNR	q <sub>e,BFA</sub>	SNR	q <sub>e,RHA</sub>	SNR	q <sub>e,CSC</sub>	SNR	q <sub>e,FJE</sub>	SNR	q <sub>e,BAEL</sub>	SNR	q <sub>e,STG</sub>	SNR
1	2	5	40	10	1.90	5.59	1.99	5.98	1.16	1.29	2.08	6.36	1.67	4.47	1.78	5.02	1.52	3.64	2.25	7.02
2	2	50	45	50	5.38	14.61	2.98	9.48	8.95	19.04	2.10	6.44	9.55	19.60	9.80	19.82	8.47	18.56	49.92	33.96
3	2	100	50	100	24.34	27.73	18.82	25.49	19.99	26.02	20.65	26.30	20.56	26.26	24.75	27.87	13.04	22.31	12.43	21.89
4	2	200	55	200	48.01	33.63	36.28	31.19	45.26	33.11	41.48	32.36	41.08	32.27	49.49	33.89	35.08	30.90	124.93	41.93
5	2	500	60	500	124.57	41.91	73.21	37.29	109.58	40.79	107.08	40.59	105.15	40.44	114.64	41.19	95.66	39.61	24.56	27.81
6	4	5	45	100	22.88	27.19	13.08	22.33	18.49	25.34	6.92	16.80	17.41	24.81	24.66	27.84	17.75	24.98	124.53	41.91
7	4	50	50	200	48.53	33.72	33.98	30.62	46.53	33.35	24.54	27.80	37.02	31.37	49.82	33.95	17.09	24.65	49.90	33.96
8	4	100	55	500	122.55	41.77	97.25	39.76	103.50	40.30	98.82	39.90	92.96	39.37	123.89	41.86	104.21	40.36	16.68	24.44
9	4	200	60	10	2.04	6.20	2.50	7.96	2.07	6.32	1.80	5.11	2.48	7.87	2.46	7.83	2.31	7.27	12.43	21.89
10	4	500	40	50	11.68	21.35	6.68	16.50	1.70	4.61	9.01	19.09	12.26	21.77	12.21	21.74	8.92	19.01	2.30	7.24
11	6	5	50	500	124.42	41.90	3.88	11.78	34.81	30.83	36.42	31.23	17.31	24.76	101.81	40.16	36.87	31.33	1.77	4.95
12	6	50	55	10	2.05	6.22	0.81	-1.83	0.39	-8.18	0.63	-4.01	0.71	-3.02	0.55	-5.12	0.62	-4.15	24.81	27.89
13	6	100	60	50	11.84	21.47	10.34	20.29	12.24	21.76	11.31	21.07	10.62	20.52	12.37	21.85	9.69	19.73	24.76	27.87
14	6	200	40	100	24.70	27.85	15.60	23.86	14.54	23.25	4.49	13.04	21.37	26.60	18.89	25.52	16.53	24.37	2.48	7.88
15	6	500	45	200	47.11	33.46	28.59	29.12	38.77	31.77	38.83	31.78	41.05	32.27	49.61	33.91	40.05	32.05	49.62	33.91
16	8	5	55	50	11.96	21.55	11.52	21.23	10.77	20.64	11.57	21.27	11.71	21.37	12.21	21.73	11.22	21.00	12.40	21.87
17	8	50	60	100	23.37	27.37	24.56	27.80	22.21	26.93	10.95	20.79	11.08	20.89	24.56	27.80	9.60	19.65	22.03	26.86
18	8	100	40	200	46.10	33.27	19.19	25.66	41.06	32.27	29.11	29.28	35.09	30.90	49.37	33.87	34.04	30.64	2.02	6.09
19	8	200	45	500	92.16	39.29	72.52	37.21	70.55	36.97	44.40	32.95	85.17	38.61	116.30	41.31	85.41	38.63	95.20	39.57
20	8	500	50	10	1.64	4.27	2.03	6.15	1.32	2.41	1.62	4.19	2.00	6.02	2.10	6.42	1.95	5.80	49.79	33.94
21	10	5	60	200	34.52	30.76	15.46	23.78	40.68	32.19	0.99	-0.09	40.12	32.07	48.17	33.66	38.23	31.65	46.88	33.42
22	10	50	40	500	118.19	41.45	104.15	40.35	0.97	-0.26	48.72	33.75	120.06	41.59	97.72	39.80	103.13	40.27	1.05	0.39
23	10	100	45	10	2.15	6.66	1.91	5.62	0.59	-4.58	1.60	4.08	1.66	4.39	0.48	-6.38	0.20	-13.98	11.18	20.97
24	10	200	50	50	11.69	21.36	10.17	20.15	7.52	17.52	9.29	19.36	10.15	20.13	12.35	21.84	7.54	17.55	124.91	41.93
25	10	500	55	100	23.26	27.33	23.08	27.26	16.19	24.18	23.12	27.28	20.53	26.25	24.58	27.81	20.51	26.24	124.54	41.91

**Table 2. Response Table for Average S/N Ratio for CR Uptake**

symbol	variables	PAC					GAC							
		mean S/N ratio												
		1	2	3	4	5	1	2	3	4	5	maximum/ minimum	process influencing rank	
pH	hydrogen ions strength	24.69	26.04	26.18	25.15	25.51	1.49	21.88	23.44	16.65	23.61	23.44	6.79	3
<i>t</i>	time (min)	25.39	24.67	26.17	25.66	25.66	1.50	17.02	21.29	23.37	24.08	23.27	7.78	2
<i>T</i>	temperature (°C)	25.90	24.24	25.79	26.10	25.54	1.85	22.47	20.76	18.84	23.53	23.43	4.69	4
<i>C</i> <sub>0</sub>	CR concentration (mg·L <sup>-1</sup> )	5.78	20.06	27.49	32.96	41.26	35.47	4.78	17.53	25.36	28.28	33.28	28.5	1
pH	hydrogen ions strength	24.05	21.98	19.88	23.84	13.80	10.24	34.67	28.21	18.33	19.53	16.74	17.93	3
<i>t</i>	time (min)	22.05	14.17	23.15	23.43	20.75	9.26	11.59	17.38	32.29	20.29	35.93	24.33	2
<i>T</i>	temperature (°C)	12.23	21.70	22.02	22.01	25.59	13.36	18.68	18.77	18.50	35.12	26.42	16.62	4
<i>C</i> <sub>0</sub>	CR concentration (mg·L <sup>-1</sup> )	-0.54	16.71	25.14	32.53	29.72	33.08	1.54	8.65	13.22	26.99	67.08	65.54	1
pH	hydrogen ions strength	24.60	25.03	20.22	23.55	24.88	4.81	25.55	26.64	23.26	26.22	23.34	3.37	2
<i>t</i>	time (min)	21.49	22.08	24.28	25.09	25.34	3.85	25.68	23.25	23.81	26.07	26.21	2.96	4
<i>T</i>	temperature (°C)	25.06	23.93	21.70	23.24	24.35	3.35	25.19	23.30	26.04	24.03	26.46	3.16	3
<i>C</i> <sub>0</sub>	CR concentration (mg·L <sup>-1</sup> )	3.94	20.67	24.96	31.77	36.95	33.0	1.55	21.39	27.37	33.85	40.86	39.30	1
pH	hydrogen ions strength	30.75	30.05	20.75	28.44	33.92	13.17	26.52	25.88	20.50	25.66	27.72	7.22	4
<i>t</i>	time (min)	21.11	27.78	32.23	29.37	33.41	12.30	21.83	24.61	20.25	30.64	28.96	10.38	3
<i>T</i>	temperature (°C)	32.82	30.37	15.29	34.32	31.09	19.03	5.72	34.06	27.33	31.60	27.56	28.34	1
<i>C</i> <sub>0</sub>	CR concentration (mg·L <sup>-1</sup> )	1.32	9.16	15.48	32.89	85.05	83.73	22.34	26.57	28.08	29.86	19.43	10.43	2

**2.2. Adsorbate.** CR was used for the preparation of synthetic aqueous solution of CR of varying concentrations  $C_o = (10 \text{ to } 500) \text{ mg} \cdot \text{L}^{-1}$ . The required quantity of the adsorbate was accurately weighed and dissolved in a small amount of DDW and subsequently made up to 1 L in a measuring flask by adding DDW. Fresh stock solution as required was prepared every day and was kept at ambient conditions in a glass stoppered glass container. The  $C_o$  was ascertained before the start of each experimental run. The pH of the different concentration solutions were adjusted using 0.1 M HCl or NaOH. All of the used chemicals were of analytical grade.

**2.3. Batch Experimental Program.** For each experiment, 50 mL of dye solution of known  $C_o$  and a known amount of the adsorbents were taken in a 100 mL airtight conical flask with a glass stopper. These mixtures were agitated in a temperature-controlled shaking water bath at a constant shaking speed of 250 rpm. The uptake ( $q_t \text{ (mg} \cdot \text{g}^{-1})$ ) of CR by all of the adsorbents at any time,  $t$ , was calculated as:

$$q_t = \left( \frac{C_o - C_t}{w} \right) V \quad (1)$$

where  $C_o$  is the initial adsorbate concentration ( $\text{mg} \cdot \text{L}^{-1}$ ),  $V$  is the volume of the wastewaters in the flask (L),  $C_t$  is the adsorbate concentration ( $\text{mg} \cdot \text{L}^{-1}$ ) after  $t$  time, and  $w$  is the mass of the adsorbent (g) used in the experiment.

**2.4. Analytical Measurements.** The concentration of CR in the aqueous solution was determined by using a spectrophotometer operating in the visible range on absorbance mode. Absorbance values were recorded at the corresponding maximum absorbance wavelength  $\lambda_{\text{max}}$ . The calibration curve of the peak area versus CR concentration was used for the determination of the unknown concentration of CR from a sample. Wherever needed, the sample was appropriately diluted to have the CR concentration in the calibration range.

**2.5. Adsorbent Characterization.** The specific surface area measurement of all of the adsorbents was carried out at Kunash Instruments Pvt. Ltd., Mumbai, by  $\text{N}_2$  adsorption using a Micromeritics instrument (Tristar 3000) and by using the Brunauer–Emmett–Teller (BET) method, using the software of Micromeritics. Nitrogen was used as a cold bath (77.15 K). The zero surface charge characteristics of all prepared adsorbents for CR removal were determined by using the solid addition method.<sup>8</sup>

**2.6. Taguchi Method.** During the past decade, the Taguchi experimental design method has been applied to several physicochemical process.<sup>9–11</sup> Taguchi designs can determine the effect of factors on characteristic properties and the optimal conditions of the factors. This method is a simple and systematic approach to optimize design for performance, quality, and cost.<sup>12</sup> In the Taguchi approach, orthogonal arrays and analysis of variance (ANOVA) are used as the tools of analysis. ANOVA estimates the effect of a factor on the characteristic properties, and the experiment can be performed with the minimum replication using the orthogonal arrays.<sup>13</sup> Conventional statistical experimental design can determine the optimal conditions on the basis of the measured values of the characteristic properties, while the Taguchi method can determine the experimental conditions with the least variability as the optimal condition. Taguchi method uses the signal/noise (S/N) ratio to measure the quality characteristic deviating from the desired value. The experimental conditions having the maximum S/N ratio are considered as the

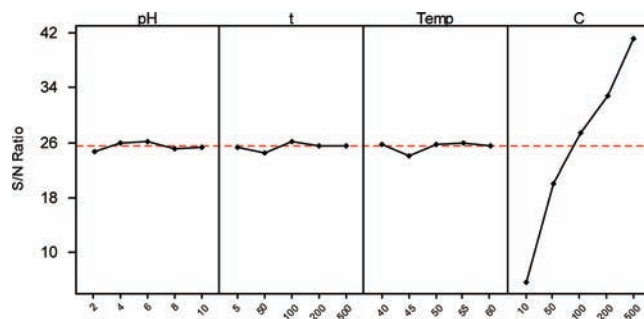


Figure 1. S/N graph for CR uptake onto PAC.

optimal conditions, and the variability characteristics are inversely proportional to the S/N ratio.<sup>14,15</sup>

**2.7. Design of the Experiment.** In the present study, the selected controlling factors are  $C_o$  of CR, pH,  $T$ , and  $t$ . Each factor varied in five levels based on the Taguchi method, and an orthogonal array (OA) was employed to reduce the number of experiments for determining the optimal removal process parameters.  $L_{25}$  OA which has 25 rows corresponding to the number of tests with four columns at five levels was chosen. Table 1 lists the OA with the design factors and their levels. The response variable for accomplishing this study was CR uptake onto commercial and low-cost adsorbents used in the present study. The parameters were optimized with the objective to maximizing the uptake of CR using commercial and low-cost adsorbents.

### 3. RESULTS AND DISCUSSION

The objective of the experiment is to optimize parameters to get a higher uptake of CR onto PAC, GAC, BFA, RHA, CSC, FJE, BAEL, and STG; the higher the ratio, the better characteristics are used. Table 1 shows the actual data for CR uptake with the computed S/N ratio, whereas Table 2 shows the mean S/N ratio for each level of CR uptake. These data were then plotted as shown in Figures 1 to 8 for all of the adsorbents used in the present work.

**3.1. Analysis of the S/N Ratio.** In the Taguchi method, the term “signal” represents the desirable value (mean) for the output characteristic, and the term “noise” represents the undesirable value [standard deviation (SD)] for the output characteristic. Therefore, the S/N ratio is the ratio of the mean to the SD. Taguchi uses the S/N ratio to measure the quality characteristic deviating from the desired value. There are three categories of quality characteristics, that is, the-lower-the-better, the higher-the-better, and the-nominal-the-better. To obtain optimal removal performance, the-higher-the-better quality characteristic was selected. The S/N ratio  $Y$  is defined as

$$Y = -10 \log \left[ \frac{1}{n} \left( \sum_{i=1}^n \frac{1}{y_i^2} \right) \right] \quad (2)$$

where  $n$  is the number of tests and  $y_i$  are the comparison variables in the  $i$ th experiment.

Taguchi recommends analyzing the means and S/N ratio using conceptual approach that involves graphing the effects and visually identifying the factors that appear to be significant, without using ANOVA, thus making the analysis simple.<sup>16</sup>

Table 1 shows the experimental results for CR removal and the corresponding S/N ratio using eq 2. Since the experimental design is orthogonal, it is then possible to separate out the effect

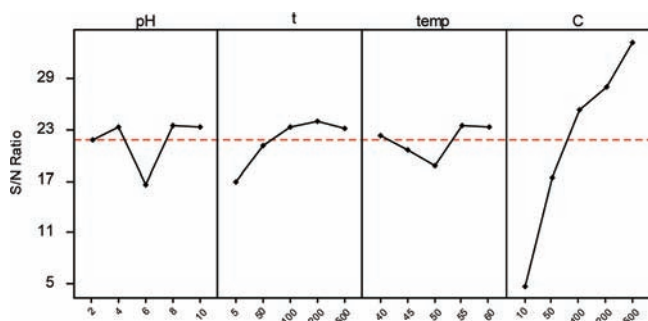


Figure 2. S/N graph for CR uptake onto GAC.

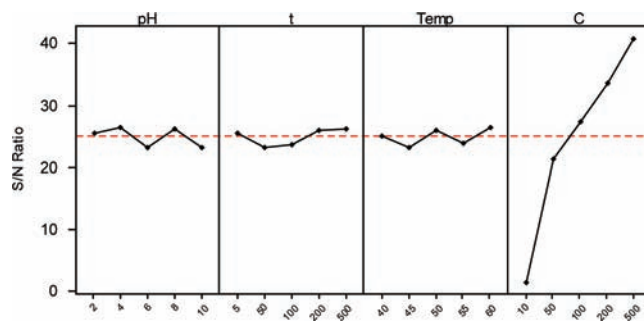


Figure 6. S/N graph for CR uptake onto FJE.

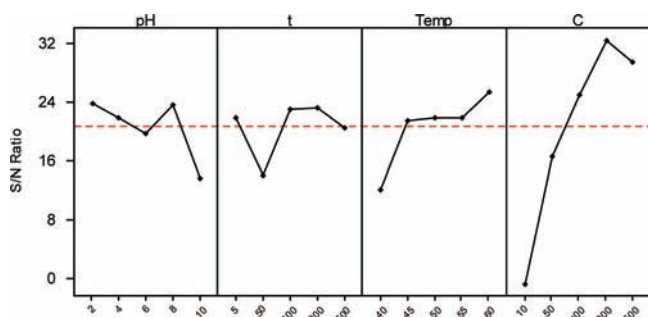


Figure 3. S/N graph for CR uptake onto BFA.

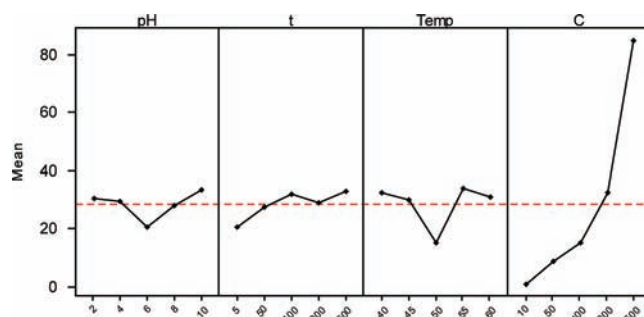


Figure 7. S/N graph for CR uptake onto BAEL.

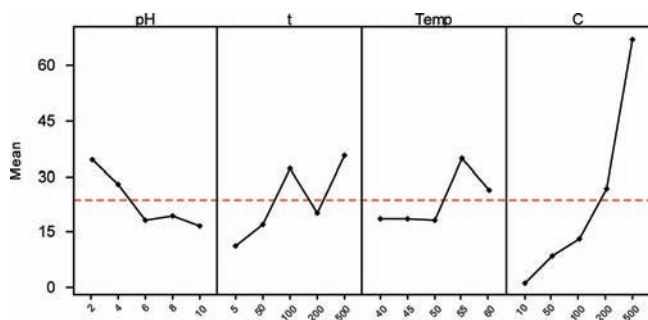


Figure 4. S/N graph for CR uptake onto RHA.

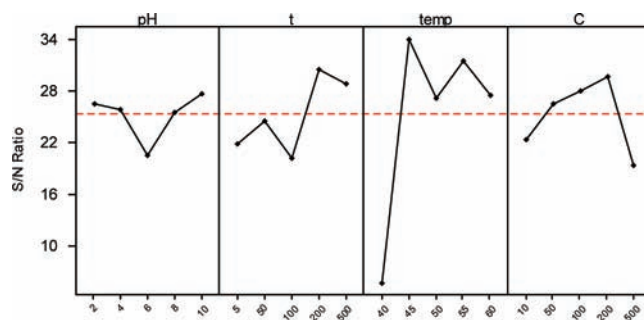


Figure 8. S/N graph for CR uptake onto STG.

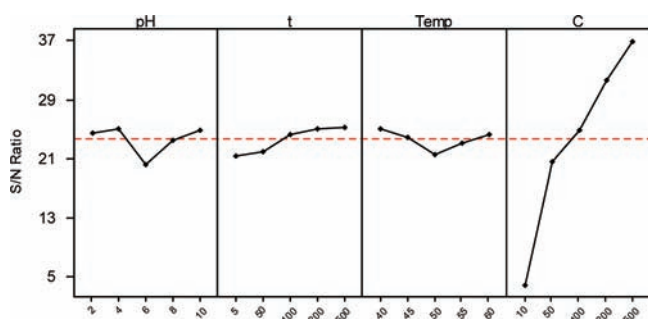


Figure 5. S/N graph for CR uptake onto CSC.

of each parameter at different levels. For example, the mean S/N ratio for pH at levels 1, 2, 3, 4, and 5 can be calculated by averaging the S/N ratios for the experiments 1–6, 6–10, 11–15, 16–20, and 21–25, respectively. In similar manner, the mean S/N ratio for each level can be computed for other parameters.

The mean S/N ratio for each level of parameters used in the present study is summarized in Table 2 and called the S/N response table. In addition, the total mean S/N ratio for the 25 experiments is also calculated and listed in same table. Through Figures 1 to 8, the S/N response graphs can be shown for CR uptake onto individual adsorbents. As shown in eq 2, the greater is the S/N ratio, the smaller is the variance of CR uptake around the desired (the-higher-the-better) value. However, the relative importance among of the process parameters for CR uptake still needs to be known so that optimal combinations of the process parameter levels can be determined more accurately. This will be discussed in the next section using the analysis of variance.

**3.2. Analysis of Variance.** The purpose of the analysis of variance (ANOVA) is to investigate which design parameters significantly affect the quality characteristic. Statistically, there is a tool called an *F* test named after Fisher<sup>17</sup> to see which design parameters have a significant effect on the quality characteristic. The *F* value for each design parameter is simply the ratio of the mean of squared deviations to the mean of squared error. Usually,

**Table 3. Analysis of Variance (ANOVA) for  $q_e$  Using Adjusted Sum of Squares (SS) for Tests**

	source	DF	seq. SS	adj. SS	adj. MS	F	contribution (%)
PAC	pH	1	75	75	75	1.76	0.17
	$t$	1	9	9	9	0.21	0.02
	$T$	1	13	13	13	0.31	0.03
	$C_o$	1	42177	42177	42177	993.25	97.80
	error	20	849	849	42		
	total	24	43123				
GAC	pH	4	1222.1	1222.1	305.5	0.91	5.75
	$t$	4	1738.5	1738.5	434.6	1.29	8.18
	$T$	4	1124.3	1124.3	281.1	0.84	5.29
	$C_o$	4	14475.8	14475.8	3619.0	10.77	68.13
	error	8	2688.0	2688.0	336.0		
	total	24	21248.7				
BFA	pH	4	1987.8	1987.8	497	1.26	8.97
	$t$	4	1370.4	1370.4	342.6	0.87	6.18
	$T$	4	2139.8	2139.8	534.9	1.36	9.66
	$C_o$	4	13488.2	13488.2	3372	8.55	60.92
	error	8	3155.3	3155.3	394.4		
	total	24	22141.5				
RHA	pH	4	1176.4	1176.4	294.1	1.66	6.07
	$t$	4	2106.6	2106.6	526.6	2.98	10.87
	$T$	4	1071.1	1071.1	267.8	1.51	5.52
	$C_o$	4	13599.9	13599.9	3400.0	19.21	70.22
	error	8	1416.0	1416.0	177.0		
	total	24	19369.9				
CSC	pH	4	1233.8	1233.8	308.5	0.89	4.41
	$t$	4	1147.7	1147.7	286.9	0.83	4.10
	$T$	4	1243.8	1243.8	311.0	0.90	4.45
	$C_o$	4	21562.9	21562.9	5390.7	15.63	77.15
	error	8	2759.7	2759.7	345.0		
	total	24	27947.9				
FJE	pH	4	140.7	140.7	35.2	1.89	0.36
	$t$	4	103.5	103.5	25.9	1.39	0.27
	$T$	4	111.6	111.6	27.9	1.50	0.28
	$C_o$	4	38300.1	38300.1	9575	514.53	98.69
	error	8	148.9	148.9	18.6		
	total	24	38804.7	38804.7			
BAEL	pH	4	482.6	482.6	120.7	0.68	1.85
	$t$	4	467.5	467.5	116.9	0.66	1.79
	$T$	4	1184.3	1184.3	296.1	1.68	4.54
	$C_o$	4	22496.8	22496.8	5624.2	31.86	86.38
	error	8	1412.0	1412.0	176.5		
	total	24	26043.3				
STG	pH	4	4330	4330	1083	0.71	9.86
	$t$	4	9737	9737	2434	1.60	21.37
	$T$	4	14009	14009	3502	2.30	30.75
	$C_o$	4	5309	5309	1327	0.87	11.65
	error	8	12159	12159	1520		
	total	24	45545				

when  $F > 4$ , it means that the change of the design parameter has a significant effect on the quality characteristic.

Table 3 shows the results of ANOVA for CR uptake onto adsorbents used in the present work. It can be found that the

**Table 4. Process Parameters for CR Batch Adsorption Study Using the Taguchi Method**

system	parameters				$q_e$ , actual value		optimized value
	pH	$t$	$T$	$C_o$	mg·g <sup>-1</sup>	S/N ratio	mg·g <sup>-1</sup>
CR-PAC	2	50	60	500	124.57	41.91	124.57 ± 2.53
CR-GAC	10	50	40	500	104.50	40.35	104.5 ± 1.73
CR-BFA	2	500	60	500	109.58	40.79	109.58 ± 0.13
CR-RHA	2	500	60	500	107.08	40.59	107.08 ± 0.41
CR-CSC	10	50	40	500	120.06	41.59	120.06 ± 0.86
CR-FJE	4	100	55	500	123.89	41.86	123.89 ± 0.24
CR-BAEL	4	100	55	500	104.21	40.36	104.21 ± 0.96
CR-STG	2	500	60	500	124.93	41.93	124.93 ± 2.57
CR-STG	4	5	45	100	124.53	41.91	124.53 ± 1.93
CR-STG	10	200	50	50	124.91	41.93	124.91 ± 0.56
CR-STG	10	50	55	100	124.54	41.91	124.54 ± 2.45

**Table 5. Surface Area and PZC of Respective Adsorbents**

material	surface area of adsorbent	
	m <sup>2</sup> ·g <sup>-1</sup>	point of zero charge
PAC	1196.85	9.7
GAC	462.67	7.8
BFA	17.38	11
RHA	24.28	9.5
CSC	431.72	8
FJE	109	9
BAEL	391.35	9.7
STG		6

concentration of CR contributes more than 60 % and other parameters contribute < 10 % when the removal pattern is observed onto PAC, GAC, BFA, RHA, CSC, FJE, and BAEL. The concentration of CR is the significant parameter. However, in the case of STG, pH,  $t$ ,  $T$ , and  $C_o$  of CR contribute 9.86 %, 21.37 %, 30.75 %, and 11.65 %, respectively. Therefore, based on the S/N and ANOVA analyses, the optimal parameters for CR uptake are provided in Table 4.

Once the optimal level of the design parameters has been selected, the final step is to predict and verify the improvement of the quality characteristic using the optimal level of the design parameters.

The estimated S/N ratio using the optimal parameters for CR uptake can then be obtained, and the corresponding to concentration can also be calculated by using eq 2. Table 4 shows the comparison of the predicted CR uptake with the actual CR uptake using the optimal CR removal parameters, indicating good agreement between the predicted and actual CR uptake being observed.

**3.3. Adsorbent Characterization.** The BET surface area of the PAC, GAC, BFA, RHA, CSC, FJE, BAEL, and STG were determined and are tabulated in Table 5. The surface area of the adsorbents used in the present work is comparable with the adsorbents available in literature.<sup>3</sup>

The PZC is an important property of carbon–water interfaces. The PZC is defined as the pH at which the carbon surface has no charge in the absence of specific adsorption. The carbon

surface is positively charged at pH values below the  $pH_{PZC}$  and negatively charged at pH values above the  $pH_{PZC}$ .<sup>18</sup> The PZC was estimated using the solid addition method for all of the commercial and low-cost laboratory prepared adsorbents, and their values obtained have also been presented in Table 5.

**3.4. Spent Adsorbents.** The exhausted low-cost adsorbents along with the sorbed CR can be separated from the solution (by filtration), dried, and used as such or as fire briquettes to recover their energy value. The resulting bottom ash blended with the cementation mixture can be used for making building blocks, or it may be used to make fire bricks.<sup>3</sup>

## 4. CONCLUSIONS

The following conclusion can be drawn from the present study:

- The high uptake of CR onto laboratory-prepared adsorbents proves the efficacy of low-cost adsorbents, when it is compared with commercial grade adsorbents used in the present work as well as available in literature.
- The higher the  $F$  value, the more the predominating factor in the CR uptake process is. The ranking of process variables can be done based on their respective  $F$ -values and mean  $S/N$  ratio.
- Among all of the tested adsorbents, STG was found to be the best one because of its high uptake capacity and its good flexibility for selecting the process variables for CR removal.
- Confirmatory tests on optimum conditions indicates that the CR uptake differ with small values.

## AUTHOR INFORMATION

### Corresponding Author

\*E-mail address: arvinpch@yahoo.co.in (A.K.). Tel.: +91 661 246 2268(O); 3268(R), +91 943 83 48807 (mobile). Fax: +91 661 247 2926; 246 2999.

### Funding Sources

The authors are highly thankful to the Director of NIT Rourkela for providing financial assistance.

## ACKNOWLEDGMENT

The authors thank the Department of Chemical Engineering, NIT, and the head of the department for providing research facilities and constant encouragement.

## REFERENCES

- (1) Namasivayam, C.; Yamuna, R. T. Removal of Congo red from Aqueous Solutions by Biogas Waste Slurry. *J. Chem. Technol. Biotechnol.* **1992**, *53*, 153–157.
- (2) Correia, V. M.; Diaper, C.; Judd, S. The use of membranes for the recycling of water and chemicals from dye house effluents: an economic assessment. *J. Soc. Dyers Colour.* **1996**, *112* (10), 273–280.
- (3) Gupta, V. K.; Suhas Application of low-cost adsorbents for dye removal—a review. *J. Environ. Manage.* **2009**, *90*, 2313–2342.
- (4) Reife, A. *Dyes, Environmental Chemistry, Kirk-Othmer Encyclopedia of Chemical Technology*, 4th ed.; John Wiley and Sons, Inc.: New York, 1993.
- (5) Sankar, M.; Sekaran, G.; Sadulla, S.; Ramasami, T. Removal of diazo and triphenylmethane dyes from aqueous solutions through an adsorption process. *J. Chem. Technol. Biotechnol.* **1999**, *74* (4), 337–344.
- (6) Tripathi, P.; Srivastava, V. C.; Kumar, A. Optimization of an azo dye batch adsorption parameters using Box–Behnken design. *Desalination* **2009**, *249*, 1273–1279.

- (7) Mane, V. S.; Mall, I. D.; Srivastava, V. C. Kinetic and equilibrium isotherm studies for the adsorptive removal of Brilliant Green dye from aqueous solution by rice husk ash. *J. Environ. Manage.* **2007**, *84*, 390–400.

- (8) Kumar, A.; Prasad, B.; Mishra, I. M. Adsorptive removal of acrylonitrile by commercial grade activated carbon: Kinetics, equilibrium and thermodynamics. *J. Hazard. Mater.* **2008**, *152*, 589–600.

- (9) Srivastava, V. C.; Mall, I. D.; Mishra, I. M. Multi-component adsorption study of metal ions onto bagasse fly ash using Taguchi's design of experimental methodology. *Ind. Eng. Chem. Res.* **2007**, *46*, 5697–5706.

- (10) Srivastava, V. C.; Mall, I. D.; Mishra, I. M. Optimization of parameters for adsorption of metal ions onto rice husk ash using Taguchi's experimental design methodology. *Chem. Eng. J.* **2008**, *140*, 136–144.

- (11) Suresh, S.; Srivastava, V. C.; Mishra, I. M. Adsorptive removal of phenol from binary aqueous solution with aniline and 4-nitrophenol by granular activated carbon. *Chem. Eng. J.* **2011**, *171*, 997–1003.

- (12) Cox, D. R.; Reid, N. *Theory of the Design of Experiments*; Chapman & Hall/CRC: London, 2000.

- (13) Roy, R. A. *Primer on the Taguchi Method*; Van Nostrand Reinhold: New York, 1990.

- (14) Park, S. H. *Robust Design and Analysis for Quality Engineering*; Chapman & Hall: London, 1996.

- (15) Farzaneh, A.; Ehteshamzadeh, M.; Ghorbani, M.; Vazifeh Mehrabani, J. Investigation and optimization of SDS and key parameters effect on the nickel electroless coatings properties by Taguchi method. *J. Coat. Technol.* **2010**, *7* (5), 547–555.

- (16) Phadke, M. S. *Quality Engineering Using Robust Design*; Prentice-Hall: Englewood Cliffs, NJ, 1989.

- (17) Fisher, R. A. *Statistical Methods for Research Workers*; Oliver and Boyd: London, 1925.

- (18) Álvarez-Merino, M. A.; Fontecha-Cámara, M. A.; López-Ramón, M. V.; Moreno-Castilla, C. Temperature dependence of the point of zero charge of oxidized and non-oxidized activated carbons. *Carbon* **2008**, *46* (5), 778–787.