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SCIENCE DIRECT
Dyes and Pigments 72 (2007) 66–74
DYES and PIGMENTS
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Optimization of process variables by the application of response surface methodology for dye removal using a novel adsorbent

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Received 4 November 2004; received in revised form 12 March 2005; accepted 11 July 2005
Available online 22 September 2005

Abstract

Decolourization of Verofix Red (Reactive Red 3GL) and Lanasyam Brown Grl (Acid Brown 29) from aqueous solution was studied by adsorption technique using a hybrid adsorbent that was prepared by pyrolysing a mixture of carbon and flyash in 1:1 ratio. A 2^4 full factorial central composite design was successfully employed for experimental design and analysis of the results. The combined effect of pH, temperature, particle size and time on the dye adsorption was investigated and optimized using response surface methodology. The optimum pH, temperature, particle size and time were found to be 10.8, 59.25 °C, 0.0525 mm, and 395 min, respectively, for Reactive Red 3GL and those for Acid Brown 29 were 1.4, 27.5 °C, 0.0515 mm and 285 min, respectively. Complete removal (100%) was observed for both the dyes using the hybrid adsorbent.

Keywords: Adsorption; Hybrid adsorbent; Dye removal; Response surface methodology; Statistical analysis

1. Introduction

The coloured dye effluents are considered to be highly toxic to the aquatic biota and affect the symbiotic process by disturbing the natural equilibrium by reducing photosynthetic activity and primary production due to the colourization of water [1]. Effluents contain significant level of organic contaminants, which are toxic as they create odour, bad taste, unsightly colour, foaming etc. These substances are often resistant to degradation by biological methods and are not removed effectively by conventional physio-chemical treatment methods [2]. Removal of dyes from effluents in an economic fashion remains a major problem for textile industries. Adsorption technique has been proved to be an excellent way to treat effluents, offering advantages over conventional process, especially from the environmental point of view [2]. Weber (1978), had identified many advantages of adsorption over several other conventional treatment methods. Carbon is being used as potential adsorbents because of its high efficiency [3]. Raise in the price of activated carbon results in economic difficulties for developing countries like India. Hence, alternate adsorbents with an equivalent potential of activated carbon are current thrust area of research.

The adsorption of dyes on various types of materials has been studied in detail. These include: activated carbon [4] by Dedrick and Beckmann, peat [5], chitin [6] by McKay et al., silica [7] by McKay, the hardwood sawdust [8], hardwood [9] by Asfour et al., bagasse pith [10] by McKay et al., flyash [11,12] by Khare et al. and Gupta et al., respectively, mixture of flyash and coal [13] by Gupta and Prasad, chitosan fiber [14] by Yoshida et al., paddy straw [15] by Deo and Ali, rice

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0143-7208/$ - see front matter © 2005 Published by Elsevier Ltd.
doi:10.1016/j.dyepig.2005.07.018

Nomenclature

C_0	initial concentration of dye solution (mg/l)
C_t	concentration of dye solution at the desired time, t (mg/l)
x_i	dimensionless coded value of the variable, X_i
X_0	value of the X_i at the center point
δX	step change
Y	predicted response
X_1	pH
X_2	temperature (°C)
X_3	particle size (mm)
X_4	time (min)
Exp.	experimental value
Pred.	predicted value

Greek letters

β_0	offset term
β_i	linear effect
β_{ii}	squared effect
β_{ij}	interaction effect
η	removal efficiency (%)

husk [16] by Lee and Low, slag [17] by Ramakrishna et al., chitosan [18] by Juang et al., acid treated spent bleaching earth [19] by Lee and Low, palm fruit bunch [20] by Nassar and bone char [21] by McKay et al.

Conventional and classical methods of studying a process by maintaining other factors involved at an unspecified constant level does not depict the combined effect of all the factors involved. This method is also time consuming and requires a number of experiments to determine optimum levels, which are unreliable. These limitations of a classical method can be eliminated by optimizing all the affecting parameters collectively by statistical experimental design such as response surface methodology (RSM) [22]. RSM is a collection of mathematical and statistical techniques useful for developing, improving and optimizing the processes and can be used to evaluate the relative significance of several affecting factors even in the presence of complex interactions. The main objective of RSM is to determine the optimum operational conditions of the system or to determine a region that satisfies the operating specifications [23]. The application of statistical experimental design techniques in adsorption process development can result in improved product yields, reduced process variability, closer confirmation of the output response to nominal and target requirements, and reduced development time and overall costs [24].

For any batch adsorption process, the main parameters to be considered are pH, temperature, particle size and time [25]. Hence, it is necessary to investigate extensively on the relationship between the adsorption efficiency and the parameters affecting it. Owing to high cost of activated carbon, an adsorbent that is cheap and easily available would be a better alternative. In the present study, a novel adsorbent consisting of 1:1 mixture of carbon and flyash was investigated for its efficiency to remove two classes of dyes namely Reactive Red 3GL and Acid Brown 29 from aqueous solution and their chemical structures are shown in Figs. 2 and 3, respectively. The interaction between the parameters was studied and optimized using response surface methodology.

2. Materials and methods

2.1. Preparation of hybrid adsorbent

Flyash was obtained from Ennore Thermal Power Plant, Chennai, Tamilnadu. The flyash was washed with distilled water, dried under sunlight and subsequently in hot air oven at 60 °C. Hybrid adsorbent was prepared by mixing carbon (supplied by SD Fine chemicals) with flyash in 1:1 ratio by pyrolysing in an isothermal reactor powered by an electric furnace. High purity nitrogen was used as the purging gas. The isothermal reactor was heated to the desired temperature of 650 °C at a heating rate of 15 °C/min, and a holding time of 3 h. After pyrolysis, the product was activated at the same temperature for 3 h using CO_2 as an oxidizing agent and subsequently used as adsorbent. The SEM (scanning electron micrograph) image (Fig. 1) shows irregular and porous structure of the hybrid adsorbent, owing to their exposure to a combustion environment, which indicates very high surface area. Chemical analysis of the hybrid adsorbent showed that carbon was the major constituent along with silica, lime and alumina. The origin of carbon constituents could be reasoned by analyzing the process and material

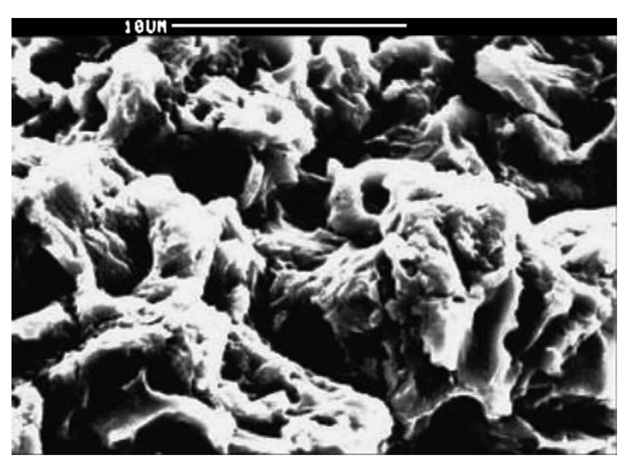

Fig. 1. SEM image of the hybrid adsorbent.

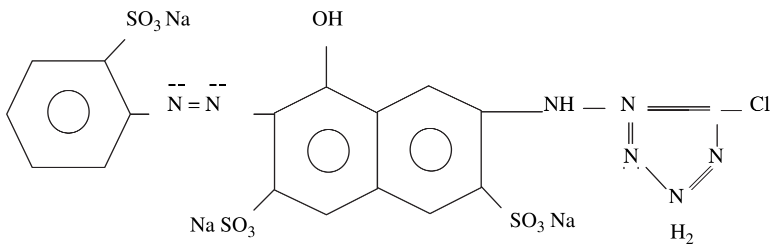

Fig. 2. Structure of Verofix Red (Reactive Red 3GL).

used for carbon manufacture. Silica and alumina content were due to the constituent present in the flyash.

2.2. Methods

2.2.1. Batch adsorption

Stock solutions of dyes (RR 3GL and AB 29) were prepared in deionized water and were diluted according to the working concentration. The required pH was adjusted by adding 0.1 N HCl or NaOH. Dye concentration was measured using UV–VIS spectrophotometer (Shimadzu UV 1600, Japan) at a wavelength corresponding to the maximum absorbance for each dye, 534 nm (λ_{max}, maximum absorbance) for RR 3GL dye and 545 nm (λ_{max}) for AB 29. One hundred milliliters of the dye solution at desired pH value, taken in 250 ml conical flasks, was contacted with 10 g/l of hybrid adsorbent.

The flasks were kept under agitation in a rotatable orbital shaker at 150 rpm for desired time. Experiments were performed according to the central composite design (CCD) matrix given in Table 3. The response was expressed as percent dye removal, calculated as $((C_0 - C_t)/C_0) \times 100$.

2.3. Factorial experimental design and optimization of parameters

Temperature, pH, particle size and time were chosen as independent variables and the efficiency of colour removal as dependent output response variable.

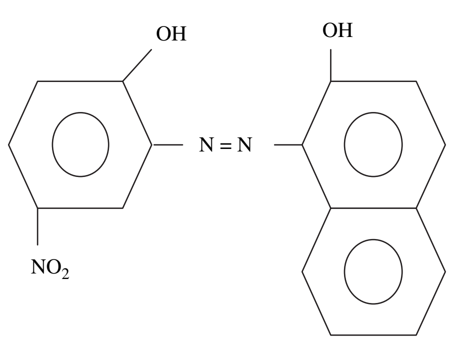

Fig. 3. Structure of Lanasyam Brown Grl (Acid Brown 29).

Independent variables' experimental range and levels for RR 3GL and AB 29 removal are given in Tables 1 and 2, respectively. A 2^4 factorial experimental design [23], with nine replicates at the center point and thus a total of 31 experiments were employed in this study. The center point replicates were chosen to verify any change in the estimation procedure, as a measure of precision property. The coded value of the variable for colour removal efficiency for RR 3GL and AB 29 and experimental plan are also given in Table 3. For statistical calculations, the variables X_i were coded as x_i according to the following relationship:

$$x_i = (X_i - X_0)/\delta X \quad (1)$$

The behaviour of the system was explained by the following quadratic equation

$$Y = \beta_0 + \sum \beta_i x_i + \sum \beta_{ij} x_i^2 + \sum \beta_{ij} x_i x_j \quad (2)$$

The results of the experimental design were studied and interpreted by MINITAB 14 (PA, USA) statistical software to estimate the response of the dependent variable.

3. Results and discussion

The most important parameters that affect the efficiency of adsorbent are time, pH and temperature of the solution and particle size of the adsorbent. In order to study the combined effect of these factors, experiments were performed at different combinations of the physical parameters using statistical experiments.

Table 1
Experimental range and levels of independent process variables for RR 3GL removal

Independent variable	Range and level				
	$-\alpha$	-1	0	1	α
Time	15	30	210	390	570
pH	3.5	6	8.5	11	13.5
Temperature	15	27	43.5	60	76.5
Particle size	0.0343	0.0518	0.3349	0.618	0.9011

Table 2
Experimental range and levels of independent process variables for AB 29 removal

Independent variable	Range and level				
	$-\alpha$	−1	0	1	α
Time	15	30	177.5	325	472.5
pH	0.5	1	4.5	8	11.5
Temperature	18	27	36	45	54
Particle size	0.0343	0.0518	0.3349	0.618	0.9011

The pH range studied was between 3.5 and 13.5 for RR 3GL, and 0.5 and 11.5 for AB 29. The temperature was between 15 and 76.5 °C for RR 3GL, and 18 and 54 °C for AB 29, the particle size was small (0.0585 mm), medium (0.198 mm), large (0.618 mm) and the time varied between 15 and 570 min for RR 3GL, and 15 and 472.5 min for AB 29.

The main effects of each of the parameter on dye removal are given in Figs. 4 and 5 for RR 3GL and AB 29, respectively. From the figures, it was observed that the maximum removal was found to be at 570 min and 325 min for RR 3GL and AB 29, respectively. This indicates that higher the contact time between dye and adsorbent, higher is the removal efficiency till the equilibrium time is reached. The same trend was found to be correct by McKay et al. [6] for dye removal on chitin.

Maximum adsorption for RR 3GL and AB 29 occurred at pH 13.5 and 1, respectively. This is due to the fact that dyes adsorb poorly when they are ionized [26]. This is because of the fact that, when the pH is such that the dyes are in ionized form, the adjacent molecules of the dyes on the hybrid adsorbent surface will repel each other to a significant degree, because of their same electrical charge. Thus, both the dyes RR 3GL and AB 29 that are anionic and cationic in nature, respectively, could not get packed very densely on the hybrid adsorbent surface and at pH value other than 13.5 and 1, respectively, the equilibrium amount of the adsorbed solute is only modest. This explains the common observation that the non-ionized form of anionic and cationic compounds adsorb much better than their ionized counterparts. Cationic species thus adsorb better at low pH and anionic species adsorb much better at higher pH. Hence, the effect of solution pH is extremely important

Table 3
Full factorial central composite design matrix for RR 3GL and AB 29

Observations	pH	Temperature	Particle size	Time	Removal efficiency			
					Reactive Red 3GL		Acid Brown 29	
					Exp.	Pred.	Exp.	Pred.
1	−1	−1	−1	−1	12.25	15.03	55.85	53.01
2	1	−1	−1	−1	44.95	43.89	35.65	26.02
3	−1	1	−1	−1	26.65	27.00	28.65	29.27
4	1	1	−1	−1	76.85	76.79	8.35	8.45
5	−1	−1	1	−1	12.45	13.39	38.25	23.04
6	1	−1	1	−1	32.45	31.95	19.75	19.04
7	−1	1	1	−1	19.45	18.87	12.45	12.26
8	1	1	1	−1	26.85	63.35	4.25	5.56
9	−1	−1	−1	1	27.45	27.57	99.95	99.51
10	1	−1	−1	1	55.75	55.74	77.75	76.03
11	−1	1	−1	1	35.15	44.55	39.15	39.58
12	1	1	−1	1	99.85	99.63	28.35	36.26
13	−1	−1	1	1	16.35	24.85	33.45	38.30
14	1	−1	1	1	48.65	47.71	62.25	61.80
15	−1	1	1	1	41.15	40.32	16.75	15.32
16	1	1	1	1	84.85	89.10	28.95	24.99
17	$-\alpha$	0	0	0	17.45	17.09	33.45	36.21
18	α	0	0	0	87.56	89.74	8.45	9.10
19	0	$-\alpha$	0	0	6.35	30.67	28.25	28.40
20	0	α	0	0	75.65	74.78	8.65	7.860
21	0	0	$-\alpha$	0	59.25	58.69	43.25	44.69
22	0	0	α	0	46.5	46.45	21.55	21.00
23	0	0	0	$-\alpha$	19.35	35.95	17.14	17.14
24	0	0	0	α	62.35	39.04	47.85	47.85
25	0	0	0	0	54.35	54.35	36.35	36.31
26	0	0	0	0	54.35	54.35	36.35	36.31
27	0	0	0	0	54.35	54.35	36.35	36.31
28	0	0	0	0	54.35	54.35	36.35	36.31
29	0	0	0	0	54.35	54.35	36.35	36.31
30	0	0	0	0	54.35	54.35	36.35	36.31
31	0	0	0	0	54.35	54.35	36.35	36.31

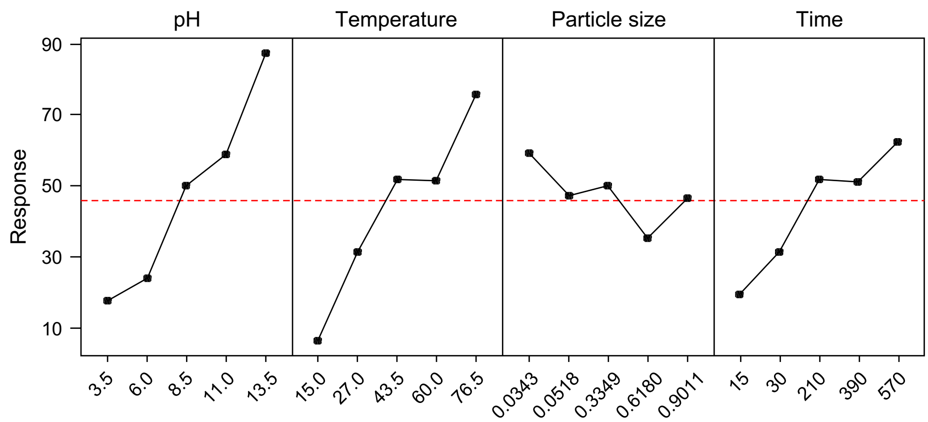

Fig. 4. Plots of main effects of parameters for RR 3GL removal.

when the adsorbing species is capable of ionizing in response to the prevailing pH.

The dyes RR 3GL and AB 29 show different behaviour for the temperature variation. The adsorption of dye at higher temperature was found to be greater compared to that at a lower temperature for RR 3GL dyes and the adsorption efficiency increased with increase in temperature. Similar temperature effects on the adsorption of reactive dye on chitosan beads had also been observed by Chiou and Li [27].

This reveals the fact that in general raising the temperature leads to increase in adsorption, which indicates the kinetically controlling process. RR 3GL is a large molecule having molecular weight of 673 cannot penetrate inside the hybrid adsorbent at the normal temperature. Increase in temperature may produce swelling effect with the internal structure of the hybrid adsorbent enabling the large dyes to penetrate further. But, room temperature (~27 °C) was found to be good for AB 29 for higher removal. This indicates the exothermic nature of the process and is due to the enhanced magnitude of the reverse step in the mechanism as the temperature increases. This reveals the fact that being the small molecules having the molecular weight of 319 have greater vibrational energies and therefore are more likely to desorb from the surface and hence adsorption will be lower if a system is run at higher temperature. Yoshida et al. [14] encountered the similar trend for the adsorption of acid dye on chitosan fiber.

Higher removal efficiency was found in particles of smaller size (0.0585 mm) in both the dyes. The relative increase in adsorption with particles of smaller size may be attributed to the fact that they have larger surface. Small particles will have a shorter diffusion path, thus allowing the adsorbate to penetrate deeper into the adsorbent particle more quickly, resulting in a higher rate of adsorption. In case of large particles, the internal diffusion path is increased and therefore the greater is the probability of encountering smaller pores.

Consequently, the dye uptake decreases with increasing particle diameter. In 2000, McKay had also observed similar results for the metal ions adsorption on bone char [21]. Using the experimental results, the regression equations (second-order polynomial) relating

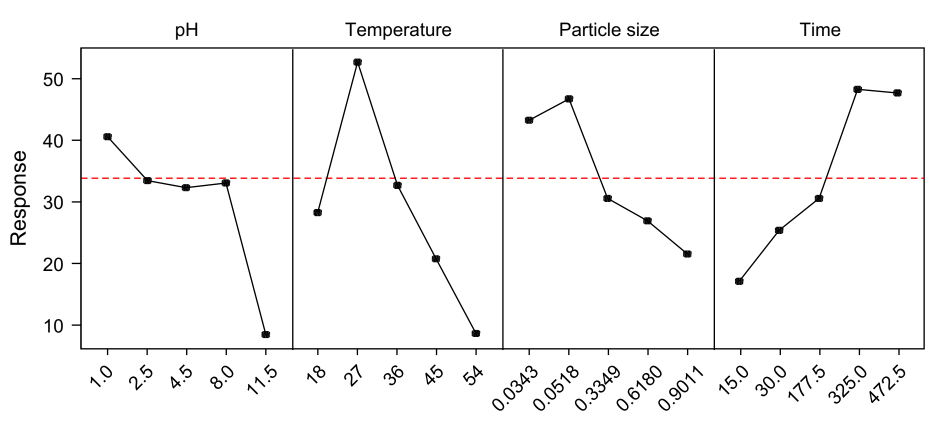

Fig. 5. Plots of main effects of parameters for AB 29 removal.

the removal efficiency and process parameters were developed and are given in Eqs. (3) and (4) for RR 3GL and AB 29, respectively. Apart from the linear effect of the parameter for the dye removal, the RSM also gives an insight into the quadratic and interaction effect of the parameters. These analyses were done by means of Fisher's 'F' test and Student's 't' test.

The Student's 't' test was used to determine the significance of the regression coefficients of the parameters. The P-values were used as a tool to check the significance of each of the interaction among the variables, which in turn may indicate the patterns of the interactions among the variables. In general, larger the magnitude of t and smaller the value of P, the more significant is the corresponding coefficient term [28]. The regression coefficient, t and P-values for all the linear, quadratic and interaction effects of the parameters are given in Tables 4 and 5 for RR 3GL and AB 29, respectively. It was observed that the coefficients for the linear effect of all the factors pH, temperature, particle size, and time ($P = 0.000$ for all) for both the dyes were highly significant except the pH ($P = 0.068$) and particle size ($P = 0.022$) for RR 3GL were slightly less significant.

Regression equation for Reactive Red 3GL

$$\begin{aligned}\eta = {} & (0.644728) + (1.1938906X_1) \\ & + (-0.4527176X_2) + (0.7475007X_3) \\ & + (0.0864869X_4) + \left(-0.0374683X_1^2\right) \\ & + \left(-0.00034454X_2^2\right) + \left(0.5560697X_3^2\right) \\ & + \left(-0.0002466X_4^2\right) + (0.1570959X_1X_2) \\ & + (-1.8743072X_1X_3) + (0.0023883X_1X_4) \\ & + (-0.0802617X_2X_3) + (0.0008422X_2X_4) \\ & + (0.0191760X_3X_4) \qquad (3)\end{aligned}$$

Table 4
Estimated regression coefficient and corresponding t and P-values for RR 3GL

Term	Coefficient	Standard deviation	t	P
Constant	0.644728	3.366	15.624	0.000
X_1	1.1938906	1.938	9.021	0.000
X_2	−0.4527176	1.993	6.505	0.000
X_3	0.7475007	2.180	−2.539	0.022
X_4	0.0864869	2.175	5.446	0.000
X_1X_1	−0.0374683	1.756	−0.337	0.740
X_2X_2	−0.00034454	1.964	−2.064	0.056
X_3X_3	0.5560697	2.314	0.063	0.950
X_4X_4	−0.0002466	2.309	−2.441	0.027
X_1X_2	0.1570959	2.373	1.388	0.184
X_1X_3	−1.8743072	2.373	−1.909	0.074
X_1X_4	0.0023883	2.373	1.546	0.142
X_2X_3	−0.0802617	2.373	−0.940	0.361
X_2X_4	0.0008422	2.373	1.714	0.106
X_3X_4	0.0191760	2.373	1.114	0.282

Table 5
Estimated regression coefficient and corresponding t and P-values for AB 29

Term	Coefficient	Standard deviation	t	P
Constant	1.0567680	3.486	9.934	0.000
X_1	0.8939271	2.471	−1.953	0.068
X_2	1.8246971	2.100	−5.857	0.000
X_3	0.9474099	2.367	−4.404	0.000
X_4	0.1884177	2.355	4.579	0.001
X_1X_1	0.0703798	2.598	−0.629	0.538
X_2X_2	−0.0252470	1.901	−1.452	0.166
X_3X_3	0.9753906	2.536	1.584	0.133
X_4X_4	−0.0001723	2.523	0.020	0.984
X_1X_2	−0.1097849	2.572	0.121	0.905
X_1X_3	0.7527198	2.572	2.134	0.049
X_1X_4	0.0133186	2.572	1.827	0.086
X_2X_3	−0.6908272	2.572	1.784	0.093
X_2X_4	−0.0022975	2.572	−1.565	0.137
X_3X_4	−0.0433983	2.572	−1.215	0.242

Regression equation for Acid Brown 29

$$\begin{aligned}\eta = {} & (1.0567680) + (0.8939271X_1) \\ & + (1.8246971X_2) + (0.9474099X_3) \\ & + (0.1884177X_4) + \left(0.0703798X_1^2\right) \\ & + \left(-0.0252470X_2^2\right) + \left(0.9753906X_3^2\right) \\ & + \left(-0.0001723X_4^2\right) + (-0.1097849X_1X_2) \\ & + (0.7527198X_1X_3) + (0.0133186X_1X_4) \\ & + (-0.6908272X_2X_3) + (-0.0022975X_2X_4) \\ & + (-0.0433983X_3X_4) \qquad (4)\end{aligned}$$

The coefficient of the quadratic effect of temperature ($P = 0.056$) and time ($P = 0.027$) was slightly significant and other two factors did not seem significant with high P-value for RR 3GL. The coefficients of the quadratic effects among the variables did not appear to be very significant in comparison to the linear and interaction effects of AB 29.

However, the interaction effect between pH and particle size ($P = 0.049$), pH and time ($P = 0.086$), and pH and temperature ($P = 0.905$) were considered as significant factors. In the interaction effect none of the variables were found significant except between pH

Table 6
Optimum values of the process parameter for maximum efficiency

Parameter	Optimum value	
	Reactive Red 3GL	Acid Brown 29
η (Efficiency)	100%	100%
X_1 (Time)	395 min	285 min
X_2 (pH)	10.8	1.4
X_3 (Temperature)	59.25 °C	27.5 °C
X_4 (Particle size)	0.0525 mm	0.0515 mm

Table 7
ANOVA for removal efficiency for Reactive Red 3GL: effect of temperature, pH, time and particle size

Source	Degree of freedom (d.f.)	Sum of squares (SS)	Mean square (MS)	$F_{Statistics}$	P
Model	14	15912.8	1136.53	82.61	0.000
Linear	4	13789.3	3588.92	39.83	0.000
Square	4	949.7	237.42	2.63	0.073
Interaction	6	1173.8	195.64	2.17	0.101
Residual error	16	1441.8	90.11	*	*
Lack of fit	10	1441.8	144.18		
Pure error	6	0.0	0.00		
Total	30	17354.6			

$R = 0.9894$; $R^2 = 0.979$.

Table 8
ANOVA for removal efficiency for Acid Brown 29: effect of temperature, pH, time and particle size

Source	Degree of freedom (d.f.)	Sum of squares (SS)	Mean square (MS)	$F_{Statistics}$	P
Model	14	10571.1	755.08	77.33	0.000
Linear	4	8464.3	2053.11	19.40	0.000
Square	4	517.9	129.46	1.22	0.340
Interaction	6	1589	264.83	2.50	0.067
Residual error	16	1693.5	169.85	*	*
Lack of fit	10	1693.5	169.35		
Pure error	6	0.0	0.00		
Total	30	12264.7			

$R = 0.9751$; $R^2 = 0.951$.

and time ($P = 0.074$). The interaction effect would have been lost by conventional method.

These two model equations were optimized using Multistage Monte-Carlo Optimization technique [29]. The optimal values of the process parameters were first obtained in coded units and then converted to uncoded units by Eq. (1) . The optimum values of the process variables for the maximum removal efficiency are shown in Table 6. These results closely agree with those obtained from the response surface analysis, confirming that the RSM could be effectively used to optimize the process parameters in complex processes using the statistical design of experiments.

Although few studies on the effects of parameters on adsorption have been reported in the literature, no attempt has been made to optimize them using statistical optimization methods. The predicted values (using model equations) were compared with the experimental result for both the dyes and the data are shown in Table 3.

The statistical significance of the ratio of mean square variation due to regression and mean square residual error was tested using analysis of variance. ANOVA is a statistical technique that subdivides the total variation in a set of data into component parts associated with specific sources of variation for the purpose of testing hypotheses on the parameters of the model [30]. According to the ANOVA, the $F_{Statistics}$ values for all regressions were higher. The large value of F indicates that most of the variation in the response can be explained by the regression equation. The associated P-value is used to judge whether $F_{Statistics}$ is large enough to indicate statistical significance. A P-value lesser than 0.01 (i.e. $\alpha = 0.01$, or 99% confidence) indicates that the model is considered to be statistically significant [31].

The P-values for all of the regressions were lesser than 0.01. This means that at least one of the terms in the regression equation has a significant correlation with the response variable. The ANOVA table also shows a term for residual error, which measures the amount of variation in the response data left unexplained by the model. The form of the model chosen to explain the relationship between the factors and the response is correct.

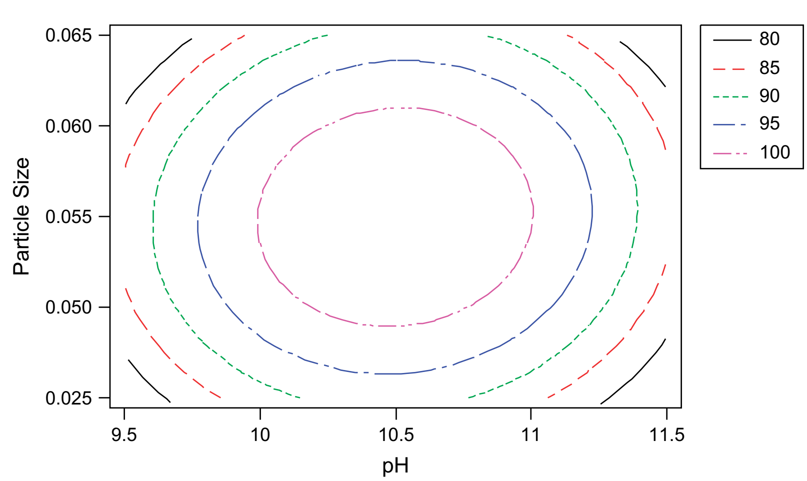

Fig. 6. Response surface contour plot of RR 3GL dye removal (%) showing interactive effect of temperature and pH.

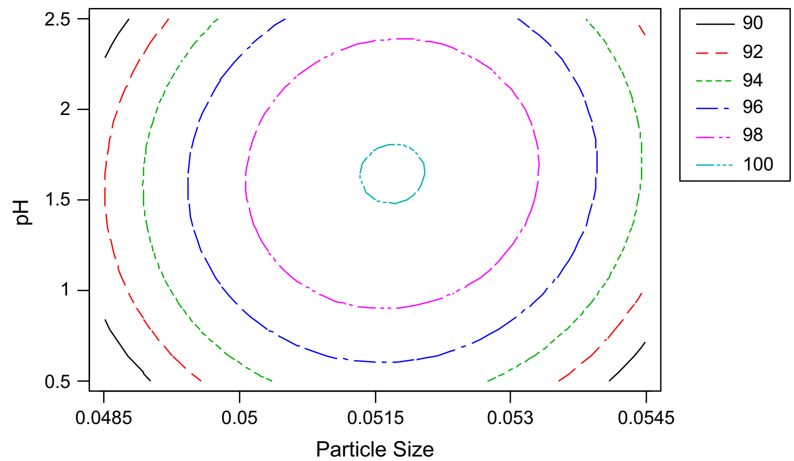

Fig. 7. Response surface contour plot of AB 29 dye removal (%) showing interactive effect of temperature and pH.

The $F_{\text{Statistics}}$ values of 82.61 for RR 3GL and 77.33 for AB 29 are greater than tabulated $F_{14,16}$, the lack of fit can be detected at the α level of significance. It indicates that the fitted model exhibits lack of fit (0.001 for both RR 3GL and AB 114) at the confidence level. The analysis of variance (ANOVA) for RR 3GL and AB 29, respectively, indicated that the second-order polynomial model (Eqs. (3) and (4)) was highly significant and adequate to represent the actual relationship between the response (percent removal efficiency) and the significant variables, with very small P-value (0.00001 for RR 3GL and 0.00001 for AB 29) and a satisfactory coefficient of determination ($R^2 = 0.979$ for RR 3GL and $R^2 = 0.951$ for AB 29) (Tables 7 and 8).

This implies that 97.9% and 95.1% of the sample variation for RR 3GL and AB 29 are explained by the independent variables and this also means that the model did not explain only about 2.1% and 4.9% of sample variation for RR 3GL and AB 29, respectively. The response surface contour plots to estimate the removal efficiency surface over independent variables pH and particle size for both the dyes are shown in Figs. 6 and 7.

The contour plots given in figures show the relative effects of pH and temperature when the remaining variables (time and temperature) are kept constant. The response surfaces of mutual interactions between the variables were found to be elliptical. A similar type of trends was observed for heavy metal removal using biosorbent by Swaminathan [32]. The stationary point or central point is the point at which the slope of the contour is zero in all directions. The coordinates of the central point within the highest contour levels in each of these figures will correspond to the optimum values of the respective constituents. The maximum predicted yield is indicated by the surface confined in the smallest curve of the contour diagram [32]. The optimum values drawn from these figures are in close agreement with those obtained by optimizing the regression equations (3) and (4).

4. Conclusion

The present study clearly demonstrated the applicability of hybrid adsorbent containing carbon and flyash in equal proportion for dye removal. Under optimal values of process parameters, complete removal (100%) was found for both the dyes using hybrid adsorbent.

This study clearly showed that response surface methodology was one of the suitable methods to optimize the best operating conditions to maximize the dye removal. Graphical response surface and contour plot were used to locate the optimum point. A 2^4 full factorial central composite design was successfully employed for experimental design and analysis of results. Satisfactory prediction equations were derived for both the dyes using RSM to optimize the parameters.

Acknowledgements

The financial support for this investigation given by Council of Scientific and Industrial Research (CSIR), New Delhi, India under the grant CSIR Lr. NO 9/468(326)/2003 – EMR-1 dated 12.05.2003 is gratefully acknowledged.

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