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RECENT ADVANCEMENT IN PROCESS OPTIMIZATION USING RSM FOR ADSORPTIVE REMOVAL OF DYES FROM AQUEOUS SOLUTIONS: A REVIEW

Chirag Y Chaware[1]*, Moni U Khobragade[2]

[1] Ph.D. Student, Department of Civil Engineering, College of Engineering Pune, India 411005,

[2] Assistant Professor, Department of Civil Engineering, College of Engineering Pune, India 411005,

* Corresponding author's

ABSTRACT: This study discusses the articles focusing on the recent advancement in adsorption process optimization using response surface methodology RSM. Optimization of independent variables (contact time, temperature, pH, initial dye concentration and adsorbent dose) and dependent variables (% removal). and Box-Behnken design BBD and Central composite design CCD are discussed along with their application and merits. Process flowchart for applying RSM is also discussed. CCD design was found to be popular and preferred design for RSM studies than BBD. Approximately 67% articles studied utilized CCD for response optimization, 30% used BBD for the process optimization also D-optimal design has been utilized.

KEYWORDS: RSM, CCD, BBD, Adsorption, Process optimization

1. INTRODUCTION

Dye wastewater is harmful for the environment as well as the human beings. The wastewater coming from textile industry and various other dye consuming industries release their effluent in the surrounding water bodies leading to contamination of the water bodies with dye. Resulting in various problems like increase in chemical oxygen demand COD, increase in acute and chronic toxicity and discoloration. Carcinogenic and mutagenic effects on aquatic beings[1][2]. To avoid such harmful effects the low cost treatment of the waste generated from such industries is necessary[3]. The treatment or extraction of colour from aqueous solution is extensively studied by different treatment methods like reverse osmosis [4], Photocatalytic degradation [5], electrochemical treatment [6], oxidation process [7], coagulation [8], membrane filtration [9] and adsorption [10]. Adsorption is establish to be effectual and low-cost alternative to treat dye wastewater[11].

The application of soft computing tools for process optimization has resulted in acceptability and application of the developed process. Response surface methodology is helpful in modelling, developing, and optimizing the response of the adsorption process [12][13][1].

In this study the recent journal articles were searched from the science direct article finder tab. The key words used for the search were "Dye removal optimization using RSM". The obtained articles were checked and used in this paper for further analysis.

2. RESPONSE SURFACE METHODOLOGY

Response Surface Methodology (RSM) is a compendium of mathematical and statistical methods used in optimization of process its design as well as improvement. RSM is useful in improvement of the existent process and products. In the field of creation of new products, their design and development RSM is applied. The optimal answer is obtained from RSM as it can explain the association between the independent variables (input variables) and responses (one or more) also the interactions of the independent variables are explained and analyse [14].

Consider any dependent variable "y" and there is a set of input variables (independent variable) x1, x2, …, xk (For e.g. y might be the Removal of colour and x1, x2, and x3might be the reaction time, the reactor temperature, and adsorbent dosage in the process and ε is error factor).

$$y = f(x1, x2, \ldots, xn) + \varepsilon \quad \ldots\ldots\ldots (1)$$

RSM can be applied to optimize the process in three phases i.e. phase zero, phase one and phase two. Starting from phase zero the preliminary screening experiments are carried out to screen and select the independent variables (selecting influential variables and omitting insignificant one). In phase one to determine the ideal response region is the main objective. To check whether the variables are consistent. The final phase two the process is near optimal, and the response functions shall lie in the region near the optimum. The confirmatory experiments are performed according to the optimal values gained from the RSM studies and compared with the predicted responses. There are various soft computing tools available to study RSM like Design expert, JMP, SAS and MATLAB. Kulkarni et al. used porous and high surface area fullers clay for the extration of methylene blue dye and indicated the industrial application of the material for contaminant removal. The process was optimized by using Artificial neural networks (ANN). pH was found to be the most important influencing parameter at 29% importance amongst, initial dye concentration, pH, agitation speed, contact time, temperature and adsorbent dose. The adsorption experiments data follows pseudo second order kinetics model, physisorption has taken place on the surface of adsorbent as suggested by kinetics and 28 kJ mol^{-1} energy of activation [15] The process flow chart of RSM application is shown in the figure 1.

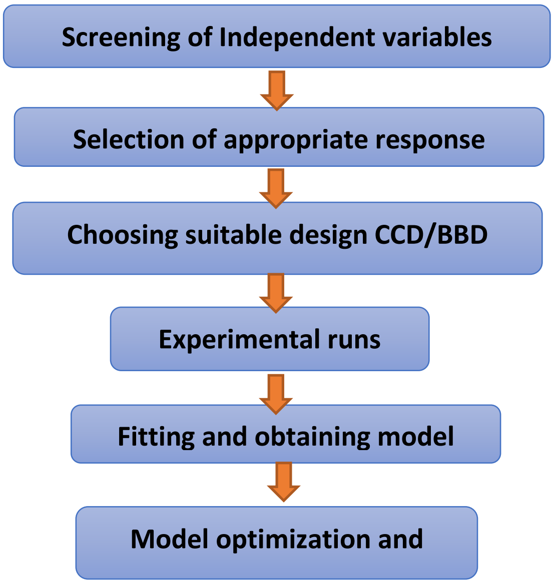

Figure 1: Process flow for application of RSM

2.1 BOX-BEHNKEN DESIGN BBD

A second order design containing three levels developed by Box and Behnken in the year 1960 is known as Box-Behnken design (BBD). The design of experiments in BBD and be done in fewer runs as compared to other designs like CCD for the same no of independent variables as in this design the design space is limited to the actual level of experimental levels. BBD was used to optimize the parameters(pH, Initial dye concentration and Adsorbent dose) affecting the adsorption process to obtain optimal removal efficiency [16]. Many researchers are using BBD design as it requires less runs and is helpful in saving the high experimental cost. BBD design avoids experimentation at extreme levels. Chabane and Bouras used extrusion method for the preparation of reinforced porous hybrid beads adsorbent. Removal efficiency was maximised using Box Behnken design in response surface methodology (RSM). ANOVA results and R^2 value indicates strong co- relation between response and independent variables (pH, initial concentration, and adsorbent dose). pH and adsorbent dose were having significant influence on removal efficiency. Optimal removal efficiency can be achieved at the following range of parameters pH 5.01, adsorbent dose 1.03 g L^{-1} and initial concentration of 92.36 mg l^{-1} [17].

Table 1:- Summary of BBD designs and the optimal responses

Dye	Adsorbent	RSM			Optimal Conditions	Optimal response	Reference
		Model	Independent variables	Response			
Methylene blue (MB), Congo red	Biochar	BBD	pH, temperature, Pyrolysis temperature	%Removal	pH-7, T-30ºC	68% and 74%	[18]
(MB)	Ageratum Conyzoides leaves	BBD	pH, Initial dye concentration, Adsorbent weight	% Removal	pH-4, m-60mg, C_0- 20 mg/l	91%	[19]
(MB)	Crocus Sativus	BBD	contact time, Initial concentration, Adsorbent dose	%Removal	Contact time-56min, C_0-176mg/l, m-1.78g/l	89.48%	[20]
Eriochrome black T	B-CuFe composite	BBD	Temperature, contact time, pH	%Removal	-	70% to 85%	[21]
Reactive blue	Kaolin clay composite	BBD	Adsorbent dose, Contact time, pH	%Removal %COD reduction	pH-4, time-30min m-0.06g/l	70.05%	[22]
Acid orange	CaO/CeO_2 composites	BBD	pH, Adsorbent dose, Initial dye concentration	%Removal	pH-2, m-0.1g, Co-10mg/l, T-301K	92.68%	[23]

2.2CENTRAL COMPOSITE DESIGN CCD

Central composite design CCD is a fractional or full factorial design developed by Box and Wilson. CCD is commonly used by researcher to optimise and the response and study the response surface methodology. [24] Used the central composite design to optimize the response (%removal of methylene blue), the independent variables were pH, bio adsorbent dose and initial dye concentration. The experiments consist of 30 runs in total. Ali et al. has utilised activated charcoal for the uptake of malachite green dye from aqueous solutions. Reaching maximum adsorption capacity of 27 mg g^{-1} for the removal of dye. Efficiency was ascending for the increase in pH of the solution above pH 5 and increase in removal efficiency for decrease in pH below pH 5. Thermodynamics study reveals that the reaction is exothermic and spontaneous and the experimental data was fitted to pseudo second order kinetic model [25]

Table 2:- Summary of CCD designs and the optimal responses

Dye	Adsorbent	RSM			Optimal Conditions	Optimal response	Reference
		Model	Independent variables	Response Removal (%)			
Malachite Green	Boron mesoporous carbon nitride	CCD	pH, Temperature, Adsorbent weight, Initial dye concentration	% Removal	pH-5, T-room temperature, m- 20mg, C_0- 20 mg/l	100%	[26]
(MB)	Ho-$CaWO_4$ nanoparticles	CCD	pH, Adsorbent dose, contact time, Initial concentration	%Removal	pH-2.03, contact time-15.16min, C_0-100.65mg/l, m-1.91g/l	71.17%	[27]
(MB)	Activated carbon	CCD	pH, Adsorbent dose, contact time, Initial concentration	%Removal	pH-11, contact time-50min, C_0-10mg/l, m-1.4g/l	87.09%	[28]
Eriochrome black-T	HCl modified clay	CCD	Temperature, contact time, Adsorbent dose, pH	%Removal	T-35ºC, m-400mg, pH-1.17	96%	[29]
(MB)	Cellulose Nanocrystals	CCD	pH, Contact time, Adsorbent dose	%Removal	pH-2, m-400mg, contact time-14min	76%	[30]
(MB)	Cashew nut shell activated carbon	CCD	pH, Adsorbent dose, contact time, Initial concentration	%Removal	pH-10, m-400mg, contact time-14min	94%	[12]

Table 3:- Summary of designs used for optimization of responses

Dye	Adsorbent	RSM			Optimal Conditions	Optimal response	Reference
		Model	Independent variables	Response Removal (%)			
Methyl Orange	Magnetic nanocomposite	CCD	Adsorbent dose, contact time, Initial concentration	Yes	contact time-24min, C_0-98.37mg/l, m-0.58g/l	99.88%	[31]
Textile Industry Effluent	CNT-Alg-Fe3O4	CCD	pH, Adsorbent dose, contact time	Yes	pH-3, contact time-85.55min, m-10g	98.43%	[32]
(MB)	Crocus Sativus	BBD	contact time, Initial concentration, Adsorbent dose	Yes	Contact time-56min, C_0-176mg/l, m-1.78g/l	89.48%	[20]
Congo Red	Activated carbon	D-optimal	Agitation speed, contact time, Initial concentration	Yes	Contact time-140min, C_0-300mg/l, speed-165rpm	79.7%	[33]

Dye	Adsorbent	RSM			Optimal Conditions	Optimal response	Reference
		Model	Independent variables	Response Removal (%)			
FD&C Red 40	CS-TiO_2-GLA beads	CCD	pH, Adsorbent dose, Initial dye concentration	Yes	pH-1.73, C_0-55.23mg/l, m-279.77mg	100%	[34]
Rhodamine B and Erythrosine B)	Activated carbon	CCD	pH, Adsorbent dose, Contact time	Yes	pH-4, m-0.3g/l, contact time-21min	99%	[35]
(MB)	Activated carbon	BBD	Adsorbent dose, Contact time, Initial dye concentration	Yes	C_0-100mg/l, time-13h, m-2g	99.99%	[36]
Rhodamine B	MIL-100(Fe)	CCD	pH, Adsorbent dose, Initial dye concentration	Yes	-	99.99%	[37]
Eriochrome black T	B-CuFe composite	BBD	Temperature, contact time, pH	Yes	-	70% to 85%	[21]
Crystal violet	Date palm leaves	CCD	pH, Contact time, adsorbent dose, Initial dye concentration, Temperature	Yes	pH-10.0, T-21.10min, m-48.64g/l, Co-16.35mg/l, Temp-55.92ºC	99.5%	[38]
Direct blue-86	Activated carbon	CCD	Adsorbent dose, Initial concentration, pH	Yes	m-24.65g/l, pH-3.1, Co-125.5mg/l	98.4%	[39]
Nile Blue	Lignocellulosic agricultural waste	CCD	Adsorbent dose, Initial Concentration, pH, Contact time	Yes	m-4.8g/l, Co-539mg/l,pH-8.88, Time-114min	-	[40]
Disperse blue 79	PACl based water treatment residuals	CCD	pH, Adsorbent dose, Initial dye concentration	Yes	pH-3, m-30g/l, Co-75mg/l	52.6%	[41]
(MB)& Acid red	Modified Oak Waste	CCD	pH, contact time, Adsorbent	Yes	pH-6.2, time-160min, m-	85.36% 41.27%	[42]

			dose, Initial dye concentration		2.0g/l, Co-70mg/l		
Remazol Brilliant blue R	Polymetric adsorbent	CCD	contact time, Adsorbent dose, Initial dye concentration	Yes	Co-60.85mg/l, m-0.04mg/50ml, time-59.91min	99.85%	[43]
Methylene blue	Agaricus campestris	CCD	Agitation speed, initial dye concentration, Temperature	Yes	Co-130.90mg/l, speed-125rpm, T-41.87ºC	95%	[44]
Reactive blue 4	GA-crosslinked CS beads	BBD	pH, Adsorbent dose, Co-5mg/l	Yes	pH-2, m-0.6g, Co-5mg/l	60.65%	[45]
Reactive orange16	Fe_3O_4 composite	BBD	pH, Adsorbent dose, Contact time, Temperature	Yes	pH-4, m-0.08g, T-30ºC, time-55min	73.1%	[46]
Reactive red 198	MSW compost ash	BBD	Contact time, Adsorbent dose, Initial dye concentration	Yes	Time-80min, m-2g/l, Co-20mg/l	92.8%	[47]

Dye	Adsorbent	RSM			Optimal Conditions	Optimal response	Reference
		Model	Independent variables	Response Removal (%)			
Malachite Green Auzamine-O	NaX nanocomposites	CCD	pH, Ultrasonic time, Adsorbent dose, initial dye concentration	Yes	pH-8, m-347mg, Co-4mg/l, U-time-11.5min	99.07% 99.61%	[48]
Acid red 18	Granular ferric hydroxide	CCD	pH, Adsorbent dose, Contact time, Initial dye concentration	Yes	pH-5, m-2g/l, Co-77.5mg/l, Time-77.5min,	78.59%	[49]

Crystal violet	Natural zeolite	CCD	pH, Temperature, Adsorbent to adsorbate ratio	Yes	pH-10, Temperature-25ºC. Adsorbent to adsorbate ratio- 0.1g/g	-	[50]
Methylene blue	Zinc oxide nanocomposite	CCD	Adsorbent dose, contact time, pH, initial dye concentration	Yes	pH-6, time-8.5min, m-0.002g, Co-5ppm	98.17%	[51]
Methylene blue	Hydrogel	CCD	Adsorbent dose, Initial dye concentration	Yes	m-550mg. C_0-5.50mg/l	95.46%	[52]

3. Trends in application of RSM for dye removal

Among the articles reviewed in this study it has come to light that researcher have preferred CCD design more than BBD and there is a start to apply D-optimal design for getting optimal responses. CCD was applied 67% and BBD 30% from the total articles reviewed. Also, most of the research have compared their RSM model results with ANN model or different soft computing tool. The figure 2 represents the pie chart explaining the utilization of different designs for optimization of adsorption process and their respective responses. Figure 3 is representing the types of dye whose removal efficiency was studied by applying RSM. And it was observed that 48% of the research focus on removal of cationic dyes (MB, MG etc). followed by azo dyes 32%, 13% for fluorescent dyes and 7% for anionic dyes. Some papers are also focusing on textile industry effluent treatment modelling using RSM.

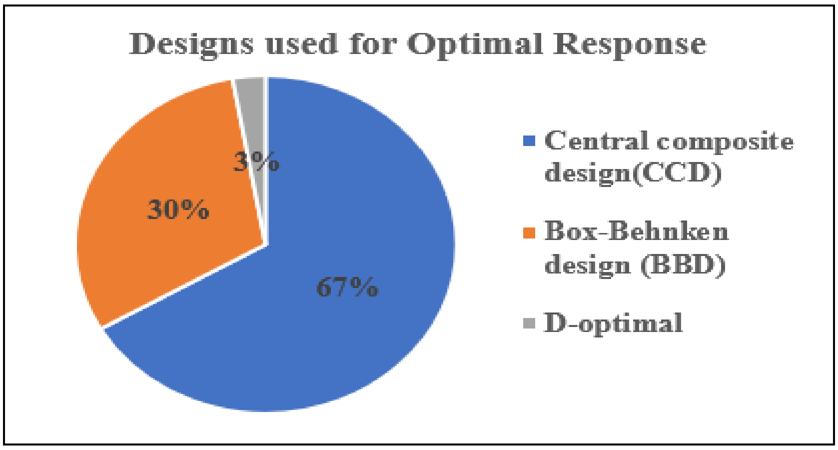

Figure 2:- Pie chart representing designs used for the optimization of responses

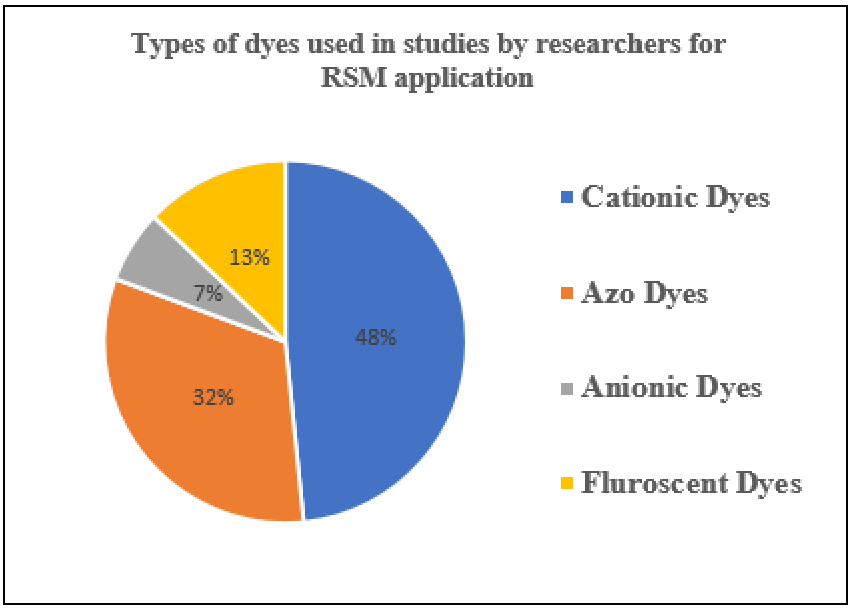

Figure 3:- Pie chart representing types of dyes used by researchers for RSM application

4. CONCLUSION

Response surface methodology is helpful in design, analyse and optimization of the adsorption process of various dyes from aqueous solutions. Both Central composite design CCD and Box-Behnken Design BBD are effective in optimization of parameters (independent variables) and prediction of responses for removal of dyes. BBD is used where there is constrained to obtain maximum information in minimum number of experiments. CCD is useful in better estimation of RSM curves due to its inclusion of the axial star points for the experimentation. Many researchers have arrived at the optimal response (% Removal) of more than 99% and verified the findings with the experimental observations. Application of RSM has helped in saving time as well as reducing cost of the experimentation. It is observed that application of RSM to optimize the adsorption process results in better understanding and graphical visualization of the process.

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