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Dye removal from textile waste water using potato starch: parametric optimization using Taguchi design of experiments

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Abstract: Typical textile waste water contains a high concentration of spent dye that can pose serious destructive impact on aquatic environment. Therefore, treatment of textile industry effluents is strictly imposed by the relevant government authorities and environmental protection agencies. During present studies, spent dye was removed using potato starch, an environmental friendly, biodegradable and cost effective coagulant, through coagulation/flocculation process. Analysis of variance (ANOVA) was performed that indicated the interaction of process parameters. It was observed that the interaction of temperature, pH and coagulant dosage were the most significant parameters that can affect the coagulation/flocculation process. So, temperature, pH and coagulant dosage were optimized by Taguchi optimization technique. The results indicated that maximum dye (about 27%) was removed when temperature, pH and coagulant dosage were kept at 55°C, 10 and 0.5% (w/v), respectively.

Introduction

Industries, especially textile and paper, use dyes in order to color their products and also consume substantial volumes of water (Gao et al. 2007). As a result, they generate a considerable amount of colored effluents. It is estimated that about 10 to 15% of the total dye produced is disposed of during dying operation (Gupta 2009). Further the presence of very small amounts of dyes in water (less than 1 ppm of some dyes) is highly visible and undesirable as they hinder to the aerobic digestion (Iqbal et al. 2007). Various methods, like coagulation, biological treatment, flotation, adsorption, oxidation and hyper filtration, were used for the treatment of waste water from dying industries (Sanghi et al. 2006). However, the coagulation attracts the attention of researchers as it is not only regarded as the most successful pretreatment but also cost effective (Kim et al. 2004, Zemaitaitiene et al. 2003). A variety of natural and synthetic adsorbents, like activated carbon, agriculture wastes, biopolymers, humin, eggshell and biocomposites were used for the removal of reactive dyes from the aqueous solution (Elkady et al. 2011, Jesus et al. 2011). Various approaches were investigated to develop efficient and cost effective coagulants consisting of natural polymers (Wu et al. 2001). Polysaccharides such as chitin and starch , and their derivatives like chitosan and cyclodextrin attracted more attention due to their particular structure, physiochemical properties, chemical stability, high reactivity and excellent selectivity towards aromatic compounds and metals and the presence of reactive groups (hydroxyl, acetamid or amino functions) in polymer chains (Wu et al. 2001, Del Valle 2004). Moreover, polysaccharides are naturally abundant and biodegradable, which have the ability to associate themselves with wide verity of molecules by physical or chemical interactions (Ciesielski et al. 2003). Hence adsorption on polysaccharide derivatives can be a low-cost procedure of choice in water decontamination, and a useful tool for protecting the environment. Efforts have been made to remove toxic compounds by adsorption using natural polymers which indicates the importance of polysaccharides. It is proposed that starch, a crossed linked biopolymer, can be used as an adsorbent for the recovery of organic pollutants from the aqueous solution (Delval et al. 2000). It is further reported that the sorption capacity of polysaccharides depends upon their origin, the degree of N-acetylation, and molecular weight (Berger et al. 2004). The size of particles has also been proved a key parameter in the control of adsorption capacity as it varies randomly due to variation in the particle size (Chiou and Li 2002). So, the adsorption capacity of an adsorbent depends on its source of raw material, which means that there is a space for the use of starch extracted from different sources for the decoloration process.

The current studies focus on the use of potato starch, as a replacement of currently used polysaccharides, for the removal of reactive dyes from textile effluent. The aim of this study is to investigate the optimum decoloration process parameters using potato starch as a coagulant. A robust experimental design, based on parameters (coagulant dosage, temperature and hydrogen ion concentration) and their levels, was developed for reducing cost and improving the decoloration of textile waste water.

Taguchi optimization process

During present studies, Taguchi method was used to determine the optimum parameters (i.e. pH, temperature and dosage of coagulant) for the removal of dye from its aqueous solution using potato starch as a coagulant. In Taguchi method, the word “optimization” means “determination of BEST levels of control factors”. The BEST levels of control factors are those that can maximize the Signal-to-Noise ratios. The Signal-to-Noise ratios are logarithmic functions of desired output characteristics. The experiments, that are conducted to investigate the BEST levels, are based on “Orthogonal Arrays”, are balanced with respect to all control factors which are minimum in number which indicates that the resources (materials and time) required for the experiments are also limited. The use of a quantitative design in the Taguchi method in order to optimize a process with multiple performance characteristics includes the following steps: (Taguchi 1987).

1) Identification of the performance characteristics and selection of process factors that should be evaluated
2) Determination of the number of quantity levels for the process and possible interaction between the process parameters
3) Selection of the appropriate orthogonal array and assignment of process factors to the orthogonal array
4) Conducting the experiments based on the arrangement of the orthogonal array
5) Calculation of the performance characteristics
6) Analyzing the experimental result
7) Conducting the verification experiments

F factor

Process factor (f factor) test is a tool to perceive which parameter has a significant effect on the performance. The f value for each process parameters is simply a ratio of mean of the squared deviations to mean of the squared error. The f test was carried out to determine the most effective and the least effective parameters. Generally, the larger the f-value, the greater is the effect of process parameter. With the ANOVA analyses, the optimum combination of process parameters can be predicted (Freund and Littell 1981). Using Taguchi process, each parameter of signal-noise graphics was drawn to determine the maximum decolouration conditions.

Material and methodology

Dye characteristics

During current investigations, the Reactive Yellow dye, also known as Yellow MERL, was collected from Sandal Dye & Dye Stuff Industries Limited Faisalabad, Pakistan. It is a single AZO class, Bi-functional (having reactive groups MCT and VS) dye, having molecular weight of 1026.25, formula $C_{28}H_{20}ClN_9Na_4O_{16}S_5$ and considered as the most useful for the dying of cotton fabric in the textile sector (Christie 2001).

Characteristics of polysaccharide

Extracted potato starch (DAE JUNG, Cat # 7662-1405), of reagent grade, in the form of white powder, having molecular formula of $[C_6H_{10}O_5]_n$, molecular weight of 342.296480 [g/mol] and Specific Gravity of 1.65.

Stock solution

Synthetic dyeing effluent was prepared to minimize the external disturbances which may be present in the effluents of a typical dye-house. 150 mg·dm^{-3} of aqueous solution of dye was prepared in RO water, which is a typical high end concentration of dye effluent (Figueiredo, Boaventura et al. 2000). Then, 10 mg·dm^{-3} of sodium carbonate was added to the dye solution followed by heating to 60°C for one hour. Further, the pH of the solution was raised to 10.5 by addition of 1M sodium hydroxide. The solution was heated at 60°C for one hour and cooled to room temperature (Blackburn 2004).

Testing

The stock solution was first filtered (with 11 μm pore size filter) and then analyzed for absorbance using a ZAR/HEC-1830/UV-2800 spectrophotometer. While the maximum wavelength of adsorption for each batch of filtered stock solution was determined in the range of 420–436 nm.

Experimental procedure

An experimental setup was developed to carry out the series of coagulation-flocculation tests. Three parameters were investigated, i.e., pH, temperature and percentage coagulant dosage, see Table 1. An orthogonal array (OA) experimental design was selected which was reported as the most suitable method, i.e., L9 (Robinson et al. 2004). Jar test procedure was adopted for coagulation & flocculation process. Initially, the absorbance of a small volume of filtered stock solution was measured using spectrophotometer to figure out the initial concentration of dye. 5 grams (0.5% w/v) of potato starch along with one liter of filtered stock solution was first agitated for 5 minutes at 100 rpm and, then, for 30 minutes at 40 rpm. Afterward, the agitation was ceased and suspension was allowed to settle for half an hour. pH of the solution was controlled by using NaOH (1 M) and HCl (1M). Finally a small solution was pipetted out to find the absorbance, which represented the final concentration of dye in the sample. The schematic diagram of the experimental setup is shown in Fig. 1.

Each experiment was repeated for three times to find out the effect of noise resources precisely. The-larger-the-better performance characteristic was chosen as the optimization criteria, which was calculated by using equation (1), (Taguchi 1987).

$$SN = -10\log\left[\frac{1}{n}\sum\frac{1}{Y_i^2}\right] \tag{1}$$

Where, larger-the-better, is performance characteristics which was calculated for the decoloration rate, n number of repetitions performed for an experimental combination, Yi is percentage eradication of dye, which is measured in terms of reduction in absorbance for ith experimental run. The SN for the larger-the-better for decoloration rate was calculated which has a higher value, determines the maximum level of

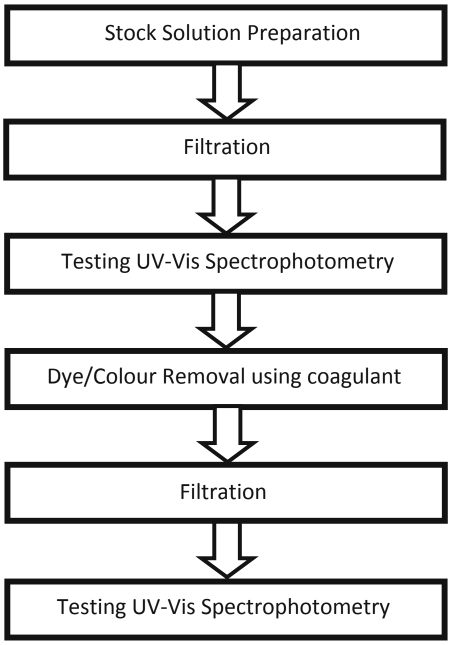

Fig. 1. Schematic diagram of experimental procedure

each factor. Only those factors were employed during present *SN* studies that can potentially affect the coagulation process. The factors and their levels, determined in the preliminary tests, are given in Table 1, while the experimental results and calculations are represented in Table 2.

Results and discussion

Effect of coagulant dosage

The statistical effect of percentage dosage of coagulant, i.e., potato starch, is represented in Fig. 2. It can be observed that the signal-noise ratio (larger-the-best, performance characteristic) is the maximum at A0, see Table 1, condition of percentage coagulant dosage, i.e., 0.5% (w/v, i.e., 5 gram of potato starch in 1000 ml of dye stock solution). Further, the percentage reduction in the absorbance, which corresponds to the dye removal from the aqueous solution, is also the maximum at A0. Moreover, the increments in the amount of the coagulant result in the reduction of the dye removal, i.e., the percentage reduction in the absorbance decreases, see Fig. 2. The initial high value of the dye removal at 0.5% (w/v) coagulant dosage was due to the availability of more surface area for adsorption of dye

Table 1. Parameters and their values corresponding to their levels for Taguchi Process

Parameters & their values corresponding to their levels				
Symbol	Parameter	Parameter Level		
		0	1	2
A	Coagulant Dosage (%)	0.5	1	2
B	pH of waste water	3	7	10
C	Temperature of waste water (°C)	25	40	55
D	Dye concentration (ppm)	Fixed at 0.15% (w/v %), 150 $mg \cdot dm^{-3}$		
E	First stage mixing speed (RPM)	Fixed at 100 (For 5 min, Coagulation time)		
F	Last stage mixing speed (RPM)	Fixed at 40 (For 30 min, Flocculation time)		

PPM = parts per million

Table 2. L9 Experimental plan and results

Experimental Results for Parameters & their values corresponding to their levels

Experiment #	Parameters & Levels			% age Colour Removed (Y)			SSQ	MSSQ	SN
	A	B	C	Y_1	Y_2	Y_3	Y_i	SSQ/n	*SN* = -10 log[*MSSQ*]
1	0	0	0	25.2	25.6	26.5	4.525E-03	1.508E-03	28.22
2	0	1	1	7.9	8.1	8	4.689E-02	1.563E-02	18.06
3	0	2	2	27.6	26.9	27	4.066E-03	1.355E-03	28.68
4	1	0	1	11.1	10.4	10.9	2.578E-02	8.593E-03	20.66
5	1	1	2	8.4	9.3	9.6	3.659E-02	1.220E-02	19.14
6	1	2	0	5.2	5.7	5	1.078E-01	3.592E-02	14.45
7	2	0	2	15.6	15.7	16.6	1.180E-02	3.932E-03	24.05
8	2	1	0	13.2	11.8	11.5	2.048E-02	6.827E-03	21.66
9	2	2	1	3	1.9	1.2	1.083E+00	3.609E-01	4.43

Y_1 = % decrease in absorbance for 1st run, Y_2 = % decrease in absorbance for 2nd run, Y_3 = % decrease in absorbance for 3rd run, SSQ = sum of squares of coefficients, MSSQ = mean of sum of squares of coefficients.

molecules. From Fig. 2 it can be deduced that as the amount of the coagulant increases the effect of reduction of percentage decrease in the absorbance was observed. It can be explained on the basis of the phenomenon of excess polymer adsorption on colloidal surfaces and producing re-stabilized colloids (Ariffin et al. 2005). When potato starch is dissolved in water, it will be ionized and produce polymer ions. The excessive amount of the polymer ions joins together under the action of van der Waals forces and results in the agglomeration. It leads to the damping of active sites required for adsorption, as a consequence the rate of dye removal deceases at high concentration of potato starch. It is worth noting that the percentage reduction in dye (plotted on Y axis of Figs 2, 3 and 4) is the reflection of percentage decrease in absorbance, since, absorbance has been taken as a key performance indicator in related studies (Blackburn 2004, Kim et al. 2004, Roussy, et al. 2005).

Effect of pH

The effect of pH on the signal-noise ratio of the performance characteristics and percentage reduction in the absorbance is depicted in Fig. 3. It can be observed that the value of signal-noise ratio, SN, is the highest at low pH value, i.e., 3. On the other hand, at pH = 3, the dye removal was the maximum but as the pH value moves to the neutral region the percentage dye removal (which is equivalent to percentage reduction in absorbance) decreases. Moreover, in the basic region (pH =10) the removal of dye increases again. The high percentage of dye removal at low pH can be explained on the fact that in acidic environment dye molecule can generate highly polar hydrophobic sites (due to the presence of aromatic rings). It helps the transportation of dye ions to the polymer (i.e., adsorbent) in an aqueous solution, which will have high concentration of proton at low pH (Hao et al. 2006). The aforementioned statement is endorsed by the findings of Yang et al. that, at pH 4, 90% of the functional groups present on the molecular surface have been protonated, when they were investigating the effect of pH on biopolymers such as chitosan. So, the molecules of biopolymer are positively charged in acidic environment resulting in the augmentation of dye removal (Amuda and Amoo 2007). Therefore, the removal of dye was maximum at B_O i.e,. pH 3, see Fig. 3. Furthermore, the percentage removal of dye molecules decreased as pH was shifted from acidic to neutral region, which was due to the decrease in degree of protonization of adsorbent. On the other hand, as pH moved from neutral to basic region, the percentage dye removal increased again, see Fig. 3. It is because, at high pH, the coagulant surface is negatively charged, while the adsorbate, i.e., dye becomes positively charged which results in high dye removal (Veeramalini et al. 2012).

Effect of temperature

The temperature is also an important parameter that has a significant effect on the dye removal, see Fig. 4. It can be deduced that at low temperature, keeping the adsorbent concentration (potato starch) constant the percentage of dye removal is high, which decreases from 25°C to 40°C. But, beyond 40°C, the value of percentage dye removal starts enhancing again. So, at elevated temperatures, there is a high adsorption of dye on potato starch, see Fig. 4. The high percentage of dye removal at elevated temperatures can be explained on the basis of inter and intra molecular hydrogen bonding of adsorbent (Shore 1995). Potato starch, similar to cellulose, requires energy to break inter and intra molecular hydrogen bonding and elevated temperature favors the bond breaking of potato starch (bio-polymer). It allows the adsorbent to bond with dye molecules. More precisely, potato starch can produce "11" hydrogen bond acceptors and "8" hydrogen bond donors. So, negative charge is dominant at elevated temperatures, within the starch molecule. It can provide a good feasible ground for hydrogen bonding to interact with positively charged functional groups of adsorbate, i.e., dye molecule (Blackburn 2004). The increment in percentage dye removal at low temperature can be explained on the fact that, in neutral environment, at pH 6.5 to 7, a few reactive sites are available on the potato starch leading to low percentage removal of dye. But, as pH moves towards the acidic region, it creates positive charge on the surface of potato starch; it can produce force of attraction with the hydrophobic sites on the dye surface. It can be deduced from Table 2 that the percentage of dye removal becomes significant at low temperature only in acidic environment, i.e., at low pH.

Optimum conditions for dye removal

The maximum and optimum values of the parameters have been analyzed on the basis of cost (Table 3). It can be observed that at low percentage of coagulant dosage (0.5 w/v %), the signal to noise ratio (statistical performance characteristics) is maximum as compared to higher dosage. Similarly, the cost of the coagulant is lower at A0 level, i.e., as quantity of

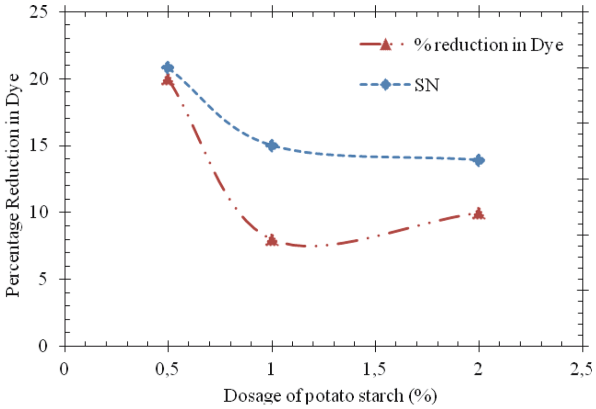

Fig. 2. The effect of potato starch dosage on the Performance statistics for spent reactive dye solution

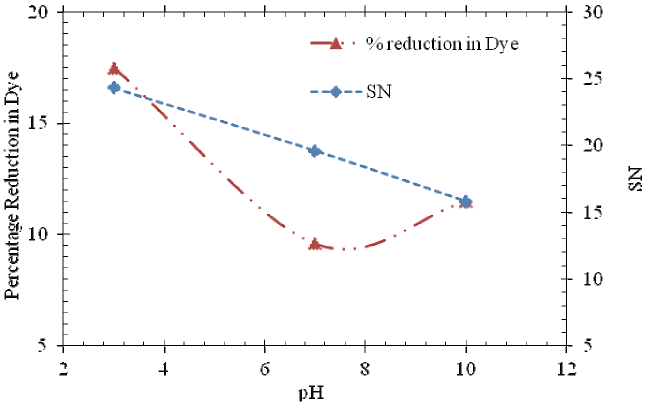

Fig. 3. The effect of pH on the Performance statistics for spent reactive dye solution

coagulant increases the cost of the coagulant also rises. So, A0 is the optimum value of coagulant dosage (i.e. 0.5% w/v). In the case of hydrogen ion concentration (pH), the maximum value of signal to noise ratio is at B0 level (i.e. pH = 3). But at this point the cost tends to the maximum. It is due to the fact that the textile effluent is available at high pH, so, an additional amount of acid is required to decrease pH. It leads to the increase of the cost of the process. As a consequence, B2 becomes the optimum parameter for the process. Similarly the temperature of the textile effluent is already at 55°C. Further, the signal to noise ratio is also the maximum at C2 (i.e. 55 °C) and the cost of dye removal is minimum at C2. Consequently, the optimum removal conditions are proved to be as A0, B2 and C2 (Possible position of Table 3).

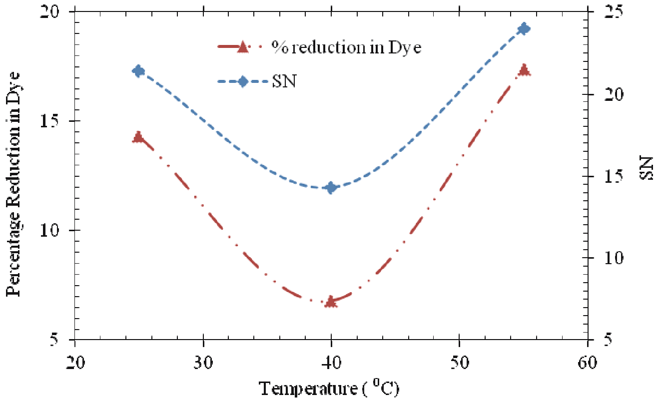

Fig. 4. The effect of temperature on the Performance statistics for spent reactive dye solution

F-factor

Table 4 represents the f-test analysis. The coagulant dosage was proved to be the most effective parameter for dye removal from waste water. It can be deduced that the percentage dosage is the most sensitive parameter (has the maximum f-value, i.e., 3.36) for the coagulation process. On the other hand, the adsorption of textile dyes on the potato starch is least sensitive towards the hydrogen ion concentration (i.e., pH).

Confirmatory test

The confirmatory test is also important in validating the experimental conclusion. So, two random average experimental values (repeated three times) of percentage dye removal were investigated and SN ratio was calculated. On the other hand, the SN value was theoretically calculated using minitab-I7 at two parametric conditions, i.e., at condition 1 (1% dosage, pH 3 and 25°C) and at condition 2 (0.5% dosage, pH 7 and 55°C). It is also worth noting that the parametric conditions were kept same for both experimental and theoretical calculations. It was found that error between experimental and calculated values was between 5 to 9% approximately.

Table 3. Maximum and Optimum “SN” conditions

Analysis for Maximum & Optimum Performance Statistic (SN)							
Parameter		**Parameters Level**		**Performance Statistics**	**Cost**	**Max S/N**	**Optimum SN**
		Level	**Value**	**SN**	min		
A	Coagulant Dose (%)	0	0.5	24.98	↓	A-0	A-0
		1	1	18.08			
		2	2	16.71			
						0.50%	**0.50%**
					max max		
B	pH	0	3	24.31	↑	B-0	B-2
		1	7	19.62			
		2	10	15.85			
						pH 3	**pH 10**
					min min		
C	Temperature (°C)	0	25	21.44	↑	C-2	C-2
		1	40	14.38			
		2	55	23.96			
						55°C	**55°C**
					min		

S/N: signal-to-noise

Table 4. The results of variance analysis using Minitab-17

	Source	DF	Adj. SS	Adj. MS	*F*-Value
A	Coagulant Dosage (%)	2	249.79	124.9	3.36
B	pH	2	99.33	49.66	1.34
C	Temperature	2	174.3	87.15	2.35
	Error	2	74.31	37.16	
	Total	8	597.73		

DF: Degree of freedom, Adj.SS: Adjusted sum of squares, Adj.MS: Adjusted mean of squares.

Conclusions

In the study optimum operating conditions for the removal of dye from textile waste water are determined using Taguchi method. Three operating parameters, namely coagulant dosage, hydrogen ion concentration and temperature, were regulated to observe their effect on the removal of dye from textile effluent by employing the naturally occurring poly-saccharide potato starch as a coagulant. It can be concluded that the percentage dosage, pH and temperature were found to be optimum at 0.5% (w/v), 10 and 55°C, respectively. It can also be deduced from analysis of variance (ANOVA) that the percentage coagulant dosage (i.e., potato starch) is the most significant parameter that can affect the dye removal process. Moreover, the error in the parametric values of signal to noise ratio has been found to be between 5 to 9%.

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