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Application of Response Surface Methodology for the Extraction of Dye from Henna Leaves

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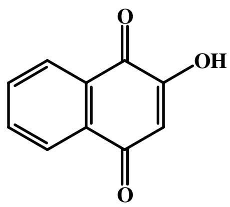
Due to toxic effect of synthetic dyes, there are growing concerns of alternative dyes which should be non toxic and environmental friendly. The dyes from natural sources can substitute the synthetic dyes. In present study, the extraction of natural dye from henna leave was evaluated. The temperature, extraction time and mass to liquor ratio (M:L) were considered for optimization. The extracted dye was applied to cotton fiber and measured the relative colour strength of extracts. The relationships between variables were established by optimal design from statistics using response surface technology to hit the target. The variables such as temperature, extraction time and mass to liquor ratio levels were 80, 90, 100 °C, 20, 40, 60 min and 1:10, 1:20 and 1: 30, respectively. The relative colour strength showed that 89 °C, 44 min and 1:23, respectively were the best levels of variable for the extraction of maximum dye form henna leaves.

Keywords: Henna leaves, Natural dye, Extraction.

INTRODUCTION

Renewable sources such as roots, stems, leaves, fruits, seeds and flowers of various plants, dried bodies of certain insects and minerals can be used for extraction of natural dyes[1]. Recently, it is reported that synthetic dyes are non-biodegradable, allergic and some are carcinogenic and there is growing concern for the use of natural dyes in textiles and food processing due to the anomalies related to synthetic dyes[2]. The use of natural dyes in textile industry was reduced to end as synthetic dyes having attraction to give variety of shade and colour[3]. Therefore, production of green dyes is a desire for dyeing fabrics with colouring compounds available naturally. It is clear that if natural dyes have to be applied in place of synthetic dye they have to manifest the same characteristics of synthetic dyes[4]. The advantages of using natural dyes including renewable sources, minimal health hazard, mild reaction conditions, no disposal problems and harmonization with nature[5]. Synthetic dyes fade in natural sunlight and, needs complex procedure for preparation and cause environmental problems also. Natural dyes only soften or mellow in sunlight, no matter how prolonged the exposure[2]. In view of importance of natural dyes, researchers have focused their efforts to evaluate the specific properties of natural dyes, *e.g.* dyeing, physicochemical properties like light fastness and washing fastness of the dyed articles[6,7].

The chemical composition of fiber and chemical nature of the dye determine their interactions[8]. Chemistry of bonding of dyes to fiber is complex, it involves direct bonding: hydrogen bond and hydrophobic interactions, although, polar interactions are important for solubility of the dye in water as well as for attraction between dye and fiber. The dye's large aromatic system is responsible for this kind of attraction. Due to hydrophobic interaction, the dye-fiber attraction is greater than the dye-water attraction despite the dye's solubility in water[2,8]. In modern textile dye houses, the use of natural dye is coupled to several necessities which have to be satisfied like adaptation of conventional processes on modern equipments, appropriate supply of dye source to dye houses and selection of material leading to product with suitable fastness properties. Lawsone (2-hydroxy-1,4-naphthoquinone) in henna leaves is considered a colouring agent[9].



2-Hydroxy-1,4-naphthoquinone (Lawsone)
(Active Principle of *Lawsonia alba*)

The present research work was performed with the explicit objective of extracting natural dye from henna by aqueous solvent and operating parameters such as temperature, time of extraction and mass to liquor ratio were optimized using central composite design through response surface methodology. The model constructed is given below (eqn. 1). where β_0, β_1, β_3, β_4 and β_5 are coefficients, while X_1 and X_2 are explanatory variables.

$$Y = \beta_0 + \beta_1X_1 + \beta_2X_2 + \beta_{12}X_1X_2 + \beta_1X_1^2 + \beta_2X_2^2 \quad (1)$$

EXPERIMENTAL

Extraction: The dried henna leaves were purchased from the cultivated source of Faisalabad and extraction was carried out according to procedure described elsewhere[2]. Distilled water was used as a solvent for extraction of dye from leaves. Dye extract was prepared by taking 25 g leaves powder and 500 mL water in a round bottom flask. The material to liquid ratio were; 1:10, 1:20, 1:30 and temperature levels were 80, 90, 100 °C and extractions time levels were 20, 40 and 60 min. After stipulated period, the contents were filtered through suction pump. Filtrate was used for dyeing the cotton fabric.

Optimization of extraction conditions: Optimization of extraction time, temperature and mass to liquor ratio was done using factorial design under RSM to obtained maximum K/S value (colour strength). The single variable optimization faces the problem that it does not take into consideration the interactive effect of other variables and in present study the interaction of among parameter was studied. The optimization of dye extraction process was carried out by chosen two variables liquor ratio (1:10, 1:20, 1:30) and temperature (80, 90, 100 °C), extraction time (20, 40 and 60 min) used in the extraction; the fabric dyed with extract and K/S value was taken as response.

Cellulosic fiber dying condition with henna leaves extracts: Fifteen samples of cotton fabric, weighing 2 g each, were dyed in separate baths of Henna leaves extracts at following dyeing conditions; L:R (30:1), time (1 h), temperature (70 °C) and pH (7). All of these dyeing were carried out in the presence of 50 g/L sodium sulphate to promote exhaustion. Finally, the colour strength of each sample was measured with Datacolour SF 600 spectrophotometer with software.

Statically analysis: All experiment was run in triplicate and data thus obtained subjected to R software (version 2.13.1) for ANOVA, RSM graphs and to measure the stationary point.

RESULTS AND DISCUSSION

The results of K/S (colour strength) were reported as percentage effect in comparison to control. The variables, their levels and runs for all parameter are given in Table-1. Based on the differences between observed and predicted responses at 95 % confidence interval of mean, the interaction effect of variable was determined. After generation of experimental matrices for each response, the experimental variables were combined to construct three-dimensional surfaces plots. In these plots the synergism between the experimental variables can be observed, resulting in an optimal zone. The response surfaces of K/S alues are shown in Figs. 1 and 2. By solving experimental matrix statistically, the model equation was developed for K/S values. The importance of the parameters in the

TABLE-1
DESIGN SHOWING THE FACTOR AND LEVELS OF VARIABLES

Temperature (°C)	M:L	Time (min)	Y
80	10	20	-
90	10	20	-
100	10	20	-
80	20	40	-
90	20	40	-
100	20	40	-
80	30	60	-
90	30	60	-
100	30	60	-
90	20	40	-
90	20	40	-
90	20	40	-

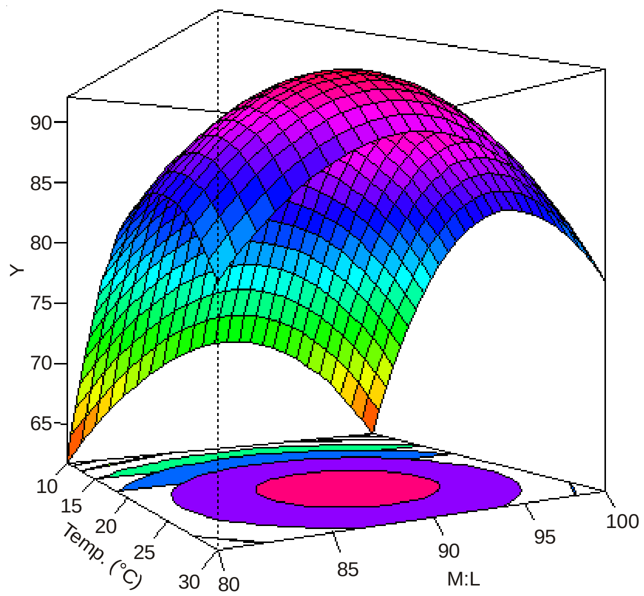


Fig. 1. Response surface graph of response between temperature (°C) and mass to liquor ratio (M:L), Y is representing K/S response

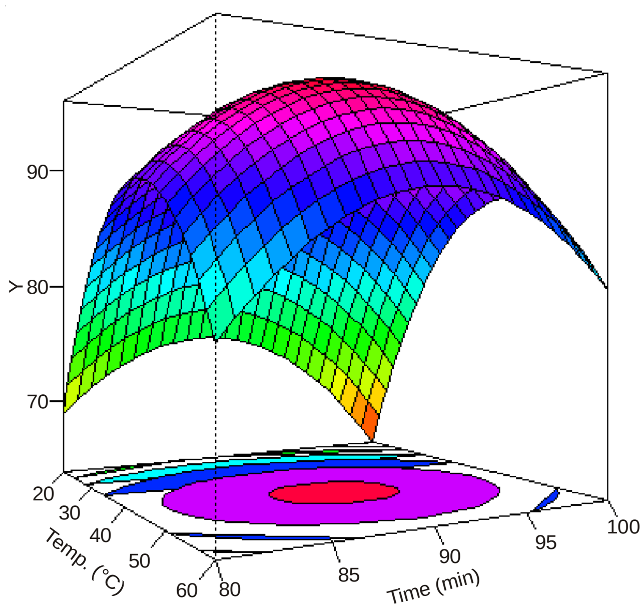


Fig. 2. Response surface graph of response between temperature (°C) and Time (min), Y is representing K/S response

mathematical solution was obtained from the experimental design, against weight of each variable (temperature, time and M:L). Thus, an empirical relationship between the response and the variables was expressed by the polynomial equations. In equations, the Y, X_1 and X_2 are representing the response and independent variables, respectively. The stationary points for K/S values in response of independent variables are given in Table-2.

TABLE-2
R^2 AND R^2 adj AND OPTIMIZED LEVELS OF VARIABLES

Parameters	Unit	R^2	R^2 adj	Optimum level
Temp.	°C	93	87	89.14
M:L		97	88	1:23.12
Time	min	97	95	44

Data analysis showed that that the first degree terms and pure quadratic terms were significant, where the polynomial equation represented the response influenced by combined effects of variables. The polynomial equations generated for K/S are shown in eqns. 2 and 3.

$$Y\ (\text{Tem and M:L}) = -7.41\ (\pm 1.92) + 4.30\ (\pm 1.71)\ X_1 + 5.55\ (\pm 1.98)\ X_2 - 9.50\ (\pm 2.38)\ X_1^2 - 1.2\ (\pm 0.02)\ X_2^2 \quad (2)$$

$$Y\ (\text{Tem and Time}) = -6.22\ (\pm 1.10) + 2.47\ (\pm 1.48)\ X_1 + 5.74\ (\pm 1.92)\ X_2 - 8.50\ (\pm 1.36)\ X_1^2 - 3.62\ (\pm 0.03)\ X_2^2 \quad (3)$$

The data analysis showed that colour strength (K/S) value was maximum at 89 °C with mass to liquor ratio 1:23 and for 44 min extraction time (Table-2). Furthermore, the observed and actual values of K/S were also found to be close to each other, which indicate the normal probability of the data Fig. 3 (temperature and M:L) and Fig. 4 (temperature and time). The ANOVA for the experimental design is shown in Table-3. The high F values with very low probability value (P) indicate the high significance of the fitted model. The goodness of fit of the model was evaluated by the coefficient of determination (R^2) (Table-2). The smaller the P value for a parameter the more significant is the parameter, hence reflecting the relative importance of the term attached to that parameter. The F value for interaction between temperature and mass to liquor ratio and extraction time are the strong evidence for significance of the model. So, the proper ratio of these parameters for the extraction of dye is suggested. Simply, it can be explained as the different combinations of temperature, extraction time and mass and these findings are in accordance reported elsewhere[10,11]. The pure quadratic model ($P < 0.0001$) made the most significant contributions to the fitted model. Furthermore, all first order interaction effects were also found statistically significant ($P < 0.05$).

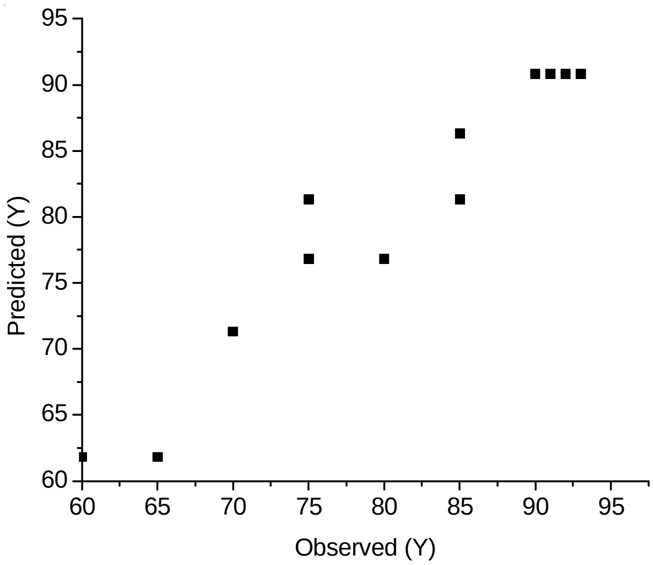


Fig. 3. Scatter plot of M:L and time actual and observed response

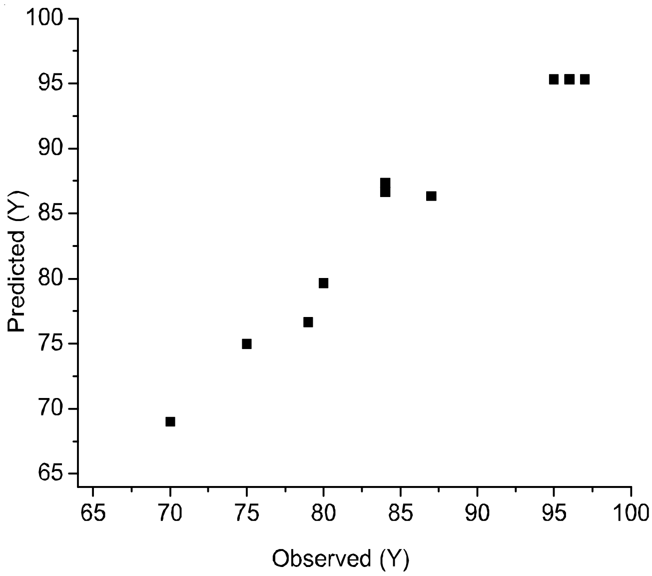


Fig. 4. Scatter plot of temperature and time actual and observed response

The results showed that the maximum dye extraction can be achieved at optimized condition of temperature, time and M:L ratio. The response surface analysis proves to be efficient for the evaluation of the optimum conditions with a statistically reliable analysis. In present study, it was found that the temperature, extraction time and mass to liquor ratio have interactive effect to extract maximum colour content form henna leaves.

TABLE-3
ANOVA FOR THE DESIGN USED FOR THE OPTIMIZATION OF VARIABLES

	DF	SS	MS	F	P
FO	2	205.67	102.83	20.56	0.002*
TWI	1	16	16	3.2	0.12
PQ	2	1094	547	109	0.00001*
Residual	6	30	5		
Lack of fit	3	28	9.33	14	0.02
Pure error	3	2	0.67		

*significant, FO, TWI and PQ are representing first order, two way interaction and pure quadratic, respectively

Preliminary results obtained indicate that renewable natural resources should be used for dye extraction due to their eco-friendly nature[12-15] and response surface methodology is an efficient tool for the optimization of independent variables since the predicted values were in agreement with observed responses[16].

REFERENCES

1. P.S. Vankar, *J. Sci. Educ.*, **5**, 73 (2000).
2. S. Ali, T. Hussain and R. Nawaz, *J. Clean. Prod.*, **17**, 61 (2009).
3. H.T. Deo and B.K. Desai, *Colour. Technol.*, **115**, 224 (1999).
4. S. Sukalyan and R.S. Bal, Natural, "Green" Dyes for the Textile Industry, The Massachusetts Toxics Use Reduction Institute, Technical Report No. 57, Lowell (2003).
5. R.L.M. Allen, Colour Chemistry, Thomas Nelson and Sons Ltd., London, pp. 11 (1971).
6. D. Cristea and G. Vilarem, *Dyes Pigments*, **70**, 238 (2006).
7. X.X. Feng, L.L. Zhang, J.Y. Chen and J.C. Zhang, *J. Clean. Prod.*, **15**, 366 (2007).
8. E.R. Trotman, Dyeing and Chemical Technology of Textile Fibers, Charles Griffin and Company Ltd., London, edn 5, pp. 456 (1975).
9. V.S. Govindarajan and W.H. Stahl, *Crit. Rev. Food Sci. Nutr.*, **12**, 199 (1980).
10. V.A. Shenai, Technology of Textile Processing II Chemistry of Dyes and Principles of Dyeing. Sevak Publications. Bombay, India (1995).
11. V.A.A. Shenai, *Textile Dyer and Printer*, **12**, 29 (1979).
12. M. Iqbal, I.A. Bhatti, M. Zia-ur-Rehman, H.N. Bhatti and M. Shahid, *Asian J. Chem.*, **26**, 4291 (2014).
13. Q. Manzoor, R. Nadeem, M. Iqbal, R. Saeed and T.M. Ansari, *Bioresour. Technol.*, **132,** 446 (2013).
14. I. Ullah, R. Nadeem, M. Iqbal and Q. Manzoor, *Ecol. Eng.*, **60,** 99 (2013).
15. N. Bilal, S. Ali and M. Iqbal, *Asian J. Chem.*, **26,** 1882 (2014).
16. M. Iqbal, I. Ahmad, S.M. Hussain, R.A. Khera, T.H. Bokhari and M.A. Shehzad, *Int. Agrophys.*, **27**, 265 (2013).