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Optimization of Dye Extraction Conditions from (*Camellia sinensis*) Green Tea Leaves Using Response Surface Methodology

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Natural dyes are potential source of colourants for textile industries. The aim of this study was to optimize the extraction conditions of natural dye from green tea leaves using response surface methodology. Extraction pH, time and material to liquor ratio (M:L) were taken as input variables in the central composite experimental design (CCD) and the colour strength (K/S) of the fabric dyed with the extracted colourant was taken as response. It was found that maximum colour strength of the extract could be obtained at 6.6 pH in about 1 h extraction time at 1:33 M:L. It was further found that effect of pH, time and M:L was significantly non-linear. The second order regression equation for K/S indicated 91 % variation in the response that could be explained by the terms included in the equation. The colourant extracted at the optimized conditions showed good washing, light and dry rubbing fastness but poor wet rubbing fastness.

Keywords: Green tea dye, Extraction, Response surface methodology, Central composite design, Optimization.

INTRODUCTION

Nature has its beauty due to the existence of colourful display all around. The rainbow, greenery, flowers, animals, birds *etc*. they all have some sort of colourful display naturally. The importance of colours cannot be denied as it is very vital part of our life. In the history of mankind, the use of colourants for many purposes has been in vogue in every civilization and various dyes and pigments have been developed using many innovative techniques and extraction processes of dyes from natural sources. These dyes range naturally extracted dyestuffs to the chemically synthesized dyes that are mostly used now-a-days[1,2].

Textile colourants whether natural or synthetic, have to be able to render some colour for value addition to product, giving it a better look according to the prospective customer's desire. Natural dyes are considered advantageous because they are cost effective, renewable and non-carcinogenic. They also do not pose any disposal problems and cause minimal skin allergic reactions[3]. Natural dyes are non-toxic and do not make ecological issues due to their bio-degradable characteristics[4]. Requirement of natural colours is improving consistently as their manufacturing does not need powerful chemicals and alkalies. Moreover of their ecologically helpful nature many natural colours have stop hypersensitive and bad odouring qualities[5]. The use of natural colours has been restricted to small scale dyers and artisan. However many dyers and exporters have began to discover opportunities of using natural colours on frequent use as alternative of synthetic colours to get over the issues of contamination due to synthetic colourants[6].

Green tea plants have been classified as *Camellia sinensis*. There are many important types of compounds present in green tea leaves such as amino acids, catechin, lipids, minerals, nucleotides, caffeine, carbohydrates, carotenoid contents, chlorophyll, some organic acids, saponins, polyphenols, unsaponifiable and many compounds with low volatility[7].

Fig. 1 shows the structure of green tea polyphenols. Among the commercially available natural dyes, green tea has unique properties due to its extensive use as drink or beverage. The use of green tea extract as a colourant has been reported specially for cellulose fabric dyeing[8]. However, there is still room for research work on optimization of conditions of dye extraction from tea leaves for maximum colour yield.

The response surface methodology (RSM) is considered better as compared to conventional methods of optimizing factors because the RSM statistical optimization modeling has the ability to consider the non-linear effects as well as interactions among the variables for optimizing multivariable and multilevel processes[8,9]. Central composite design (CCD) is one of the most powerful tools of RSM[10]. This CCD method suits well in fitting a quadratic surface model, the effective factors are

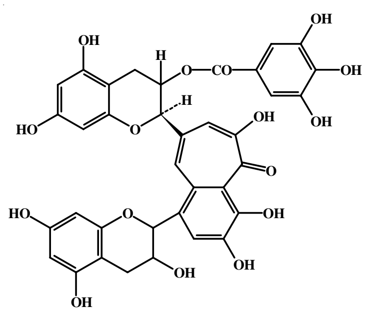

Fig. 1. Green tea polyphenols

optimized with minimum experiments and the interactions among all the parameters are also analyzed through this method[11].

The present work is aimed at optimizing dye extraction conditions from green tea leaves using central composite experimental of response surface methodology.

EXPERIMENTAL

Dry leaves of green tea (*Camellia sinensis*); sodium hydroxide, used for pH adjustment; dilute hydrochloric acid solution, used for pH adjustment; Gluaber's salt, used as dye exhausting agent; industrially scoured and bleached cotton fabric; lab-scale dyeing machine, for exhaust dyeing; Datacolor SF 600 spectrophotometer with software, for colour measurement of the dyed fabric; Crockmeter, for testing rubbing fastness of the dyed fabric; Launder-o-meter, for testing washing fastness of the dyed fabric; Fadometer with xenon arc lamp, for testing light fastness of the dyed fabric.

Selection of experimental variables: Based on the literature survey and preliminary trials, extraction pH, time and material to liquor ratio (M:L) were selected as the experimental variables or factors. The actual and coded levels of these factors are given in Table-1.

TABLE-1
EXPERIMENTAL FACTORS AND THEIR LEVELS

Factors	Levels				
	-alpha	-1	0	+1	+alpha
pH	2	4	7	10	12
Time (min)	30	42	60	78	90
M:L	20	26	35	44	50

Design of experiment: A critical facet of RSM is the design of experiment (DOE)[12,13]. The type of design of experiment used in this study was central composite design (CCD) comprising total 20 experiments, including 8 factorial points, 6 axial points and 6 center points. The experimental factors were the dye extraction pH, extraction time and material to liquor ratio (M:L) and the response variable was the colour strength (K/S) obtained by the extracted dye after dyeing on cotton fabric. All the design and analysis of experiments was performed using Design-Expert 7.0 software package.

Dye extraction: The dye was extracted from the dry green tea leaves using reflux boiling methodology. Water was used as the dye extraction medium. The choice of the dye extraction medium is critical for the efficiency and kinetics of the extraction process[14]. Twenty samples of the dye liquor were prepared through extraction according to the experimental design at different pH, time and M:L combinations. pH values ranged between 2, 4, 7, 10 and 12, extraction time ranged between 30, 42, 60, 78 and 90 min. The w/w ratio between the green tea leaves and the extraction liquor ranged from 20, 26, 35, 44 and 50.

Dye application on cotton fabrics: All the twenty samples of the extracted dye were used separately for dying cotton fabric samples in a lab-scale dyeing machine at 100 °C for at least 60 min. At the completion of dyeing, the dyeing baths were cooled to room temperature; the dyed cotton samples were removed and washed with tap water followed by air drying under the shade[15].

Assessment of colour strength of the dyed fabrics: All the fabric samples dyed with dye liquors extracted at twenty different set of conditions were assessed for their colour strength (K/S) using spectrophotometer.

Determination of fastness properties: Colour fastness refers to ability of any material to resist any variation in its colour characteristics or the extent of colour transfer to adjacent light coloured or white substrate due to some physical contact or losing colour upon washing, rubbing, dry cleaning or heat/light exposure of the fabric[16].

The dyed cotton samples were passed through fastness tests in accordance with ISO standard procedures[17]. Colour fastness to rubbing was determined using crockmeter according to ISO 105-X12 method. Colour fastness to washing was determined using launder-o-meter according to ISO 105-CO2 method and colour fastness to light was determined using Fade-o-meter according to ISO-B-02 method.

RESULTS AND DISCUSSION

The colour strength (K/S) values of the cotton fabric samples dyed with the dye extracted from green tea leaves at different extraction conditions, are given in Table-2. A higher value of K/S indicates higher colour strength, thus better dyeing extraction efficiency.

TABLE-2
CENTRAL COMPOSITE EXPERIMENTAL DESIGN WITH FACTORS AND RESPONSE

No.	Factors			Response
	pH	Time (min)	M:L	K/S
1	10	78	44	3.5
2	7	60	35	3.8
3	7	60	35	3.9
4	4	78	44	3.1
5	10	42	26	3.2
6	2	60	35	3.4
7	4	42	26	3.4
8	10	78	26	3.3
9	7	60	20	3.6
10	7	60	35	3.8
11	7	60	35	3.9
12	7	60	35	3.8
13	7	90	35	3.7
14	7	30	35	3.5
15	4	78	26	3.2
16	7	60	35	3.9
17	10	42	44	3.0
18	4	42	44	3.2
19	12	60	35	2.7
20	7	60	50	3.0

T2

TABLE-3
ANOVA FOR COLOUR STRENGTH (K/S VALUES) OF THE DYES FABRICS

Source	Sum of squares	DF	Mean square	F value	p-Value	Remarks
Model	2.200	9	0.240	11.50	0.0003	Significant
A-pH	0.085	1	0.085	3.99	0.0737	
B-time	0.030	1	0.030	1.39	0.2653	
C-M:L	0.047	1	0.047	2.23	0.1666	
AB	0.100	1	0.100	4.76	0.0542	
AC	0.011	1	0.011	0.53	0.4840	
BC	0.631	1	0.031	1.47	0.2536	
A^2	1.480	1	1.480	69.72	< 0.0001	Significant
B^2	0.230	1	0.230	10.83	0.0081	Significant
C^2	0.460	1	0.460	21.82	0.0009	Significant
Residual	0.210	10	0.021			
Lack of Fit	0.210	5	0.083			
Pure Error	0	5	0			
Total	2.42	19				

Analysis of variance (ANOVA): Analysis of variance (ANOVA) was performed to statistically analyze the results. The ANOVA results for K/S values are given in Table-3. The model F-value of 11.5 implies that the model is significant. There are only 0.03 % chance that a "model F-value" this large could occur due to noise. P-values < 0.05 indicate significant model terms. In this case A^2, B^2 and C^2 are significant model terms. This implies that the effect of extraction pH, time and M:L is significantly non-linear.

Regression coefficients for the second order equation: The regression equation for colour strength (K/S) is given in eqn. 1:

$$\text{Colour strength (K/S)} = + 3.90 - 0.079*A + 0.047*B - 0.059*C + 0.11*A*B + 0.038*A*B*C + 0.063*B*C - 0.32*A^2 - 0.13*B^2 - 0.18*C^2 \quad (1)$$

R^2 for eqn. 1 was found to be 0.9119, which indicates that 91.19 % change in colour strength is explained by the terms included in the equation.

Effect of extraction conditions on colour strength: The 3D surface plots and contour plots are presented in Figs. 2 and 3. The graphical representation of three dimensional response surface is a function of two independent variables keeping the third variables at a constant or fixed level. The main purpose of these plots is to create understanding of the main effects and interactions among the independent variable for the response. Fig. 2 depicts the effect of extraction pH and time on the colour strength of the dye extracted from green tea leaves. The pH shows a very vivid influence on the colour strength. During extraction process pH was maintained using 1 M solution of HCl for acidic pH adjustment and 1 M NaOH solution was used to adjust the pH on alkaline side. After the initial adjustment, the pH did not significantly vary during the extraction procedure as the original pH of green tea was supposed to be neutral because of the presence of polyphenols and tanins.

The optimized extraction pH for green tea dye was found to be almost neutral (pH 6.6). It was also found that at pH higher or lower than 7 there was an obvious decline in the values of colour strength. The decline of colour strength at pH higher or lower than than 7 may be due to degradation of major polyphenolic colourant in green tea. The optimum extraction time was nearly 1 h (63 min to be exact). At lower times, the colourant was not sufficiently extracted while the higher times may have resulted in the degradation of the extracted colourant.

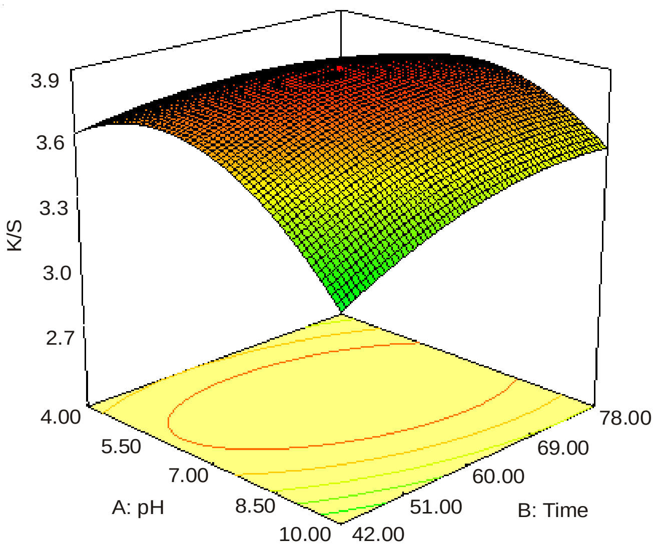

Fig. 2. Effect of extraction pH and time on colour strength (K/S)

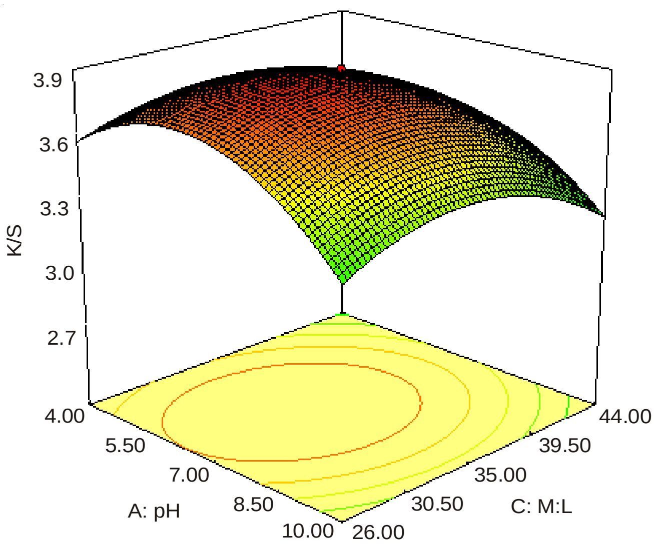

Fig. 3. Effect of extraction pH and M:L on colour strength (K/S)

Fig. 3 shows the effect of extraction pH and L:R on colour strength. The optimum L:R was found to be 1:33. With higher material to liqour ratio, the dilution may have resulted in the loss of colour strength while at lower liquor ratio, the colourant may not have properly extracted from the leaves into the liquor.

Colour fastness of the fabric dyed with the optimized extract: Colour fastness results of the fabric dyed with the optimized extract of green tea leaves are given in Table-4. Colour fastness to washing, light and dry rubbing was found to be good whereas wet rubbing fastness was found to be poor. Further research work is required to enhance the wet rubbing fastness and to further improve the washing fastness properties of the dye obtained from green tea leaves.

TABLE-4
COLOUR FASTNESS OF THE FABRIC DYED WITH THE OPTIMIZED EXTRACT

Washing fastness	Light fastness	Rubbing fastness	
		Dry	Wet
4-5	5-6	4	2-3

Conclusion

Parameters for the extraction of dye from green tea leaves were optimized using central composite design of response surface methodology. The highest colour strength of the extracted dye was obtained at 6.6 extraction pH after 1 h at material to liquor ratio of 1:33. The analysis of variance (ANOVA) showed significant non-linear effect of pH, time and M:L on the colour strength. The second order regression equation for colour strength was obtained with 91 % R-sq. The optimized extract of the green tea leaves gave good washing, light and dry rubbing fastness properties on cotton dyed without using any mordant. The wet rubbing fastness needs further improvement. The washing fastness also needs further enhancement for more durable applications.

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