Pol. J. Environ. Stud. Vol. 28, No. 4 (2019), 2145-2150
DOI: *10.15244/pjoes/85125* ONLINE PUBLICATION DATE: 2019-01-25

Original Research

Statistical Modeling for the Extraction of Dye from Natural Source and Industrial Applications

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Received: 17 January 2018
Accepted: 7 February 2018

Abstract

This study is intended to scrutinize the consequence of extraction conditions of *Lawsonia Inermis* (henna) leaves on the color strength values of dyed fabric. Extraction pH, M:L ratio, and time were optimized using central composite design (CCD) under response surface methodology (RSM). We investigated how the maximum colour strength values were obtained at pH 6.64 in about 78 minutes extraction with an M:L ratio of 1:44. ANOVA results showed that all the extraction parameters significantly affect the colour strength values of dyed samples. 99% variation in the response was indicated in the 2nd-order regression equation for K/S. The fastness properties with respect to light, washing, and rubbing were quite satisfactory of dyed samples under optimized extraction conditions.

Keywords: central composite design, henna leaves, natural dye, response surface methodology, extraction

Introduction

Beautiful and attractive displays of the color of nature have captivated our attention since prehistoric times. In ancient times humans used natural resources (plants, animals, and minerals) to extract dye or colorant and used them for dyeing textile fibers and ornamental purposes, thus marking the start of a colorful lifestyle [1]. However, in the mid-18th century the innovation of synthetic dye (Mauveine) completely replaced the use of natural dyes [1-2]. These dyes were cheap and give a bright reproducible range of colors and have excellent fastness properties [2-3]. Synthetic dyes are widely used in various industries, e.g., textile, leather, petroleum, and wood protective chemicals [4]. However, large amounts of effluents released from textile or leather industries are directly wasted in the water, where it causes water

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pollution. About 10-35% of the dye is wasted in the water during the dyeing process [5-18]. Therefore environmental pollution due to synthetic dyes is a severe problem due to their pessimistic effect on the ecosystem and bioaccumulation in flora and fauna [19]. Water pollution due to synthetic dyes affects the environment and has detrimental impacts on living organisms [20-21]. Due to the negative aspects of these dyes, people have been moving toward natural dyes as they are less harmful, environmentally friendly, and have colorful eye-catching shades. Natural dyes have considerably reduced the amount of poisonous waste released due to dyeing methods and they have successfully replaced synthetic dyes.

Natural dyes are acquired from different sources including plants and animals without the application of any chemical handling. This group of dyes is documented to be eco-friendly and slightly poisonous as compared to synthetic dyes [22-24]. However, it has been reported that some natural dyes may cause mutagenic effects such as safflower yellow, and elderberry color and continuous inhalation of carmine can cause asthma, but it can be said that some even have a curative impact such as turmeric with its antibacterial properties due to its coloring component, curcumin [25-30].

Lawsonia inermis L., generally known as henna, is a small plant commonly cultivated in Iran, India, Egypt, and Pakistan. The aqueous paste of powdered leaves of the plant is employed for coloring hair and hands. Basically it contains lawsone, i.e., red-orange pigment (also known as hennotannic acid – the chief constituent of henna leaves). The dyeing, UV absorption, and corrosion inhibition characteristics of henna leaves are associated with the presence of lawsone (2- hydroxy-1, 4-naphthoquinone) [31-34]. For keratin, lawsone acts as a substantive dye and produces orange color due to the presence of the hydroxyl group in naphthoquinone structure [34-35].

It is quite important to optimize extraction conditions in order to obtain maximum coloring components solubilized into solvent. There were three main factors with different levels to evaluate the interactions among the factors by the approach of an experimental design. RSM is a statistical approach for the optimization process as compared to full-factorial approach [36]. CCD is a very reliable model fitting tool of RSM and was used to recognize the connection between yield and all the process variables such as extraction pH, extraction time, and M:L [37-39]. The purpose of this investigation was to examine the outcomes of different extraction parameters, i.e., pH, time, and material to liquor ratio on the extraction of natural dye from henna leaves.

Material and Methods

The sample was obtained from the botanical garden at the University of Agriculture in Faisalabad, Pakistan.

Table 1. Experimental variable and their levels.

Variables	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

After washing and drying the leaves at about 37-40ºC, the leaves were crushed to fine powder of uniform particle size that can pass through 25 mesh size. This powdered material was employed for extraction under different conditions.

On the basis of previous studies and initial trials, we identified three experimental variables for study, including pH of extraction, time of extraction, and ratio between material and liquor (M:L). The data is recorded in Table 1. In this experimental design, low and high levels are designated by -alpha and +alpha, respectively. The axial points are positioned at a distance of α (alpha) from central point and thus make the design rotatable [40].

The relationship between the experimental variables (pH, extraction time, and M:L) and response variable of color strength values (K/S) of cotton fabric dyed at different extraction conditions) was developed by two level-three factor (2^3) full factorial CCD in RSM. The statistical software package, design-expert 7.0, was used to design experimental conditions (Table 2).

Reflux boiling methodology was used to extract natural dye from henna leaves in distilled water. 5 g powdered sample were added in round-bottomed flasks and pH of the extraction liquors was adjusted. The extract from the sample was taken at different time spans (30, 42, 60, 78, and 90 min) and different L:R (Table 2). Bleached cotton fabric was dyed by following the method given in Ali et al. [41]. The dyed cotton samples were then washed and air-dried under the shade. The colour strength (K/S) values of dyed samples were measured by the method described in Kannan et al. [42].

Most dyes are organic compounds that can be affected by natural destructive agents that can fade and destroy these colours. This is further supported by the use of certain chemicals used in finishing treatments that may affect the fastness properties of dyed fabrics. Washing fastness (ISO 105-CO2method), rubbing fastness (ISO 105-X12 method), and light fastness (ISO-B-02) tests were performed to evaluate fastness properties.

Results and Discussion

Table 2 shows the colour strength values of fabric dyed extracted material from leaves of henna under different experimental/extraction conditions.

Table 2. Colour strength (K/S) values under different extraction conditions.

S. No.	pH	Time (min)	M:L (ml)	K/S
1	7	60	35	6.5
2	10	42	26	5.7
3	12	60	35	4.3
4	7	60	35	6.5
5	4	42	26	6.4
6	10	42	43	5
7	7	30	35	6.2
8	7	60	20	6.9
9	2	60	35	5
10	7	90	35	7
11	10	78	44	5.9
12	7	60	35	6.5
13	7	60	35	6.5
14	4	78	44	6.5
15	10	78	26	6.1
16	4	42	44	6
17	4	78	26	6.2
18	7	60	35	6.5
19	7	60	35	6.5
20	7	60	50	6.7

A higher K/S magnitude indicates higher dye extraction proficiency. The analysis of variance (ANOVA) results (Table 3) show that the model is significant with p-value less than 0.0001, and all terms are significant except B^2.

The 2nd-order quardatic equation for colour strength is given as follows:

$$K/S = +6.50 - 0.26\,A + 0.22B - 0.098C + 0.13AB - 0.100AC + 0.15BC - 0.66A^2 + 0.033B^2 + 0.10C^2$$

The process parameters pH, time, and M:L are designated as A, B, and C, respectively. The positive and negative signs with each term exhibit synergistic and antagonistic effects, respectively. The R^2 value (0.9941) indicates that 99% change in colour strength can be explained by the terms included in the equation, and only 0.59% of the variance was not explained by the equation.

Fig. 1a) displays the effect of pH and extraction duration on strength of colour on the dyed fabric (cotton). pH plays a significant role in the extraction of coloring component from natural dyes. Initially for the experimental set up, the acidity and alkalinity levels were adjusted by employing 0.1M HCl and 0.1M NaOH, accordingly. pH was not significantly altered after initial adjustment as the original pH of henna leaves was found to be neutral. Although color strength of dye extracts obtained in distilled water is not appreciable, maximum color strength in distilled water was obtained at pH 6.46 – approximately near neutral. Because the coloring component of henna dye is acidic hydroxylated (a-hydroxynaphthaquinone) [42], color strength values decrease in acidic pH while hydroxy-naphthaquinone

Table 3. ANOVA for colour strength.

Source	Sum of Squares	Mean df Square value	FP Value	Prob>F	Remarks
Model	8.82	9	0.98	186.80	< 0.0001 significant
A-pH	0.94	1	0.94	178.67	< 0.0001
B-Time	0.64	1	1	121.13	< 0.0001
C-M:L	0.13	1	1	24.93	0.0005
AB	0.13	1	1	23.83	0.0006
AC	0.080	1	0.080	15.25	0.0029
BC	0.18	1	0.18	34.32	0.0002
A^2	6.21	1	6.21	1184.96	<0.0001
B^2	0.015	1	0.015	2.95	0.1168
C^2	0.15	1	0.15	29.41	0.0003
Residual	0.052	10	5.245E-003		
Lack of Fit	0.052	5	0.010		Insignificant
Pure Error	0.000	5	0.000		
Cor Total	8.87	19			

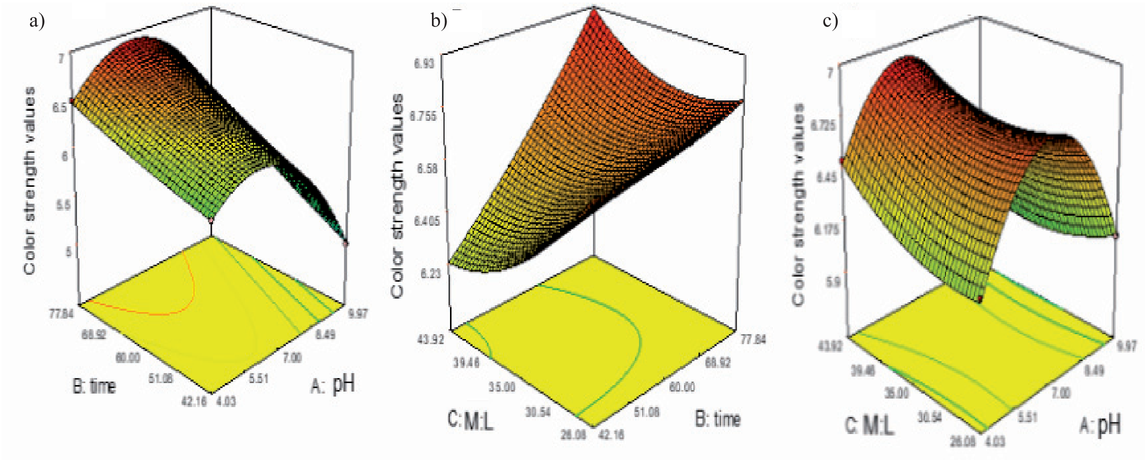

Fig. 1. Effect of extraction a) pH and time, b) time and M:L, and c) pH and M:L on color strength (K/S).

have high reactivity at basic pH due to which coloring components have strong interaction with the solvent and transfer of coloring component toward the cotton fabric is reduced, whereby colour strength values also decrease at basic pH.

The K/S values increase with extraction span and touch a maximum value at 78 minutes. This might be due to the fact that as the extraction time increased, the interaction between plant materials and extracting solvent increased, hence leading to increased yield of natural dye in extraction media. It can be concluded from the results that the *K/S* values were highly dependent on extraction time. However, further increases in extraction time resulted in decreases in colour strength values due to the decomposition of the coloring component [43].

Fig. 1b) shows the extraction time and M:L effect on color strength of dyed fabric. A low M:L (1:44) ratio was found to be appropriate for higher colour strength. Increasing M:L ratios caused a decrease in extraction of the required component. Low M:L ratio and longer extraction time improved the extraction of the coloring component from henna. This comment can be associated with the idea that the saturation point of the colouring component was achieved at initial M:L ratio.

Fig. 1c) shows the effect of pH and M:L on K/S of the dyed cotton fabric. Colour strength was high at neutral pH and at low M:L ratio. The significant decrease in colour yield by increasing the M:L from 44 to 78 was also supported by ANOVA results.

The colour fastness to washing, light, and rubbing (dry and wet) of the dyed fabric with the optimized extract of henna leaves was 4, 4-5, 4-5, and 3-4, respectively. Colour fastness to various parameters was found to be good. The superior washing fastness properties of the colorant might be owing to the affinity of colouring component caused by hydrogen bonding and Vander Waals forces for the dyed sample. The light fastness of the dye was fairly good due to hydrogen bonding between the hydroxyl and carbonyl groups of naphthaquinone, hence shielding the chromophoric group from fading [44-50]. The fastness with respect to light washing and rubbing were satisfactory of dyed samples at optimized extraction conditions and the technique was ecofriendly and could be used to reduce pollution [51-57].

Conclusions

RSM has been used to make a model of colour strength values of dyed samples with henna leaves extract. ANOVA showed that extraction pH, extraction time, and M:L have a significant impact on colour strength values. The values of the optimized factors of extraction for henna leaves are: pH 6.64, M:L 1:44 ml, and extraction time 78 minutes. The pH of the solution markedly influenced the dye extraction from dried henna leaves and found maximum at pH 6.46, while extraction time and M:L are also statistically significant. Dyed samples not only show good colour strength values but also show good fastness properties without using any mordant.

Conflict of Interest

The authors declare no conflict of interest.

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