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Ultrasound-assisted extraction of natural dye from *Swietenia mahagoni* and its application on silk fabric

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The current study deals with the extraction of natural dyes from the flower of the *Sweitenia mahagoni* plant by ultrasound-assisted extraction method using ethanol as solvent. Box-Behnken design has been employed to optimize and investigate the effect of three independent variables (sample-solvent ratio, sonication time and extraction temperature) on the natural dye yield. The results reveal that the experimental data are fitted to a second-order polynomial equation using multiple regression analysis with high coefficient of determination value (R^2> 0.98, Adj. R^2 >0.96 & Pred. R^2 >0.81). Optimal extraction conditions for the dyes yield are: sample-solvent ratio 1/20 g/mL, sonication time 30 min and extraction temperature 50°C. Under these conditions, the highest dyes yield is predicted to be 0.855%. FTIR spectroscopy has been used to identify the major chemical group in the extracted dye. Further, dyeing of silk fabric has been carried out by an exhausted dyeing method and the dyeing property and fastness properties of the dyed samples are also assessed.

Keywords: Dyeing, Natural dyes, Response surface methodology, Silk, *Swietenia mahagoni*, Ultrasound-assisted extraction

1 Introduction

The use of non-allergic, non-toxic, and eco-friendly natural dyes on textiles has become a matter of significant importance due to the increased environmental awareness in order to avoid some hazardous synthetic dyes[1]. Recently, interest in the use of natural dyes has been growing rapidly due to the result of stringent environmental standards imposed by many countries in response to toxic and allergic reactions associated with synthetic dyes and growing interest on the application of natural dyes [2,3] on natural fibre due to worldwide environmental consciousness[4,5]. The use of the least toxic natural dyes for the dyeing of textiles can be considered a very suitable alternative, since such compounds are already environmentally compatible, have biodegradability and low toxicity [6,7]. Bangladesh has a rich repository of dye producing plants and *Swietenia mahagoni* (Mahagoni) is one of them. It is a medium to large deciduous tree [8]. Antioxidant and antimicrobial activities of the flower of *S. mahagoni* was investigated by Rahman *et al.*[9]. Several workers have reported the extraction of natural dyes from different plant species[1,10,11]. Even today, many species remain unexplored. So, it is crucial to explore the extraction of natural dyes from abundantly occurring plant sources.

The extraction of natural compounds from permeable solid plant materials using solvents is an important step in the manufacture of natural color products. Conventional methods of color extraction such as maceration and soxhlet extraction, are very time-consuming and require relatively large quantities of solvents. Nowadays, various modern extraction methods have been developed for the extraction of active components from plants, such as ultrasonic-assisted extraction (UAE), supercritical fluid extraction, enzymatic extraction, and dispersive liquid-liquid micro-extraction [12,13]. In comparison with other extraction techniques, the UAE is less expensive and easier in practice. Ultrasound-assisted extraction has been employed for several studies of plant materials, antioxidants and natural dyes [10,14,15]. Ultrasound is a special type of sound wave beyond human hearing. It can pass through a medium by creating expansion and compression. The process produces a phenomenon called cavitation, which is the production, growth, and collapse of bubbles. The implosion of cavitation bubbles can hit the surface of the solid matrix, break the cells and release the compounds[16,17]. The advantages of ultrasound-assisted

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extraction include reduction of extraction time, energy and use of solvent [12].

The present research was performed with the explicit objective of extracting natural dye from the flower of *S. mahagoni* by aqueous extraction method and to investigate the combined effect of different parameters such as sample-solvent ratio, sonication time, and extraction temperature on the dye extraction process using response surface methodology (RSM). RSM is a collection of mathematical and statistical techniques based on the fit of a polynomial equation to the experimental data that describe the behavior of a data set with the objective of making statistical previsions. It is useful for analyzing the effects of several independent variables on the response. The main objective of RSM is to simultaneously optimize the levels of these variables, determine the optimum operating conditions to obtain the desirable response[18]. One of the advantages of RSM is the reduction in a number of experimental trials required to evaluate multiple parameters and their interactions. Box-Behnken designs (BBD) are a class of rotatable or nearly rotatable second-order designs based on three-level incomplete factorial designs. BBD suggests how to select points from the three-level factorial arrangements, which allows the efficient estimation of the first and second-order coefficients of the mathematical model. In comparison with other response surface designs such as central composite and three-level full factorial design, the BBD is slightly more efficient[19]. The specific objectives of this research include to examine the effects of three independent extraction parameters (ratio of sample-solvent ratio, sonication time and extraction temperature) on the dye yield; to apply a three-factor Box-Behnken design combining with RSM for maximizing the dye yield and to verify the validity of the RSM model. The study also includes the characterization of natural color and dyeing properties onto silk fabric.

2 Materials and Methods

2.1 Materials

Flowers (without causing any harm to the plant) of the mahogoni (*S. mahagoni*) plant was collected from the University of Rajshahi, Bangladesh (24°22'16.3"N latitude and 88°38'18.0"E longitude). The obtained plant material was washed under flowing water to remove dust particles and shade dried at room temperature (24-25 °C). Then it was oven-dried at a temperature below 40 °C and this processed sample was ground into powder form through electrically operated high-efficiency grinder. The screened powder was stored in an air-tight container with identification and kept in a cool, dark and dry place for future use. Degummed mulberry silk yarn (20/22 denier) was used for dyeing. The silk was purchased from Sopura Silk Mills Limited, Rajshahi, Bangladesh. All the chemicals were analytical grade and procured from Merck Germany.

2.2 Solvent Extraction of Natural Colorant by UAE Method

Natural dyes extraction was performed by using ultrasound assisted extraction (UAE) method[20] in an ultrasonic bath "krisbow KW1801033" (maximum power 120W, maximum heating power 150W, maximum frequency 40 kHz and capacity 5L). About 1g of the processed flower was taken in 100 ml Erlenmeyer flask. Different volumes of solvent (15 mL, 20 mL, and 25 mL) were added to each flask in order to keep the plant materials fully immersed in the solvent. The flasks were then incubated at different temperatures (40 °C, 50 °C and 60 °C). Extracted samples were taken out at different time intervals (20 min, 30 min, and 40 min). Thus, dye from the powder was extracted by ultrasound-assisted extraction method. The extraction process was carried out at a different temperature, different extraction time and with a different mass of ground flowers in order to obtain the optimal extraction conditions[21]. All experiments were performed at least in triplicate and results were expressed as means of ±S.D. (Standard Deviation). The extracts were centrifuged for 10 min and filtered through a filter paper (Whatman No. 1, England). The color was dried and the solvent was recovered (90-95%) by rotary vacuum evaporator. The extracts were placed in clean, dried and pre-weighed glass petri dishes. The extracts were then dried in a hot-air oven until all the solvent was evaporated and only the extract was left. The dishes were then cooled in a desiccator and weighed. The drying, cooling and weighing procedures were repeated to get a constant weight and thus the weight of the extract was determined. The weight of the colorant extract obtained per gram of the flower used was then calculated. The extraction yield of color was calculated using the following equation

$$\text{Color yield (\%)} = \frac{W_3 - W_2}{W_1} \times 100$$

where W_1 is the weight of flower sample (g); W_2, the weight of petri dish (g); W_3, the weight of petri dish + extracted dye (g).

2.3 Experiment Design and Statistical Analysis

The present work involves the optimization of different parameters governing the extraction process. The general practice of determining these optima is by varying one parameter while keeping the others at an unspecified constant level. The major disadvantage of this single variable optimization is that it does not take into consideration the interactive effects among the variables; thus it does not depict the net effects of various parameters on the reaction rate. In order to overcome this problem, optimization studies have been done using Response Surface Methodology (RSM)[21]. RSM is an effective statistical technique for optimizing complex processes. RSM reduces the number of experimental trials required to evaluate multiple parameters and their interactions. It is less laborious and less time-consuming than other approaches.

In this study, the optimization of the dye extraction process was carried out by three chosen independent process variables including sample-solvent ratio (S/L), sonication time, and extraction temperature used in the extraction. The ranges and levels of variables investigated in the research are given in Table 1. The total amount of dye extracted was taken as the response of the system.

Experimental data were analyzed using Design-Expert 11.1.2.0 (State-Ease Inc. Minneapolis, MN, USA) statistical package. A Box-Behnken response surface experimental design with three factors was used to optimize and investigate the individual and interactive effects of process variables on the color yield from the flower of Mahagoni. The experiments were conducted in a randomized order and the data were analyzed by multiple regression analysis in order to develop a quadratic second-order regression polynomial mathematical model, which exhibits the relationships between response and independent variables. The quadratic equation model for predicting the optimal point was expressed according to the following equation:

$$Y = \beta_0 + \beta_1 X_1 + \beta_2 X_2 + \beta_3 X_3 + \beta_{12} X_1 X_2 + \beta_{13} X_1 X_3 + \beta_{23} X_2 X_3 + \beta_{11} {X_1}^2 + \beta_{22} {X_2}^2 + \beta_{33} {X_3}^2$$

where Y is the estimated response; β_0, the constant; β_1, β_2 and β_3, the linear coefficients; X_1, X_2, and X_3, the independent variables; β_{12}, β_{13} and β_{23}, the interaction coefficients between the three factors; and β_{11}, β_{22} and β_{33}, the quadratic coefficients.

The construction and analysis of the experimental design included Analysis of Variance (ANOVA) to obtain the interaction between the process variables and the response, quality of the fit of the polynomial model (coefficient of determination (R^2), adjusted coefficient of determination (Adj. R^2) and predicted coefficient of determination (Pred. R^2) and optimization of process condition. After optimization, triplicate experiments were performed under the optimal conditions and the average value of the experiments was compared with the predicted values of the developed model equation.

2.4 Method of Dyeing

The crude dye was sparingly soluble in water. The dyeing of silk fabric was carried out by an exhausted dyeing method. The dye baths were prepared by adding 0.2% dye and 1% mordant (Alum) on the basis of the weight of the fabric. Dyeing was performed with fabric-liquor ratio maintained at 1:30 at 70°C for 30 min with occasional stirring, and then allowed for further 30 min as the bath cools down.

2.5 Colorfastness Test

The fastness ratings were evaluated by comparing with the Grey Scale Standard ISO-1976. The changes in shades were related to the standard Grey Scale rating (rating is 1-5; 1 - poor; 5 - excellent).

3 Results and Discussion

3.1 Box-Behnken Experimental Design

In this study, the extraction process parameters have been optimized using RSM. Three variables BBD has been used to evaluate the main and interaction effects of the factors including sample-solvent ratio, sonication time and extraction temperature on the natural dye yield (Table 1). Three values in the proper range of each factor were chosen

Table 1 — Symbols and coded level of three variables chosen for Box-Behnken design

Variables	Symbols	Coded levels		
		-1	0	+1
Sample-solvent (S/L) ratio, g/mL	X_1	1/15	1/20	1/25
Time, min	X_2	20	30	40
Temperature, °C	X_3	40	50	60

and used in the subsequent Box-Behnken experiments[18]. The number of experiments (17th Run) required to investigate the above three parameters using BBD are given in Table 2. In which, 12 are factorial experiments and 5 are zero point tests performed to estimate the pure errors. Each experiment has been performed in triplicate and the average natural dye yield is taken as the response.

Based on the Box-Behnken experimental design model, an empirical relationship between the input variables and the obtained experimental results are expressed by a second-order polynomial equation with interaction terms. The model obtained (equation) in terms of coded factors is given below:

$$Y = 0.8430 - 0.0096\,X_1 + 0.0965\,X_2 + 0.0024\,X_3 + 0.0145\,X_1X_2 - 0.0182\,X_1X_3 - 0.0115\,X_2X_3 - 0.0051\,X_1^2 + 0.0216\,X_2^2 - 0.0071\,X_3^2$$

where Y is the predicted response natural dye yield; X_1, X_2 and X_3, the coded terms for three independent test variables, sample-solvent ratio, sonication time and extraction temperature respectively.

The statistical significance and fitness of the model are evaluated by the analysis of variance (ANOVA) as presented in Table 3. The model F-value of 44.6 with low p-value ($p < 0.0001$) indicates that the model is highly significant. The lack of fit F-value of 2.37 and the associated p-value of 0.212 are found insignificant due to relative pure error which indicates that the model equation is adequate for predicting the dye yield. The goodness of fit of the model is evaluated by the determination coefficient (R^2), adjusted determination coefficient (Adj. R^2), predicted determination coefficient (Pred. R^2) and coefficient of variance (CV) (Table 3). The $R^2 = 0.9829$ indicates that only 1.71% of the total variations are not explained by the model. The value of Adj. R^2 is also found very high (0.9608), confirming that the model is highly significant. The higher the Adj. R^2, the better is the degree of correlation between the values observed from the experiments and the values

Table 2 — Box-Behnken design matrix for three variables, and the observed and predicted response

Run No.	Sample-solvent ratio, g/mL	Time min	Temperature °C	Color yield, %	Predicted value, %
1	1/15	40	50	0.955	0.951
2	1/20	30	50	0.834	0.843
3	1/20	30	50	0.852	0.843
4	1/15	30	60	0.864	0.861
5	1/15	20	50	0.773	0.787
6	1/25	30	40	0.834	0.837
7	1/20	40	60	0.938	0.945
8	1/25	40	50	0.975	0.961
9	1/20	30	50	0.858	0.843
10	1/20	20	40	0.754	0.747
11	1/20	20	60	0.786	0.775
12	1/25	30	60	0.798	0.805
13	1/20	30	50	0.837	0.843
14	1/20	30	50	0.834	0.843
15	1/15	30	40	0.827	0.820
16	1/20	40	40	0.952	0.963
17	1/25	20	50	0.735	0.739

Table 3 — Analysis of variance (ANOVA) for the fitted quadratic polynomial model of color yield

Source	Coefficient estimate	Sum of squares	df	Standard error	Mean Square	F-value	p-value	Remarks
Model	0.843	0.0802	9	0.0063	0.0089	44.6	< 0.0001	Significant
X_1	-0.0096	0.0007	1	0.005	0.0007	3.71	0.0955	
X_2	0.0965	0.0745	1	0.005	0.0745	372.96	< 0.0001	
X_3	0.0024	0	1	0.005	0	0.2259	0.649	
X_{12}	0.0145	0.0008	1	0.0071	0.0008	4.21	0.0793	
X_{13}	-0.0182	0.0013	1	0.0071	0.0013	6.67	0.0363	
X_{23}	-0.0115	0.0005	1	0.0071	0.0005	2.65	0.1477	
X_1^2	-0.0051	0.0001	1	0.0069	0.0001	0.5537	0.4811	
X_2^2	0.0216	0.002	1	0.0069	0.002	9.86	0.0164	
X_3^2	-0.0071	0.0002	1	0.0069	0.0002	1.07	0.3353	
Residual		0.0014	7		0.0002			
Lack of Fit		0.0009	3		0.0003	2.37	0.212	Not significant
Pure Error		0.0005	4		0.0001			
Cor Total		0.0816	16					

Std. Dev.	0.0141	R^2	0.9829
Mean	0.8474	Adjusted R^2	0.9608
C.V. %	1.67	Predicted R^2	0.815
Adeq Precision	20.688		

predicted from the model. The predicted determination coefficient (Pred. R^2) is a measure of how good the model predicts a response value. The difference between Adj. $R^2$and the Pred. R^2 should be less than 0.20; otherwise, there may be a problem with either the data or the model. In this case, the Pred. R^2 is 0.8150, which is in reasonable agreement with the Adj. $R^2$of 0.9608.

Furthermore, the coefficient of variance (CV) is the ratio of the standard error of the estimate to the average value of the observed response defining by the reproducibility of the model. The low CV (1.67) clearly states that the deviations between experimental and predicted values are a low, and a high degree of precision and also show good reliability of the conducted experiments. The lack of fit F-value of 2.37 implies that the lack of fit is not significant relative to the pure error. There is 21.20% chance that a lack of fit F-value this large could occur due to noise. Adequate precision is the measure of signal to noise ratio and greater than 4 of this value is desirable. In this study, the ratio is found to be 20.688, which indicates an adequate signal and confirms that the model is significant for this present extraction process.

3.2 Diagnostics of Model Adequacy

Generally, it is important to confirm that the fitted model gives a sufficient approximation to the actual values. Unless the model shows a satisfactory fit, proceeding with an investigation and optimization of the fitted response surface likely gives poor or misleading results. In addition to the determination coefficient, the adequacy of the models is also evaluated by the residuals (the difference between the observed and the predicted response value) and the influence plots for the experimental data obtained. Residuals are thought as elements of variation, unexplained by the fitted model and then it is expected that they occur according to a normal distribution. Normal probability plots are a suitable graphical method for judging residuals normality. The observed residuals are plotted against the expected values, as they lie reasonably close on a straight line and show no deviation of the variance (Fig. 1a). This can confirm the normal distribution of the data. Diagnostic plots such as predicted versus actual (Fig. 1b) help us to evaluate the model suitability and to find out the relationship between predicted and experimental values. By constructing an externally studentized residuals plot, a check is made to analyze the experimental data to find out the satisfactory fit of the developed models and the plot shows that all the data points lie within the limits (Fig. 1c). The data points on this plot lie reasonably close to the straight line and indicate that this is an adequate agreement between real data and the data obtained from the models. Figure 1d shows that all the data points lie within the limits. The difference of fits plot is a measure of the influence of each point on the predicted value. Figure 1e suggested that there are no high deviations in the experimental data. Hence, trends observed in Fig. 1 reveal that no obvious patterns are found and residuals appear to be randomly scattered.

3.3 Influence of Process Variables on Extraction

Three factors at three-level BBD have been used in this study to investigate the influence of process variables such as sample-solvent ratio, sonication time and extraction temperature by the ultrasound-assisted extraction method of natural pigment from the flower of *S. mahagoni*. To understand the interaction between the independent variables and to estimate the color extraction efficiency over independent variables, three dimensional (3D) response surface plots are made from the developed model (Fig. 2). In this study, the model has more than two factors. So, the 3D plots have drawn by maintaining two factors at a constant level (in turn at its central level), whereas the other two factors are varied in their range ,in order to understand their main and interactive effects on the dependent variables. It is also used to locate the optimum conditions.

3.3.1 Influence of Sample-solvent Ratio

The sample-solvent ratio is one of the most critical factors during the mass transfer because a larger volume of solvent helps to accelerate the diffusion process[22]. It is ideal to provide the amount of solvent required to enter the cells, thereby improving the permeation of the natural color compounds[23]. Therefore, in this study, the influence of the sample-solvent ratio on the extraction yield has been evaluated. It is observed that the extraction yield of dye increases with increasing sample-solvent ratio (Figs 2a and b). A higher volume of solvent enhances the efficiency of extraction by creating a concentration difference between the interior plant cell and the exterior solvent, which in turn favors mass transfer. Too much solvent would not change much of the driving force anymore as the limitation to

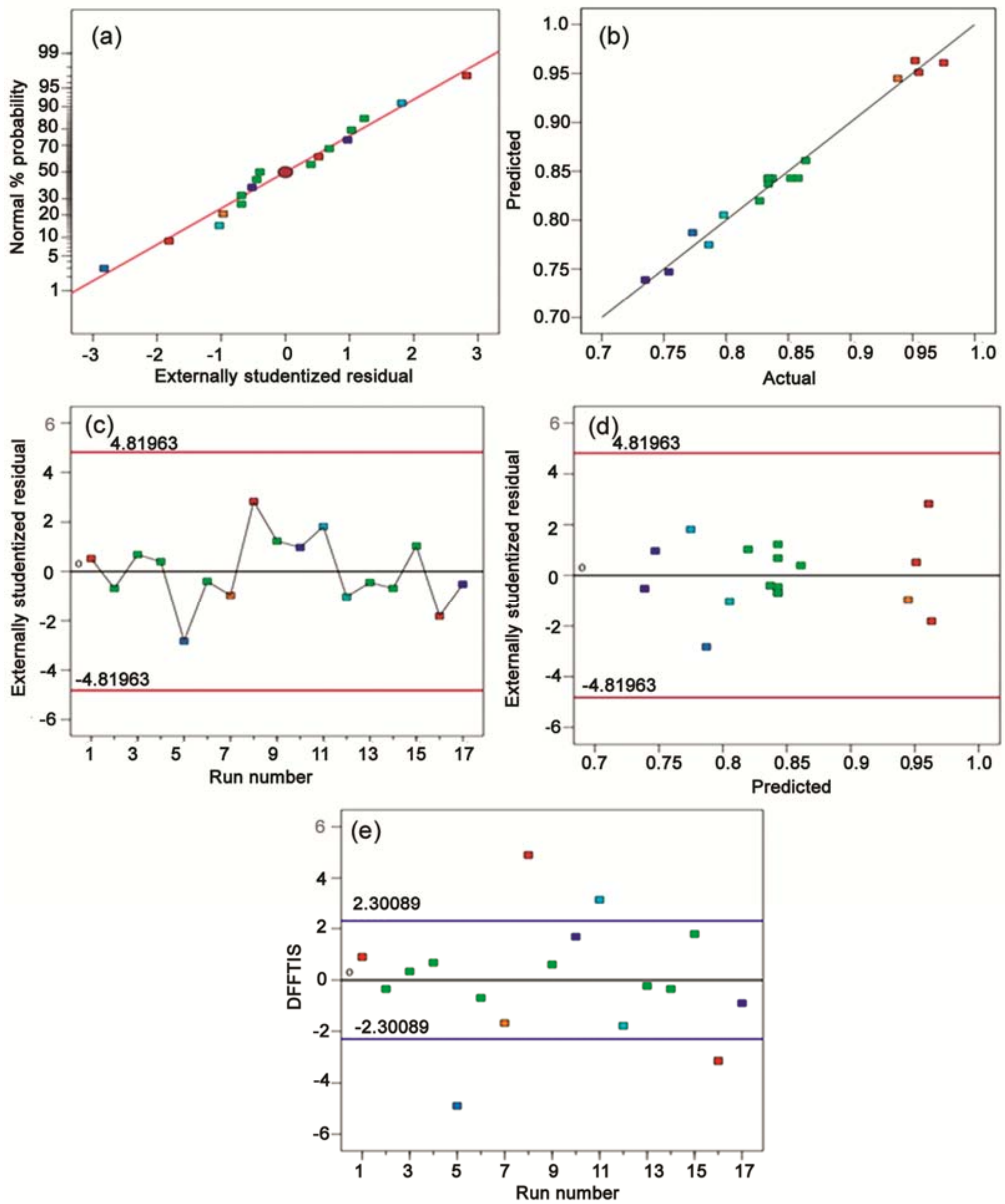

Fig. 1 — Diagnostic plots for the medel adequecy (a) residuals against the expected values; (b) predicted versus actual values; (c) externally studentized residuals plot; (d) predicted values against externally studentized residuals; and (e) standard error estimation plot

mass transfer is more confined to the solid interior. However, the combined effect of sample-solvent ratio (more than 1:20 g/mL) with the optimum conditions of other process variables (at extraction temperature 50 °C, and sonication time 30 min) observed in this study would not cause any further increase in the natural dye yield. Taking all these factors into consideration, a sample-solvent ratio of 1:20 g/mL is considered as optimal for this present extraction process.

3.3.2 Influence of Sonication Time

During the ultrasound-assisted extraction process, the solutes are in contact with the solvent, so the extraction efficiency is greatly influenced by the interaction time between the two phases[24]. The content of natural color compounds extracted by ultrasound increases as a function of time due to the solvent and sample matrix contact. As shown in Figs 2a and c, the extraction yield is increased steadily and reaches the maximum at 30 min. This phenomenon could be explained by the fact that all the plant cells will be completely cracked because of acoustic cavitation effects caused by the ultrasonic waves in the earlier period of extraction[25]. So, larger contact area between solvent and material is created and the collapse of the bubbles will promote the interpenetration of the solvent into the plant cells to dissolve most of the color present in it and increases the extraction yield. The number of cavitation micro-bubbles created by ultrasound increases with the duration of extraction extended. The asymmetric collapse of micro-bubbles near surfaces is also found associated with micro-jets that could scour surfaces and damage substance in solution[26]. However, when the plant cells rupture, various compounds such as insoluble substances are also found suspended in the extraction liquid, resulting in the lower permeability of the solvent. So, the yield is decreased slowly when the extraction time is extended. Thus, the optimum extraction time is 30 min.

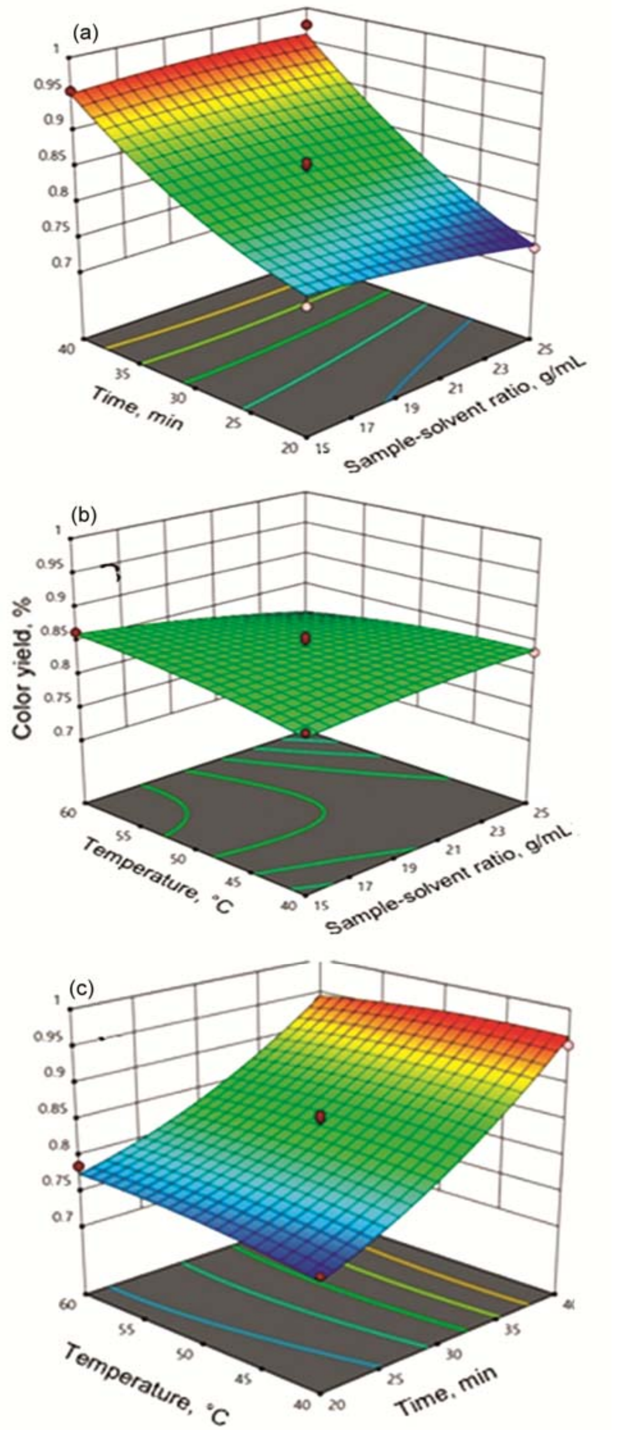

Fig. 2 — Response surface plots representing the effect of process condition on the extraction of natural dye yield

3.3.3 Influence of Extraction Temperature

Besides the effect of extraction sample-solvent ratio and sonication time, the temperature also influences the effectiveness of the extraction process. The study reveal the effect of temperature over the extraction yield of natural pigment from the flower of *S. mahagoni*. It is observed that the yield of color is increased linearly with increasing temperature from 40 °C to 60 °C (Figs 2b and c). The effect of increasing temperature is related to increase in the number of cavitation bubbles and in the surface contact area. Ultrasound irradiation will induce the cavitation on the disruption of cell wall which increases permeability of solid materials, so that the solvent can penetrate into the inner area. Increasing temperature also decreases the surface tensions of the solvent, and hence cavitation is conveniently formed. The influence of relative greater force ruptureand erupte the formed cavitational nucleus and disrupte the cell tissues during extraction, which, in turn, enhances the mass transfer rate and increases the extraction efficiency (Figs 2b and c). Additionally, solvents have a greater capacity to solubilize the analytes at higher temperatures, while surface tension and solvent viscosity decrease with increasing temperature, which will improve sample wetting and matrix penetration respectively[27]. As the solvent moves deeper, its area of exposure increases which ends up with higher extraction efficiency. Based on the results, 50 °C is chosen as the optimum extraction temperature.

3.4 Optimization of Extraction Parameters and Validation of Optimized Conditions

According to the Box-Behnken design results, the optimal extraction conditions to obtain maximum

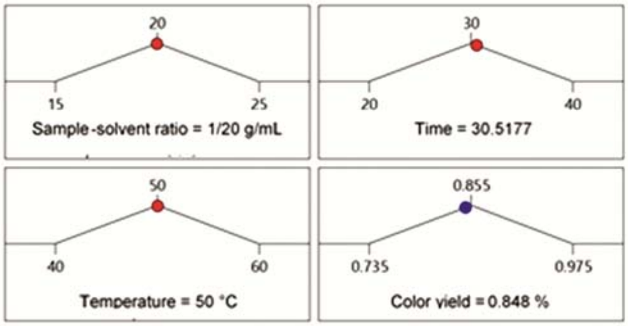

Fig. 3 — Desirability ramp for optimization of dye yield

extraction yield of dye from the flower of *S. mahagoni* are the sample-solvent ratio of 1:20 g/mL, sonication time 30 min and extraction temperature 50 °C. Under these conditions, the predicted extraction yield of dye is found 0.855% with a desirability value of 0.972. A desirability ramp is developed from optimal points via numerical optimization technique (Fig. 3). The suitability of the optimized conditions for predicting the optimum response values is tested using the selected optimal conditions. The experimental mean value of natural dye yield (0.853) is obtained at optimal conditions. The good correlation between these observed results and predicted values indicates the reliability of BBD incorporate desirability function method and it could be effectively used to optimize the extraction parameters on the maximum extraction yield of natural dye.

3.5 Characterization of Extracted Dye

FTIR is particularly useful for the identification of organic molecular groups and compounds due to the range of functional groups, side chains and cross-links involved; they have characteristic vibrational frequencies in the infrared range[21]. The broadband at 3411 cm^{-1} can be attributed to -OH stretching vibrations due to the presence of alcohol or phenol. The weak peaks at 2926cm^{-1}, 1520 cm^{-1} and 1260 cm^{-1} are indicative of C-H stretching vibration of alkane, C=C stretching of alkene, and C-H waging vibration of alkyl halide respectively. The strong band at 1614 cm^{-1} is ascribed to N-H bending vibration for presenting primary amine. At the range, 1446 cm^{-1} and 1384 cm^{-1} are represented for C-H stretching vibration in ring. Finally, the strong peak at 1069cm^{-1} is revealed for C-N stretching due to the presence of an aliphatic amine. Hence, FTIR spectral analysis has described the presence of different chemical functional groups like -OH, C=C, N-H, and C-H in the ring of extracted dye.

3.6 Fastness Property Evaluation

The simultaneous mordanting method impartes good shades on silk yarn. The color fastness data that the extracted dye from the plant species *S. mahagoni* furnish different color hues with a very good affinity for silk yarn in the presence of metal salt (alum) as mordant. The color intensity is found to be maximum and glazy when treated with Al(III) as compared to unmordanted silk fabric. The results of colourfastness show that these properties has been improved by dyeing followed by mordanting. The wash fastness of dyed silk fabric reveals that mordanting improves the rating from 4 to 5. It is the case observed during light fastness. The brightness of the shades on silk yarn might be due to the better absorption of dye and the easy formation of metal complexes with the yarn. The results are at par with the previous studies[28]. The different mixtures of these natural colorants have been subjected to dyeing silk yarns and have revealed the synergetic impact on colors.

4 Conclusion

In the current study, major emphasis has been laid on the extraction of eco-friendly natural dyes from flowers of the *S. mahagoni* plant by ultrasound-assisted solvent extraction. The results reveal that ultrasound-assisted extraction is an effective technique for natural color extraction in comparison with the conventional extraction method. The benefit of this technique includes less solvent requirment, lower heat consumption, less extraction time and more satisfactory color obtained in both quantity and quality. Box-Behnken design is successfully employed to optimize the values and to study the individual and interactive effect of process variables such as sample-solvent ratio, sonication time, and extraction temperature on the extraction yield of natural dye from the flower of the *S. mahagoni* plant by UAE. The results indicate that the factors selected in this study have a significant effect on the extraction of color. The yield of dye increases with increasing sample-solvent ratio, sonication time, and extraction temperature. From the ANOVA results, a high correlation second-order polynomial regression model is developed and this could be employed to optimize dye extraction from the flower of *S. mahagoni* by ultrasonic technology. The optimum conditions are found to be sample-solvent ratio 1/20 g/mL, sonication time 30 min, and extraction temperature 50 °C. Under these conditions, the total amount of dye extracted from the plant flower is estimated to be

0.853%. The extracted dye is characterized by FTIR spectroscopic analysis, and the spectra have revealed the presence of different chemical functional groups, such as -OH, C=C, N-H; and C-H stretching in ring inside the extracted dye. The silk yarn dyed with natural colorants exhibits acceptable fastness properties. Based on the results presented in this investigation, we can provide the opportunity to initiate a project for scaling up of the above process. The current findings clearly demonstrate that the extraction of natural colorants from flowers of the plant is a sustainable technique towards waste utilization. However, the data generated from the present study may foresee the economic viability of dye production in commercial-scale for the utilization of bio-resources in developing countries for their socio-economic growth.

References

1 Narayana Swamy V, Ninge Gowda K N & Sudhakar R, *J Inst Eng Ser E*, 95 (2014) 11.

2 Samanta A K & Agarwal P, *Indian J Fibre Text Res*, 34 (2009) 384.

3 Vadwala Y & Kola N, *Int J Home Sci*, 3 (2017) 175.

4 Uddin M G, *Text Cloth Sustain* 1 (2015) 7.

5 Hasan M M, Nayem K A, Azim A Y M A & Ghosh N C, *J Text Inst*, 2015 (2015) 1.

6 Mirjalili M, Nazarpoor K & Karimi L, *J Clean Prod*, 19 (2011) 1045. 7.

7 Ibrahim N A, El-Gamal A R, Gouda M & Mahrous F, *Carbohydr Polym*, 82 (2010) 1205.

8 Haque M A, Arifuzzaman G M, Razzaque S M, Khatun K, Chakraborty A K & Alam M S, *Indian J Fibre Text Res,* 38 (2013) 280.

9 Rahman S M A, Maksuda H, Tasnuva E M, Yeunus A, Iftekhar R & Muhammed M, *J Pharmacogn Phytochem,* 2 (2014) 185.

10 Sivakumar V, Vijaeeswarri J & Anna J L, *Ind Crops Prod*, 33 (2011) 116.

11 Khan A A, Naeem A, Shahid A, Muhammad B, Fatima B & Ijaz A, *Dye Pigment*, 103 (2014) 50.

12 Salarbashi D, Mortazavi S A, Rezaei K, Rajaei A & Karimkhani M M , *Food Sci Biotechnol*, 21 (2012) 1005.

13 Wang L & Weller C L, *Trends Food Sci Technol*, 17 (2006) 300.

14 Yolmeh M, Habibi Najafi M B & Farhoosh R, *Food Chem*, 155 (2014) 319.

15 Santos D T, Veggi P C & Meireles M A A, *J Food Eng*, 101 (2010) 23.

16 Azmir J, Rahman I S M, Sharif M M, Mohamed K M, Sahena A, Jahurul F, Ghafoor M H A, Norulaini K & Omar A K M, *J Food Eng*, 117 (2013) 426.

17 Joana G G, G Villa, José A F A, Basilio H, Sepulveda D Y, Elhadi M & Gustavo A, *Food Sci Food Saf*, 12 (2013) 5.

18 Almeida M, Erthal R, Padua E, Silveira L & Am L, *Talanta*, 76 (2008) 965.

19 Ferreira S L C, Bruns R E, Ferreira H S, Matos G D, David J M, Brand G C, Silva E G P, Reis P S, Souza A S & Santos W N L , *Anal Chem Acta*, 597 (2007) 179.

20 Ying Z, Han X & Li J, *Food Chem*, 127 (2011) 1273.

21 Sinha K, Das P & Datta S, *Dye Pigment*, 94 (2012) 212.

22 Muñiz-Márquez D B, Martínez-Ávila G C, Wong-Paz J E, Belmares-Cerda R, Rodríguez-Herrera R & Aguilar C N, *Ultrason Sonochem J*, 20 (2013) 1149.

23 Sousa A D M, Ana I V R, Tigressa H S C, Kirley M R, Paulo R V, Rita V, Roberto F B & Edy Sousa, *Ind Crops Prod*, 79 (2016) 91.

24 Hossain M B, Brunton N P, Patras A T, Brijesh O D C P, Martin-Diana A B & Barry-Ryan C *Ultrason Sonochem,* 19 (2012) 582.

25 Maran J P, Mekala V & Manikandan S, *Carbohydr Polym*, 92 (2013) 2018.

26 Vilkhu K, Mawson R, Simons L & Bates D, *Innov Food Sci Emerg Technol*, 9 (2008) 161.

27 Xie J, Xie M, Shen M & Nie S, *J Sci Food Agricalture*, 90 (2010) 1353.

28 Prusty A K, Das T, Nayak A & Das N B, *J Clean Prod*, 18 (2010) 1750.