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Extraction of lutein dye from Tagetes erecta garland waste for green dyeing of hemp fabric using response surface methodology

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Abstract

Large quantities of discarded flowers from religious observances are left at temples and other places of worship, causing global disposal and environmental issues. Recycling and transforming such organic waste into value-added products is one of the most effective and beneficial solutions to the problem. The main goal of this study is to convert the most abundant temple wastes of marigold (*Tagetes erecta*) flowers into an eco-friendly dyestuff for the textile industry. Our study assessed the suitability of dye extract from garland waste for dyeing hemp fabric and valuated indicators including color strength (K/S) and fastness properties using tannic acid as a bio-mordant. Response surface methodology (RSM) was used for optimization of the dyeing process and evaluation of the interaction effects of various operating parameters. The optimal conditions were determined to be pH of 4.23, dyeing temperature of 99.98°C, and dyeing time of 82.64 min. To validate the optimal conditions identified by RSM, performance evaluations were conducted, including color fastness properties of the dyed hemp fabrics as well as the total color difference after repeated standard washing. These results demonstrate the use of aqueous extract from temple garland waste combined with bio-mordant represents a promising approach for textile dyeing.

1. Introduction

Waste disposal is the most challenge issue in the globalized field of research. Garland waste is one of the major concern [1,2]. Flowers are regularly used in religious places, for celebrating various social occasions as well as used as a raw material in different industries which generate large quantities of waste. The safe disposal of flower waste has been a concern for the temple's management. However, the management of this garland waste is poorly addressed all over the globe [2]. In general, temple waste is disposed of directly into landfills without any treatment, such as waste recycling. However, sometimes it is found that the temple waste is discarded into the rivers or dumped in an open place. The improper disposal of such waste creates an array of environmental and human health problems [2,3]. According to the WHO, 36% of the chemicals used in flower farms are extremely toxic, and when discarded in landfills, they can produce 22 times more environmentally destructive greenhouse gases than carbon dioxide [4]. The appropriate management of such waste could provide as converting them into useful or value-added products such as biochar, biofertilizer, dye extraction, essential oil, color powder, biogas, etc. [2,5,6].

Marigold (*Tagetes erecta*) from *Asteraceae* family is one of the most widely used flowers for ornamental purposes universally. It is the most commonly used for religious and ritual garlands, as shown in Figure 1. Large amounts of marigold flowers are discarded after use, and hence natural dyeing is an alternative to using this wasted biomass, which can be utilized as dyeing or finishing materials on textiles. The main colorant components of marigold flower are carotenoid compounds such as lutein and lutein esters. Lutein ($C_{40}H_{56}O_2$) (Figure 2) is a bright yellow xanthophyll pigment and is very popularly used in medical and food industries [7,8]. A study has shown that lutein is a pigment, which is water soluble and behaves like a dye [9]. Studies on dyeing of various textiles such as wool, silk, and cotton with marigold flower extracts have been reported [9-14]. To the best of our knowledge, the dyeing properties of marigold flower extracts on hemp textiles have never been described. Generally, natural dyes are known to have some drawbacks, such as poor color fastness and a narrow shade range for the dyed fibers. To overcome the problems, textile industry requires mordants for increasing the dye affinity to the textile fibers [15]. Metal salts are the most commonly used mordants, but most are non-biodegradable and harmful, leading to water pollution and health risk. Therefore, replacement of conventional metal ions with natural mordant is necessary in the development of natural colorants for sustainable textile industry [16,17]. With the growing trend of using green materials, tannins have received a lot of attention. Tannin is a naturally occurring polyphenol that is abundant in the tissues of vascular plants [18,19]. As a renewable resource, tannin can reduce the demand for chemical

Figure 1. Marigold garlands in Buddhist temple in Thailand.

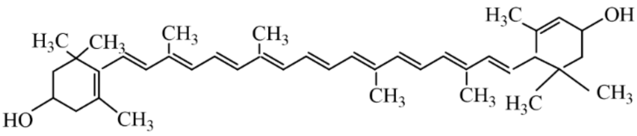


Figure 2. Chemical structure of Lutein.

additives and has the benefits of being cost-effective and nontoxic. Multiple phenolic hydroxyl groups in tannin lead to the formation of complexes with proteins, metal ions, and other macromolecules such as polysaccharides. As a result, tannin is utilized in textile dyeing as a colorant compound and a bio-mordant to increase the uptake of the dyes and improve their fastness properties [20]. Tannin can be divided into two types based on its chemical structure: condensed tannin and hydrolyzable tannin [21]. Tannic acid is one of the simple and special forms of hydrolyzable tannin. When tannic acid is used as a mordant, natural dyes bind to fibers more effectively, enhancing the quality of the dyeing process [22,23].

Response surface methodology (RSM) is an effective tool for identifying the optimum set of operational variables for process development and optimization [24]. RSM can also identify the interactions, thoroughly covers the design space, and is able to establish the solution with minimal usage of the resources [25]. RSM has been extensively utilized to optimize the extraction process of natural dyes from a variety of sources [26-30]. In addition, there are studies that demonstrate the advantages of using RSM to optimize variables in the textile dying process, but most of these studies have focused on the dyeing cotton, wool, silk, and some synthetic textiles [31-34].

Our study aimed at exercising the leverage of temple waste, especially discarded flowers, into natural colorants through recycling, the majority of which are marigolds. Although the most popular cellulosic fabric is cotton, the climate of Thailand and Southeast Asian countries is unsuitable for its commercial production. Offering sustainability and economic value, hemp (*Cannabis sativa*) has been recently designated as an industrial crop in Thailand and can be cultivated in the northern region. Hemp fibers are composed of cellulose, hemicellulose, pectin, lignin, ester wax, water-soluble matter, and a small amount of ash [35]. The presence of cellulosic substances in hemp fibers offers characteristics such as good water absorption, comfort, and stability. To encourage the use of hemp fabrics in high-added value products, the color yield and color fastness must be improved. Specifically, the purpose of our study was to investigate the suitability of dye extract from *Tagetes erecta* garland waste for dyeing hemp fabric. Besides, modelling and optimization of the operating parameters were investigated in order to improve the performance of this dyeing process. Traditionally, the optimization of textile dyeing processes has been done by observing the impact of one factor on response output in the absence of changes to other factors (one-factor-at-a-time methodology). However, in textile dyeing processes, the effect of one process variable depends on the others, and the interactions between factors need to be taken into account. Thus, the effects of the dyeing process factors (pH, dyeing temperature, and dyeing time) on the color strength (K/S) of the dyed hemp fabrics will be investigated using RSM and central composite design (CCD). The optimal amount of each effect for attaining the highest color yield will be based on the experimental results and the established model. To validate the optimal conditions identified by RSM, performance evaluations were conducted, including color fastness properties according to the AATCC Test Method standard and ISO standard and total color difference after repeated standard washing. The goals of this study are: (1) to promote the usage of temple garland waste in textile dyeing through scientific approaches; (2) to improve dye crafts by bio-mordant through three mordanting methods; and (3) to investigate a yellow colorant derived from garland waste on hemp fibers, a new commercial crop in Southeast Asia.

2. Material and experimental

2.1 Materials

Plain weave hemp fabric (mass per unit area: 250 $g{\cdot}m^{-2}$, warp and weft densities: 140 ends/inch, and 75 picks/inch) was purchased from a local market in Chiang Mai Province, Thailand. The tensile strengths in the warp and weft directions of the hemp fabric are 556.8 N and 542.6 N, respectively. The scouring was done with 2 $g{\cdot}L^{-1}$ AATCC detergent at 70°C for 30 min to remove the dirt and impurities from the fabric. Marigold flowers were obtained from local Buddhist temple, Prathumthami Province, Thailand. Tannic acid ($C_{76}H_{52}O_{46}$), sodium hydroxide (NaOH), and hydrochloric acid (HCl) of analytical grade were purchased from Sigma-Aldrich (Thailand) Company Limited.

2.2 Aqueous extraction of natural dye

Firstly, the fresh marigold flowers (Tagetes erecta) were collected. The petals were separated, washed thoroughly, and sun-dried before use. The dried petals were ground to make a dye powder. To extract the dye, 100 g of the dye powder in 1 L of DI water were refluxed for 15 min and heated at 100 °C for 2 h. Solid residues were removed by filtration, and the solution concentration was adjusted to 10 wt% and had a pH of 4.58, as shown in Figure 3.

Figure 3. The aqueous extract of *Tagetes erecta* flowers.

2.3 Dyeing

According to the experimental design (Table 1), the untreated and treated hemp samples were dyed in an infrared dyeing machine (Starlet DL-6000), which contained the concentration of *Tagetes erecta* flower extract (10 wt%). The pH value was adjusted to (3 to 12) by adding 1 M HCl or NaOH solutions, and the temperature of the dye bath was raised to (60°C to 110°C). The dyeing was conducted at a material to liquor ratio of 1:30, following the procedure presented in Figure 4. Finally, the dyed samples were then washed with 2 $g{\cdot}L^{-1}$ of the 1993 AATCC Standard Reference Detergent without optical brightener at 95°C for 30 min to remove excess and unfixed dyes, followed by repeated water washing, and then dried at room temperature.

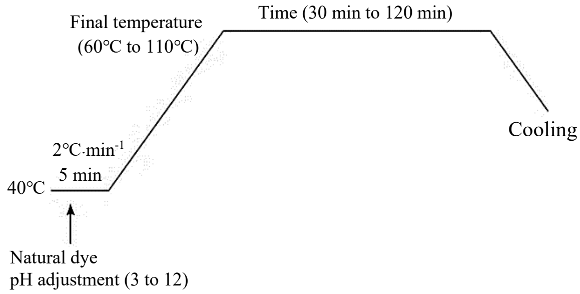


Figure 4. Dyeing procedure.

2.4 Mordanting

The mordanting process was conducted for 60 min at 70°C with tannic acid (5% on weight of fabric, owf) at a material to liquor ratio of 1:30. Methods for pre-, post-, and meta-mordanting were investigated. For the pre-mordanting method, the fabric was first put into the mordanted solution and then into the dyed solution, and for post-mordanting, the sequence was reversed. For the meta-mordanting method, the dyed solution and mordanted solution are mixed together for the dyeing process.

2.5 UV-visible spectroscopy

The UV-visible spectral of the natural dye extracted from the marigold flowers was examined by the Shimadzu UV 1800 spectrophotometer. The UV-Vis spectrum of the dye was measured in the range of 300 nm to 800 nm. The dilution of the extracted dye was by 1 mL of dye extract to 80 mL of deionized water.

2.6 Color measurement and color fastness properties

Each dyed sample was measured for color coordinates and color strength (*K/S*) values on a spectrophotometer (GretagMacbeth LLC, Switzerland). The settings of the apparatus were as follows: illuminant D65, 10° standard observer, and specular and UV included. Each measurement was made three times, and the average values were recorded. The *K/S* values were assessed by using the Kubelka-Munk equation as shown in Equation (1).

$$\frac{K}{S} = \frac{(1-R)^2}{2R} \quad (1)$$

Where R is the reflectance of the samples at maximum absorption wavelength, K is the absorption coefficient, and S is the scattering coefficient.

The color coordinates of the dyed samples are expressed in terms of the CIELab coordinates (L^*, a^*, b^*), where L^* corresponds to the brightness (100 = white, 0 = black), a^* to the red-green coordinate (+ve = red, -ve = green), and b^* to the yellow-blue coordinate (+ve = yellow, -ve = blue). The C^* value representing chroma (vividness or dullness), h^o denoting hue angle, and total color difference (ΔE) were calculated using Equation (2), Equation (3), and Equation (4), respectively.

$$C^* = [(a^*)^2 + (b^*)^2]^{1/2} \quad (2)$$

$$h^o = arctan(b^*/a^*) \quad (3)$$

$$\Delta E = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2} \quad (4)$$

Where $\Delta L^* = L^*_{unwashed} - L^*_{washed}$, $\Delta a^* = a^*_{unwashed} - a^*_{washed}$ $\Delta b^* = b^*_{unwashed} - b^*_{washed}$

The percentage of dye uptake (*%E*) is calculated using Equation (5), where A_o and A_r are the absorbance of the dyebath before and after dyeing at λ_{max} of the extracted dye used.

$$\%E = \frac{A^0 - Ar}{A_o} \times 100 \quad (5)$$

Color fastness properties to washing, crocking, light, and perspiration were evaluated following AATCC Test Method 61-1A:2013, AATCC Test Method 8-2013, ISO 105-B02:1994, and ISO 105-E04:1994, respectively.

2.7 Experimental design and data analysis

In order to identify the best combination of experimental dyeing process parameters for optimal color strength of the dyed material, RSM was used. RSM is applied to model the relationship between multiple experimental parameters and one or more responses by modeling linear and nonlinear relationships between these parameters and responses, as well as their interactions. Three important dyeing process parameters (dyebath pH, dyeing time, and dyeing temperature)

Table 1. Experimental factors and range for CCD.

Symbol	Factor	Unit	Lower limit	Upper limit
A	dyebath pH	-	3	12
B	dyeing time	min	30	120
C	dyeing temperature	°C	60	110

were assessed using central composite design (CCD) in conjunction with RSM. Preliminary trials were conducted to identify the range of each input parameter and the minimum number of experimental runs, as shown in Table 1. The experimental design, statistical analysis, and optimization of the dyeing process parameters were performed by Minitab software (version 20). A total of 20 experiments were performed according to CCD. Then, the optimum values of the selected parameters were generated by solving the regression model equation and by analyzing the response surface contour plots. In RSM, a polynomial response surface is employed to present the relationship between a response Y and predicted variables X. The functional relationship for a quadratic model with four experimental variables is presented in Equation (6).

$$Y = b_0 + \sum_{i=1}^{3} b_i X_i + \sum_{i=1}^{3} b_{ii} X_i^2 + \sum_{i=1}^{3} b_{ij} X_i X_j \quad (6)$$

Where Y represents the response function, b_0 is the intercept, b_i, b_{ii} are the coefficients of the quadratic model with four experimental parameters.

To evaluate the significance of each parameter in the predicted model, analysis of variance (ANOVA) was performed and a *p*-value of less than 0.05 was determined as statistically significant, which means the value indicates statistical significance with a 95% confidence level.

3. Results and discussion

3.1 Absorbance of the aqueous extract of *Tagetes erecta*

An UV-vis absorption spectrum of *Tagetes erecta* flower after aqueous extraction is shown in Figure 5. It shows the principal absorption peaks at 443 nm and 475 nm, which are similar to the report data for lutein pigment [36,37].

3.2 Response surface methodology regression

The experimental results of CCD based on three factors with five levels are shown in Table 2. Three major parameters were selected to determine the optimal conditions of the hemp dyeing using RSM. In this study, the considered response or dependent variable was the color strength (K/S) whereas the independent variables were the pH of the dye bath, dyeing time, and dyeing temperature. Then, the results were analyzed, and a regression analysis by a quadratic model led to the following equation:

$$\begin{aligned}\text{Response (K/S)} = &-25.1 + 3.73*A + 0.193*B + 0.611*C \\ &- 0.2416*A^2 - 0.0023*B^2 - 0.0036*C^2 \\ &- 0.0053*A*B - 0.0117*A*C + 0.0021*B*C\end{aligned}$$

Where, Response is K/S, A is dyebath pH, B is dyeing time, and C is dyeing temperature.

The statistical significance of the fitted quadratic model equation based on ANOVA is shown in Table 3. To identify the significant variables and interactions in the dyeing process, the model term having a *p*-value higher than 0.05 was considered as insignificant and eliminated from the final model. The quality of the model fit was expressed by the correlation coefficient (R^2), and its statistical

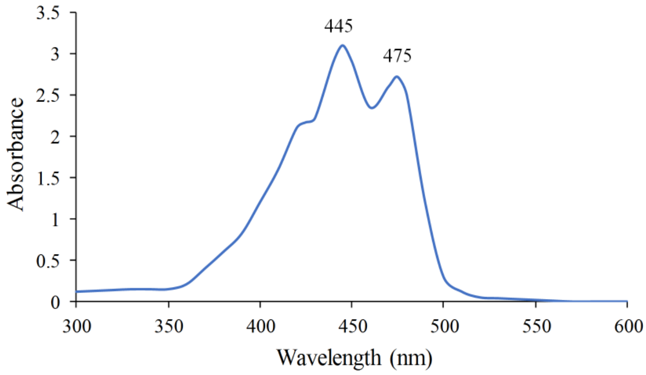


Figure 5. Ultraviolet-visible spectrum of *Tagetes erecta* flower under aqueous extraction.

significance is confirmed using the *p*-value. The closer the value of R^2 is to 1, the better the model fit. When the *p*-value for the model is at the 5% level (p <0.05), the regression model is accepted. Otherwise, the *p*-value for lack of fit is higher than 0.05 [27]. In this study, the predicted model represents a R^2 of 0.9714, which implied that only 3% of the variation could not be explained by this model. The predicted R^2 demonstrates the accuracy of a regression model in prediction of responses for new trials. The adjusted R^2 is a modified version of the correlation coefficient (R^2) for comparing the ability of models with different number of variables to predict the response. If the difference between adjusted R^2 and predicted R^2 is less than 0.2, the model is sufficiently accurate [31]. In this study, the adjusted R^2 and predicted R^2 were 0.9456 and 0.7822, respectively. In addition, the Fisher's *F*-value (37.70) with a low probability value (p <0.05) suggested that the model was statistically significant, thus the experimental values agreed very well with the predicted ones, providing a good predictability of the model. So, it can be concluded that the obtained model has a good predictability within the range of the chosen variables.

The ANOVA results of the applied quadratic model to navigate the design space of hemp fiber dyeing with *Tagetes erecta* are shown in Table 4. The statistically significant or insignificant factors are determined based on the *p*-values of the independent variables. The *p*-value less than 0.05 suggest that its effect on response is significant. Therefore, it can be inferred that the dyebath pH (A), dyeing time (B), and dyeing temperature (C) were the significant factors on the color strength of dyed hemp fiber with *Tagetes erecta,* due to their low probability values (<0.05) and high F value. Moreover, their interactions such as BC, A^2, B^2, and C^2 were significant model terms. Overall, the ANOVA analysis signifies the applicability of the model for color strength of dyed hemp with *Tagetes Erecta* extract at the time of the dyeing process within the limits of the experimental factors. A regression analysis of the model equation indicates that the main, square, and the interaction effects of independent variables were significant.

3.3 Effect of dyeing conditions on dyeing quality

In this study, the effects of pH, temperature, and time on the performance of the dyeing process were investigated. The results were evaluated by measuring the color strength (K/S) of the dyed samples.

Table 2. CCD experiments of samples and corresponding response.

Run	Coded level of variables			Actual level of variable			Response	Color coordinates				
	X1	X1	X1	pH	time (min)	Temp (°C)	K/S	*L**	*a**	*b**	*C**	*h*°
1	-1	1	-1	5	102	70	7.20	53.79	3.65	20.2	20.53	79.77
2	-1	-1	-1	5	48	70	6.57	55.62	3.21	20.18	20.43	80.97
3	-α	0	0	3	75	85	8.78	51.82	4.24	23.86	24.23	79.92
4	0	0	α	7.5	75	110	7.50	52.1	4.66	20.71	21.22	77.33
5	0	0	0	7.5	75	85	8.16	53	4.88	21.9	22.5	77.5
6	1	-1	1	10	48	100	6.06	55.59	3.85	19.93	20.3	79.07
7	1	1	-1	10	102	70	4.20	55.99	2.97	16.05	16.32	79.52
8	0	0	0	7.5	75	85	7.88	62.52	1.29	23.58	23.61	86.87
9	-1	-1	1	5	48	100	9.00	60.36	1.73	26.25	26.31	86.23
10	0	0	0	7.5	75	85	7.42	54.82	3.51	19.52	19.84	79.8
11	0	0	-α	7.5	75	60	7.28	55.6	3.78	21.44	21.77	80.01
12	0	0	0	7.5	75	85	7.96	49.79	4.06	20.98	21.37	79.05
13	0	0	0	7.5	75	85	7.79	51.23	4.98	20.55	21.14	76.36
14	1	1	1	10	102	100	6.05	54.54	4.15	19.76	20.19	78.14
15	-1	1	1	5	102	100	8.83	56.73	1.71	25.38	25.44	86.14
16	0	α	0	7.5	120	85	6.00	51.81	3.83	17.54	17.95	77.67
17	1	-1	-1	10	48	70	5.63	58.3	3.14	20.16	20.4	81.14
18	0	0	0	7.5	75	85	7.93	59.63	2.68	25.57	25.71	84.03
19	0	-α	0	7.5	30	85	6.60	53.66	4.18	20.19	20.62	78.3
20	α	0	0	12	75	85	3.60	62.11	3.64	18.16	18.52	78.66

Table 3. ANOVA results of fitted quadratic model to the experimental data.

Source	DF	Sum of square	Mean square	F-value	P-value
Model	9	271.027	30.114	37.70	0.000
Linear	3	178.406	59.469	50.55	0.000
Square	3	78.534	26.178	22.25	0.000
Interaction	3	35.094	11.698	2.42	0.124
Residual	10	25.389	5.078		
Lack of fit	5	11.764	2.353	0.001	0.000
Pure error	5	0.051	0.017		
Total	19	277.243			

Table 4. Regression analysis for the established model.

Source	Coefficient estimate	Standard error	*F*-value	*P*-value
A-pH	-3.449	0.242	138.07	0.000
B-Time	-0.326	0.242	11.35	0.007
C-Temperature	1.150	0.242	12.24	0.006
AB	-0.375	0.383	0.96	0.351
AC	-0.466	0.383	1.48	0.252
BC	0.842	0.383	4.82	0.023
A^2	-1.730	0.286	36.67	0.000
B^2	-1.651	0.286	33.40	0.000
C^2	-0.796	0.286	7.77	0.019

3.3.1 Effect of pH in dyeing process

Figure 6 illustrates the effect of dyebath pH (3-12) on K/S values of the dyed fabrics. It is observed that the optimum K/S is obtained at acidic pH of 4. The fiber polymer and the dye molecule became more anionic at their hydroxyl sites due to the alkaline pH, creating repulsion between them and thus resulting in a lower K/S value [9].

3.3.2 Effect of dyeing time

Figure 7 illustrates the effect of dyeing time (30-120 min) on K/S values of the dyed fabrics. It shows that the K/S values increase as the dyeing time increases up to about 80 min, and then slightly decreases. It seems that the dyed fabrics had reached the saturation and can no longer absorb more dyes.

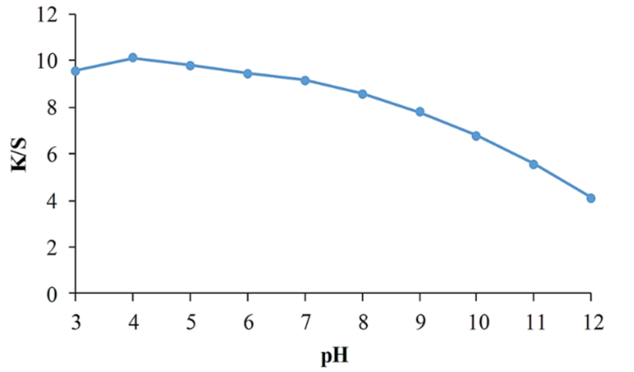


Figure 6. Effect of dye bath pH on color strength of dyed fabrics at dyeing temperature of 100°C and dyeing time of 80 min.

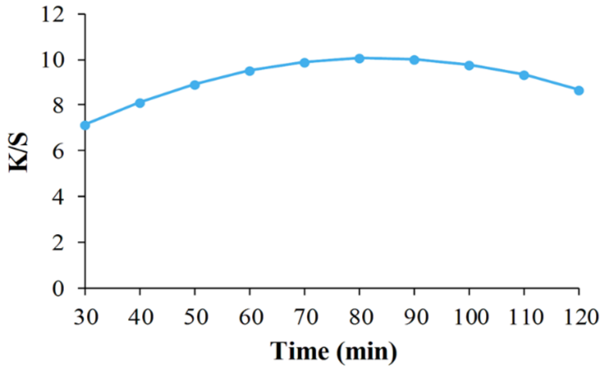


Figure 7. Effect of the dyeing time on color strength of dyed fabrics at pH of 4 and dyeing temperature of 100°C.

3.3.3 Effect of dyeing temperature

Figure 8 illustrates the effect of dyeing temperature (60°C to 110°C) on the K/S values of the dyed fabrics. As can be observed, it is indicated that the percentage of dye uptake increases with an increase in dye temperature and reaches a maximum value at approximately 100°C. The result may be due to the higher kinetic energy of the dye molecules and fiber swelling effects that enhance the dyeing capacity of the hemp fiber. Furthermore, the dye solution is comprised of single molecules as well as aggregates. Larger aggregates cause lower diffusion rates and surface adsorption, resulting in a decrease in the color strength and fastness properties upon the detachment from the surface during washing. Increasing temperature usually causes aggregates to break down, and dye exhaustion and color strength to increase, resulting in complete and even dyeing [9,10]. Higher dyeing temperatures (>100°C) decreased the K/S values, which might be attributed to the reduction of dye molecular stability [9].

3.4 Contour and 3D response surface plots

To examine the interaction among the varied independent variables and their corresponding effect on the response, the contour and response surface plots were made [32,38,39]. The contour plot is a graphical representation of a two-dimensional plane as a function of two independent variables where all points that have the same response are connected to produce contour lines of constant responses. The contour plots would indicate that the greatest shade depth is obtained from optimized values of dyeing variables, which are dyebath pH, dyeing time, and dyeing temperature. In the response surface and contour plots, the K/S values of the dyed fabrics are obtained along with two continuous variables, while another is fixed at some level. In this study, all the response surface plots (Figure 9-11) confirm that there is an optimum for the dyeing variables, maintaining the low and high levels in order to maximize the color strength of the dyed hemp fabrics. And, all of the contour plots (Figure 9-11) show that all three independent dyeing variables have significant positive effects on the K/S values of the dyed materials to achieve the maximum color yield.

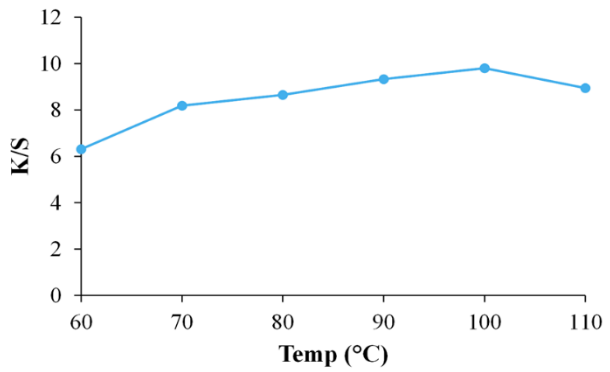


Figure 8. Effect of the dyeing temperature on color strength of dyed fabrics at pH of 4 and dyeing time of 80 min.

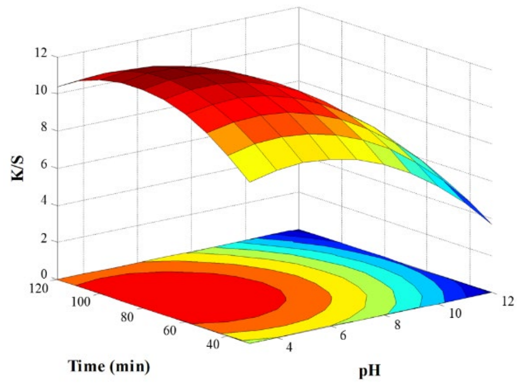


Figure 9. Contour and response surface plot showing the effect of pH and dyeing time.

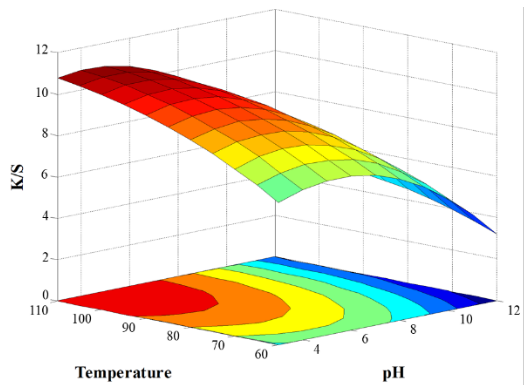


Figure 10. Contour and response surface plot showing the effect of pH and dyeing temperature.

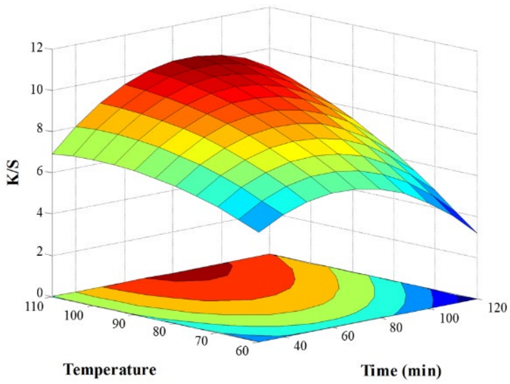


Figure 11. Contour and response surface plot showing the effect of dyeing time and temperature.

3.5 Response optimization and validation of the model

The dyeing conditions of hemp fabrics with *Tagetes erecta* flower extract were optimized using the optimization function of Minitab. The optimization function examines a combination of factor levels that simultaneously satisfy the goal placed on response and each factor. In this study, the dyeing condition was chosen in the proposed range that obtained the highest value of color strength. Due to that, the proposed optimized conditions for hemp fabric dyeing with flower extract are presented in Table 5. The results obtained when the optimal dyeing conditions are used are consistent with the theoretical results. Indeed, an experimental K/S value of 10.54 was obtained vs. a predicted value of 11.95. Therefore, the model proposed by this study is validated and confirmed. The maximum K/S value obtained was greater than 10, indicating a successful color buildup for the yellow hue. This demonstrates efficient utilization of lutein, the primary carotenoid pigment in marigold flowers. Hence, the obtained dyeing conditions were chosen for the dyeing of samples for evaluation of color fastness properties as well as the total color differences after repeated standard washing.

3.6 Effect of mordanting methods

Three conventional modanting methods, pre-mordanting, meta-mordanting, and post-mordanting, were used to dye hemp fabrics with the optimum condition obtained. Tannic acid is used as a bio-mordant. Table 6. represents the effects of various mordanting methods on dyeing hemp fabrics with the aqueous extract of Tagetes erecta flowers on K/S and the colorimetric data (L^*, a^*, b^*, C^*, and $h°$). Hemp fabric dyed without mordant had a shade of yellowness, while fabric mordanted with tannic acid had a dark yellow color shade. It was shown that types of modanting methods led to a variation of shade from light yellow to dark yellow. In the case of the pre-mordanting method, tannic acid gave the highest yield of shade value (K/S = 11.65). The K/S values of the dyed fabrics increased in the following order: pre-mordanting> meta-mordanting> post-mordanting> non-mordanting. It can be explained that cellulose fiber pre-mordant with tannic acid provides carboxylic acid groups (-COOH) and provides additional hydroxyl groups (-OH) in the dyeing system [23,32]. Tannic acid contains hydroxyl groups, unsaturated double bonds, and carboxyl groups, which chemically combine with groups on natural dyes and fibers or form hydrogen bonds and other intermolecular interactions to improve the color yield and color fastness of dyed cellulose fabrics. Tannic acid acts as a mordant in dyeing hemp with natural dye, using its –OH groups to make hydrogen bonds with cellulose while –COOH and –OH groups react with natural dye extract. This leads to the formation of a stable compound and fixes the color, as shown in Figure 12.

Table 5. Optimal dyeing conditions for obtaining the highest color strength.

A: pH	B: Time (min)	T: Temperature	K/S (predicted)	K/S (actual)
4.23	82.64	99.98	11.95	10.54

Table 6. Color parameters of hemp dyed with marigold extracts without and with bio-mordant.

Method	K/S	L^*	a^*	b^*	C*	h°	% dye uptake	Image
control	-	87.14	-1.15	13.09	13.14	95.03	-	
Without mordant	10.54	47.02	2.14	34.01	34.08	86.40	65.6	
Pre-mordant	11.65	50.95	4.04	40.64	40.84	84.33	81.7	
Meta-mordant	11.29	48.34	3.15	37.24	37.37	85.17	74.6	
Post-mordant	11.10	49.02	2.56	38.26	38.35	86.18	71.7	

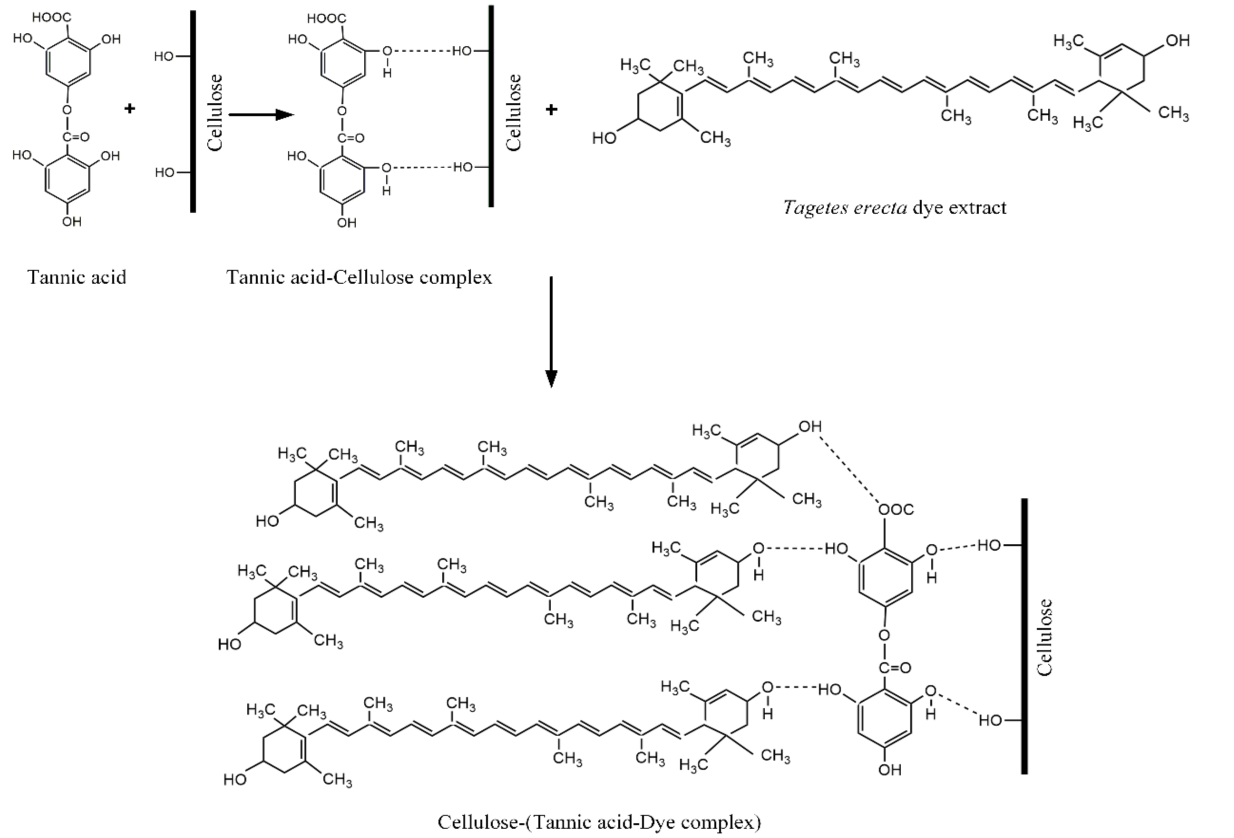


Figure 12. Proposed fixation of *Tagetes erecta* dye extract on pre-mordanted hemp.

3.7 Color fastness properties

Color fastness is the ability of colored textile to maintain its original color over the course of normal use and is a fundamental requirement for dyed goods marketed commercially [40,41]. Color fastness is defined by the degree of color change. Color stability is also evaluated from color staining, which is defined as excess pickup of dye by a substrate due to exposure to a contaminated medium or by direct contact with a dyed material [42,43]. Table 7-10 shows the washing, perspiration, light, and crocking fastness properties of both hemp fabrics dyed under the optimal condition of this study with and without mordant. Tannic acid is used as a bio-mordant and contains hydrophobic (aromatic), carboxylic acid, and hydrophilic (-OH) groups, while hemp fabrics also contain hydrophilic cellulose groups. The washing fastness was evaluated on the basis of color change based on the rate of diffusion of dye molecules. Tannic acid exhibited increased color change and staining properties in washing fastness. There was no substantial difference in washing fastness between the three different modanting methods. Table 9 highlights the light colorfastness ratings of dyed fabrics with and without mordant. Actually, it is generally known that textiles dyed with most natural dyestuffs have poor color fastness to light, but the light colorfastness of dyed hemp fabric with marigold flowers was moderate to good. A possible explanation for the result maybe the strong absorbance in ultra-violet area produced by lutein pigments [36], which improves the stability of dyed fabric exposed to sunlight. The colorfastness to crocking was also evaluated as shown in Table 10. This involved rubbing the dyed samples. The control sample (without mordant) and experimental samples were rated as good-excellent (4-5) for dry crocking fastness. On the other hand, after applying tannic acid, wet crocking fastness change from fair (3) to good (4), except dyed hemp fabric mordanted with post-mordanting method. Overall, the control samples (without mordant) showed comparatively good washing, and perspiration colorfastness properties, which were further improved with mordanting. Also, there was no significant variation observed in the fastness with the three different mordanting methods. Similar findings were reported in the previous work by [15,36]. As a conclusion, compared with dyeing without mordant, tannic acid as a natural mordant in three different mordanting processes significantly improved color fastness to washing, perspiration, and crocking.

3.8 Durability of color under multiple washing

The total color difference (ΔE) and color strength (K/S) of both hemp fabrics dyed under the optimal condition of this study with and without mordant (tannic acid) under repeated laundering were investigated. The results in Table 11 show that the non-mordant dyed sample suffered color losses and reduced color strength as the number of washings increased. After washing, color fastness property was also improved by using bio-mordant. Specifically, compared to dying without mordant, tannic acid was shown to improve color fastness property after 20 washing cycles with all three mordanting methods. In conclusion, pre-mordanting with tannic acid can improve the dye fixation of hemp fabrics dyed with lutein pigments, resulting in the highest color yield.

Table 7. Color fastness to washing (AATCC Test Method 61-1A:2013).

Color fastness to washing	**Mordanting**			
	None	**Pre**	**Meta**	**Post**
Color change	3-4	4-5	4-5	4-5
Color staining				
- Acetate	4-5	4-5	4-5	4-5
- Cotton	4-5	4-5	4-5	4-5
- Nylon	4-5	4-5	4-5	4-5
- Polyester	4-5	4-5	4-5	4-5
- Acrylic	4-5	4-5	4-5	4-5
- Wool	4-5	4-5	4-5	4-5

Table 8. Color fastness to perspiration (ISO 105-E04:1994).

Color fastness to washing	**Mordanting**			
	None	**Pre**	**Meta**	**Post**
Acid				
Color change	2	4	4	4
Color staining				
- Acetate	3-4	4-5	4	4-5
- Cotton	3-4	4-5	4	4-5
- Nylon	3-4	4-5	4	4-5
- Polyester	3-4	4-5	4	4-5
- Acrylic	3-4	4-5	4	4-5
- Wool	3-4	4-5	4	4-5
Alkaline				
Color change	2-3	4	4	4
Color staining				
- Acetate	4	4-5	4	4-5
- Cotton	4	4-5	4	4-5
- Nylon	4	4-5	4	4-5
- Polyester	4	4-5	4	4-5
- Acrylic	4	4-5	4	4-5
- Wool	4	4-5	4	4-5
- Cotton	3-4	4-5	4	4-5
- Nylon	3-4	4-5	4	4-5

Table 9. Color fastness to light (ISO 105-B02:1994).

Color fastness to light	**Mordanting**			
	None	**Pre**	**Meta**	**Post**
Dyed fabric	5	5	5	5

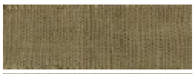
Table 10. Color fastness to crocking (AATCC Test Method 8-2013).

Color fastness to crocking	**Mordanting**			
	None	**Pre**	**Meta**	**Post**
Dry	4-5	4-5	4-5	4-5
Wet	3	4	4	3

Table 11. Total color difference and color strength of dyed hemp fabrics after multiple washes.

Sample	**Washing cycle**	***L****	***a****	***b****	**ΔE**	**K/S**	**Image**
	0	50.95	4.04	40.64	0.00	10.54	
Non- mordant	10	51.55	3.05	38.99	2.02	8.56	
	20	51.94	3.45	38.54	2.39	7.99	

Table 11. (Continued).

Sample	Washing cycle	*L**	*a**	*b**	ΔE	K/S	Image
Pre-mordant	0	50.95	4.04	40.64	0.00	11.65	
	10	51.24	3.85	39.74	0.96	10.94	
	20	51.56	3.93	39.59	1.21	9.95	
Meta-mordant	0	48.34	3.15	37.24	0.00	11.29	
	10	49.32	3.00	37.04	1.01	9.88	
	20	49.74	3.01	36.99	1.42	9.16	
Post-mordant	0	49.02	2.56	38.26	0.00	11.10	
	10	49.87	2.49	37.45	1.16	9.01	
	20	50.33	2.34	37.37	1.60	8.76	

4. Conclusions

This study presents the utilization of natural dye recovery from garland temple wastes. The *Tagetes erecta* colorant was successfully extracted and applied onto hemp fabrics by an eco-friendly and economical method. The natural dyeing processes were optimized for the highest color yield of hemp fabric dyed by RSM. RSM was shown to be effective and reliable in finding the optimal conditions for this dyeing process. Indeed, it was determined and confirmed that the best dyeing quality (K/S = 10.54) was obtained for a dyebath pH of 4.23, a dyeing time of 82.64 min, and a dyeing temperature of 99.98°C with the dye concentration of 10 wt%. The maximum K/S value obtained was greater than 10, indicating a successful color buildup for the yellow shade. This demonstrates efficient utilization of lutein, the primary carotenoid pigment in *marigold* flowers. The use of tannic acid as a bio-mordant enhances color strength and color fastness to washing, perspiration, and crocking of hemp fabrics dyed with *Tagetes erecta* flower extract. It can be inferred that this natural dye can be used to dye hemp fibers with a high color yield and satisfactory fastness properties through 20 wash cycles. Furthermore, because of their good light fastness, we encourage the use of garland waste marigold flowers in the production of yellow-hued curtains, as well as an eco- and health-friendly dyed fabric for use in medical applications.

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