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OPTIMIZATION ON DYEING UPTAKE EXHAUSTION PERCENTAGE OF BETACYANIN PIGMENT EXTRACTED FROM *HYLOCEREUS POLYRHIZUS* PEEL ONTO THE SPUN SILK YARN USING CENTRAL COMPOSITE DESIGN

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ABSTRACT

The prior motivation of this research study is to determine the ideal condition for dyeing process of spun silk yarn with natural dye extracted from dragon fruit peel using a central composite design. The peel of dragon fruit is contained betacyanin pigment can be utilized to be a natural dye which normally the peel of dragon fruit was discarded as a waste. As the betacyanin pigment was considered to be a newest red dye color in textile application, it is necessary to determine the optimal value for its specific dyeing process. In this experiment, natural dye was extracted from dragon fruit peel using water extraction with the acidified water as a medium. The natural dye was then applied onto the spun silk yarn at different dyeing time (60 – 90 min), pH (3 – 5) and dye concentration (100 –120 g/L). The dye-uptake of natural dye was measured using UV-Vis spectrophotometer. The result of dye-uptake percentage was employed using analysis of variance (ANOVA). By ANOVA, the dyeing condition was determined as pH of 2, dyeing time of 95 min and the dye concentration of 110 g/L which is generated from the mathematical model. Using the ideal condition, the dye uptake percentage was determined to be 51.58% to a desirability value of 0.995. So, it is concluded that dyeing condition for the optimal value of dyeing uptake exhaustion of betacyanin pigment onto the spun silk can be determined using CCD.

Keywords: natural dye, exhaustion, central composite design, spun silk dyeing, betacyanin extract

1.0 INTRODUCTION

Nowadays, the demand for natural colorants is growing worldwide because they are more eco-friendly and non toxic compared to synthetic colorants (Guesmi *et al*., 2012). The use of the natural dye/ colorant has numerous advantages including high bio-degradability and compatibility with the environment (Erkan *et al*., 2011). A red-violet color of dragon fruit peel is contributed from the betacyanin pigment (Kunnika and Prancee, 2011). Betacyanin pigment extracted from the various plants has been reported to be used as a natural colorant in a food and cosmetic industry (Britton *et al*., 2002). Yet, there is no such application in the textile industry. So, it is suggested to apply the betacyanin pigment extracted from dragon fruit peel in a usage of dyeing textile application. Dragon fruit (*Hylocereus* spp.) is a one of the cactus fruit from the Cactaceae family which has been commercially cultivated in tropical climate especially in Malaysia. Dragon fruit is rich with the nutrients, minerals and furthermore abundantly antioxidant properties (Zainoldin *et al*., 2009). The peel from the dragon fruit is usually discarded by the food manufacturing industries and end up as a waste. The peel of dragon fruit can be exploited by extracting its vibrant color to be a natural dye as an alternative to replace the synthetic dye, especially in textile application. In modern textile dye houses, the insertion of natural dyes necessitates the classifications of products in terms of standardized quality including the shade of dyeing, the depth of the color and also natural properties (Nasirizadeh *et al*., 2012).

Optimization of dyeing is definitely the substantial method to determine the optimum condition in order to get highest dyeing uptake exhaustion. As the betacyanin pigment extracted from the dragon fruit peel had never been used in the textile application, it is necessary to study the significant variables with the certain conditions that affect the dyeing exhaustion. Response surface methodology (RSM) is one of a perfect method to simulate the variables simultaneously (Karthikeyan *et al*., 2010). It additionally can decide ideal restrictive for multi parameters and the interactives impacts. Thus, it has widely used in process and product improvement because it can optimize the complex process and minimixe the experimental numbers of trials (Sinha *et al*., 2012). The key purpose of this study is to optimize the dyeing conditions between betacyanin pigments extracted from dragon fruit peel onto of the spun silk yarn. The significant factors such as dyeing time, dye-bath concentration and pH of the solution have been identified and proceed further using central composite design (CCD) under RSM. The CCD is perceived the association among the parameters utilizing the generated statistical models (Demirel and Kayan, 2012). In the last part of the CCD, the optimal conditions were determined to achieve the maximum dyeing uptake percentage during the dyeing process.

2.0 EXPERIMENTAL METHODS

Spun Silk Preparation

The spun silk yarn at about 100g was soaked in boiling water of 2 L for about 1 hr, in the purposed of eliminating the dirt and dust. After that, the spun silk was removed and rinsed with the cold water. The excess water was squeezed and lastly, dried in the air (Chairatanaphani, 2008).The dried spun silk yarn was then weighted at about 1 g to use in the experiment.

Betacyanin Extraction

The raw material, dragon fruit was purchased from the regional cultivator near to Gambang, Pahang and kept at 4 °C before proceed for the extraction process. The peel was separated manually from the fruit and washed thoroughly. The peel was then cut into a small pieces at about 1 mm and macerated by using a blender (Guesmi *et al*., 2013). The macerated juice was subjected to the dye extraction. The extraction was carried out by applying the solid to liquid ratio (SLR) of 1:5 equivalents to 10 g of macerated juice into 50 mL of water in the water-bath shaker for 30 minutes in the temperature of 45 °C.

The dye solution was detached from the plant tissue with the help of a Buchner funnel lined with the filter paper, linked to the vacuum pump (Tang & Norziah, 2007). To make sure the dye solution is clear from the plant tissue, the filtered solution was centrifuged for 15 minutes at the speed of 9000 rpm. The extracted solution was kept in the dark brown storage box for the next experiments.

Instruments

The absorbance measurements were recorded using UV-Visible spectrophotometer by using quartz cells of path length 1 cm. The dye uptake percentage was measured using the absorbance of dye solution before and after dyeing in three measurements. The adsorbance was measured at maximum wavelength which is 538 nm.

The percentage of dye uptake exhaustion was calculated (Nasirizadeh *et al.,* 2012) by using the relation given in Equation 1:

$$E\% = \left({Abs\ (before) - Abs\ (after)} \middle/ {Abs\ (before)} \right) \times 100 \tag{1}$$

For pH measurement of the betacyanin solution, a pH meter (Mettler Delta 320) was used.

Experimental Design

The RSM method was used to determine the optimum conditions for a multi-variable system and to anticipate the collective influence of few variables. The CCD under RSM was utilized to improve the screened medium components that influencing the dye exhaustion. The CCD was used to examine the importance effects of pH, dyeing time and dye-bath concentration. The software Design Expert was used to the model the experimental data obtained from a laboratory-scale setup. The optimization process using CCD was involved 17 runs with three-factor of five levels. Three of the screened factors from the factorial analysis process were used as the independent factors. The three factors were further optimized namely dyeing time, dye-bath concentration and pH. In this design, the temperature and salt concentration were maintained at 45^{o}C and 0.3 g/L, respectively, due to its lower significance for dyeing uptake exhaustion. the range for the significant factor are stated as following; the dyeing time represented as 'A' ranged from 80 min to 100 min, followed by dye-bath concentration represented as 'B', which ranged from 80 g/L to 120 g/L and the pH represented as 'C' ranged between 1 to 5. The ranges of independent variables are summarized in Table 1. An analysis of variance (ANOVA) and R^2 (coefficient of determination) statistical methods were implemented to confirm the adequacy of the developed model (Subroto *et al*., 2015).

Table 1: Independent variables and concentration levels for response surface study

Factors	Unit	Levels				
		-2	-1	0	1	2
A (x_1) dyeing time	Minute	80	85	90	95	100
B (x_2) dye concentration	g/L	80	90	100	110	120
C (x_3) pH	pH	1	2	3	4	5

Verification of Optimized Conditions and Predictive Models

The verification step is comparing the value from the experiment with the predicted values under the optimal conditions (Prakash et al., 2012). The proposed dyeing conditions were 95 minutes for dyeing time, the dye concentration of 110 g/L and the pH of 2. The residual and percentage error from both actual and predicted values were calculated using Equation 2 and Equation 3 respectively, as given below:

$$\textit{Residual} = (\textit{actual value} - \textit{predicted value}) \quad (2)$$

$$\textit{Error \%} = (\textit{Residual} / \textit{actual value}) \times 100 \quad (3)$$

3.0 RESULTS AND DISCUSSIONS

Fitting of Second Order Polynomial Equations and Statistical Analysis

The quadratic model was found statistically substantial to signify the dye uptake exhaustion percentage response, as shown in the Fit summary of output analysis. The adequacy of the quadratic model was examined by F-test, "Prob > F" values and the determination coefficient R^2. The generated F and Prob > F are recorded in Table 2. The model was highly significant because the F value is 422.37 and the Prob > F is indicating less than 0.0001 as observed in Table 2. The values of "probability more than F" for the terms of the model under 0.05 (95% confidence level) determine that the generated model is recognized to be statistically significant at 95% confidence level which is required. It reveals that the terms of the model have significance over the response (Ojha and Achwal, 2014). Similarly, the correlation coefficient R^2 was calculated to be 0.9987. Also, an acceptable agreement with the adjusted determination coefficient is essential. The *Adj-*R^2 value of 0.9963 was found in this research study. The values of R^2 and *Adj-*R^2 are close to 1.0 signifying a high correlation between the experimental and predicted values. Furthermore, the value of 'Lack of fit' turned out insignificant (Prob > F = 0.7720) indicating an acceptably fit model. Moreover, as shown in Table 2, the primary impact towards the rate of dyeing uptake are dyeing time (A) and pH (C) were emerged to be the most substantial factors. Followed by the second order effect of dyeing concentration (B^2), pH (C^2) and dyeing time (A^2) and the two-level interactions between dyeing time and pH (AC). Furthermore, the primary influence of dyeing concentration (B), the two-level interaction between dyeing time and dye concentration (AB) and two-level interaction between dyeing time and pH (BC) were found to be corresponding to the secondary effect on the dyeing uptake percentage.

With the assistance of the responses for the relating coded estimation of the three distinctive process factors, the polynomial regression modelling was executed, and the outcomes were evaluated. The subsequent equation was employed to obtain the predicted response (Y) for the percent of dye uptake exhaustion:
Final empirical model in terms of coded factors as given in Equation 2:

$$Dye\ uptake\ (\%) = 47.60*A - 12.10*B + 4.24*C - 4.17*A^2 - 3.29*B^2 - 2.23*A*B - 1.28*A*C + 0.034*B*C \quad (4)$$

Final empirical model in terms of actual factors

$$Dye\ uptake\ (\%) = -1725.54 + 30.48*dyebath\ concentration + 112.18*pH + 15.98*dyeing\ time - 0.17*(dyebath\ concentration)^2 - 13.14*(pH)^2 - 0.092*(dyeing\ time)^2 - 0.89*(dyebath\ concentration*dyeing\ time) + 0.013*(pH*dyeing\ time) \quad (5)$$

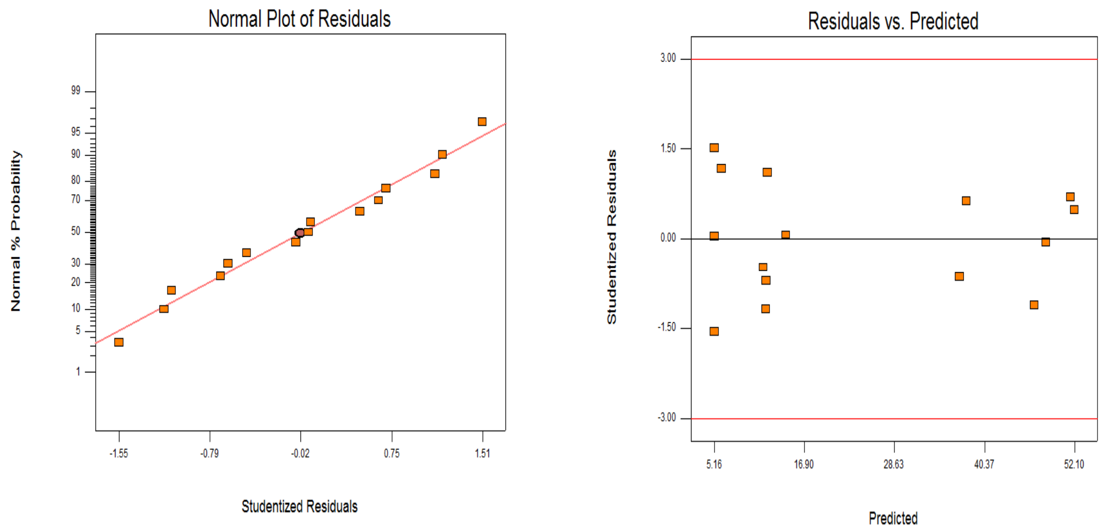
The empirical model equation is the mathematical correlation model that could be used in the optimization and prediction of the dyeing uptake percentage within the range of variable factors in this experiment.

Table 2: ANOVA analysis and statistical parameters of the model

Source	Sum of squares	DF	Mean squares	F-value	Prob > F	
Model	5103.21	9	567.01	422.37	<0.0001	Significant
A	33.64	1	33.64	25.06	0.0041	
B	0.78	1	0.78	0.58	0.4794	
C	2772.89	1	2772.89	2065.49	<0.0001	
A^2	17.33	1	17.33	12.91	0.0157	
B^2	1259.71	1	1259.71	938.34	<0.0001	
C^2	1053.94	1	1053.94	785.06	<0.0001	
AB	2.10	1	2.10	1.57	0.2663	
AC	3.62	1	3.62	2.70	0.1616	
BC	1.81	1	1.81	1.34	0.2986	
Residual	6.71	5	1.34			
Lack of Fit	2.51	3	0.84	0.40	0.7720	Not significant

Adequacy of the Models

Figure 1 (a) shows a normal probability plot of the residuals, which demonstrates the residual distribution was closely to a straight line. The straight line implies the even distribution of errors and the adequacy of the least square fit (Zularism *et al.*, 2009). The data points also indicate that the normality is normal and the responses change is unnecessary. Figure 1 (b) is a plot of residuals against the predicted response of dyeing uptake rate which demonstrates that the proposed models is certainly satisfactory. As studentized residuals are equally tabulated within the red line along the x-axis, it means that the data is free from any desecration of the independence or constancy variance assumption. Thus, the model is sufficiently to be utilized as a part of upgrading the dyeing condition for betacyanin pigment onto the spun silk.



(a) (b)

Figure 1: (a) The studentized residuals versus normal probability plot of betacyanin extracted dyeing of spun silk, (b) Residuals versus predicted plot of betacyanin extract dyeing of spun

Interaction of Process Variables

The effect of pH, dye-bath concentration and dyeing time process variables on dyeing uptake percentage was further analyzed with the assistance of three-dimensional response surface plots in the views of the quadratic model. The effect of dyeing time and dye-bath concentration, illustrated in Figure 2, demonstrated that the dye uptake percentage decreased when dye-bath concentration changed from 75 g/L to 85 g/L and as dyeing time increased from 55 min to 65 min. The findings of this study show that the maximum dye uptake percentage of 57.63% can be obtained when dyeing time and initial dye bath concentration was 65 min and 75 g/L respectively. From this result, it is suggested that the percentage of dye uptake is likely depending on the initial of dye-bath concentration. As reported by Saleh *et al*., (2013), the effect of the initial dye-bath concentration relies on two factors which are the direct interaction between the free particle of color in the dye-bath solution and also the availability of active-sites on the yarn surface (Saleh *et al.,* 2013). At lower dye-bath concentration, the ratio between the dye particle and fiber sites is higher which resulted the higher dye uptake as all the dye particles are attached to the active site on the yarn surface. Meanwhile, the higher concentrations resulted lower dye-uptake percentage may be due to the saturation of adsorption sites and the abundance of free molecule dye that unoccupied (Jaikumar, 2007). Similar trends were also reported by Liu *et al*., (2013) who studied the adsorption properties of natural dye lac dye onto the chitosan fiber (Liu *et al*., 2013). The surface plot of the interaction between pH and dyeing time at fixed initial dye-bath concentration (80 g/L) is illustrated in Figure 3.

As the time increasing, the dye uptake percentage also increased from 49.89% to 58.30%

at the same value of pH 2. However, the dye uptake percentage was decreased drastically at about 24.2% from 58.30% to 34.17% when the pH value was changed from pH 2 to pH 3. It is clearly shown that pH significantly gives effects to the dye uptake percentage. It is suggested that the changes of dye uptake percentages may due to the relationship between dye structures; the type of fiber used and dye stability. Guesmi *et al*. (2012) reported that the betacyanin's charge in the aqueous solution depends on the pH value. The betacyanin molecule may be present in cationized or monoanionic form under the intense acidic environment (Guesmi *et al*., 2012). Since the betacyanin dye encloses carboxyl group, it may interact ionically with the protonated terminal amino groups of spun silk fibers at acidic pH via ionic interaction. The anion of the dye has a complex character, and when it interacts with the fiber, the ionic force takes place. The dyeability of the dye molecule to the yarn surface was depends to this ionic attraction (Guesmi *et al*., 2013).

The 3D surface plot of the interaction between pH and dye-bath concentration, at a constant time of dyeing is presented in Figure 4. When the concentration of dye-bath is 75 g/L and the pH of 2, the dye-uptake percentage pointed the highest percentage of 57.63%. The dye uptake percentage decreased from 57.63% to 46.84% with the increase of initial dye-bath concentration from 75 g/L to 85 g/L at the pH of 2. This result indicates that the dyeing mechanism between betacyanin pigment with the spun silk is more likely to be affected by the changes of dye molecule structure in different pH conditions. It can be concluded that the dyeing process is influencing by the interaction between the dye molecule and the active-site of yarn surface. So, it is indicated that the dye attached to the yarn surface is preferably of chemical adsorption or chemisorption (Miyake *et al*., 2013) Chemisorption is a stronger perturbation of molecular structure with the formation of chemical bonds with the substrates. The connection between dye molecule and the active site of yarn is either covalent, ionic or metallic bonding (Yagub *et al*., 2014).

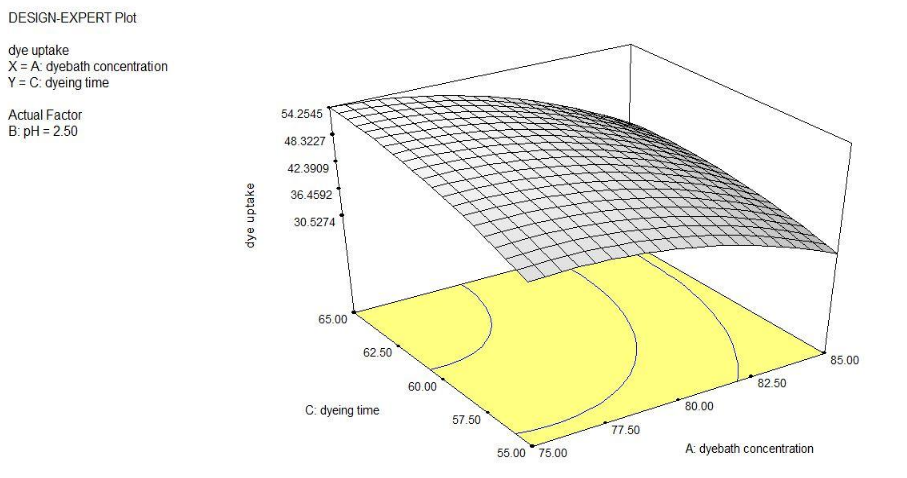


Figure 2: The effect of dyeing time and dye-bath concentration on the dye uptake percentage at a fixed pH of 2.5

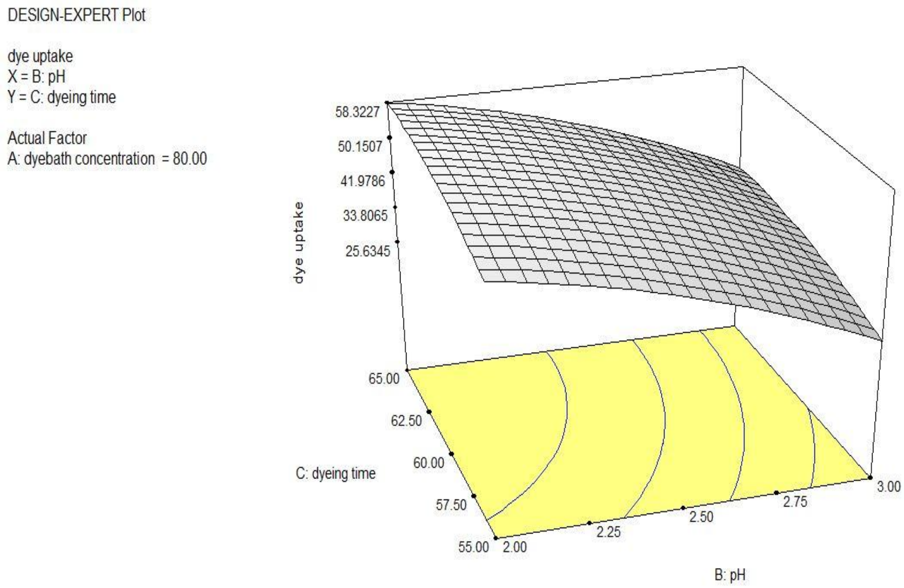


Figure 3: The effect of dyeing time and pH on the dye uptake percentage at a fixed dye-bath concentration of 80 g/L

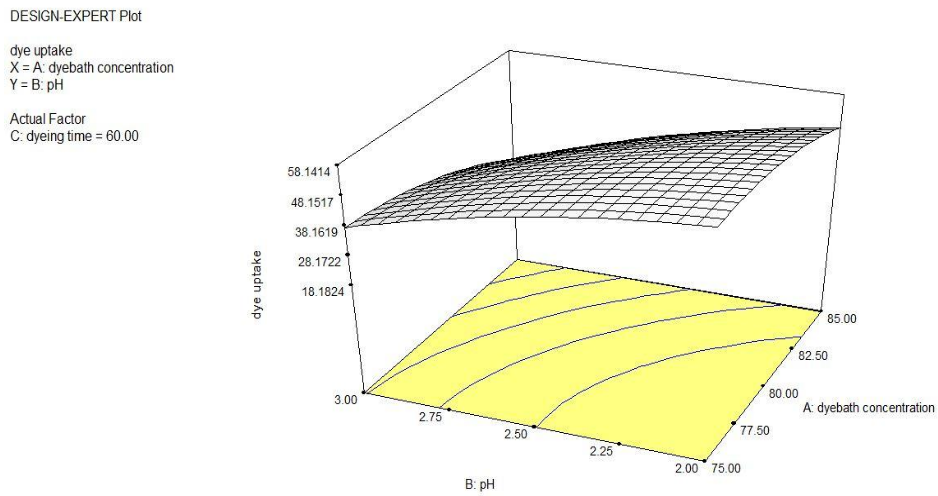


Figure 4: The effect of pH and dye-bath concentration on the dye uptake percentage at a fixed dyeing time of 60 min

Verification of Optimized Conditions and Predictive Model

The experimental of verification or validation is crucial to be carried out to affirm the validity of the generated empirical model (Karthikeyan *et al*., 2010). Consequently, the analysis for approval was done to compare the estimated values of responses with the experimental data. The acquired actual values and their related predicted values from the selected experiments were compared. The residuals and percentage errors between the actual and predicted values of both responses over a selected range of operating levels are listed in Table 3. From Table 3, the result showed that the calculated percentage error is between 1.7 % to 4.35 %. It could be deduced that the developed empirical model is substantially precise for a response, as the percentage error between the actual and predicted values was within the limit of 5% (Zularism *et al.*, 2009). It implies that the model is satisfactory at 95% of prediction interval, thus, it is adequate to approach the operational process for optimal dyeing uptake according to this model.

Table 3: Results of operating conditions with experimental design in confirmation runs

Run	**Factor**			**Dye uptake percentage**
	X_1 (minutes)	X_2 (g/L)	X_3 (pH)	Error %
1	110	95	2	3.39
2	90	95	2	1.7
3	120	90	3	1.8
4	110	85	2	2.245
5	90	85	2	4.35

In the view of prior examination, the significant factors which are dyeing time, dye concentration and pH were dominant for the response of dyeing uptake percentage. The prediction of response at 95.0% prediction interval is possible by employing the optimization mode capability of the software. The optimization procedure has been executed for the dyeing condition of betacyanin extracted onto the spun silk yarn and prediction was set within the defined level. The results from the model were determined to be as: dyeing time= 95 min), dye concentration= 110 g/L and pH= 2. The maximum value of dyeing uptake percentage was 51.58% under the suggested condition that generated from the model. In order to validate the predicted optimized conditions, the experimental confirmation run was executed by using the proposed model conditions. The result of the dyeing uptake percentage calculated from the experimental run was 52.45%. The error percentage between the predicted and actual value is within 5% as it can be confirmed that the projected mathematical models are practically consistent and precise.

4.0 CONCLUSIONS

As the conclusion, the optimum conditions for the dyeing uptake of betacyanin pigment extracted onto the spun silk yarn can be reliably employed by using the central composite design under response surface methodology. Analysis of variance showed a high coefficient of determination value (R^2) of 0.9986 for dyeing uptake exhaustion percentage. It indicated that the experimental data is adequately fit with the second-order polynomial regression model. The optimum conditions for dyeing process are as follows: dyeing time = 95 minutes, dye-bath concentration = 110 g/L and pH 2. By using the optimum conditions, the dyeing uptake percentage was found to be 52.45%. The percentage errors for experimental validation showed a minimal difference which is 1.68%. Hence, the generated model using CCD in this study is considered to be a valid optimization.

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