



Research Article

Central composite design for the optimisation of silk yarn dyeing with natural extract from *Melastoma malabathricum* L. fruit

Nazifah Mohd Adham¹, Nurul Nadhirah Mohd Shukri¹, Wan Khartini Wan Abdul Khodir^{1,2}, Ahmad Farid Abdul Jalal³, Shafida Abd Hamid^{1,2*}

¹Department of Chemistry, Kuliyah of Science, International Islamic University Malaysia, Bandar Indera Mahkota, 25200 Kuantan, Pahang, Malaysia

²SYNTOF, Kuliyah of Science, International Islamic University Malaysia, Bandar Indera Mahkota, 25200 Kuantan, Pahang, Malaysia

³Lembaga Muzium Negeri Pahang, Jalan Sultan Ahmad, 26600 Pekan, Pahang, Malaysia

*Corresponding author: shafida@iium.edu.my

Received: 13 November 2024; Revised: 13 February 2025; Accepted: 15 March 2025; Published: 21 April 2025

Abstract

Natural dyes have gained interest in sustainable textile applications. However, the potential of *Melastoma malabathricum* as a silk dye source remains unexplored. Pigments extracted from *M. malabathricum* L. fruit using acidified methanol were used for silk yarn dyeing through the meta-mordanting process with stannous chloride (2%) as the mordant. A four-factor, face-centred composite design from response surface methodology was applied to optimise the dyeing process. The effect of extract weight, temperature, pH, and dyeing duration on colour intensity was analysed. The optimum conditions ($R^2 = 0.9517$) were found using 1 g of dye extract, 30 °C, pH 3, and 120 min, yielding a colour intensity of 28.99. The ultraviolet-visible spectra indicated the highest peak absorbance of the dye bath at pH 3, aligned with the highest colour intensity. Higher temperatures and amount of dye extract increased the colour intensity, while lower temperatures and longer durations had the opposite effect. This study contributes to sustainable silk dyeing by utilising *M. malabathricum* fruit as a natural dye source and provides a foundation for its systematic optimisation. The findings also highlight its potential for large-scale applications, offering an eco-friendly alternative to synthetic dyes in the textile industry.

Keywords: *M. malabathricum*, natural dye, RSM, colour intensity, silk yarn

Introduction

Most of the colours or dyes used in the textile industry are synthetic, with over 20% of these dyes ending up in the environment and water sources [1]. The textile industry widely uses synthetic dyes for their colour fastness and vibrancy. However, the toxicity and non-biodegradability of synthetic dyes pose risks to people, animals, and the environment, leading to issues such as skin allergies, respiratory problems, hormone imbalances, and cancer [2]. Furthermore, the pollution of water bodies resulting from textile industry waste can disrupt the ecosystem and harm living organisms. As a result, natural dyes derived from plants, animals, or minerals are experiencing a resurgence, as they offer a safer alternative to their synthetic counterparts. Recent advancements in plant-based dyeing techniques, including the use of natural additives and bio-mordants, have significantly

improved dye uptake and fabric performance [3]. Additionally, new extraction protocols for anthocyanins have been developed to enhance pigment stability and dyeing efficiency [4]. Moreover, the application of natural dyes in a variety of applications not only promotes environmental sustainability but also provides a more ethically responsible approach to create different colour schemes [5].

Natural dyeing techniques, such as eco-friendly solvent extraction and optimised mordanting, have been shown to improve dye uptake and colour fastness on various fabrics [6,7]. In another study, the integration of inorganic ultraviolet (UV) absorbers and reducing agents has been shown to inhibit photofading in anthocyanin-based dyes, enhancing their longevity on silk yarns [8]. Significant

advancements in sustainable dyeing techniques in recent years also highlight innovative methods, such as ultrasonic and microwave-assisted dyeing, which improve dye uptake and reduce environmental impact [6]. Although *Melastoma malabathricum* fruit extract has been reported for fabric dyeing [9], studies specifically investigating its application as a textile dye remain limited. Furthermore, there is currently no systematic study on its optimisation for textile applications.

Melastoma malabathricum Linn. (also known as senduduk) is a shrub tree, with pink or mauve-coloured flowers. It is typically found along roadsides in tropical and subtropical climates and is often regarded as a weed. This plant produces small, berry-like fruits with numerous dark purple seeds coated with pink-purple pulp when ripe (**Figure 1**). The flowers and fruits of *M. malabathricum* plant contain anthocyanins (cyanidin dihexoside, cyanidin hexoside, and delphinidin hexoside), which provide natural colours for various applications, including food and cosmetics [10,11]. The flower extract of the plant has been used as a source of natural dyes for fabric dyeing [12], while its fruit extract has also been reported as a natural colouring agent in cosmetic products, such as lip cream, giving a purplish-red colour [13].

Anthocyanins are water-soluble flavonoid pigments found in the fruits, leaves, and flowers of various plants, such as purple sweet potatoes [14], mangosteen skin [15], and hibiscus flowers [16]. Natural dyes are sensitive to several factors, such as pH, temperature, and light, which complicate the achievement of consistent and optimal dyeing outcomes. Depending on the pH of the surrounding environment, anthocyanins contribute to a wide range of colours, including red, purple, and blue. Anthocyanins are highly soluble in water at low pH, producing red

pigment, predominantly due to flavylum cations, while high pH values lead to blue colour shades [17,18]. Traditional methods often fail to capture the relationships among various elements influencing the dyeing process. Therefore, response surface methodology (RSM), which allows for the simultaneous interaction of multiple process variables to determine their relationships, can be utilised to optimise the dyeing process [19]. In this study, central composite design (CCD) of RSM was utilised to evaluate the effect of four operating variables: the amount of *M. malabathricum* fruit extract, duration, temperature, and pH, on the colour intensity of the dyed silk yarn. By identifying the key parameters that influence dye uptake and colour intensity, this study may provide a foundation for the potential large-scale application of this natural dye in sustainable textile manufacturing.

Materials and Methods

Stannous chloride (SnCl_2), aluminium potassium sulphate dodecahydrate (alum), methanol, acetic acid (96%), and sodium hydroxide were purchased from Merck and used without any purification. The 120/2 100% silk yarn was purchased from TCB Batik & Songket Sdn. Bhd., Malaysia. *M. malabathricum* L. fruits were collected from the surrounding areas of the International Islamic University Malaysia, Kuantan, Malaysia, and identified by a botanist. The voucher specimens (IUM486) were deposited in the Herbarium, Kulliyah of Science, International Islamic University Malaysia. The fruits were peeled, dried in an oven at 40 °C for 12–24 h, and stored in a zipper bag until further use. All experiments were conducted under controlled laboratory conditions to enhance reproducibility as much as possible. However, due to the natural variability of plant-based extracts, slight variations in dye composition and performance may still occur.



Figure 1. Ripe fruits of *M. malabathricum* Linn

Extraction

The dye pigment from the fruits was extracted using two solvents: distilled water and methanol. The extraction of dye pigment using distilled water was carried out by heating the preweighed fruits in 50 mL of distilled water at 60 °C for 2 h. The mixture was then filtered, and the filtrate was used as the dye bath solution. A preweighed (0.50 g) crushed *M. malabathricum* fruit was extracted in 50 mL of acidified MeOH (methanol with 0.05% acetic acid) for 2 h at 60 °C while stirring, with the beaker covered with aluminium foil. The resulting mixture was filtered, and the solvent was evaporated. The crude extract obtained (26.5%–55.7%) was collected and kept in a dark, dry, and cool place.

Scouring

The silk yarn (1 g) was soaked completely in distilled water for 1 h. Subsequently, the yarn was washed with a soapnut solution for 30 min at 50–60 °C, maintaining a material-to-liquor ratio of 1:40. The scoured sample was then thoroughly washed with distilled water and kept wet until use.

Mordanting and dyeing

The dye bath was prepared by diluting the dye extract (1.0, 1.5, and 2.0 g) in 40 mL of distilled water along with the mordant. The silk yarn was treated with 2% SnCl₂ through a meta-mordanting process, in which the scoured silk yarn was simultaneously immersed in the dye bath solution and the mordant. The material-to-liquor ratio was maintained at 1:40 in all methods. The yarn was kept in the dye bath for specific durations (30, 75, and 120 min) at the determined temperatures (30, 60, and 80 °C). The pH was maintained at the required value (3, 6, and 9) by adding a buffer solution (acetic acid or sodium hydroxide). The dyed yarn was then washed with distilled water and dried at room temperature.

Ultraviolet-visible spectroscopic analysis

The UV-vis absorption spectra of dye solutions were measured using the Shimadzu UV-1900i spectrophotometer. Dye bath solutions (containing 0.50 g of crude extracts diluted in 40 mL of distilled water) at different pH (3, 6, and 9) and distilled water (as reference) were prepared. For each dye bath solution, 1 mL was measured and diluted in 9 mL of distilled water before being transferred into cuvettes. The wavelength range was set to 400–700 nm, and the baseline was adjusted to zero absorbance. The absorbance of the dye solutions was recorded and the spectrum was obtained for analysis.

Colour intensity measurement

The colour intensity of the dyed samples was analysed using a CHN Spec CS-10 colourimeter to measure the L^* , a^* , and b^* values (CIELAB) under standard

illumination conditions of D65 and 10 observers. The colourimeter was calibrated by measuring the black and white colour. Subsequently, the CIELAB colour space was selected prior to measuring the dyed yarn samples. L^* is a measure of lightness or darkness of the fabric, ranging from 100 (white) to 0 (black). a^* measures the redness (positive (+) values) or greenness (negative (-) values), whereas b^* measures the yellowness (positive (+) values) or blueness (negative (-) values) [20]. Measurements were conducted in triplicate, and the colour intensity (C_{ab}) for each dyed yarn was calculated using the following equation:

$$C_{ab} = \sqrt{a^2 + b^2}$$

Response surface methodology

The statistical software package Design-Expert 13 (v13.0.5, Stat Ease Inc., Minneapolis, MN, USA) was used for the design and analysis of experimental parameters. Central composite design was used to investigate the effect of pH, temperature, time, and amount of sample extraction on the colour intensity (C_{ab}) of the dyed silk yarn. A four-factor, face-centred composite design resulting in 28 runs was employed to optimise the dyeing parameters. The response of colour intensity for all runs was measured, and the main effects and interactions among the factors were determined. The adequacy of the model was assessed in terms of the values of coefficient of determination (R^2) and adjusted R^2 . Analysis of variance (ANOVA) and graphical optimisations were performed to determine the optimal conditions of the process variables.

Results and Discussion

Extraction and mordant selection for the dyeing process

Extraction of anthocyanins usually requires an acidified solvent because the flavylium ion, a key component of anthocyanins, is stable in an acidic condition [21]. The extraction process was carried out at 60 °C to ensure the solubilisation of the pigment while preventing the thermal degradation of anthocyanins, which are known to be heat-sensitive. Previous studies indicate that anthocyanins degrade at temperatures exceeding 70 °C, while extraction within the 50–70 °C range preserves pigment stability [22]. Similar findings have been reported for anthocyanin extractions from *Hibiscus sabdariffa* and *Garcinia mangostana* fruit hulls, where temperatures around 60 °C yielded optimal pigment recovery while preventing degradation [15,23]. A recent study has also optimised natural anthocyanin extraction protocols to improve their dyeing potential, resulting in higher washing fastness and longer-lasting colour [4]. Apart from the solvents, optimisation of solvent-to-solid

ratio, extraction time, and temperature is also needed to achieve the highest product yield. Generally, highly polar solvents (e.g., water) are effective solvents to extract polar anthocyanins as the hydroxyl groups of anthocyanins can form hydrogen bonds with the water molecules. However, water extraction may introduce additional compounds, such as polyphenols, sugars, carbohydrates, and also other pigments (e.g., chlorophylls and carotenoids). Several studies have reported that the extraction of anthocyanins from parts of plants using alcohol acidified with citric acid produced a good yield and better colour than using hydrochloric acid [10,23–24]. In addition, citric acid is less corrosive than hydrochloric acid. A less polar solvent, such as methanol, can be applied to minimise the co-extraction of interfering compounds [25].

There are relatively few studies published on the dye extraction from the *M. malabathricum* plant. An earlier study by Wan Ahmad et al. [12] described the methanolic extraction of nano-sized powder form of *M. malabathricum* flower for silk fabric dyeing using several mordants. Their results indicated that different solvents used for extraction and the type of mordant used for dyeing produced different colour shades on

the fabric. The same group extended their investigation using the natural dye extracted from *Melastoma malabathricum* L. fruit on polyester fabric [26]. Among the anthocyanidins, delphinidin is the most soluble in methanol (58.61 ± 0.01 – 168.64 ± 0.02), followed by water (53.53 ± 0.06 – 163.71 ± 0.02). *M. malabathricum* fruit, which turns dark purple when ripe, has been reported to contain primarily cyanidin and delphinidin [10]. In our study, the dye pigment from *M. malabathricum* fruit was preliminarily extracted using distilled water and methanol with 0.05% acetic acid (acidified MeOH). The silk yarn dyed with the dye bath prepared using the acidified MeOH extract exhibited a darker shade compared to the yarn dyed using the distilled water dye bath (**Figure 2**). Husain et al. [27] conducted high-performance liquid chromatography analysis to quantify anthocyanin solubility in different solvents and revealed that methanol enhanced delphinidin extraction while minimising interference from non-pigment compounds. This finding aligns with our observation that acidified methanol produced a more intense dye shade than water extraction. Therefore, for this study, the extraction was performed using methanol with 0.05% acetic acid as the solvent, at 60 °C for 120 min.

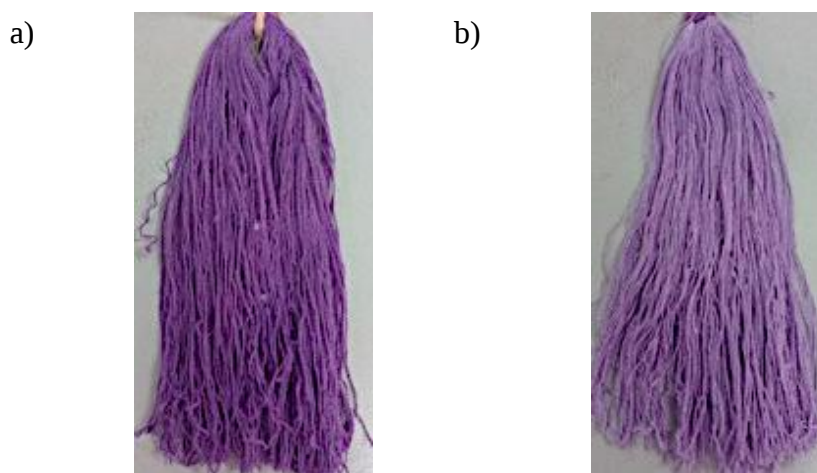


Figure 2. The silk yarns dyed using different solvent extraction methods: a) acidified methanol and b) distilled water

The use of mordants is essential in fabric dyeing, as they can enhance colour stability, depth of dyeing, and the fastness properties of the fabrics. In the current study, the dyeing process was initially conducted using SnCl_2 as a mordant, employing different mordanting methods (pre-, meta-, and para-mordanting). The results showed that the meta-mordanting method produced the highest colour intensity value (28.57), followed by pre-mordanting (23.28). This observation is similar to Vankar &

Shukla [24], who also determined that the pre-mordanting method using SnCl_2 resulted in the highest colour intensity in the silk yarn dyeing using anthocyanins extracted from *Hibiscus rosa-sinensis* flowers compared to alum. Hence, the SnCl_2 (2%) meta-mordanting method was chosen in the current study. The challenge encountered in the meta-mordanting process was to ensure uniform dye uptake. Furthermore, the choice of pH was critical in preventing excessive hydrolysis of the mordant, which

could affect its coordination with dye molecules. Studies on other anthocyanin-rich plant dyes, such as *Diospyros kaki* (persimmon) and *Clitoria ternatea* (butterfly pea), have shown that metal mordants improve colour fixation by forming coordination bonds with fabric protein [21,28]. The stannous ions (Sn^{2+}) form the mordant solution complex with the silk fibre by interacting with the functional groups present in the silk protein, such as amino groups ($-\text{NH}_2$) and carboxyl groups ($-\text{COOH}$). These interactions create coordination bonds between stannous ions and silk fibres as a preparation for receiving the dye.

Response surface methodology analysis

Optimisation of the dyeing process was conducted using RSM with the following input variables: amount of dye extract (1.0, 1.5, and 2.0 g), pH (3, 6, and 9), dye bath temperature (30, 60, and 80 °C), and duration of silk yarn dyeing (30, 75, and 120 min). The output for the analysis is the C_{ab} of the dyed silk yarn. A four-factor, face-centred composite design with 28 runs was employed to optimise the dyeing parameters. The colour intensity for all runs was measured and tabulated in **Table 1**, while the colour intensity for the experimental runs generated is presented in **Table 2**.

Table 1. The 28 runs computed based on RSM with colour intensity as the response variable

Run	Mass (g)	pH	Dye Duration (min)	Temperature (°C)	Colour Intensity (C_{ab})
1	1	3	120	80	26.46
2	1	3	30	80	23.09
3	1	9	120	30	6.23
4	1.5	3	75	55	19.5
5	2	9	120	80	14.47
6	2	3	120	30	25.04
7	1	9	30	80	10.99
8	1.5	6	75	30	15.59
9	1	3	120	30	28.99
10	1.5	6	75	55	18.46
11	1.5	6	120	55	18.27
12	1.5	6	30	55	18.1
13	1	9	30	30	5.77
14	2	3	30	80	23.82
15	2	6	75	55	17.7
16	2	3	120	80	25.62
17	1.5	6	75	55	17.06
18	1.5	9	75	55	5.66
19	1.5	6	75	55	18.15
20	2	9	30	30	8.44
21	2	3	30	30	24.20
22	1.5	6	75	80	18.14
23	2	9	30	80	13.65
24	2	9	120	30	15.93
25	1	6	75	55	12.85
26	1.5	6	75	55	16.93
27	1	9	120	80	13.05
28	1	3	30	30	23.76

Table 2. The CIELAB values, colour intensity, and dyed silk yarn for the 28 runs

Run	CIELAB			Colour Intensity	Dyed Silk Yarn
	L^*	a^*	b^*		
1	32.61	20.89	-16.23	26.46	
2	41.33	20.47	-10.67	23.09	
3	62.01	6.06	-1.08	6.23	
4	47.48	18.82	-5.08	19.50	
5	56.81	10.43	10.03	14.47	
6	43.52	21.08	-13.51	25.04	
7	64.45	8.06	7.46	10.99	
8	52.33	12.94	-8.69	15.59	
9	38.18	22.27	-18.55	28.99	
10	42.42	16.51	-8.23	18.46	
11	44.02	14.72	-10.82	18.27	
12	52.65	17.94	-2.37	18.10	
13	61.40	5.57	-1.51	5.77	
14	34.17	21.82	-9.55	23.82	
15	43.97	17.44	-2.70	17.70	
16	37.51	22.06	-13.03	25.62	
17	43.52	15.45	-7.21	17.06	
18	59.77	5.22	2.18	5.66	
19	42.71	16.53	-7.31	18.15	
20	64.31	6.50	5.33	8.44	

Run	CIELAB			Colour Intensity	Dyed Silk Yarn
	L^*	a^*	b^*		
21	40.19	20.32	-13.13	24.20	
22	39.62	14.81	-10.48	18.14	
23	59.23	10.34	8.92	13.65	
24	64.06	5.63	14.9	15.93	
25	57.13	11.94	-4.58	12.85	
26	42.89	15.80	-6.05	16.93	
27	56.71	10.06	8.32	13.05	
28	44.55	19.91	-12.96	23.76	

Table 3. Analysis of variance for the response surface quadratic model

Source	Sum of Squares	df	Mean Square	F-value	P-value	
Model	1,044.80	14	74.63	18.30	<0.0001	significant
A-Mass sample extraction	17.37	1	17.37	4.26	0.0596	
B-pH	886.06	1	886.06	217.26	<0.0001	
C-Time	27.48	1	27.48	6.74	0.0222	
D-Temperature	13.07	1	13.07	3.21	0.0967	
AB	25.18	1	25.18	6.17	0.0274	
AC	0.0018	1	0.0018	0.0004	0.9835	
AD	1.49	1	1.49	0.3664	0.5554	
BC	0.0105	1	0.0105	0.0026	0.9603	
BD	22.07	1	22.07	5.41	0.0368	
CD	2.23	1	2.23	0.5462	0.4730	
A ²	0.0006	1	0.0006	0.0002	0.9902	
B ²	18.95	1	18.95	4.65	0.0504	
C ²	21.61	1	21.61	5.30	0.0385	
D ²	6.39	1	6.39	1.57	0.2326	
Residual	53.02	13	4.08			
Lack of Fit	51.25	10	5.12	8.67	0.0509	not significant
Pure Error	1.77	3	0.5909			
Cor Total	1,097.82	27				
Model Statistic						
Std Dev.	0.074			R-Squared	0.9982	
Mean	5.82			Adj R-Squared	0.9965	
C.V %	1.28			Pred R-Squared	0.9897	
PRESS	0.47			Adeq Precision	99.158	

Analysis of variance was used to investigate the interaction among the variables and to assess the significance of the analysis (Table 3). A p -value of less than 0.05 indicates the significance of each variable. From the results, variables A (amount of extract), B (pH), and C (time) were found to be significant. The p -value of B was very small ($p < 0.0001$), suggesting that pH highly influenced the changes in colour intensity. Variable A was slightly higher than 0.05, while D showed the most insignificant value at 0.0967 ($p > 0.05$). The results indicate that the dye bath temperature does not significantly affect the colour intensity of the dyed silk yarn. For the cross-product coefficients, AB and BD are significant with p -values of 0.0222 and 0.0368, respectively, while the quadratic coefficients of B^2 and C^2 are significant with p -values of 0.0504 and 0.0385, respectively. Thus, A, B, C, AB, BD, B^2 , and C^2 are the most important factors that can affect the C_{ab} values of the dyed silk yarn. The predicted R^2 of 0.9897 and the adjusted R^2 of 0.9965 are in reasonable agreement with each other. The predicted versus

actual graph plots were clustered tightly around the line of best fit (**Figure 3**), thereby verifying the goodness of model fitting. The model F -value of 18.30 implies that the model is significant, with only a 0.01% chance that it occurs due to noise. Thus, it can be deduced that the model obtained has very good predictability and can be used to navigate the design space.

Effect of dyeing parameters on colour intensity

The influence of four process variables on the colour intensity of the dyed silk fabric can be visualised in the three-dimensional response surface plots (**Figures 4(a)–4(d)**). The surface plots (**Figures 4(a)–4(c)**) illustrate the interactive effect of pH on the colour intensity of the dyed silk yarn. The response factor shows a rapid decrease as the pH of the dye solution increases, with the highest colour intensity at pH 3. The optimal range of colour intensity is likely between pH 3 and 6.

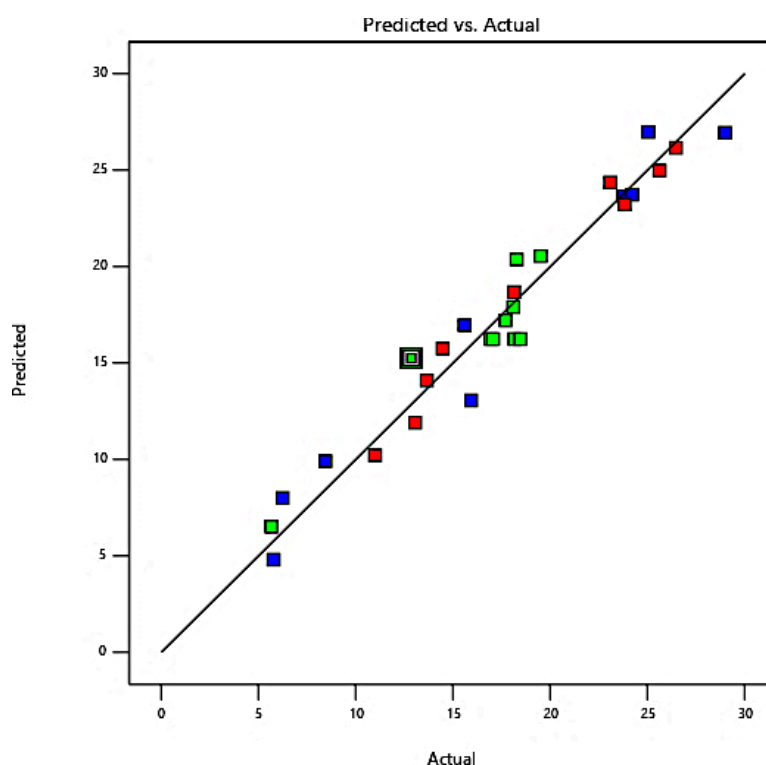


Figure 3. The predicted versus actual data for colour intensity

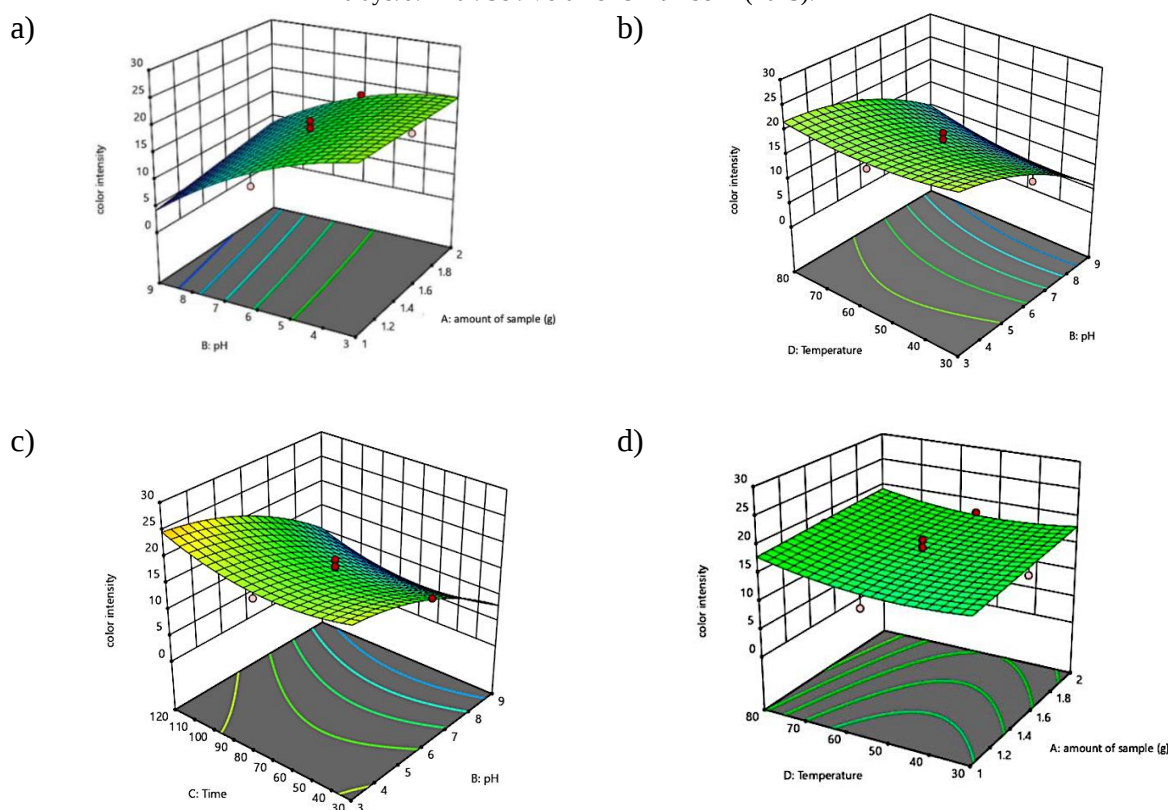


Figure 4. Three-dimensional response surface plots illustrating the interactive effect of key dyeing parameters on the C_{ab} of silk yarn dyed with *M. malabathricum* fruit extract: (a) the relationship between pH and dye extract mass, b) the relationship between temperature and pH, c) the relationship between dyeing duration and pH, and d) the relationship between temperature and dye extract mass

The dye bath was prepared by diluting the resulting extract with distilled water. The observed decrease in colour intensity as the pH of the dye bath increases is attributed to the shift in anthocyanin forms (**Figure 5(a)**). The UV-vis absorption spectra for the dye baths at pH 3, 6, and 9 showed the $\lambda_{\max}$ peak at about 550 nm (**Figure 5(b)**), which is within a typical absorption range for anthocyanidins, including delphinidin in the visible spectrum [29]. The highest peak absorbance at pH 3, followed by pH 6 and pH 9, indicates that the dye pigment is the highest at pH 3. The decline in colour intensity at pH 9 may suggest the degradation of anthocyanins at higher pH.

Co-pigmentation of the anthocyanin-metal complex may also contribute to the formation of the rich colour. Colour amplification due to complexation is maximum at a low pH domain (pH 2–4) [24]. Delphinidin, a purple-coloured pigment, is known to be stable under acidic conditions [17]. As delphinidin

is the main constituent of the colour pigment in *M. malabathricum* fruit, it likely contributes to the observed purple colouration at pH 3. The combined effect of temperature and pH on colour intensity is shown in **Figure 4(b)**, with the maximum colour intensity at the highest temperature and pH 3. This is consistent with findings from previous studies, indicating that the flavylium cation form of anthocyanins predominates under acidic conditions, producing more intense red-purple shades [17]. In contrast, higher pH levels shift the equilibrium towards the quinoidal base and chalcone forms, leading to colour fading and reduced stability [29]. The observed trend is also in agreement with studies on blueberry and camu-camu [30–31], where anthocyanin-rich extracts demonstrated higher colour retention in acidic environments. These studies demonstrate the importance of maintaining an acidic dye bath to achieve optimal colour intensity in *M. malabathricum*-dyed silk.

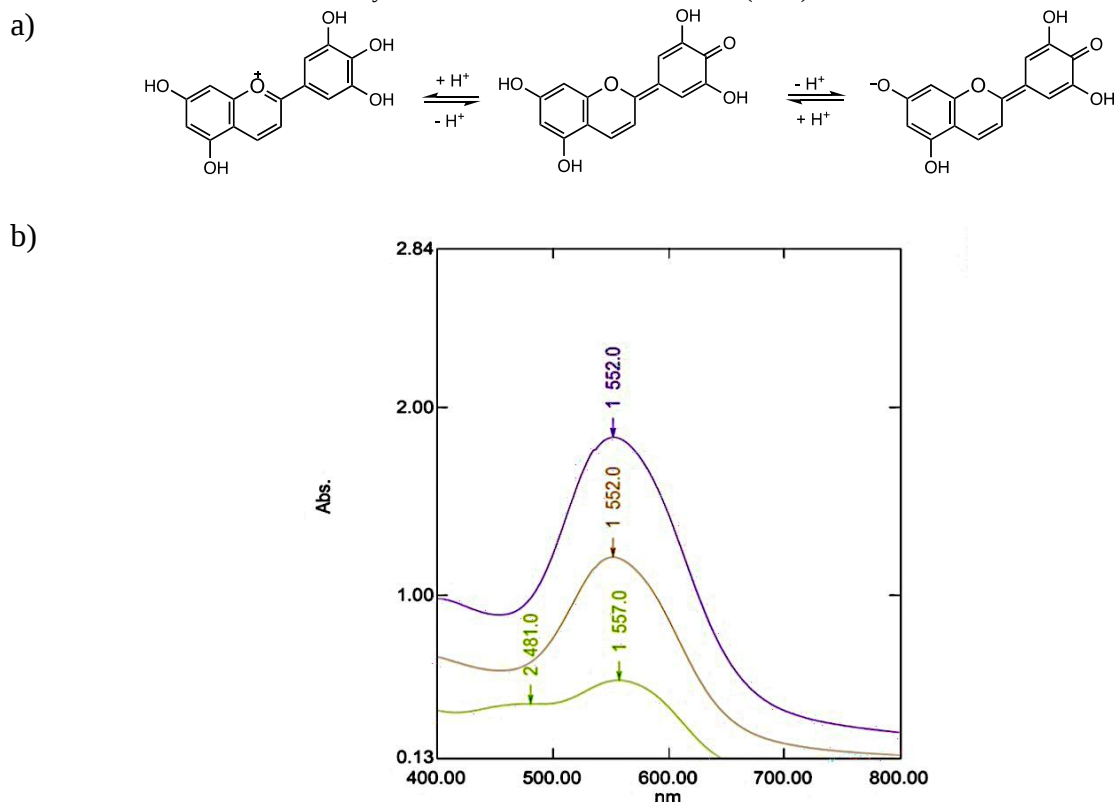


Figure 5. a) The equilibrium of anthocyanins under acidic and basic conditions and b) UV-vis spectra of dye bath at pH 3, 6, and 9

Figure 6 illustrates the colour representation of the 28-silk yarn dyeing runs for easier visualisation. The best result for the silk yarn dyeing process was achieved by using 1 g of the *M. malabathricum* fruit extract for 120 min at 30 °C and pH 3. Several factors may contribute to the observed results. Increasing the amount of fruit extract generally increases the colour intensity, although a longer dyeing duration (120 min) at 30 °C significantly decreases the colour intensity (**Table 1**). At higher concentrations or amounts of dye extract, longer dyeing durations may cause the dye bath to reach a dynamic equilibrium faster, thus limiting further dye uptake. This trend is consistent with previous reports on anthocyanin-based dyes, where excessive dye concentrations can lead to dye aggregation and reduced fabric uptake efficiency due to saturation effects [16]. In addition, studies on *Punica granatum* (pomegranate) dyeing indicate that prolonged immersion times can result in dye desorption and uneven colouration due to equilibrium shifts in dye-fibre interactions [32]. These findings suggest that optimising extract concentration and dyeing duration is essential to achieving a balance

between high colour intensity and uniform dye uptake. The phenomenon potentially leads the dye to desorb back into the solution, reducing the overall colour intensity. Higher temperature (80 °C vs. 30 °C) runs were found to reduce the colour intensity (runs 2 vs. 28 and 14 vs. 21). At higher temperatures, the energy of the dye-fibre bonds increases, leading to a higher rate of desorption. Furthermore, the equilibrium may be shifted, favouring the dye to remain in the solution rather than binding to the fibres. In addition, anthocyanins may also undergo thermal degradation at elevated temperatures, resulting in a reduction in their effectiveness of binding with the fibres. The study also indicates that while prolonged dyeing times can enhance colour uptake, extended exposure may lead to dye desorption and wastage of dye materials, which is a key factor to consider when scaling up natural dyeing processes. By balancing the optimum extract concentration, pH, and duration, textile manufacturers can optimise their resource utilisation to maintain the consistency of fabric colouration.

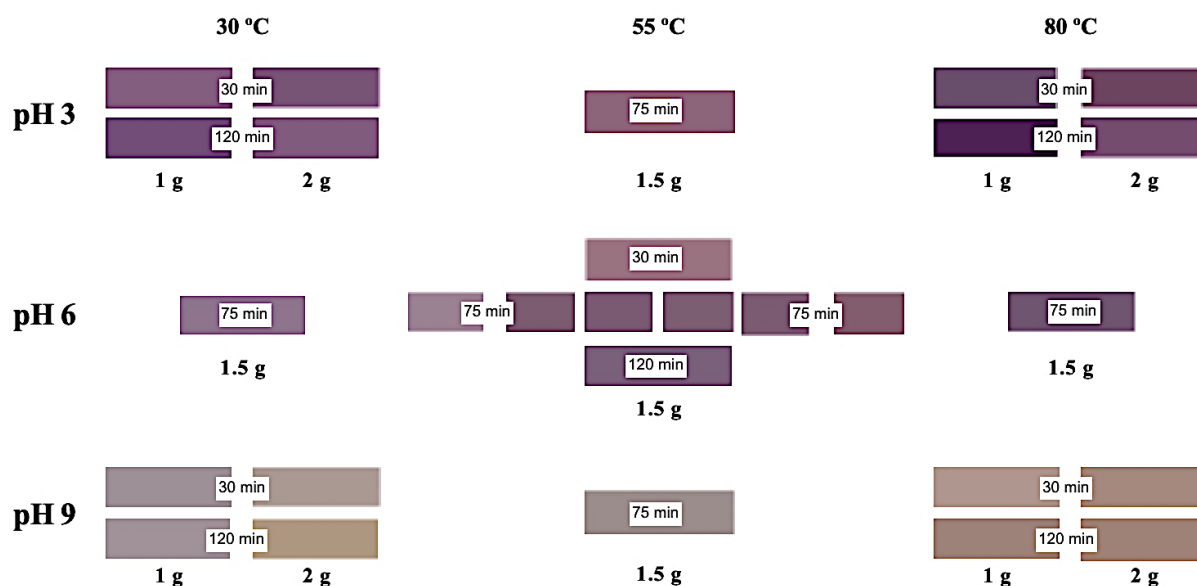


Figure 6. Summary of the colour shades of the dyed silk yarn

Conclusion

This study explores the potential of anthocyanins extracted from *M. malabathricum* fruit as a natural dye for silk yarn. The dye pigment was extracted using methanol with 0.05% acetic acid at 60 °C for 120 min and was used in the dyeing process of silk yarn through the meta-mordanting method with SnCl₂ (2%) as the mordant. The optimal dyeing conditions that yielded the highest colour intensity (28.99) were identified as using 1 g of fruit extract at pH 3 and 30 °C for 120 min. The acidic environment played a crucial role in stabilising the anthocyanin pigments. Generally, the colour intensity increased with an increased amount of extract. However, the amount of dye extract must be balanced with the weight of silk yarn to avoid saturation and aggregation of dye. The duration of dyeing depends on the temperature of the dye bath during the dyeing process, while the temperature of the dyeing process should be controlled to avoid the degradation of anthocyanins. As no prior research has investigated the use of *M. malabathricum* L. fruits in silk dyeing, this study introduces a sustainable approach while establishing its potential as a natural textile dye. These findings hold promise for eco-friendly textile applications, particularly as a biodegradable alternative to synthetic dyes. The low-temperature (30 °C) dyeing process enhances energy efficiency and cost-effectiveness, making it viable for industrial adoption. However, the variability of natural dye composition remains a challenge as it may affect batch-to-batch reproducibility, requiring standardisation strategies for industrial scalability. Furthermore, the light fastness and wash

fastness of the dyed fabric require further investigation to ensure long-term durability. Future research should explore alternative bio-mordants, improved fixation methods, and applications to different fabric types to expand the commercial feasibility of this natural dyeing process.

Acknowledgement

The authors extend their appreciation to the Al-Sultan Abdullah History & Civilisation Research Centre (Al-ASAR) - Pahang Museum for funding this research through project number SPG23-092-0092. The authors would also like to thank Miss Muezzah Afkarah Salleh for her assistance in collecting the fruit for this project.

Competing interests

The authors declare no competing interests.

References

1. Ardila-Leal, L. D., Poutou-Piñales, R. A., Pedroza-Rodríguez, A. M., and Quevedo-Hidalgo, B. E. (2021). A brief history of colour, the environmental impact of synthetic dyes and removal by using laccases. *Molecules*, 26(13): 3813.
2. Anderson, S. E., and Meade, B. J. (2014). Potential health effects associated with dermal exposure to occupational chemicals. *Environmental Health Insights*, 8: EHI.S15258.
3. Brudzyńska, P., Sionkowska, A. and Grisel, M. (2023). Silk textiles dyeing by plant-derived

- colorant in the presence of chitosan and shellac. *Fibers and Polymers*, 24: 2761-2771.
4. Gecchele, E., Negri, S., Cauzzi, A., Cuccurullo, A., Commisso, M., Patrucco, A., Anceschi, A., Zaffani, G., and Avesani, L. (2021). Optimization of a sustainable protocol for the extraction of anthocyanins as textile dyes from plant materials. *Molecules*, 26(22): 6775.
5. Elshahida, K., Fauzi, A. M., Sailah, I., and Siregar, I. Z. (2019). Sustainability of the use of natural dyes in the textile industry. *IOP Conference Series. Earth and Environmental Science*, 399: 012065.
6. Pizzicato, B., Pacifico, S., Cayuela, D., Mijas, G., and Riba-Moliner, M. (2023). Advancements in sustainable natural dyes for textile applications: A Review. *Molecules*, 28(16): 5954.
7. Darmawan, A., Widowati, R. A., Muhtar, H., and Kartono, A. S. (2024). Enhancing cotton fabric dyeing: Optimizing Mordanting with natural dyes and citric acid. *International Journal of Biological Macromolecules*, 276: 134017.
8. Chairat, M., Thongsamai, P., Meephun, T., Pantanit, S., Samosorn, S., Sajomsang, W., Gonil, P., and Bremner, J. B. (2024). Silk dyeing with anthocyanin dye extract from *Melastoma malabathricum* L. fruits using metal oxides and reducing agents to ameliorate photo-fading. *ES Materials & Manufacturing*, 25: 1213.
9. Ahmad, M., Azahari, S., and Rahim, F. (2021). Kearifan masyarakat melayu terhadap pokok senduduk dan dapatan kajian saintifik terhadap spesies ini. Sempo, M. W., Yabi, S., A. S., Syed Hassan, S. N., Syamila Sulaiman, N. A. (Eds.), *E-Prosiding Konvensyen Kearifan Nusantara Ke-3 Arif 2021*, 207-225.
10. Anuar, N., Mohd Adnan, A. F., Saat, N., Aziz, N., and Mat Taha, R. (2013). Optimization of extraction parameters by using response surface methodology, purification, and identification of anthocyanin pigments in *Melastoma malabathricum* fruit. *The Scientific World Journal*, 2013: 810547.
11. Janna, O. A., Khairul, A., Maziah, M., and Mohd, Y. (2006). Flower pigment analysis of *Melastoma malabathricum*. *African Journal of Biotechnology*, 5(2): 170-174.
12. Wan Ahmad, W. Y., Mohd Nor, M. A., Saim, N., Ab Kadir, M. I., and Ahmad, M. R. (2012). Nano natural dyes from *Melastoma Malabathricum* L. *Advanced Materials Research*, 545: 59-63.
13. Zulaicha, A.S., Munawaroh, K., Saputra, I.S., and Setiajaya, A. (2023). Utilization of senduduk fruit extract (*Melastoma malabathricum*) for natural color in lip cream. *Walisongo Journal of Chemistry*, 6(2): 215-223.
14. Sankaralingam, B., Balan, L., Chandrasekaran, S., and Muthu Selvam, A. (2023). Anthocyanin: A natural dye extracted from *Hibiscus sabdariffa* (L.) for textile and dye industries, *Applied Biochemistry and Biotechnology*, 195(4): 2648-2663.
15. Chairat, M., Bremner, J. B., and Chantapromma, K. (2007). Dyeing of cotton and silk yarn with the extracted dye from the fruit hulls of mangosteen, *Garcinia mangostana* linn., *Fibers and Polymers*, 8(6): 613-619.
16. Bechtold, T., Mahmud-Ali, A., and Mussak, R. (2007). Anthocyanin dyes extracted from grape pomace for the purpose of textile dyeing. *Journal of the Science of Food and Agriculture*, 87(14): 2589-2595.
17. Khoo, H. E., Azlan, A., Tang, S. T., Lim, S. M. (2017). Anthocyanidins and anthocyanins: colored pigments as food, pharmaceutical ingredients, and the potential health benefits. *Food & Nutrition Research*, 61(1): 1361779.
18. Affat, S. (2021). Classifications, advantages, disadvantages, toxicity effects of natural and synthetic dyes: A review. *University of Thi-Qar Journal of Science (UTsci)*, 8(1): 130-135.
19. Chelladurai, S. J. S., Murugan, Ray, A. P., Upadhyaya, M., and Narasimharaj, V., G. (2021). Optimization of process parameters using response surface methodology: A review. *Materials Today: Proceedings*, 37: 1301-1304.
20. Punyachareonnon, P., Deerattrakul, V., Luepong, K. (2021). The influence of pH, temperature and time on dyeing of silk fabric by black bean anthocyanin-rich extract as colorant. *Progress in Color, Colorants and Coatings*, 14(3): 179-186.
21. Naczki, M., and Shahidi, F. (2004). Extraction and analysis of phenolics in food. *Journal of Chromatography A*, 1054(1-2): 95-111.
22. Oancea, S. (2021). A review of the current knowledge of thermal stability of anthocyanins and approaches to their stabilization to heat. *Antioxidants*, 10(9):1337.
23. Saptarini, N. M., Fathi, and Sofian, F. F. (2013). The effect of acetic acid in anthocyanins extraction from mangosteen (*Garcinia mangostana* L.) pericarp. *Research Journal of Pharmaceutical, Biological and Chemical Sciences*, 4(2): 213-220.
24. Vankar, P. S., and Shukla, D. (2011). Natural dyeing with anthocyanins from *Hibiscus rosa sinensis* flowers. *Journal of Applied Polymer Science*, 122(5): 3361-3368.
25. Nunes, A. N., Borges, A., Matias, A. A., Bronze, M. R., and Oliveira, J. (2022). Alternative extraction and downstream purification processes for anthocyanins. *Molecules*, 27(2): 368.
26. Wan Ahmad, W. Y., Tuan Zainal Abidin, T. M. S., Ahmad, M. R., Ab Kadir, M. I., and Mohd Yusof, N. J. (2014). Dyeing of polyester using natural colorant from *Melastoma malabathricum*

- L. In: Ahmad, M., Yahya, M. (eds) *Proceedings of the International Colloquium in Textile Engineering, Fashion, Apparel and Design 2014 (ICTEFAD 2014)*. Springer, Singapore.
27. usain, A., Chanana, H., Khan S. A., Dhanalekshmi, U. M., Ali, M., Alghamdi, A. A., and Ahmad A. (2022). Chemistry and pharmacological actions of delphinidin, a dietary purple pigment in anthocyanidin and anthocyanin forms. *Frontiers in Nutrition*, 9: 746881.
28. Uddin, M. G. (2013). Effects of different mordants on silk fabric dyed with onion outer skin extracts. *Journal of Textiles*, 2014(1): 405626.
29. Skaar, I., Adaku, C., Jordheim, M., Byamukama, R., Kiremire, B., and Andersen, M. (2014). Purple anthocyanin colouration on lower (abaxial) leaf surface of *Hemigraphis colorata* (Acanthaceae). *Phytochemistry*, 105: 141-146.
30. He, R., Li, Z. M., and Wang, B. C. (2010). The stability and antioxidant activity of anthocyanins from blueberry. *Food Technology and Biotechnology*, 48(1): 42-49.
31. Zanatta, C. F., Cuevas, E., Bobbio, F. O., Winterhalter, P., and Mercadante, A. Z., (2005). Determination of anthocyanins from camu-camu (*Myrciaria dubia*) by HPLC-PDA, HPLC-MS, and NMR. *Journal of Agricultural and Food Chemistry*. 53(24): 9531-9535.
32. Adeel, S., Ali, S., Bhatti, I. A., Zsila, F. (2009). Dyeing of cotton fabric using pomegranate (*Punica granatum*) aqueous extract. *Asian Journal of Chemistry*, 21(5): 3493-3499.