Journal of Sustainable Development of Energy, Water and Environment Systems

http://www.sdewes.org/jsdewes

Year 2025, Volume 13, Issue 3, 1130576

Original Research Article

Sustainable Cotton Dyeing with Banana Floral Stem Extract Using Microwave Irradiation and Chitosan Optimised by Response Surface Methodology

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Cite as: Amalya Melati, H., Dwi Wahyuningsih, T., Rahayuningsih, E., Kartini, I., Sustainable Cotton Dyeing with Banana Floral Stem Extract Using Microwave Irradiation and Chitosan Optimised by Response Surface Methodology, J. sustain. dev. energy water environ. Syst., 13(3), 1130576, 2025, DOI: https://doi.org/10.13044/j.sdewes.d13.0576

ABSTRACT

Natural dyes offer a sustainable alternative to synthetic dyes, addressing environmental concerns in textile production. Currently, the dyeing and finishing stages typically consume large amounts of water, energy, and chemicals. A microwave-assisted dyeing process using chitosan and banana floral stem extract has been developed for cotton fabrics to enhance sustainability. This process was optimised using Response Surface Methodology and Box-Behnken Design to minimise lightness and maximise colour difference. The optimal conditions include a radiant power of 554.12 W, radiation time of 7.10 min, and chitosan concentration of 0.80%. These conditions result in a lightness value of 62.04, a colour difference of 35.36, and a desirability coefficient of 0.856. Cotton samples treated under such conditions exhibited superior colourfastness compared to conventional methods, with ratings of 4 (good) for washing, 4–5 (good to excellent) for lightfastness, and 4–5 (good to excellent) for perspiration. The colourfastness scale ranges from 1 (very poor) to 5 (excellent), indicating the level of resistance to fading or colour change. The study demonstrates the eco-friendly and efficient potential of microwave-assisted dyeing with chitosan, promoting sustainable textile practices.

KEYWORDS

Microwave irradiation, Chitosan, Banana floral stem, Box-Behnken Design, Lightness, Colour difference.

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INTRODUCTION

The utilisation of natural dyes as replacements for synthetic counterparts in the textile sector is progressively burgeoning, driven by the goal of mitigating environmental consequences by adopting secure and suitable alternative materials [1]. Natural dyes derived from renewable sources, like plants, algae, and fungi, are valued for their biodegradability, lack of toxicity and carcinogenicity, and environmental compatibility. For example, lawsone from the henna plant offers an environmentally sustainable alternative to synthetic dyes, as it does not require costly effluent treatment systems [2]. Similarly, a dye derived from green algae is biodegradable and free from harmful chemicals, making it a sustainable choice for textile industries [3]. The fungus *Talaromyces verruculosus* produces a natural pigment that is non-cytotoxic and safe for use, providing an eco-friendly dyeing solution [4]. Methylating brazilein from second wood with dimethyl carbonate (DMC) enhances its colour stability and resistance to pH changes, while DMC's environmentally benign properties align with sustainable chemistry principles [5]. Another source of natural dyes is banana plants, which are classified as biowaste following the harvest of the fruit. Phytochemical analysis indicates that the aqueous extract of the banana floral stem (BFS) from the Ambon variety (*Musa sapientum*) comprises chromophore compounds potential as natural dyes, including condensed tannins, flavonoids, anthraquinones, and anthocyanins, rendering it a viable option for multifunctional textile applications [6]. The findings further validated that BFS can yield significant colour strength ($K/S = 0.71$) in cotton fabric [7].

Another concern for natural dyes is the weak adherence to the fabrics. For example, the most famous natural fibre, cotton, has a high density of negatively charged groups from exclusively hydroxyl groups and ether bonds, which makes it difficult to dye with natural dyes [8]. Natural dyes in aqueous solutions are repelled by these charges, resulting in weak colourfastness and poor colour absorption [9]. Numerous techniques, including surface modification using metal and biological mordant, have been carried out to enhance cotton fabric dyeing performance [8]. Amongst these, biological mordants, such as chitosan, are used to modify the surface of cotton to increase its affinity for anionic dyes. Chitosan polysaccharides are highly recommended among natural materials due to their distinctive chemical and biological features, as well as their accessibility to industrial applications [10]. Silk fabrics coloured with natural *Rubia cordifolia* L. colours using the pad-dry method and treated with chitosan have shown better colourfastness of 5 (excellent) to wash, rub, and light [11]. This cationisation technique will be applied in the dyeing process for BFS on cotton fabrics.

Furthermore, research on applying environment-safe radiation technology to dyeing is ongoing. Gamma radiation at 30 kGy (kiloGray, a unit of absorbed dose of ionising radiation) enhances the dyeing affinity of cotton fabric with lutein from marigold, improving colour strength and fastness, particularly when tannic acid is used as a pre-mordant and copper as a post-mordant [12]. Electron beam irradiation efficiently modifies synthetic fabrics like polypropylene and polyester by introducing functional groups that enhance natural dye absorption and colour retention [13]. Ultraviolet radiation also improves lutein uptake, increasing colour strength and fade resistance, especially when applied for 90 min under optimal conditions [14]. Plasma treatment enhances fabric properties, including antibacterial activity, while reducing the use of hazardous chemicals, contributing to more sustainable dyeing processes [15]. Microwave radiation in linen dyeing with natural lac optimises processing by reducing both time and energy consumption, offering a safer and more eco-friendly alternative to conventional heating methods [16].

Among the mentioned techniques, microwave irradiation, not only for surface modification but also for the dyeing process simultaneously, provides numerous benefits owing to its uniform heating, low cost, short processing time, and low energy consumption with mass transfer dynamics [17]. Recent research indicates that combining natural dye from

the insect *Laccifer lacca* with microwave heating is effective for linen dyeing, lowering dyeing time to approximately 7 min while ensuring sufficient colourfastness and conserving energy [16]. Stimulation of dyeing with microwave assistance for 4 min yields excellent colour strength [18]. Another study used microwave heating for reactive dyeing and obtained superior results at a fixing duration of just 3 min, unlike the conventional technique, which takes 45 min to complete the process. Additionally, the colour strength of the samples dyed using the microwave method was greater than that of those dyed via the conventional method [19].

The prospective microwave dyeing combined with chitosan cationisation should enhance the adherence of the BFS on cotton fabrics. In the present study, the process will be optimised using a statistical approach, specifically the Design of Experiments (DoE) approach with Response Surface Methodology (RSM). Such an approach is instrumental in constructing empirical models, optimising processes, and analysing interactions among numerous factors to enhance process outcomes [20]. With Box-Behnken Design (BBD), RSM enhances efficiency by enabling the concurrent optimisation of many process variables, thus minimising the number of experimental trials needed when compared with other RSM techniques, such as Central Composite Design (CCD) [21]. The absence of corner points in BBD avoids extreme conditions, especially in the dyeing process using the microwave. This technique has been successfully implemented in industrial-scale optimisation of various process parameters [22].

To the best of our knowledge, there have been no previous reports on using microwave and chitosan to dye cotton with banana (*Musa paradisiaca* var. Nipah) floral extract. In the present study, the BBD was employed to refine the cotton dyeing process. The optimisation of process variables, including radiant power, radiation time, and chitosan concentration, enabled attaining optimal lightness levels and colour differences. Furthermore, a comparative assessment of treated and untreated cotton samples was performed to evaluate differences in colour strength and fastness.

MATERIALS AND METHODS

This section provides a comprehensive description of the methodology employed in the present study. The process begins with preparing natural dyes, followed by applying chitosan to fabric samples. The dyeing process, along with its optimisation, is then carried out. Afterwards, the samples are analysed using Fourier Transform Infrared Spectroscopy (FTIR) and Scanning Electron Microscopy (SEM) to characterise their properties. Finally, the colour evaluation of the samples is conducted, providing a thorough assessment of the effects of the treatments applied.

Materials

Plain 100% cotton fabric (96 g m^{-2}), supplied by the company PT Primissima of the Special Region of Yogyakarta, was purchased from the local online market. Banana (*Musa paradisiaca* var. Nipah) floral stems, selected as a natural dye source, were gathered from the indigenous habitat of Sungai Kakap, Kuburaya, Indonesia. Other materials were chitosan ($C_{56}H_{103}N_9O_{39}$) with medium molecular weight (GR for analysis, CAS number 9012-76-4), acetic acid (CH_3COOH, Merck for analysis EMSURE®, CAS number 64-19-7), and non-ionic detergent Triton X-100 ($C_{16}H_{26}O_2$, GR for analysis, CAS number 9036-19-5).

Laboratory procedures

Dye Extraction. A crude bio-source extract was formulated by boiling 50 g of banana floral stem powder with 1000 mL of distilled water. The extract was then heated at 60 °C for 60 min, followed by filtration, and the resulting extracted material was preserved for subsequent dyeing procedures.

Chitosan Treatment. Before dyeing process, a clean cotton fabric (5 cm × 10 cm) was washed using non-ionic detergents was impregnated for 1 h in a solution containing 0.5–1% (w/v) of chitosan and 1% (v/v) of acetic acid in distilled water, then padded 100% wet pick up with a padder by a pad-dry-cure method, dried at 80 °C for 5 min, and cured at 120 °C for 3 min.

Dyeing Process. It was conducted in a laboratory-based domestic microwave oven (Electrolux, model EMM2308X, with an output power of 800 W operating at 2450 Hz). Using hard liquor with a fabric ratio of 40:1, variable factor settings of the microwave's radiant power and radiation time were adapted based on the experimental design. For comparison purposes, the conventional dyeing technique involved treating cotton by heating the dye solution to a temperature of 60 °C for 1 h, also utilising a liquor-to-material ratio of 40:1.

Measurement techniques

Fourier Transform Infrared Analysis. The functional groups present in material samples were identified using Attenuated Total Reflectance-Fourier Transform Infrared (ATR-FTIR) spectroscopy using Thermo Scientific Nicolet iS10 with a resolution of 4 cm^{-1} and a spectral range of 4000−550 cm^{-1}. The investigation included samples of cotton, chitosan, chitosan-treated cotton, dyed cotton using conventional, and dyed cotton using microwave-assisted methods.

Scanning Electron Microscopy. The surface morphology of untreated, treated with chitosan, and treated with both chitosan and natural dye cotton was analysed using a scanning electron microscope (SEM) JEOL JSM-6510LA operating at 10 kV.

Experimental design

The experimental design and statistical response analysis were conducted using the statistical software Design Expert 13.0 (Stat-Ease, Minneapolis, USA) to obtain optimal radiant power, radiation time, and chitosan concentration variables that yield the lowest lightness and highest colour difference. The Box Behnken Design, an RSM tool, optimised those three process variables. The ranges of experimental factors representing the independent input parameters, their coded factors, and actual factors are tabulated in **Table 1**. The statistical software IBM SPSS Statistics (IBM Co., Ltd., America, United States) was utilised to evaluate the difference between the predicted values and the actual or experimental values by a sample-independent t-test for lightness and colour difference. Lightness and colour difference were designated as the corresponding output responses to the input parameters and expressed by second-order polynomial relationship as shown in eq. (1):

$$Y = \beta_0 + \sum_{i=1}^{n} \beta_i X_i + \sum_{i=1}^{n} \beta_{ii} X_{i^2} + \sum_{1 \le i \le j}^{n} \beta_{ij} X_i X_j + \varepsilon \quad (1)$$

Where β_0, β_i, β_{ii}, and β_{ij} represent regression coefficients; X_i and X_j are coded independent variables; Y is the response of lightness and colour difference, and ε is the model error.

Table 1. Experimental factors and their ranges

Code factor	Actual factor	Unit	Low level	High level
A	Radiant power	[W]	100	800
B	Radiation time	[min]	1	12
C	Chitosan concentration	[%]	0.5	1.0

Dyeing assessment

The colour analysis of the cotton fabric samples at each stage was conducted. The optimisation of three parameters of dyeing (radiant power, radiation time, and chitosan concentration) used lightness values from colour coordinate measurement, especially lightness (L^*) and colour difference (ΔE) as responses.

The Colourimeter HFBTE AMT516 was used to measure CIE colour space parameters L^*, a^*, and b^*. The symbol of L^* represents the level of lightness, a^* ranges from redness to greenness, and b^* ranges from yellowness to blueness. The value of ΔE means the colour difference between the sample and reference, which are the treated cotton and untreated cotton fabrics. The colour difference was obtained using eq. (2):

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

The absorbance (Abs) and reflectance coefficient (R) of some selected dyed cotton were measured utilising a Shimadzu UV–2401–PC Spectrophotometer. The Kubelka-Munk formula in eq. (3) was employed to compute the colour strength value, that is, the ratio of absorbance coefficient K and diffusion coefficient S:

$$K/S = \frac{(1-R)^2}{2R} \quad (3)$$

Further, the colour fastness of the dyed cotton was evaluated by International Standards, utilising the grey scale for colour change and staining, rated on a scale of 1 to 5. A grey scale value of 1 indicates very poor fastness, while a rating of 5 signifies excellent fastness [23]. The dyed cotton samples were tested under ISO 105-C10:2006, ISO 105-B01:2014, and ISO 105-B07:2009 to assess wash fastness, light fastness, and perspiration fastness levels with colour changes assessed. The schematic illustration of the methodology is expressed in Figure 1.

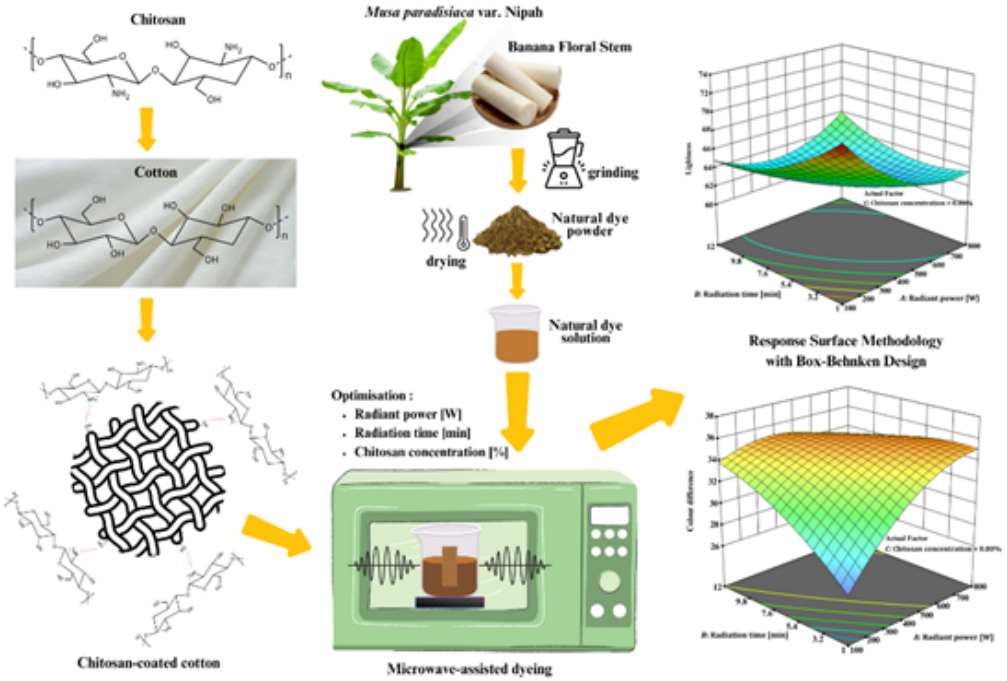


Figure 1. Schematic illustration of microwave-assisted dyeing of chitosan-coated cotton using banana floral stem extract and optimisation of the dyeing process utilising RSM with BBD

RESULTS AND DISCUSSION

This section begins by demonstrating the effects of chitosan and the dyeing process assisted by microwave radiation on the colour coordinate values (L^*, a^*, b^*), the colour difference (ΔE), colour strength (K/S), and absorbance (Abs) of dyed samples. Subsequently, optimisation is conducted to determine the optimal values of radiation power, radiation time, and concentration used in the dyeing process, along with its mathematical model, followed by experimental validation. The section concludes with the evaluation of the colour fastness of both treated and untreated samples towards wash, light, and perspiration.

Effect of chitosan and microwave-assisted dyeing treatment

The cotton fabrics were subjected to chitosan treatment through the pad-dry-cure method to impart a cationic charge, facilitating dyeing with banana (*Musa paradisiaca* var. Nipah) floral stem dye without requiring metallic mordants. The infrared spectrophotometer was employed to determine the functional groups of the cotton samples and confirm the modification of the cotton surface with chitosan. The spectra of cotton fabrics, chitosan, and chitosan-coated cotton fabrics are presented in Appendix **Figure A1**. After the dyeing process, the microwave-dyed and conventionally dyed cotton fabrics were evaluated and compared. **Figure 2** shows the absorbance spectra of the fabrics dyed with and without chitosan using microwave-assisted and conventional dyeing methods. Chitosan coating as a bio-mordant on cotton before dyeing enhances the cotton's ability to absorb dyes compared to untreated cotton. This is indicated by the high absorbance value in **Figure 2**. More effective dye absorption occurs in cotton fabrics with higher absorbance levels **[24]**. Conversely, dyeing treatment using microwave radiation influences the absorbance value. In microwave-assisted dyeing, the absorbance value is higher compared to conventional dyeing. The data shown in **Table 2** further support this observation.

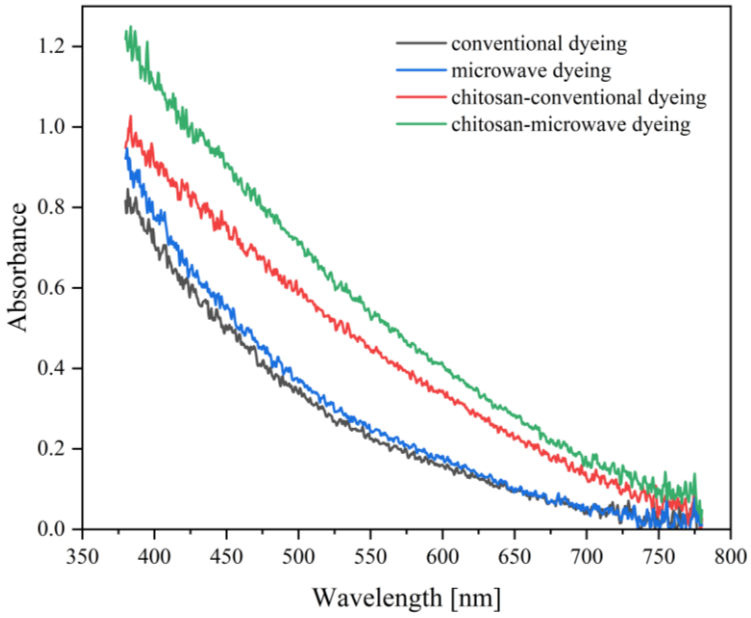


Figure 2. Absorbance spectra of the dyed cotton with conventional dyeing, microwave dyeing, conventional dyeing with chitosan-coated, and microwave dyeing with chitosan-coated

Table 2 shows that the dyeing of cotton fabrics treated with chitosan shows better colour difference, colour strength, and colour absorption ability than untreated cotton fabrics. This indicates that chitosan aids in absorbing and retaining the colours of banana flower stem extracts on cotton fabrics. Lower L^* values suggest that the sample darkens relative to the control sample.

Table 2. Effect of microwave irradiation and chitosan treatment on colour coordinates (L^*, a^*, b^*), colour difference (ΔE), colour strength (K/S), and absorbance (Abs) of dyed samples

Sample	L^*	a^*	b^*	ΔE	K/S	Abs	Coloured image
Cotton fabrics	92.84	0.77	–2.28	–	–	–	
Cotton dyeing with conventional dyeing	71.29	4.52	10.06	25.11	0.36	0.25	
Cotton dyeing with microwave dyeing	70.35	5.33	14.55	28.46	0.44	0.28	
Chitosan-coated with conventional dyeing	61.46	8.65	15.14	36.74	0.86	0.42	
Chitosan-coated with microwave dyeing	59.12	8.77	14.77	38.62	1.43	0.52	

The colour strength (K/S) value of the dyed material exhibits a direct relationship with the dye concentration within the material [23]. Chitosan-coated cotton fabrics exhibit a higher K/S value than uncoated cotton. Amino groups of chitosan that are slightly protonated in acidic environments may enhance the immobilisation of chitosan molecules onto cotton via electrostatic attraction [25]. **Figure 3** illustrates the possible mechanism of dye adherence to cotton coated with chitosan. The diverse functional groups in the cotton, chitosan, and dye structures allow an electrostatic force among the three [26].

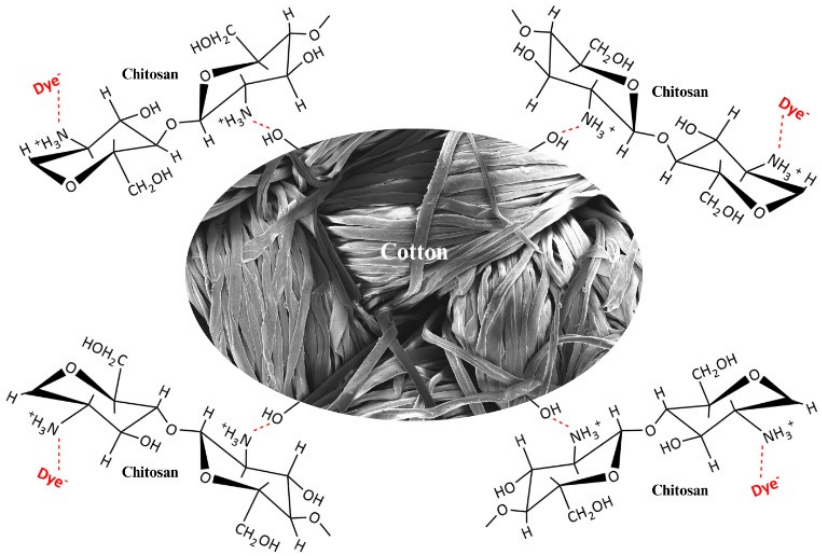


Figure 3. Possible mechanisms that generate electrostatic attraction between cotton, chitosan, and dye

Table 2 also demonstrated that chitosan-coated cotton dyeing assisted by microwave irradiation yields superior results, with the lowest lightness value and the highest colour difference and colour strength (K/S). Microwave-assisted heating is a more efficient method for chemical reactions, significantly reducing reaction times. This method directly heats the reaction mixture, providing uniform temperature throughout the system [27]. Unlike conventional

heating, which only affects the surface, microwave irradiation heats materials volumetrically. Polarised molecules, such as water, reorient with the changing electromagnetic field, generating heat through friction and facilitating dye penetration into the fabric. The microwave heating rate and dye fixation reach equilibrium at a specific point, optimising dye adherence to the fabric and producing superior colour depth in a shorter time than conventional methods [16]. The enhancement in colour immersion and dye absorption on textile materials can be attributed to the efficient mass transfer kinetics promoted by microwave-assisted extraction, which accelerates the process while preserving the integrity of the cotton structure [28]. Additionally, microwave radiation plays a dual role by altering fibre surfaces, which further enhances dye penetration and improves colour fastness, ensuring better retention without damaging the cotton material [29]. As seen in the FTIR spectra presented in Appendix **Figure A2**, microwave irradiation did not exert a notable impact on the chemical structure of cotton fibres, as also observed by [30]. The surface morphology of cotton fabrics also displays no differences, as examined using Scanning Electron Microscopy (SEM) in Appendix **Figure A3**.

Microwave-assisted dyeing provides significant environmental and economic advantages, including reductions in water, chemical, and energy consumption, while promoting eco-friendly dyes [31]. When compared to other radiation-based dyeing methods, such as ultrasonic and UV radiation, microwave-assisted dyeing offers superior efficiency and environmental benefits. This method reduces dyeing time, lowers chemical consumption, and enhances colour strength and durability [32]. Microwave-assisted dyeing is more time-efficient than ultrasonic dyeing, while UV radiation is typically restricted to post-dyeing treatments [33]. However, the industrial-scale implementation of microwave dyeing faces challenges related to high initial investment, the necessity for specialised equipment, and the need for uniform heating across large volumes of fabric [34]. Process optimisation is crucial to maintaining operational efficiency and product quality.

Model fitting and statistical analysis

Significant processes of variables (A) radiant power, (B) radiation time, and (C) chitosan concentration were chosen for dyeing process optimisation of the operating conditions using the Box-Behnken approach for minimising lightness (L^*) and maximising colour difference (ΔE) of dyed samples (**Table 3**). Box Behnken Design generated a total of 27 experiments. All experiments' lightness and colour difference values are presented in **Table 4**.

Table 3. Independent variables and levels according to Box-Behnken

Variables	Levels		
	−1	0	1
Radiant power [W]	100	450	800
Radiation time [min]	1.0	6.5	12.0
Chitosan concentration [%]	0.50	0.75	1.00

Analysis of variance (ANOVA) was employed to assess the significance of the factors and their interactions with the response. The p-values below 0.05 indicate the statistical significance of the model and its terms. Insignificant interactions were eliminated through a model reduction in this study to enhance the model. **Table 5** demonstrates that the model is adequate for exploring the design space. The significance of the interaction terms (AB, AC, BC) indicates whether a meaningful interaction effect exists between the two dependent variables. In this case, the ANOVA model results indicate that A, B, C, AB, A^2, B^2, and C^2 are significant model terms as the p-values are less than 0.05. This table also confirmed that only the interaction between A and B is significant, and the relationship between A and the responses L^* or ΔE is influenced by term B or remains constant across different levels of the term B.

Table 4. Experimental design of the process factors and responses

Sample	*A*: Radiant power [W]	*B*: Radiation time [min]	*C*: Chitosan concentration [%]	Response 1: Lightness L^*	Response 2: Colour difference ΔE
1	450	6.5	0.75	61.04	36.52
2	450	6.5	0.75	61.71	35.26
3	100	6.5	0.5	69.28	28.17
4	450	1.0	0.5	69.44	28.22
5	800	12.0	0.75	66.28	32.01
6	100	6.5	1.0	68.64	29.41
7	450	6.5	0.75	60.31	36.57
8	800	1.0	0.75	63.17	35.85
9	800	6.5	0.5	68.63	28.30
10	450	6.5	0.75	62.17	34.68
11	450	6.5	0.75	62.32	34.60
12	450	6.5	0.75	63.86	33.20
13	450	6.5	0.75	63.39	33.83
14	450	12.0	0.5	65.34	32.23
15	100	12.0	0.75	65.56	32.87
16	450	6.5	0.75	63.92	33.41
17	100	1.0	0.75	71.22	26.06
18	450	1.0	1.0	66.68	30.87
19	450	6.5	0.75	62.71	35.84
20	450	6.5	0.75	61.24	35.96
21	450	12.0	1.0	65.30	32.23
22	450	6.5	0.75	63.85	33.73
23	450	6.5	0.75	60.87	36.73
24	800	6.5	1.0	63.70	33.68
25	450	6.5	0.75	61.36	35.87
26	450	6.5	0.75	62.74	35.05
27	450	6.5	0.75	64.79	32.52

Similarly, the significance of the quadratic terms (A^2, B^2, C^2) suggests the presence or absence of a significant quadratic relationship between the variables (A, B, C) and the responses variables. This indicates that the relationship between the dependent variables and the response variables is non-linear, exhibiting a curved pattern [35]. The sum of the square model provided the most significant model ($p < 0.0001$) with an insignificant lack of fit for both L^* and ΔE, which were 0.4562 and 0.3386, respectively; the lack of significance in lack-of-fit testing implies that the model is accurate. The quadratic model in terms of coded factors is provided (after removing insignificant terms) in equations (4) and (5).

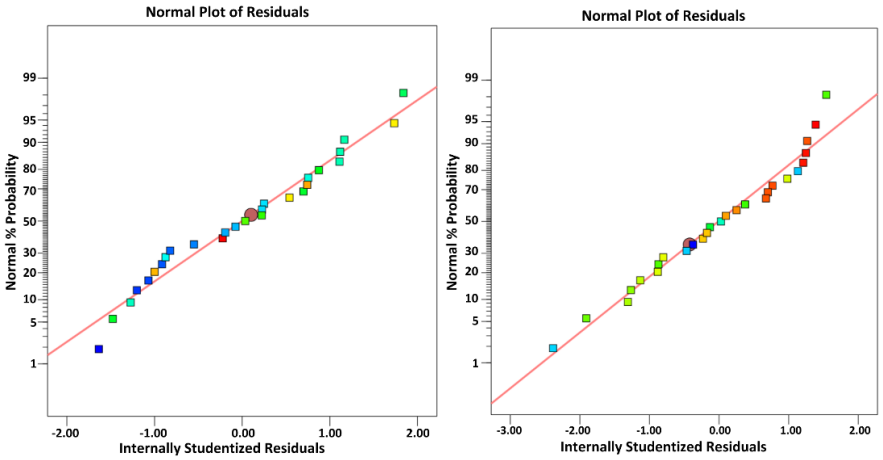
$$\text{Lightness } (L^*) = 62.42 - 1.62A - 1.00B - 1.05C + 2.19AB + 2.51A^2 + 1.63B^2 + 2.64C^2 \quad (4)$$

$$\text{Colour difference } (\Delta E) = 34.92 + 1.67A + 1.04B + 1.16C - 2.67AB - 2.11A^2 - 1.11B^2 - 2.92C^2 \quad (5)$$

Where A is the radiant power of the microwave, B is radiation time, C is chitosan concentration, and AB is the term of interaction between the radiant power and the radiation time.

Table 5. ANOVA results of the established model

Source	Mean square		F-value		p-value		
	L^*	ΔE	L^*	ΔE	L^*	ΔE	
Model	27.91	26.91	15.68	13.98	< 0.0001	< 0.0001	significant
A – Radiant power [W]	20.87	22.26	11.72	11.57	0.0028	0.0030	
B – Radiation time [min]	8.06	8.71	4.53	4.53	0.0467	0.0467	
C – Chitosan concentration [%]	8.76	10.74	4.92	5.58	0.0390	0.0290	
AB	19.23	28.42	10.80	14.77	0.0039	0.0011	
A^2	30.75	21.75	17.27	11.30	0.0005	0.0033	
B^2	13.06	6.03	7.34	3.14	0.0139	0.0927	
C^2	34.09	41.81	19.15	21.72	0.0003	0.0002	
Lack of Fit	1.77	2.26	0.99	1.25	0.4562	0.3386	not significant



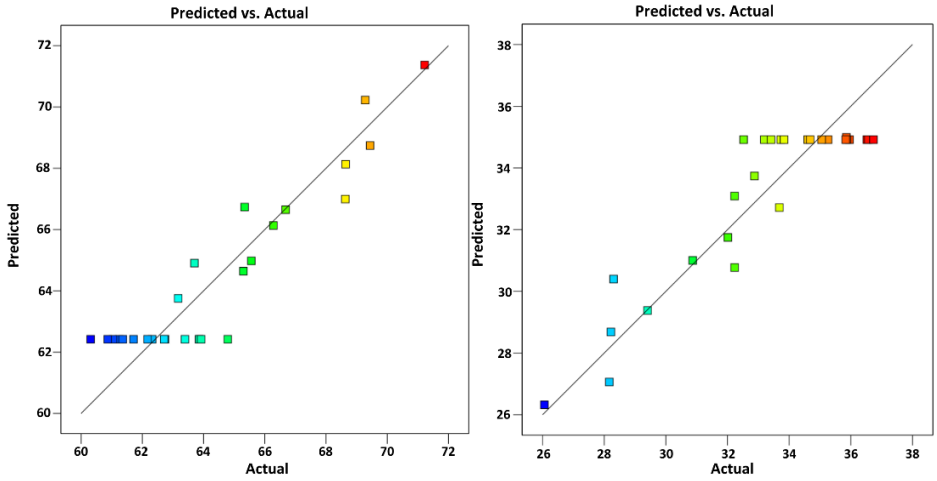
(a) (b)

Figure 4. The studentised residuals vs. normal percentage probability plot for L* responses (a) and ΔE responses (b)

The regression coefficients (R^2) for the two responses (lightness and colour difference) are 0.8524 and 0.8374, respectively, indicating that only 0.1476% (lightness) and 0.1626% (colour difference) are not explained by the model due to errors. The predicted R^2 and adjusted R^2 values for lightness and colour difference are 0.7981 and 0.6823 and 0.7776 and 0.5830, respectively (**Table 6**). The predicted R^2 agrees with the adjusted R^2 for lightness and colour difference since the difference is less than 0.2. These high R^2 values are quite consistent with each other and enhance the significance of the model. After plotting normal percentage probability plots, data points that deviate from the model are scattered along the straight line, and very few points are outside the line (**Figure 4**). A strong correlation between the dependent variables and responses was observed from a simplified quadratic model, which was validated through a normal probability plot of the residuals. The assumption of normality was confirmed, as the residual plots closely followed a linear correlation for both lightness and colour difference, indicating that the simplified quadratic model is reliable for the selection process **[36]**. The same observation is seen in the actual versus predicted lightness (**Figure 5a**) and colour difference values (**Figure 5b**). Therefore, this relationship between experimental responses and model outputs is significant.

Table 6. ANOVA results of fit statistics

Measuring item	*L**	*ΔE*
Std. Dev.	1.33	1.39
Mean	64.43	33.10
C.V. [%]	2.07	4.19
R^2	0.8524	0.8374
Adjusted R^2	0.7981	0.7776
Predicted R^2	0.6823	0.5830

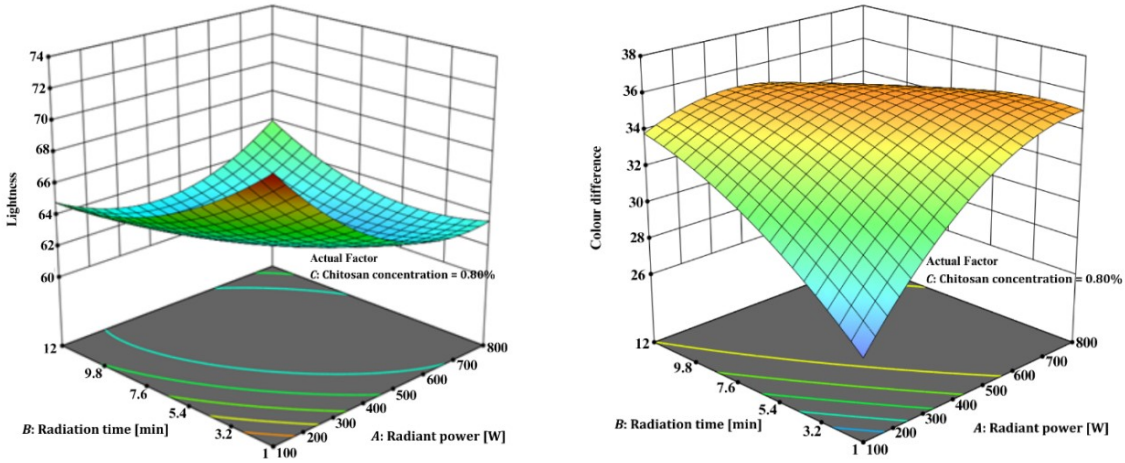


(a) (b)

Figure 5. The actual versus predicted plot for L* responses (a) and *Δ*E responses (b)

Effects of dyeing parameters on lightness and colour difference

Figure 6 illustrates how both radiant power (A) and radiation time (B) influence the lightness and colour difference of dyed samples. Equations (4) and (5) were employed to simplify the plotting of response surfaces, with two variables represented simultaneously while the third variable remained constant at a specified value within each graph.



(a) (b)

Figure 6. Response surface plots showing the effect of each significant parameter on the dyed samples: L* response (a) and *Δ*E response (b)

The figure demonstrates that higher radiant power and longer radiation times directly correlate with increased colour differences and decreased lightness. An increase in radiant power and radiation time decreases the lightness value and increases the colour difference value until reaching the optimum power of 554.12 W and radiation time of 7.10 min. This relationship is attributed to the rise in system temperature as radiation time extends. The elevated temperature enhances the diffusion of dye substances, leading to more significant fibre swelling in cotton [37]. In low temperatures, the quantity of dye absorbed is small, leading to a diminished value of colour difference. At elevated temperatures, more dye molecules rapidly migrate to the surface from the dye bath and permeate the fabric through the open pores of the cellulose material, resulting in a higher colour difference value. The solubility of the dye, the rate of dye absorption, and the absorption of the dye are all improved at elevated temperatures, leading to accelerated diffusion of dye molecules into the fabric through its open pores, yielding a higher colour difference value [23].

On the other hand, further increases in power >554.12 W and radiation time >7.10 min have the opposite effect. Increased microwave power and radiation time cause more significant damage to the natural dye matrix, and intense absorption of microwave energy leads to a swift temperature rise within the dyeing vessel [38]. Increasing radiant power and radiation time accelerates temperature rise, leading to dye molecule degradation. The decline in colour difference values beyond a specific dyeing temperature (more than 80 °C) may be attributed to the desorption of dye molecules upon attaining equilibrium at this temperature, leading to diminished dye absorption and, consequently, reduced colour difference values [23]. The chromophore compound from banana floral stem extract is significantly retained within the cellulose matrix at 80 °C, which expands the cotton pores, facilitated by the attractive interactions between the cellulose of cotton and the chromophore anchoring sites. Nonetheless, elevating the temperature beyond 80 °C results in the hydrolytic destruction of the chromophore compound of banana floral stem extract [7]. Insufficient heating levels fail to expedite the movement of dye molecules toward the changed fabric surface.

In contrast, excessive heating induces the desorption of dye molecules or leads to dye degradation, culminating in diminished colour strength [16]. The higher kinetic energy of the dye substances and the breakdown of dye aggregates occur at higher temperatures [39]. In the microwave mechanism, internal electric fields are produced through the interaction of energy with substances at the molecular level. This ability converts electromagnetic energy into thermal energy within the irradiated material [40].

Optimisation and validation of the model

The optimal conditions for dyeing cotton fabrics with banana floral stem extract were predicted using Design Expert 13.0 on the optimisation function. The study aimed to discern the variance between observed outcomes and projected results based on one or more criteria of the dyeing process. More specifically, the aim was to achieve the lowest lightness and the highest colour difference, considering microwave dyeing radiant power, radiation time, and chitosan concentration within a specified range. The findings are outlined in Table 7.

Table 7. Optimised conditions for obtaining the lowest lightness and the highest colour difference

Name	Goal	Lower Limit	Upper Limit	Optimum
A: Radiant power [W]	in range	100	800	554.12
B: Radiation time [min]	in range	1	12	7.10
C: Chitosan concentration [%]	in range	0.5	1	0.80
Lightness (L^*)	minimise	60.31	71.22	62.04
Colour difference (ΔE)	maximise	26.06	36.73	35.36

The optimal conditions for dyeing were achieved at a radiant power of 554.12 W, radiation time of 7.10 min, and chitosan concentration of 0.80% with 0.856 desirability. The ideal expected value for minimum lightness is around 62.04, and for maximum colour difference is around 35.36.

Cotton fabric samples were arranged under the optimised conditions to validate the experimental model. **Table 8** shows that the predicted lightness and colour difference are closely aligned with the experimental or exact values, further validating the experimental model's accuracy. The predicted values exhibit significant agreement with the exact (experimental) values and are deemed statistically insignificant, with $p > 0.05$. The values for prediction data exhibit negligible variance from the experimental data. This implies that there are no significant deviations from the objective distribution, suggesting that the applied model is appropriate and sufficiently adaptable for accurate optimisation **[41]**.

Table 8. Predicted and exact values of the dependent variable under optimum conditions of microwave-assisted dyeing; ns denotes an insignificant difference between the predicted and the exact value ($p > 0.05$)

Dependent variable	Microwave-assisted dyeing		Sig.
	Predicted	Exact	
Lightness (L^*)	62.04	61.54	0.575^{ns}
Colour difference (ΔE)	35.36	36.46	0.474^{ns}

Colourfastness properties

This study assessed the quality of dyeing on the cotton fabric via a colourfastness test for wash, light, and perspiration. The evaluation compares the colour change with a colour change standard. Proven standards include those published by the International Standards Organisation (ISO), including a grey scale for evaluating colour change in cotton samples. **Table 9** presents the colourfastness values of cotton fabrics dyed using conventional and microwave methods, both with and without chitosan treatment, using an aqueous extract from a banana (*Musa paradisiaca* var. Nipah) floral stem. The coloured images of colourfastness properties are shown in Appendix **Table A1**, **Table A2**, and **Table A3**.

Table 9. The colourfastness properties of dyed cotton (1–2 very poor to poor, 2 poor, 3–4 fair to good, 4 good, 4–5 good to excellent)

Sample	Wash fastness (ISO 105-C10:2006)	Light fastness (ISO 105-B01:2014)	Perspiration (ISO 105-B07:2009)
Conventional dyeing	1–2	4–5	2
Microwave dyeing	1–2	4–5	2
Conventional dyeing with chitosan-coated	3–4	4–5	4–5
Microwave dyeing with chitosan-coated	4	4–5	4–5

Cotton fabrics treated with chitosan exhibit good wash, light, and perspiration fastness properties, scoring between 3–4, 4–5, and 4–5 on the grey scale, respectively, with 5 being the highest rating **[42]**. In contrast, cotton samples without chitosan treatment exhibit poor wash and light fastness. The highest colourfastness is achieved when the cotton fabric is coated with chitosan, possibly owing to the ionic interaction between chitosan's amine groups and the natural dye molecules' hydroxyl groups **[43]**. Additionally, there are slight differences in wash fastness

between conventional dyeing methods and microwave-assisted dyeing on chitosan-coated cotton, with microwave assistance resulting in slightly better results. This improvement can be ascribed to the ability of microwave radiation to modify the fabric surface, enhancing the sorption of colourants without altering their chemical nature [44], thus suggesting the application of a microwave-assisted process for cotton dyeing. The results highlight the effectiveness of microwave irradiation and chitosan in improving colour difference, colour strength, and the colourfastness of cotton fabrics dyed with banana floral stem extract.

Based on the literature, the aqueous extract of BFS (*Musa sapientum*) contains several compounds, including condensed tannin, flavone, anthraquinone, and anthocyanin, contributing to its dyeing properties. Among these, flavone and anthraquinone showed stronger exhaustion and fixation on cotton than tannin and anthocyanin. Tannin, with its higher molecular weight, exhibited lower fixation after dyeing and washing. The better fixation of flavones and anthraquinone is likely due to their ability to form stronger interactions with cotton fabrics through hydrogen bonding and van der Waals forces [7]. Adding chitosan further enhances colourfastness by introducing functional groups (–OH, $–NH_2$), which improve the binding of these compounds to the cotton matrix [45].

Microwave-assisted dyeing technology offers significant advantages in terms of dye uptake, colour strength, and fastness, making it a versatile solution for various fibre types [46]. While the technology provides notable thermal energy savings and operational cost reductions through faster and more uniform heating, the high initial capital investment remains a substantial barrier to large-scale implementation [46]. Despite this, long-term cost savings in water, chemicals, energy, and labour can offset these upfront expenses [47]. However, challenges persist, including the need for specialised technical expertise and difficulties in scaling the process for industrial use, particularly for small-scale manufacturers [48]. While the technology reduces the need for additional chemicals and water, certain operational costs, such as electricity consumption and maintenance, may partially diminish the anticipated energy savings. Therefore, it is crucial to carefully evaluate the economic feasibility of adopting microwave-assisted dyeing on a large scale, considering both initial and ongoing operational costs [49].

CONCLUSION

The growing demand for eco-friendly products has led to their widespread adoption. This study optimises microwave-assisted dyeing of cotton fabric using chitosan and banana floral stem extract. The dyeing method was validated and optimised using Response Surface Methodology (RSM) with Box-Behnken Design (BBD). Results show banana floral stems are an excellent natural dye source, enhancing cost-effectiveness, energy efficiency, and time savings. Optimal conditions were 554.12 W radiant power, 7.10 min radiation time, and 0.80% chitosan concentration.

Applying the optimisation tools to other botanical dye sources is recommended. Compared to conventional methods, microwave dyeing significantly reduces dyeing time. Chitosan, used as a bio-mordant, improves colour strength and colour fastness, thus reducing environmental impact. A combined chitosan-treated cotton and microwave dyeing technique shall pave the way for green dyeing of natural dyes on cotton fabrics.

ACKNOWLEDGMENT

The first author (H.A.M.) acknowledges the Indonesian Education Scholarship (BPI), Center for Higher Education Funding and Assessment (PPAPT) Ministry of Higher Education, Science, and Technology of Republic Indonesia, and Indonesia Endowment Fund for Education (LPDP) for providing funding support of this research with the Awardee BPI ID Number 202101120971.

STATEMENT AND DECLARATION

During the preparation of this manuscript, the first author utilised ChatGPT to conduct grammar and style checks, enhancing the clarity and readability of the text. After using this tool/service, the authors reviewed and edited the content as needed and took full responsibility for the content.

NOMENCLATURE

Symbols

a^*	colour space coordinate ranging from redness to greenness	
A	radiant power	[W]
Abs	absorbance	
b^*	colour space coordinate ranging from yellowness to blueness	
B	radiation time	[min]
C	chitosan concentration	[%]
ΔE	colour difference	
K/S	colour strength	
L^*	lightness	
p-value	probability value	
R^2	regression coefficient	
X_i and X_j	coded independent variables	
Y	response of lightness and colour difference	

Greek letters

β_0, β_i, β_{ii}, and β_{ij}	regression coefficients
ε	the model error

Subscripts and superscripts

ns	not significant

Abbreviations

ANOVA	Analysis of Variance
ATR-FTIR	Attenuated Total Reflectance-Fourier Transform Infrared
BBD	Box-Behnken Design
BFS	Banana Floral Stem
CCD	Central Composite Design
C.V.	Coefficient of Variation
DoE	Design of Experiments
RSM	Response Surface Methodology
SEM	Scanning Electron Microscopy
Sig.	Significance
Std. Dev.	Standard Deviation

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APPENDIX

This appendix provides additional data on the effect of chitosan and microwave-assisted dyeing treatment, as well as the optimisation of the dyeing process using Box-Behnken Design (BBD). It includes infrared spectra of untreated, chitosan-coated, and dyed cotton, alongside surface morphology images obtained through scanning electron microscopy (SEM). The appendix also presents the optimal dyeing conditions derived from BBD, showing the best parameters for microwave-assisted dyeing. Additionally, the colourfastness properties, including wash, light, and perspiration fastness, of the dyed cotton samples are summarised with detailed grading results.

Effect of chitosan and microwave-assisted dyeing treatment

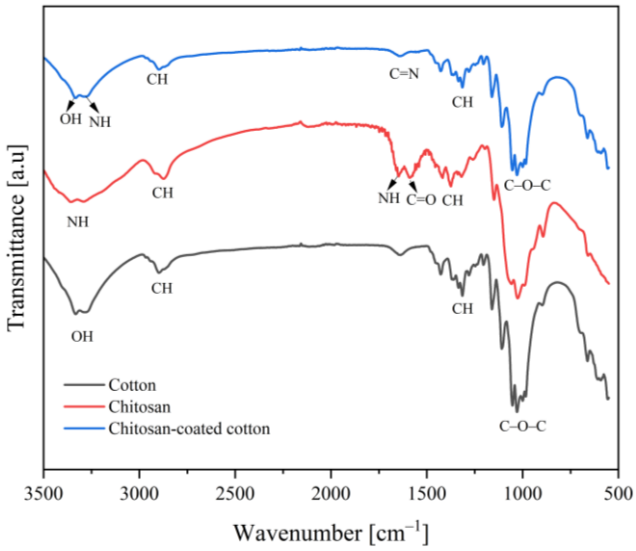


Figure A1. Infrared spectra of cotton (a) chitosan (b) and chitosan-coated cotton (c)

The distinctive peaks of chitosan were challenging to differentiate from those of cotton in the Infrared spectra of chitosan-coated cotton due to similarities in their chemical structures. The similarity between the chitosan backbone and cotton has been suggested as the basis due to their mutual affinity. Chitosan has a free amine group at the C_2 position of its glucosamine residue that is protonated in acidic conditions, resulting in a positively charged chitosan molecule [50]. In **Figure A1**, black line (a), the broad spectrum observed within the interval 3500–3000 cm^{-1} indicates the –OH stretching vibration of cotton, while the asymmetric C–H stretching is evident around 2900 cm^{-1}. There is a spectrum corresponding to the CH range of wagging at wavenumber 1314 cm^{-1}. The complex absorption observed around 1029 cm^{-1} is linked with the cotton's C–O–C stretching groups [9]. This indicates that cotton contains a cellulose functional group. In **Figure A1**, red line (b), the absorption band between 3500–3100 cm^{-1} represents NH_2 groups of chitosan, with amino groups (N–H) appearing between 1650–1580 cm^{-1}, which is a characteristic absorbance band of chitosan, depending on the groups it connects [51]. The C=O stretching vibration exhibits an absorption band around 1646 cm^{-1}. Furthermore, the spectrum depicted in **Figure A1**, blue line (c), reveals a broad spectrum at 3500–3100 cm^{-1} assignable to NH_2 and –OH groups, with a peak at 1641 cm^{-1} corresponding to forming a C=N group between the aldehydic group and chitosan. There are peaks at wave numbers 1161 cm $^{-1}$ and two peaks at wave numbers 1054 cm^{-1} and 1031 cm^{-1}, which are the peaks of the C–O–C stretching group and C–O stretching group, respectively. This suggests successful binding between chitosan and cotton, affirming the effective coating of the cotton [42].

Infrared spectra were obtained to observe the chemical properties. **Figure A2** shows the infrared spectra of dyed cotton using conventional and microwave-assisted methods. Microwave irradiation did not exert a notable impact on the chemical structure of cotton fibres [30]. Even after radiation exposure, the intensity of the functional peaks of –OH groups and bonds remained unchanged, suggesting that microwave irradiation remains unchanged in the chemical composition of the fabric. This underscores another noteworthy benefit of microwave radiation, as it preserves the fabric's chemical properties while offering energy, time, and cost savings [20]. The distinctive peak associated with hydrogen bonding within the cellulose structure of cotton fabric remains unchanged following microwave treatment lasting up to 4 min. Infrared spectral analysis conducted on treated and non-treated cotton fabric reveals that the peaks corresponding to –OH at 3300 cm^{-1}, CH at 1200 cm^{-1}, and C=O at 1075 cm^{-1} exhibit no alteration. The persistence of the characteristic hydroxyl linkage peak in the cellulosic fabric suggests that the chemical composition remains unaffected by microwave radiation, offering significant advantages within the textile processing sector [52].

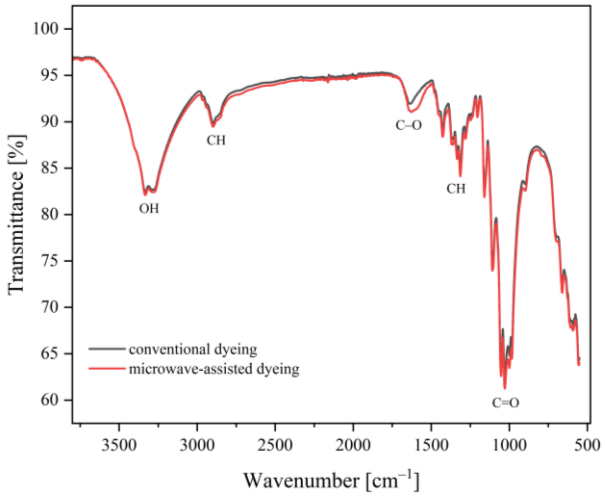
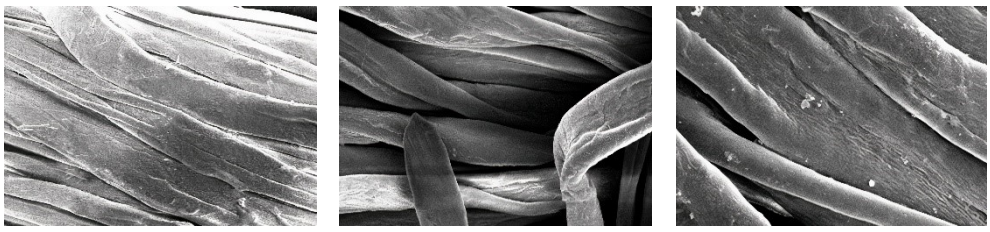


Figure A2. Infrared spectra of dyed cotton with different dyeing methods, conventional dyeing and microwave-assisted dyeing

The surface morphology of cotton fabrics was examined using Scanning Electron Microscopy (SEM). The sample was coated with a thin layer of gold (Au) to ensure proper conductivity and reduce charging effects during SEM imaging. This allowed for high-quality images with minimal distortion. **Figure A3** presents images of cotton fabrics in three conditions, including untreated, treated with chitosan, and treated with both chitosan and natural dye.



(a) (b) (c)

Figure A3. Scanning Electron Microscope images of undyed cotton (a), chitosan-coated cotton (b), and chitosan coated-dyed cotton (c)

Profiles of optimisation dyeing process use Box-Behnken Design

Figure A4 is a ramp function graph illustrating the main dependent variables (red dots) and the independent variables or their responses (blue dots). The dot's position height relative to the base is the criterion anticipated to be attained during the optimisation procedure. The peak response value is predicted to emerge through a positive ramp gradient (lightness and colour difference), whereas a flat ramp exhibits a uniform desired value (radiant power, radiation time, and chitosan concentration). A desirability achievement of 85.6% was attained under the specified criteria when the cotton dyeing process employed a microwave with a radiant power of 554.12 W, a radiation duration of 7.10 min, and a chitosan concentration of 0.80%, yielding the lowest lightness (62.04) and the highest colour difference (35.36).

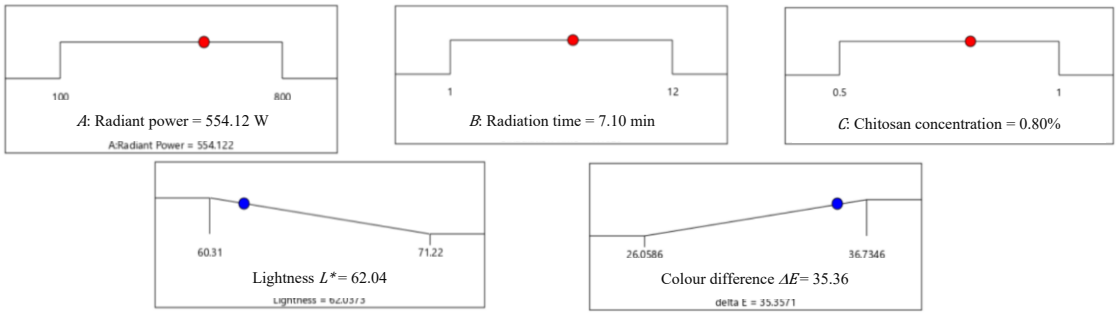


Figure A4. Desirability ramps for optimisation variable – the optimal values of the design variables (red points – optimal dependent variables values; blue points – optimal responses the predicted values)

Colourfastness properties of dyed cotton

Table A1. The colourfastness properties to wash (1–2 very poor to poor, 3–4 fair to good, 4 good)

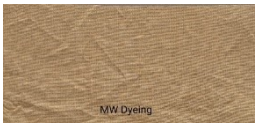
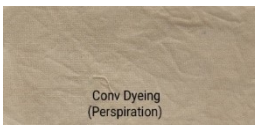
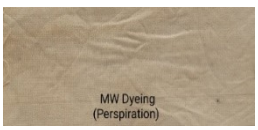
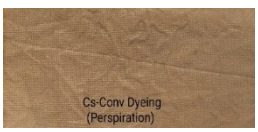
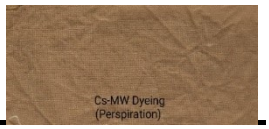
Samples	Greyscale	Before	After
Conventional dyeing	1–2		
Microwave dyeing	1–2		
Conventional dyeing with chitosan-coated	3–4		
Microwave dyeing with chitosan-coated	4		

Table A2. The colourfastness properties to light (4–5 good to excellent)

Samples	Greyscale	Before	After
Conventional dyeing	4–5		
Microwave dyeing	4–5		
Conventional dyeing with chitosan-coated	4–5		
Microwave dyeing with chitosan-coated	4–5		

Table A3. The colourfastness properties of perspiration (2 poor, 4–5 good to excellent)

Samples	Greyscale	Before	After
Conventional dyeing	2		
Microwave dyeing	2		
Conventional dyeing with chitosan-coated	4–5		
Microwave dyeing with chitosan-coated	4–5		

Paper submitted: 29.11.2024
Paper revised: 25.03.2025
Paper accepted: 26.03.2025