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Optimization of indigo dyeing on modal fabrics using thiourea dioxide as an environmentally friendly reducing agent

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Abstract

This investigation examines the potential of thiourea dioxide as an environmentally reducing agent for dyeing of natural indigo on modal fabrics. Although traditional sodium dithionite is effective, its sulfur by-products generate environmental concerns. Thiourea dioxide, which provides a more environmentally friendly alternative, was studied under various dyeing conditions. In this study, six dyeing parameters, including thiourea dioxide and sodium hydroxide concentrations, reduction temperature, reduction duration, dyeing time, and reduction-oxidation cycles, were optimized to enhance color strength. Results indicated that the most favorable reduction conditions were achieved when 80 $g \cdot L^{-1}$ of thiourea dioxide and 2 $g \cdot L^{-1}$ of sodium hydroxide were heated to 50°C for 30 min and at the dyeing duration of 30 min. Moreover, the repeated reduction and oxidation cycles continuously improve color strength of the dyed fabric. The indigo dyed fabrics had excellent wash fastness, good light fastness, and slightly lower rub fastness.

1. Introduction

Nowadays, sustainability is the most prioritized concept in many industries, including textiles and clothing [1,2]. With its complex supply chain, the textile industry, particularly the dyeing sector, contributes significantly to pollution, driving interest in natural dyes. This interest was motivated by concerns about water pollution, sustainably sourced raw materials, dye biodegradability, and ecological safety [3]. A comprehensive study of natural dyes over the past decade has confirmed this trend. Major brands, such as Levi's, now support sustainable fashion by incorporating plant-based indigo dyes into their collections.

Natural dyes come from sustainable sources such as plants, minerals, and insects. Among these, natural indigo (C.I. 75780, C.I. Natural Blue 1) is one of the most historically and culturally significant blue pigments, widely used for dyeing cellulose fibers, particularly in denim. *Indigofera* species, particularly *Indigofera tinctoria*, is the primary source of this dye. The indigo dye is not directly present in the leaves but is synthesized from glucoside indican, a colorless and water-soluble compound that constitutes 3% to 4% of the plant's weight. Indican undergoes a natural fermentation process where it is first hydrolyzed to indoxyl by the action of glucosidase enzymes found within the leaves. Subsequently, indoxyl dimerizes into indigo upon exposure to atmospheric oxygen [4,5]. Figure 1 illustrates the enzymatic transformation.

As a vat dye, dyeing with indigo is therefore based on a reduction reaction to convert water-insoluble indigo (non–fiber-affinity form, which cannot interact with the fibers) to a water-soluble leuco compound (fiber-affinity form, which can absorb into the fibers). After an adequate dyeing time, the fabric undergoes air-oxidation, where the dyes are converted back to original insoluble form. Figure 2 depicts indigo dyeing reactions in an alkaline condition [5,6].

Sodium dithionite ($Na_2S_2O_4$) is most wildly used as a reducing agent for its strong capabilities and low cost. However, the use of sodium dithionite poses environmental challenges due to the formation of sulfites, sulfates and sulfide within effluents [7,8]. Additionally, the high level of sulfates can accelerate the corrosion of effluent drainage systems.

Due to the factors mentioned above, the current research emphasizes the development of environmentally friendly technologies for indigo reduction as an alternative to sodium dithionite. They include α-hydroxy ketones [9,10], sodium borohydride [11], biological reduction [12,13], glucose-based reducing agents [14,15], electrochemical reduction [15-18], plant extract [19,20], and catalytic hydrogenation. However, formamidine sulfinic acid, also known as thiourea dioxide, is a potent reducing agent that offers a safer and more environmentally friendly alternative to sodium dithionite. Because thiourea dioxide has a lower relative molecular weight than sodium dithionite, the decreased bath

effluent contains less waste products based on sulfur [21]. Figure 3 illustrates the formation of the active reducing form during the conversion of thiourea dioxide under alkaline conditions [21]. Initially, thiourea dioxide converts into formamidinesulfinic acid. It then decomposes into two key components: urea and sodium hydrogen sulfoxylate ion, which act as the active reducing species.

Cotton, widely valued for its versatility, has significant environmental and health impacts due to extensive use of fertilizers and persistent chemicals such as pesticides and herbicides. With growing consumer awareness about harmful substances in textiles, the demand for sustainable fibers has increased. Modal, a second-generation regenerated cellulose fiber created by Lenzing AG, provides a more environmentally friendly option. Made from renewable forest sources and produced through sustainable viscous-spinning technology, modal fiber is known for its strength, stability, and softness, making it ideal for high-quality clothing, intimate apparel, and home textiles [22].

Although the application of thiourea dioxide in textile has been reported, its primary application is in the fields of textile printing particularly as an effective discharge agent [23-25]. Broader studies on thiourea dioxide as a reductive compound for indigo are still limited. Mongkholrattanasit *et al.* [26] investigated its application in cotton printing with natural indigo, utilizing a thickening agent from wild taro corms and thiourea dioxide, resulting in a successful dark-blue print. Chollakup *et al.* [27] designed fabric structures for home textiles dyed with natural indigo and thiourea dioxide, with a primary focus on product design rather than reduction efficacy.

This study aims to further explore the dyeing efficiency of natural indigo on modal fabrics, substituting thiourea dioxide for sodium dithionite as a more sustainable reducing agent. We systematically optimized the significant dyeing parameters that affect the color depth, such as thiourea dioxide concentration, alkali concentration, the reduction conditions, the dyeing time, and the exhaustion-oxidation cycles. Furthermore, the investigation was extended to assess the colorfastness characteristics of the dyed fabrics, including their resistance to washing, crocking (rubbing), and exposure to light, to ensure their color durability during usage.

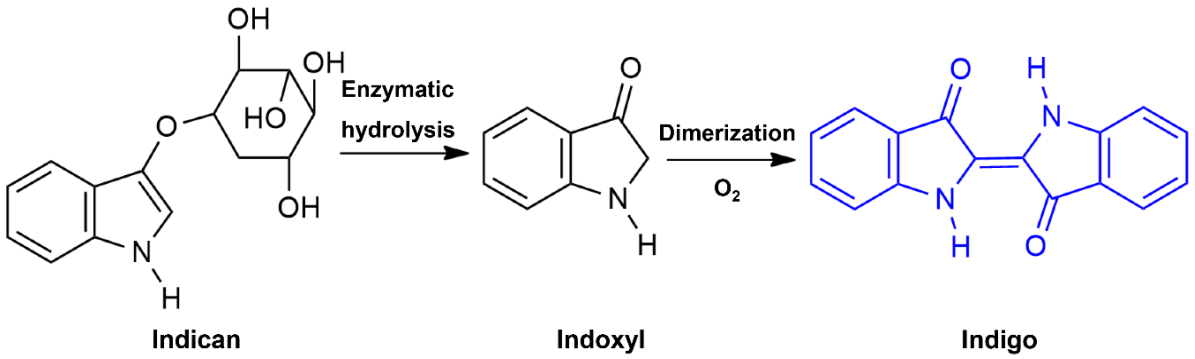

Figure 1. Enzymatic transformation of natural indigo from indicant [4,5].

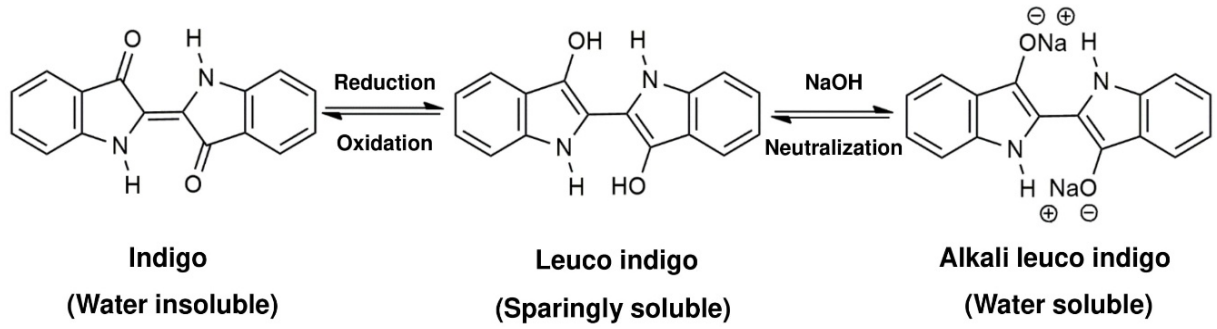

Figure 2. The indigo dyeing reactions [5,6].

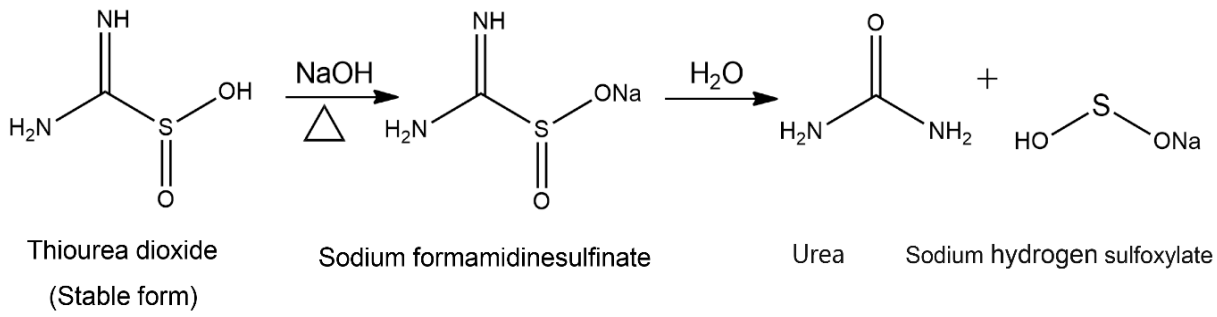

Figure 3. Conversion of thiourea dioxide into the active reducing agent [21].

2. Experimental

2.1 Materials and chemicals

Natural indigo paste was obtained from Nakhon Pathom province, Thailand. Thiourea dioxide was obtained from Star Tech Chemical Industrial Co., Ltd., Thailand. Sodium hydroxide (NaOH), sodium dithionite ($Na_2S_2O_4$), and hydrogen peroxide (H_2O_2, 30% w/w) of laboratory reagent grade were purchased from Ajax Finechem Pty Ltd., Australia. Soaping agent (non-ionic) was obtained from Boonthawee Chemephan Co., Ltd., Thailand. Plain weave modal with a mass per unit area of 178 $g{\cdot}m^{-2}$, warp density of 96 ends/inch, and weft densities of 97 picks/inch was sourced from 863 Textile, Bangkok, Thailand.

2.2 Natural indigo dyeing process

The natural indigo dyebaths were prepared following the dyeing parameters shown in Table 1, with the dyeing profile depicted in Figure 4. The dyeing parameters were preliminarily studied for determining the appropriate ranges for the process variables. The dyeing started with the preparation of a 100 mL dyebath containing natural indigo, a reducing agent, and alkali (sodium hydroxide). The dyebath was then heated to the desired reduction conditions. Following the reduction stage, modal fabric was then dyed for a specific duration. After that the fabric was then oxidized with hydrogen peroxide (H_2O_2; 10% w/v) for 5 min and soaped (2 $g{\cdot}L^{-1}$) at 50°C for 5 min.

In the study of the dyeing-oxidation cycles, after the first reduction-oxidation step (referred to as the first cycle), the fabric was immersed back in the reduced bath for 10 min, followed by oxidation for 5 min. This dyeing and oxidation process were repeated until the desired number of cycles was achieved.

2.3 Color determination

A spectrophotometer (GretagMacbeth LLC, Switzerland) was used to measure the color parameters of the dyed samples. These included the CIE color coordinates (L^*, a^*, b^*) and the color strength (K/S) at the wavelength of maximum absorption. The value of K/S was determined based on the Kubelka-Munk equation as shown Equation (1). The spectrophotometer settings included illuminant D65, a 10° standard observer, and the inclusion of both specular and UV light. A total of three measurements per sample were conducted, with average values recorded.

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

Where, R = reflectance of the samples at the maximum absorption wavelength (λ_{max})
K = absorption coefficient
S = scattering coefficient

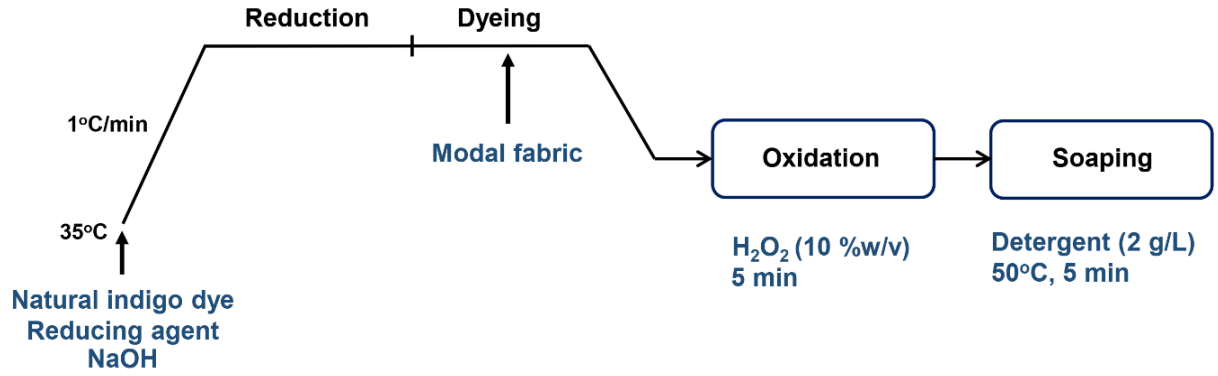

Figure 4. Dyeing procedure of modal fabric with natural indigo.

Table 1. The dyeing parameters of natural indigo dye on modal fabrics.

Dyeing variables	Values
1. Thiourea dioxide ($g{\cdot}L^{-1}$) (indigo 50 $g{\cdot}L^{-1}$, NaOH 2 $g{\cdot}L^{-1}$, reduction condition: 50°C, 30 min, dyeing time 30 min)	20, 40, 60, 80, 100
2. Sodium hydroxide (($g{\cdot}L^{-1}$) (indigo 50 $g{\cdot}L^{-1}$, thiourea dioxide 80 $g{\cdot}L^{-1}$, reduction condition: 50°C, 30 min, dyeing time 30 min)	0.25, 0.50, 1, 2, 3
3. Reduction temperature (°C) (indigo 50 $g{\cdot}L^{-1}$, thiourea dioxide 80 $g{\cdot}L^{-1}$, NaOH 2 $g{\cdot}L^{-1}$, reduction time 30 min, dyeing time 30 min)	40, 50, 60, 70, 80
4. Reduction time (min) (indigo 50 $g{\cdot}L^{-1}$, thiourea dioxide 80 $g{\cdot}L^{-1}$, NaOH 2 $g{\cdot}L^{-1}$, reduction temperature 50°C, dyeing time 30 min)	5, 10, 20, 30, 40
5. Dyeing time (min) (indigo 50 $g{\cdot}L^{-1}$, thiourea dioxide 80 $g{\cdot}L^{-1}$, NaOH 2 $g{\cdot}L^{-1}$, reduction condition: 50°C, 30 min)	10, 20, 30, 40, 60
6. Dyeing-oxidation (cycles) (indigo 50 $g{\cdot}L^{-1}$, thiourea dioxide 80 $g{\cdot}L^{-1}$, NaOH 2 $g{\cdot}L^{-1}$, reduction condition: 50°C, 30 min, dyeing time 30 min)	1, 2, 3, 4, 5, 6, 7, 8
7. Indigo concentrations ($g{\cdot}L^{-1}$) (thiourea dioxide 80 $g{\cdot}L^{-1}$, NaOH 2 $g{\cdot}L^{-1}$, reduction condition: 50°C, 30 min, dyeing time 30 min)	10, 50, 100, 200, 300

2.4 Statistical analysis

The *K/S* data were analyzed using SPSS software (Version 27). The statistical significance of the dyeing variables was assessed using one-way analysis of variance (ANOVA) to determine the probability values (p-values).

2.5 Color fastness tests

The color fastness properties of the dyed samples were evaluated, including wash fastness (ISO 105-C06 Test No.A1S:2010) using a Gyrowash (James H. Heal & Co. Ltd., England), rub fastness (ISO 105-X12:2016) using a crockmaster (James H. Heal & Co. Ltd., England), and light fastness (ISO 105-B02:2014) using a Suntest CPS xenon arc lamp tester (Atlas, USA). Gray scale (grades 1 to 5) was used to assess color changes and staining for wash and rub fastness, while the blue wool scale (grades 1 to 8) was used to evaluate light fastness. All assessments were conducted under a D65 color assessment cabinet (VeriVide, UK).

2.6 FTIR spectroscopy

A Nicolet iS50 FTIR spectrometer (Thermo Scientific, USA) was used to record FTIR spectra in attenuated total reflection (ATR) mode. For optimal signal acquisition, the samples were placed directly on the diamond ATR crystal, ensuring firm and uniform contact with the crystal surface. The transmittance spectra were collected within the wavenumber range of 400 cm^{-1} to 4000 cm^{-1}, with a resolution of 4 cm^{-1}. Three spectral scans were conducted for each sample.

2.7 Scanning electron microscopy (SEM)

The JEOL JSM-5410LV scanning electron microscope (SEM) was employed to examine the surface morphology of the samples. Before testing, the samples were carefully placed on brass stubs and coated with fine layer of gold to improve conductivity. The SEM analysis was conducted at a working distance of 20 mm and an accelerating voltage of 10 kV. Observations were made at magnifications of 100x and 1000x for both low and high magnification levels.

3. Results and discussion

3.1 Effect of thiourea dioxide

Modal was dyed with natural indigo using thiourea dioxide concentrations ranging from 20 $g{\cdot}L^{-1}$ to 100 $g{\cdot}L^{-1}$. Figure 5(a) illustrates that thiourea dioxide was an essential factor affecting the color intensity. The color strength sharply rose with thiourea dioxide concentrations, achieving its maximum at 80 $g{\cdot}L^{-1}$. The dyed fabric turned darker blue as thiourea dioxide concentration increased. This suggests that thiourea dioxide effectively acts as a reducing agent, converting insoluble indigo into soluble forms that are substantive to fiber. In the presence of alkali, thiourea dioxide decomposes to yield sodium hydrogen sulfoxylate ion, which serves as a powerful reducing agent [21]. However, beyond 80 $g{\cdot}L^{-1}$, the color strength decreased slightly due to over-reduction of the indigo dye, which reduced the dye's affinity for the fiber. Therefore, the optimum thiourea dioxide concentration of 80 $g{\cdot}L^{-1}$ was used in subsequent experiments.

3.2 Effect of sodium hydroxide

The effect of sodium hydroxide (NaOH) concentration on the color strength of the dyed fabric was the second factor evaluated. Figure 5(b) shows that as the NaOH concentration increased, the color strength initially rose and reached an optimum value at 2 $g{\cdot}L^{-1}$ of NaOH (pH = 11.5). This suggests that the alkali concentration effectively generates the fiber-affinity mono-sodium phenolate form of reduced indigo. The color strength slightly decreased above 2 $g{\cdot}L^{-1}$ of NaOH, indicating that higher alkalinity might cause the formation of di-sodium phenolate forms, which had less fiber affinity and lower color strength. The dyed samples visually confirmed these observations, showing darker blue shades at the optimal NaOH concentration. The role of alkali is important in the indigo reduction process; in which it affects the solubilization of indigo [28]. Indigo exists in four distinct forms depending on the pH of the dyebath: (1) nonionic form (very low alkali condition), (2) reduced but non-ionic form (moderate alkali; pH less than 10.5), (3) mono-sodium phenolate form (pH 10.5 to 11.5, with fiber affinity), and (4) di-sodium phenolate form (pH greater than 11.5). Additionally, alkali is essential in the dyeing of indigo in order to convert thiourea dioxide to sodium hydrogen sulfoxylate ion, an active reducing species.

3.3 Effect of reduction temperature

Figure 5(c) shows the influence of the reduction temperature on indigo dyeing. The results show that 50°C gave the maximum color strength and produced the deepest blue of the dyed fabric. This is due to the characteristics of indigo, which belong to the IK subclass of vat dyes. Dyes in the IK subclass typically require relatively low reduction and dyeing temperatures, as well as low concentrations of alkali and electrolyte. The small molecular structure of indigo makes it easy to reduce, and therefore effective reduction typically occurs at lower ranging from 35°C to 50°C. Beyond these optimum ranges, the color strength drastically decreased, as shown by the fabric swatches turning colorless. This decline was caused by over-reduction of indigo at high temperature [29].

3.4 Effect of reduction time

According to Figure 5(d), the reduction reaction of natural indigo occurred rapidly within 10 min. This was due to the high efficiency of thiourea dioxide as a reducing agent. Although differences across 10 min to 30 min may be statistically minimal, a dyeing time of 30 min was selected to ensure complete reduction reaction and enhance dye uniformity.

3.5 Effect of dyeing time

Figure 5(e) displays the effect of the dyeing time on the color strength of the natural indigo dyed fabric. The color strength increased

rapidly within the first 20 min of dyeing application and reached its maximum depth of shade within 30 min. Although the *K/S* values at 20 min to 30 min may not be statistically significant, 30 min was chosen as the optimal dyeing time to achieve level dyeing. Since the reduced form of indigo dye has low fiber substantivity due to its small molecular structure, extending the dyeing time beyond 30 min does not further improve color depth.

3.6 Effect of dyeing-oxidation cycles

To enhance the dye uptake of natural indigo, the fabric underwent multiple dyeing cycles in a reduced indigo dyebath, followed by oxidation for up to 8 cycles. Figure 5(f) shows that the numbers of dyeing-oxidation cycles significantly impacted the dye uptake of the indigo-dyed samples. The results demonstrate the effective build-up properties of indigo dyeing using this technique.

Statistical analysis was performed using ANOVA to identify significant variables influencing the optimization of indigo dyeing on modal fabrics. Variables with p-values below 0.05 were considered statistically significant. The analysis confirmed that three primary factors had a substantial impact on dyeing performance, particularly in terms of color strength: the concentration of the reducing agent, the reduction temperature, and the number of dyeing-oxidation cycles.

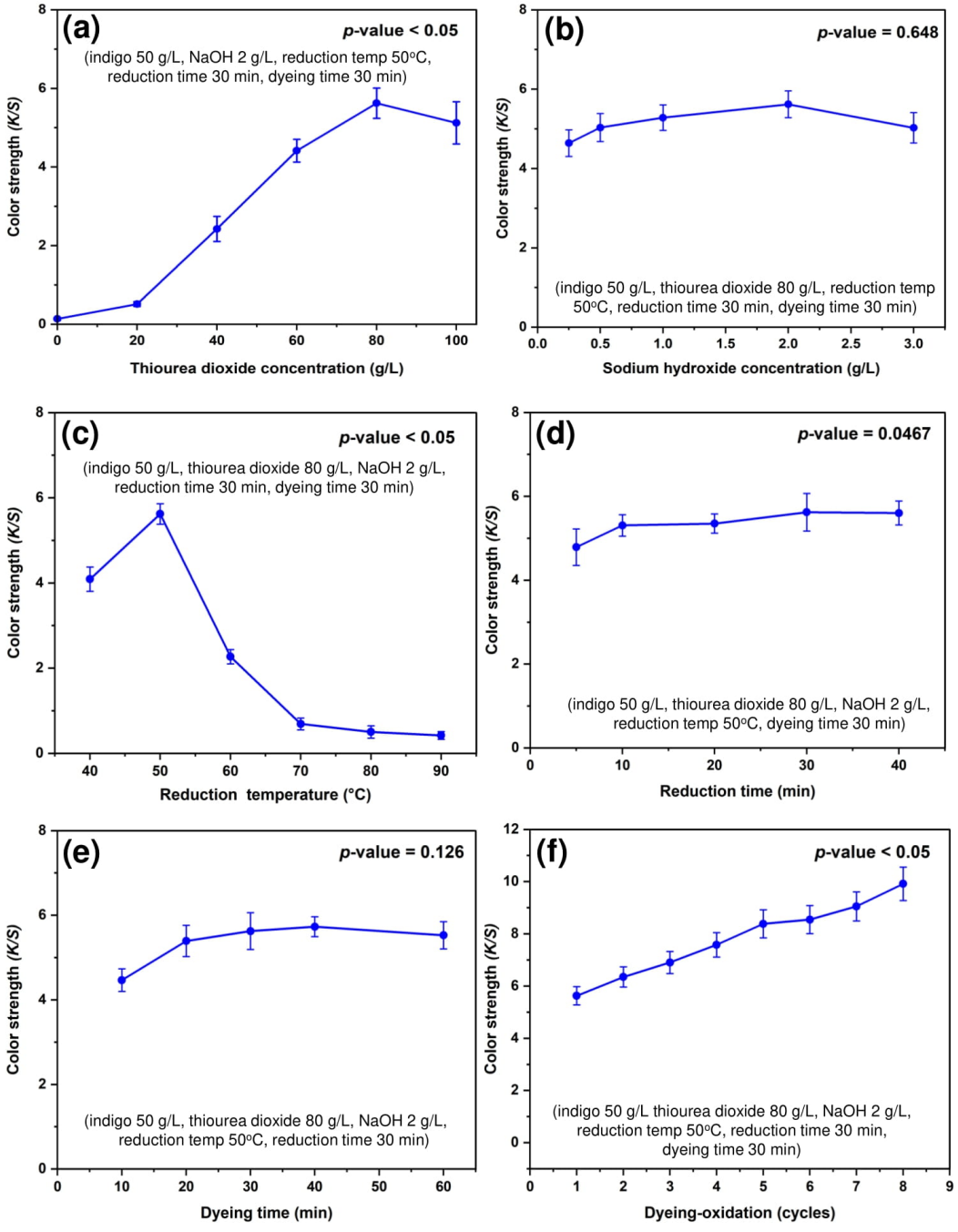

Figure 5. Effect of dyeing parameters on natural indigo on modal (a) effect of thiourea dioxide, (b) effect of alkali, (c) effect of reduction temperature, (d) effect of reduction time, (e) effect of dyeing time, and (f) effect of dyeing-oxidation cycles.

3.7 Colorimetric properties and SEM analysis of natural indigo dyed fabrics

After optimizing the conditions for natural indigo dyeing (thiourea dioxide at 80 g·L^{-1}, NaOH at 2 g·L^{-1}, reduction temperature of 50°C, reduction time of 30 min, and dyeing time of 30 min), the colorimetric measurement of dyed samples at various natural indigo concentrations was further investigated. Figure 6 shows the *K/S* spectra of dyed fabrics at different dye concentrations across the visible wavelength region. All dyed samples exhibited the maximum absorption wavelength (λ_{max}) at 650 nm, which indicates the blue color obtained after dyeing. The degree of color strength steadily increased across all wavelengths as the dye concentration increased. This effect is anticipated due to a greater number of indigo dye molecules at increasing dye concentrations. Table 2 presents the color measurement of the dyed samples using CIE coordinates (*L**, *a**, *b**), whereas Figure 7 illustrates the images of the indigo-dyed fabric. The results demonstrate that increasing the concentration of natural indigo dye produces darker shades, as evidenced by lower *L** and higher *K/S* values. Additionally, increasing the dye concentration from 100 g·L^{-1} to 300 g·L^{-1} led to *b** values that were slightly less negative, indicating a shift toward a lighter or less intense blue hue.

Table 3 summarizes the dyeing comparison between the traditional reducing agent (sodium dithionite) and thiourea dioxide. According to the colorimetric measurements, dyeing with thiourea dioxide produced better dyeing performance. It had a darker shade (lower *L**), a redder (less negative *a**), a bluer (more negative *b**), and a higher *K/S* value than sodium dithionite. The oxidation-reduction potential (ORP) of thiourea dioxide was more negative, indicating that it was a stronger reducing agent than sodium dithionite.

Figure 8 compares the morphological analysis of undyed and dyed modal samples using SEM at magnifications 100x and 1000x. The undyed sample displayed a smooth, ribbon-like surface. In contrast, the dyed sample showed deposits of indigo dye on the fiber surface, resulting from the characteristic reduction–oxidation process of vat dyeing, during which the leuco form reverts to its insoluble indigo state.

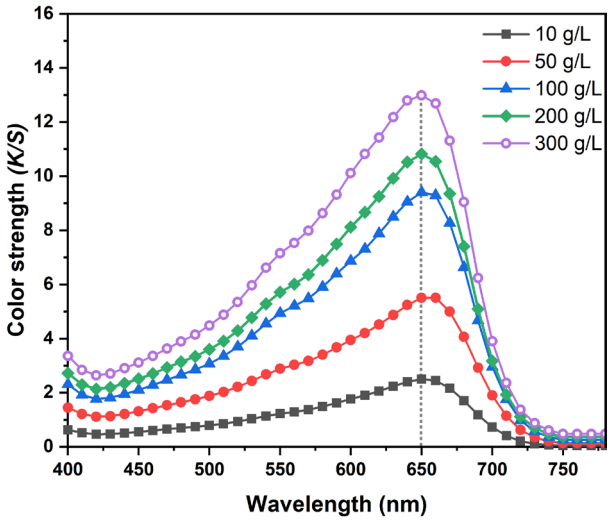

Figure 6. *K/S* spectra as a function of wavelength at varying indigo concentrations (thiourea dioxide 80 g/L, NaOH 2 g/L, reduction temperature 50°C, reduction time 30 min, and dyeing time 30 min).

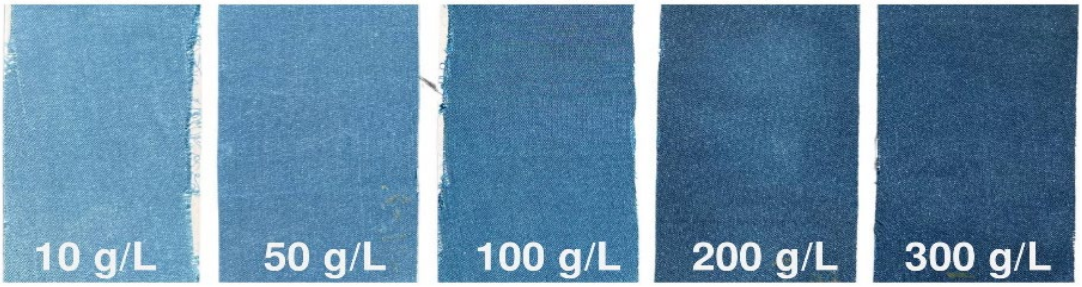

Figure 7. Images of natural indigo dyed modal fabrics at varying indigo concentrations.

Table 2. Colorimetric measurements of modal fabric dyed with natural indigo at different dye concentrations.

Natural indigo concentrations [g·L^{-1}]	*L**	*a**	*b**	*K/S*
10	56.36	-4.87	-17.43	2.50
50	44.25	-3.17	-18.19	5.62
100	37.62	-2.27	-18.62	9.40
200	33.99	-2.68	-18.27	10.81
300	30.95	-2.18	-17.97	12.98

(thiourea dioxide = 80 g·L^{-1}, NaOH = 2 g·L^{-1}, reduction condition: 50°C, 30 min, dyeing time = 30 min)

Table 3. CIE(*L*a*b**) and *K/S* comparison of natural indigo dyed fabrics treated with sodium dithionite and thiourea dioxide.

Reducing agent	ORP [mV]	*L**	*a**	*b**	*K/S*
Sodium dithionite	-861	53.25	-4.70	-19.80	3.30
Thiourea dioxide	-932	44.25	-3.17	-18.19	5.62

(indigo 50 g·L^{-1}, reducing agents = 80 g·L^{-1}, NaOH = 2 g·L^{-1}, reduction condition: 50°C, 30 min, dyeing time = 30 min)

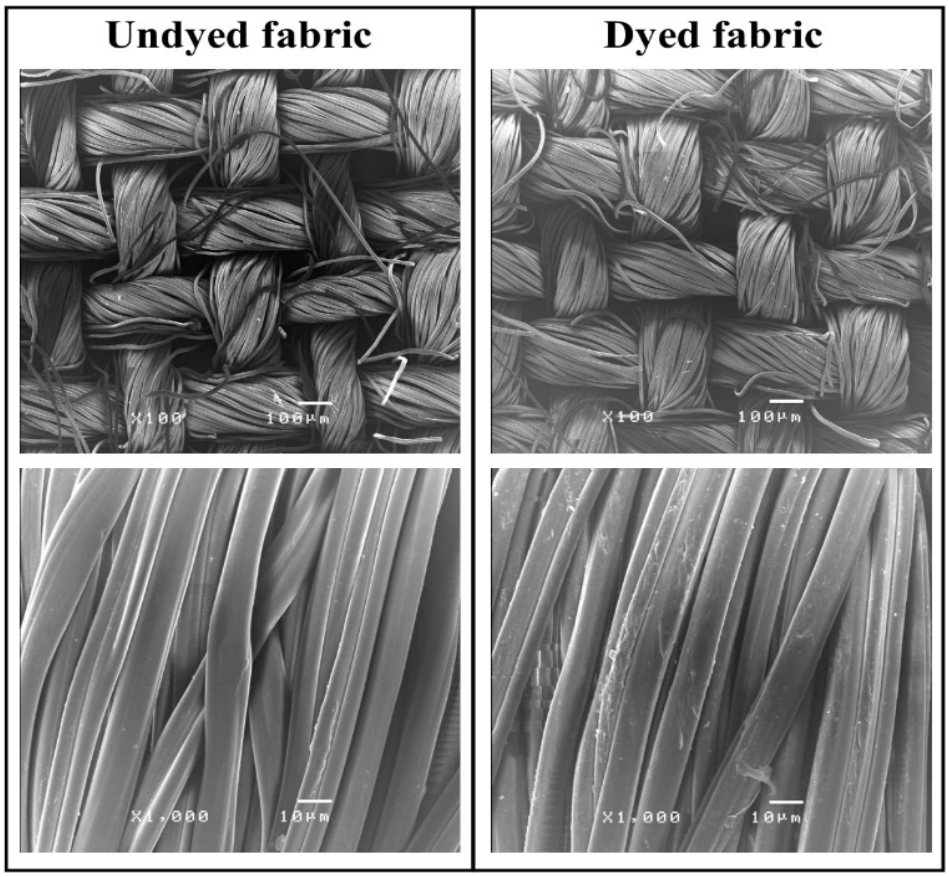

Figure 8. SEM images of undyed and indigo dyed fabrics at dye concentration of 300 g·L^{-1}.

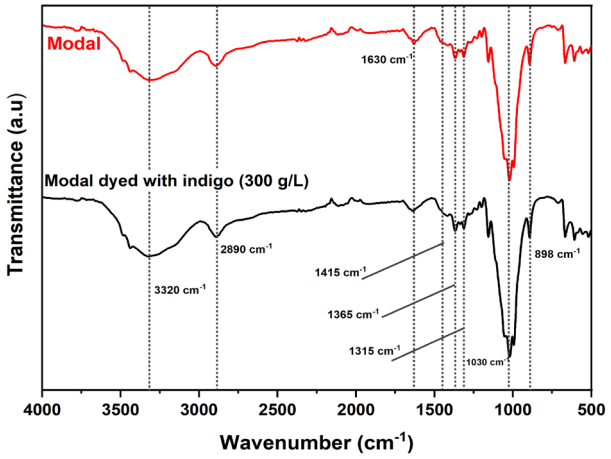

Figure 9. FTIR spectra of undyed and indigo dyed modal fabric.

3.8 FTIR analysis

FTIR analysis was conducted to examine the undyed and indigo-dyed modal fabrics, as shown in Figure 9. Both samples showed the distinctive peaks associated with cellulose. The broad peak around 3320 cm^{-1} is attributed to –OH stretching vibrations, indicative of the hydroxyl groups in cellulose. Additionally, the peak at approximately 2890 cm^{-1} represents the C–H stretching vibrations associated with CH_2 groups in cellulose. The characteristic peaks around 1160 cm^{-1} and 1050 cm^{-1} are associated with C–O stretching vibrations in the cellulose backbone, further confirming the presence of cellulose in both samples. However, there were no important differences seen between the samples that had not been dyed and those that had been dyed with indigo. This is most likely because the strong cellulose absorption peaks were stronger than the signals from the dye molecules.

3.9 Fastness properties

The wash fastness of dyed samples was assessed at three different indigo concentrations, corresponding to pale (50 g·L^{-1}), medium (100 g·L^{-1}), and dark shades (300 g·L^{-1}). Table 4 shows that the wash fastness ratings, based on color change and color staining, ranged from good to excellent (rated 4 to 5) across all three samples. The formation of the insoluble original form of indigo during oxidation, which leads to the trapping of insoluble compounds within the fiber, accounted for the high color fastness after washing. The color fastness properties in terms of rubbing and light are shown in Table 5. As the concentration of indigo dye increased, more dye molecules became susceptible to color loss during the rubbing test, resulting in a lower rating. When performed in wet conditions, the color loss was more noticeable. Another factor contributing to the low rub fastness rating is the dyeing characteristic of indigo, known as ring dyeing effect, where majority of the dyes are located at the fiber surface. This could facilitate dye loss. However, the light fastness was superior in samples dyed with a higher indigo concentration. This was due to the aggregations of indigo dye molecules, enhancing the light resistance properties.

Table 4. Color fastness of natural indigo-dyed fabrics to washing.

	Natural indigo concentration [g·L^{-1}]		
Color fastness to washing	50	100	300
Color change	4-5	4-5	4
Color staining			
Acetate	4-5	4-5	4-5
Cotton	4-5	4-5	4-5
Nylon	4-5	4-5	4-5
Polyester	4-5	4-5	4-5
Acrylic	4-5	4-5	4-5
Wool	4-5	4-5	4-5

Table 5. Color fastness of natural indigo-dyed fabrics to rubbing and light.

	Natural indigo concentration [g·L^{-1}]		
Color fastness	50	100	300
Rubbing (dry)			
Warp	4-5	4	3-5
Weft	4-5	4	3-5
Rubbing (wet)			
Warp	4	3-5	2-5
Weft	4	3-5	2-5
Light	3	3-5	4

4. Conclusion

The study effectively optimized the indigo dyeing process on modal fabrics by utilizing thiourea dioxide as a more environmentally friendly reducing agent in place of sodium dithionite. Through systematic optimization of key dyeing parameters, including thiourea dioxide and sodium hydroxide concentrations, reduction temperatures, and dyeing times, the research identified optimal conditions that yielded high color strength and excellent colorfastness properties. The findings revealed that thiourea dioxide, at an optimal concentration of 80 g·L^{-1}, effectively reduced indigo, producing vibrant and durable blue shades. Additionally, the study showed that sodium hydroxide concentration of 2 g·L^{-1} (pH 11.5) was most effective in facilitating the reduction process. The reduction conditions at 50°C for 30 min and a dyeing time of 30 min provided the best results in this research study. However, for large-scale dyeing, it is advisable to conduct the process at room temperature to avoid over-reduction, which may lead to decreased color strength and uneven dyeing. The repeated exhaustion-oxidation cycles further enhanced dye uptake. The overall results support the use of thiourea dioxide in textile dyeing and suggest further exploration of its application across various fabrics and dyes to validate its versatility and efficacy.

Acknowledgments

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