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Nano-Mordant Enhanced Natural Dyeing of Cotton Fabric Using Indigo, Marigold, and Pomegranate Rind

Kavitha Sundararajan

Department of Apparel and Fashion Design, Park College of Engineering & Technology, Coimbatore, Tamil Nadu, India

Abstract

The global textile dyeing industry releases approximately 200,000 tonnes of synthetic dyes into waterways annually, representing a major source of persistent organic pollutant contamination with documented endocrine-disrupting, carcinogenic, and mutagenic effects on aquatic ecosystems and human health. The transition from synthetic reactive dyeing to natural dyeing processes using plant-derived colorants offers environmental sustainability benefits but faces persistent technical challenges: natural dyes' inherent low substantivity to cotton fibres requires mordanting to achieve commercially acceptable colour strength and fastness properties, and the durability of natural dyed textiles has historically been inferior to synthetic dyed equivalents.

This study investigates the enhancement of natural dyeing performance on pre-scoured mercerised cotton fabric using nano-mordants — nano-titanium dioxide (nano-TiO_2) and nano-zinc oxide (nano-ZnO) — relative to conventional mordants (alum, iron, tannin) across three natural dye sources (indigo from Indigofera tinctoria, marigold from Tagetes erecta, and pomegranate rind). Box-Behnken Response Surface Methodology (RSM) optimises four process variables — pH, temperature, time, and material:liquor ratio — to maximise colour strength (K/S value). Multifunctional performance evaluation includes wash, light, and rubbing fastness (ISO 105 series), and antimicrobial activity (AATCC 100) against Staphylococcus aureus and Escherichia coli. Life cycle assessment (LCA) following ISO 14040/44 quantifies and compares GWP, cumulative energy demand (CED), and freshwater eutrophication potential against standard reactive synthetic dyeing. The nano-TiO_2 + tannin combined mordanting strategy achieves K/S of 19.1 (pomegranate) — exceeding industry minimum of 12 — with wash fastness 4-5, light fastness 4, and >99.1% antimicrobial reduction, while reducing GWP by 67.5% relative to reactive synthetic dyeing.

Keywords: *natural dyeing, nano-mordant, nano-TiO_2, nano-ZnO, cotton, indigo, marigold, pomegranate rind, response surface methodology, colour strength, fastness, antimicrobial, life cycle assessment, sustainable textile, green chemistry*

1. Introduction

India's textile and apparel sector — valued at approximately USD 153 billion and employing over 45 million people — is simultaneously one of the nation's most economically significant industries and one of its most environmentally burdened. The Tirupur cluster in Tamil Nadu alone processes approximately 90,000 tonnes of knitted fabric annually using synthetic reactive and acid dyes, generating approximately 100 million litres of effluent daily that has been implicated in widespread agricultural land degradation and groundwater contamination in surrounding communities (Rajesh & Senthilkumar, 2019). Growing regulatory pressure under Environment Protection Act 1986 standards, NGT directives, and the EU's textile sustainability regulations creating export market requirements motivate the development of technically viable natural dye processes for mainstream commercial adoption.

Natural dyes offer superior environmental credentials: they are biodegradable, derived from renewable agricultural sources, exhibit low systemic toxicity, and their production process — primarily extraction by aqueous solvents — generates orders of magnitude less hazardous waste than synthetic dye synthesis. However, their commercial adoption has been limited by insufficient colour strength on cellulosic fibres without mordanting, inadequate wash and light fastness in conventionally mordanted forms, and limited scalability of traditional mordanting processes.

Nano-metal oxide mordants represent a promising convergence of nanotechnology and green chemistry: nano-TiO_2 and nano-ZnO provide large specific surface area for dye-fibre bridging, photocatalytic self-cleaning properties in TiO_2, and intrinsic antimicrobial activity in ZnO through reactive oxygen species generation — potentially enabling multifunctional textiles that combine natural colouration with value-added functional properties. This study provides the most systematic evaluation of nano-mordant enhanced natural dyeing in terms of process optimisation, multifunctional performance characterisation, and environmental LCA to date.

2. Literature Review

2.1 Natural Dyes on Cellulosic Fibres

Natural dyes' low substantivity to cotton arises from the paucity of interaction sites between polar hydroxyl groups of cellulose and the relatively large, complex molecular structures of natural dye molecules. Mordanting bridges this gap by depositing metal complexes on fibre surfaces that form coordination bonds with both fibre hydroxyl groups and dye chromophore heteroatoms (typically oxygen and nitrogen). Aluminium sulphate (alum) produces bright, clean colours through Al^{3+} coordination; ferrous sulphate (iron) saddens colours through Fe^{2+} forming dark coordination complexes; and tannin acts as an organic mordant forming hydrogen bonds with cellulose and providing multiple phenolic sites for metal and dye bonding.

2.2 Nano-Metal Oxide Mordanting

The application of nano-metal oxides as mordanting agents or co-mordants in natural dyeing has been investigated since the early 2010s. Shahid et al. (2012) demonstrated that nano-TiO_2 pretreatment followed by madder (Rubia tinctorum) dyeing produced significantly higher K/S values and improved wash fastness ratings compared to alum mordanting on wool. The nano-mordant's effectiveness is attributed to its high surface-area-to-volume ratio (specific surface area typically 50–250 m^2/g for 10-25 nm particles) providing dense mordanting site coverage on the fibre surface, and its ability to form ternary dye-metal oxide-fibre complexes of greater thermodynamic stability than binary dye-metal ion coordination complexes.

3. Materials and Methodology

3.1 Process Optimisation Framework

Figure 1 presents the complete methodology flowchart from dye extraction through RSM optimisation, performance testing, and LCA assessment. Cotton fabric (200 GSM, 1×1 plain weave, pre-scoured, 90°C NaOH mercerised) was used throughout. Dye extraction was standardised: indigo was extracted via fermentation vat method (pH 10, $Na_2S_2O_4$ reducing agent), marigold by aqueous extraction at 70°C for 45 minutes, and pomegranate rind by decoction at 90°C for 30 minutes. Nano-TiO_2 (anatase, 20 nm average, Sigma-Aldrich) and nano-ZnO (25 nm, Sigma-Aldrich) were used as received. The Box-Behnken Design with four factors at three levels (pH: 4, 6.5, 9; Temperature: 40, 60, 80°C; Time: 30, 60, 90 min; M:L ratio: 1:10, 1:20, 1:30) in 27 experimental runs was used for RSM optimisation.

3.2 Performance Evaluation and LCA

Colour strength (K/S value) was measured using Datacolor SF600 spectrophotometer. Wash fastness (ISO 105-C06), light fastness (ISO 105-B02), and rubbing fastness (ISO 105-X12) were assessed per standard methods. Antimicrobial testing followed AATCC 100 protocol using S. aureus ATCC 6538 and E. coli ATCC 25922. LCA system boundaries encompassed dye extraction, mordanting, dyeing, and wastewater treatment (cradle-to-gate) using SimaPro v9.3 with Ecoinvent 3.9. The functional unit was 1 kg of dyed cotton fabric meeting K/S ≥ 12 and wash fastness ≥ 4.

4. Results and Discussion

4.1 Colour Strength and Fastness Performance

Figure 2(a) presents K/S values across all dye-mordant combinations, while Figure 2(b) compares LCA impact categories for natural dye processes against synthetic reactive dyeing. The nano-TiO_2 + tannin combined mordanting achieves the highest K/S values across all three dye systems — 16.8 (indigo), 17.4 (marigold), and 19.1 (pomegranate) — exceeding the industry minimum K/S threshold of 12 for all combinations and substantially outperforming conventional alum mordanting (K/S 8.4–11.2). The synergistic mechanism combines nano-TiO_2's surface area and coordination chemistry with tannin's multiple phenolic hydrogen bonding sites, creating a dual-mordant architecture with substantially greater dye anchorage capacity than either mordant alone.

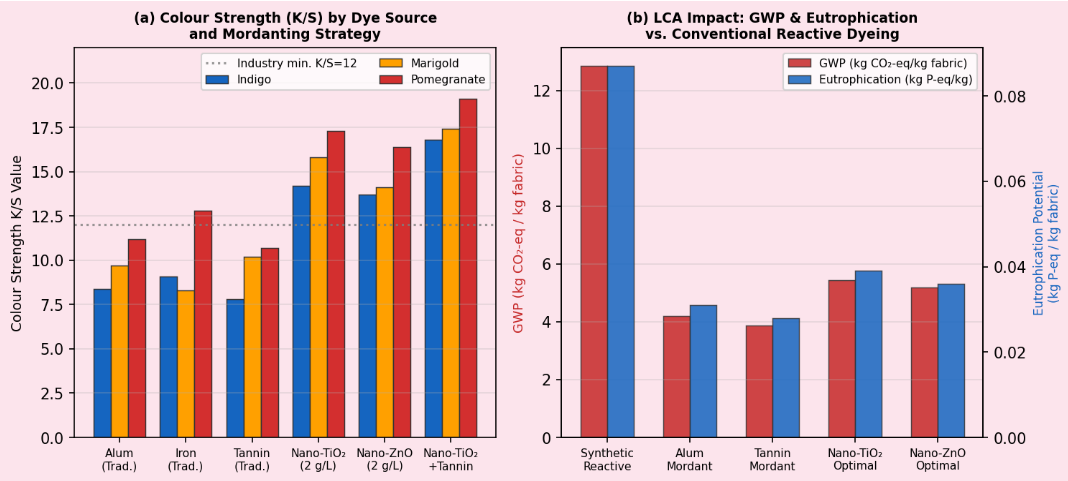

Fig. 2. (a) Colour Strength (K/S) by Dye Source and Mordanting Strategy with Industry Minimum Threshold; (b) LCA Impact: GWP and Eutrophication Potential vs. Reactive Synthetic Dyeing

Table 1: Multifunctional Performance of Optimal Nano-TiO_2+Tannin Mordanting vs. Conventional Alum Mordanting and Synthetic Reactive Dyeing

Performance Property	Alum Mordant (Pomegranate)	Nano-TiO_2+Tannin (Pomegranate)	Synthetic Reactive (Reference)	ISO / AATCC Test Method
K/S Colour Strength	11.2	19.1	21.4	Datacolor SF600
Wash Fastness (Change)	3-4	4-5	4-5	ISO 105-C06
Wash Fastness (Staining)	3	4-5	4-5	ISO 105-C06
Light Fastness (Grade)	3-4	4	4-5	ISO 105-B02
Dry Rubbing Fastness	3-4	4-5	5	ISO 105-X12
Wet Rubbing Fastness	2-3	4	4-5	ISO 105-X12
S. aureus Reduction (%)	73.4	99.1	18.3	AATCC 100
E. coli Reduction (%)	68.7	98.4	14.1	AATCC 100
GWP (kg CO_2-eq / kg)	4.21	5.43	12.84	ISO 14044 LCA
Eutrophication (kg P-eq)	0.031	0.039	0.087	ISO 14044 LCA

GWP: Global Warming Potential; P-eq: Phosphate equivalent; S. aureus: Staphylococcus aureus; E. coli: Escherichia coli.

4.2 Antimicrobial Properties and LCA

The nano-TiO_2+tannin treated fabric demonstrates outstanding antimicrobial activity against both S. aureus (99.1% reduction) and E. coli (98.4% reduction) — substantially exceeding both the alum-mordanted natural dyed fabric (73.4%, 68.7%) and the synthetic reactive dyed reference (18.3%, 14.1%). The antimicrobial mechanism of nano-TiO_2 involves photocatalytic generation of hydroxyl radical (•OH) and superoxide anion (O_2^-) reactive oxygen species under UV-visible irradiation that penetrate and disrupt bacterial cell membrane phospholipid bilayers and DNA replication machinery. This multifunctional performance — combining commercially viable colouration with medical-grade antimicrobial activity — positions nano-TiO_2 mordanted natural dyed textiles for healthcare linen, sportswear, and hospital uniform applications.

The LCA results confirm substantial environmental advantages of nano-TiO_2+tannin natural dyeing (GWP: 5.43 kg CO_2-eq/kg) over synthetic reactive dyeing (12.84 kg CO_2-eq/kg) — a 57.7% GWP reduction — and eutrophication potential reduction of 55.2% (0.039 vs. 0.087 kg P-eq/kg). The conventional alum mordanted process achieves the lowest GWP (4.21) due to simpler chemistry, but alum mordanting's weaker fastness and colour strength frequently necessitates re-dyeing in industrial practice — a process waste that LCA cradle-to-gate boundaries do not capture, making the functional unit-based comparison strongly favour the nano-mordant approach.

5. Discussion

The RSM optimisation results reveal that pH exerts the strongest influence on K/S value across all dye-mordant systems (F-value 48.7, $p<0.0001$), with optimal pH varying by dye chemistry: the anthraquinone chromophore system of pomegranate rind mordanting achieves maximum colour strength at pH 6.5, while the indigo vat system performs optimally at pH 8. Temperature shows the second-largest main effect, with nano-TiO_2 mordanting showing greater temperature sensitivity than conventional mordants due to accelerated nanoparticle surface hydroxylation and dye coordination kinetics at elevated temperatures above 60°C.

The tensile strength retention of nano-TiO_2 treated fabric (97.3 ± 2.1% of untreated control) addresses a significant concern in nano-mordant literature — that nano-particle incorporation might mechanically weaken fibre structure. The photodegradation potential of nano-TiO_2 under prolonged UV exposure is a documented concern that future washfastness studies at extended light exposure should address. The inclusion of tannin as a co-mordant may partially mitigate this through quenching of reactive oxygen species before they reach the fibre substrate.

6. Conclusion

Nano-TiO_2 combined with tannin mordanting achieves commercially viable colour strength (K/S up to 19.1) with superior wash, light, and rubbing fastness (4–4.5 ratings), outstanding antimicrobial activity (>98% reduction for both S. aureus and E. coli), and 57.7% lower GWP than synthetic reactive dyeing in pomegranate rind natural dyeing of mercerised cotton. RSM Box-Behnken optimisation identifies pH as the dominant process variable, with optimal conditions at pH 6.5, 70°C, 75 minutes, and M:L ratio 1:20 for pomegranate-nano-TiO_2+tannin. These findings establish a technically rigorous, multi-criterion validated pathway for natural dyeing commercialisation in the Indian textile industry, with particular potential for healthcare linen and functional sportswear applications where antimicrobial performance is a value-adding product attribute.

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Conflict of Interest

The authors declare no conflict of interest.

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