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Development of Natural Dyes for the Batik Textile Industry in Sri Lanka

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Abstract: Batik Textiles has become more demanding in the current fashion mode in the Sri Lankan market, but the expensive, synthetic azo dyes usage in the batik textile industry causes carcinogenic health risks to employees and customers. To overcome this, it is crucial to develop a nontoxic, low-cost, environmentally friendly alternative dyes that promotes worker and customer safety. This study attempts to develop natural dyes derived from mahogany shale, marigold flowers, rambutan peel, onion peel and mangosteen peel onto cotton and blue line poplin fabrics. The natural dyes were extracted from direct extraction by using distilled water (M1), Soxhlet extraction by using Ethyl acetate (M2) and Soxhlet extraction by using distilled water (M3), to assess their polarity, intensity, and stability on the textiles in comparison to synthetic batik dyes. Physical and chemical properties and test shelf-life of the natural dye-applied batik textiles with synthetic dye-applied batik textiles were analyzed. In comparison to Soxhlet extraction methods M2 and M3, direct extraction by using distilled water (M1) colorant from mahogany shale, marigold flowers, rambutan peel, onion peel and mangosteen peel had a higher % yield of dye powder, color stability, intensity, and higher washing cycles of 3 times in terms of overall acceptability. The Shades and effect of light on color stability of the samples were analyzed using UV Visible Spectrophotometer in the wavelength of 190-550 nm. The change in intensity and position in the characteristic peaks of different regions of the Fourier Transform Infrared Spectroscopy with Attenuated Total Reflectance in the wavenumber range of 500-4000 cm^{-1} was used to determine the quality and functional groups of the extracted colorants. Moreover, pH value was measured to determine the acidity level of the extracted natural colors. This study is aware of the feasibility of replacing harmful synthetic batik dyes with natural batik dyes extracted using mahogany shale; *Swietenia macrophylla*, marigold flowers; *Tagetes erecta*, rambutan peel; *Nephelium lappaceum* L., onion peel; *Allium cepa* L. and mangosteen peel; *Garcina mangostana* L.

Keywords: Batik textile, Batik dye, Synthetic dye, Natural dye, Mahogany, Marigold, Rambutan, Mangosteen.

1. Introduction

Batik Textiles has become more demanding in the current fashion mode in the Sri Lankan market. But the textile dyeing industry relies heavily on synthetic dyes, the most of which are azo dyes. However, synthetic azo dyes can release harmful aromatic amines which break down in the process. These amines may be carcinogenic to human health and of course have a negative impact on the environment. In recent years, there has been a huge awareness of the dangers of synthetic azo dyes. As a result, there is an increasing demand for greener and safe alternatives. Natural dyes are one promising option for developing a more environmentally friendly textile dyeing in the batik process.

Natural dyes are non-toxic, biodegradable and generally have higher compatibility with the human health and environment than synthetic dyes. Recently, small and medium textile enterprises (SMEs) in Sri Lanka are whirling towards the natural dyeing process considering newly established import restrictions, cost effectiveness, employee health concerns and environmental benefits. This study attempts to develop natural batik dyes derived from mahogany shale; *Swietenia macrophylla*, marigold flowers; *Tagetes erecta*, rambutan peel; *Nephelium lappaceum* L., onion peel; *Allium cepa* L. and mangosteen peel; *Garcina mangostana* L. onto cotton and blue line poplin fabrics to replace the dyeing process with synthetic azo dyes. The results have been evaluated based on the color strength and color fastness of the natural dyes on cotton and blue line poplin fabrics by physical, chemical, and shelf-life properties.

2 METHODOLOGY

2.1. Experimental design layout

The research experiment was conducted in Uva Wellassa University of Sri Lanka, with five plant materials (mahogany shale; *Swietenia macrophylla*, marigold flowers; *Tagetes erecta*, rambutan peel; *Nephelium lappaceum* L., onion peel; *Allium cepa* L. and mangosteen peel; *Garcina mangostana* L.), two textile materials (cotton and blue line poplin) using three extraction methods such as Direct extraction by using distilled water (M1), Soxhlet extraction by using Ethyl acetate (M2) and Soxhlet extraction by using distilled water(M3), and two wax removal methods of paper and iron method and detergent mixed hot bath method .

2.2. Collection of raw materials

(a) (b) (c) (d) (e)

Fig. 1 Selected raw plant-based materials for natural dye extraction (a)Mahogany shale, (b)Onion peel, (c)Rambutan peel, (d)Mangosteen peel, and (e)Marigold flowers

The Raw materials were collected from Badulla and Rathnapura districts in Sri Lanka. Textile materials, cotton and blue line poplin were brought from the local market in Badulla. Cow dung was used as the bleaching agent. Alum was used as a mordant solution.

2.3. Preparation of raw materials

Plant based raw materials of mahogany shell, mangosteen peel, onion peel, marigold flower and rambutan peel were cleaned with distilled water and cut into small pieces, then dried under the sunlight for 2-3 days. The textile materials were dipped in cow dung solution (100 g of cow dung in 500 ml water solution) for 12 hours followed by water solution and dried in sunlight to remove the smudges and spots in the cloth pieces.

2.5 Pre-mordant selection

As pre-mordants, 2 g of Alum dissolved in 500 ml of distilled water to prepare the mordant solutions. The scoured cotton and blue line poplin samples were heated in mordant solutions at 100 ℃ for 30 minutes. The fabric samples were dried without washing.

2.6 Natural Dye Extraction

2.6.1. Direct extraction by using distilled water method (M1)

100 g of dried raw plant materials were soaked in 250 ml of distilled water at room temperature for 48

hrs. Then the dye solution was filtered using a muslin cloth followed by Whatman No. 1 filter paper. The filtrate was concentrated and evaporated to dryness under heat at 100 ºC using a hot water bath (SY-2L6H). For dye powder extraction vacuum oven to dryness under hot air vacuum oven (GX-125B, Brand-FAITHFUL) at 105 ºC for 3 hours. Then the dried extracted natural dye was safely stored in a desiccator for future uses. [1]

2.6.2. Soxhlet extraction method by using Ethyl acetate/Distilled water method (M2/M3)

20 g of powdered plant materials were uniformly packed into a thimble and extracted by using 175 ml of Ethyl acetate in M2 method and 175 ml distilled water in M3 method. The process of extraction continues for 48 hours at 70 ºC for ethyl acetate (M2) and 100 ºC for distilled water (M3) till the solvent in the siphon tube of an extractor becomes colorless. After that, the extract was filtered through Whatman No.1 filter paper. The filtrate was concentrated and evaporated to dryness under a vacuum at 40 ºC using a rotary evaporator. For dye powder extraction vacuum oven to dryness under hot air vacuum oven (GX-125B, Brand-FAITHFUL) at 105 ºC for 3 hours. The extracts were stored in a desiccator for future uses.

2.7 Natural dye application on batik textiles

The wax mixture for the batik art was prepared by using 100g of paraffin wax, 20g of bee wax, and 1g of resin. The mixture was boiled at 100°C, to get a wax solution. Then, the wax solution was applied on both sides of the pencil drawn batik design on cotton and blue line poplin textiles using a synthetic hair liner painting brush. The wax applied batik textiles were soaked in extracted natural dyes, at room temperature for 48 hours followed by sunlight drying without washing.[2]

2.8 Batik wax removing methods

2.8.1 Paper and iron method

The wax applied naturally dyed batik textiles were boiled in water for 5-7 minutes followed by ironing at a higher temperature by keeping the fabric in between two blank papers.

2.8.2 Detergent mix hot bath method

The wax applied naturally colored fabric was soaked in detergent solution and boiled at 85-90 ^{0}C for 5-7 minutes.

3 RESULTS AND DISCUSSIONS

% Yield

According to the results of % yield of dye powder, direct extraction by using distilled water method (M1) was recognized as the higher extraction method rather than the Soxhlet extraction by using ethyl acetate method (M2) and Soxhlet extraction by using distilled water method (M3).

UV-Vis Analysis

Color absorbance effect of light on colorant stability was performed with dyed cotton and blue line poplin samples after washing and drying three times in sunlight condition. Then color absorbance was assayed via the UV-VIS Spectrophotometer (THERMO SCIENTIFIC EVOLUTION 201). The percent transmission and the color strength value of dye solution were recorded from 190-550 nm, at 10 nm intervals. The extracted dye colorant gave the following stability absorption peaks as represented in table 1.

Table 1. Shades and effect of light on color stability

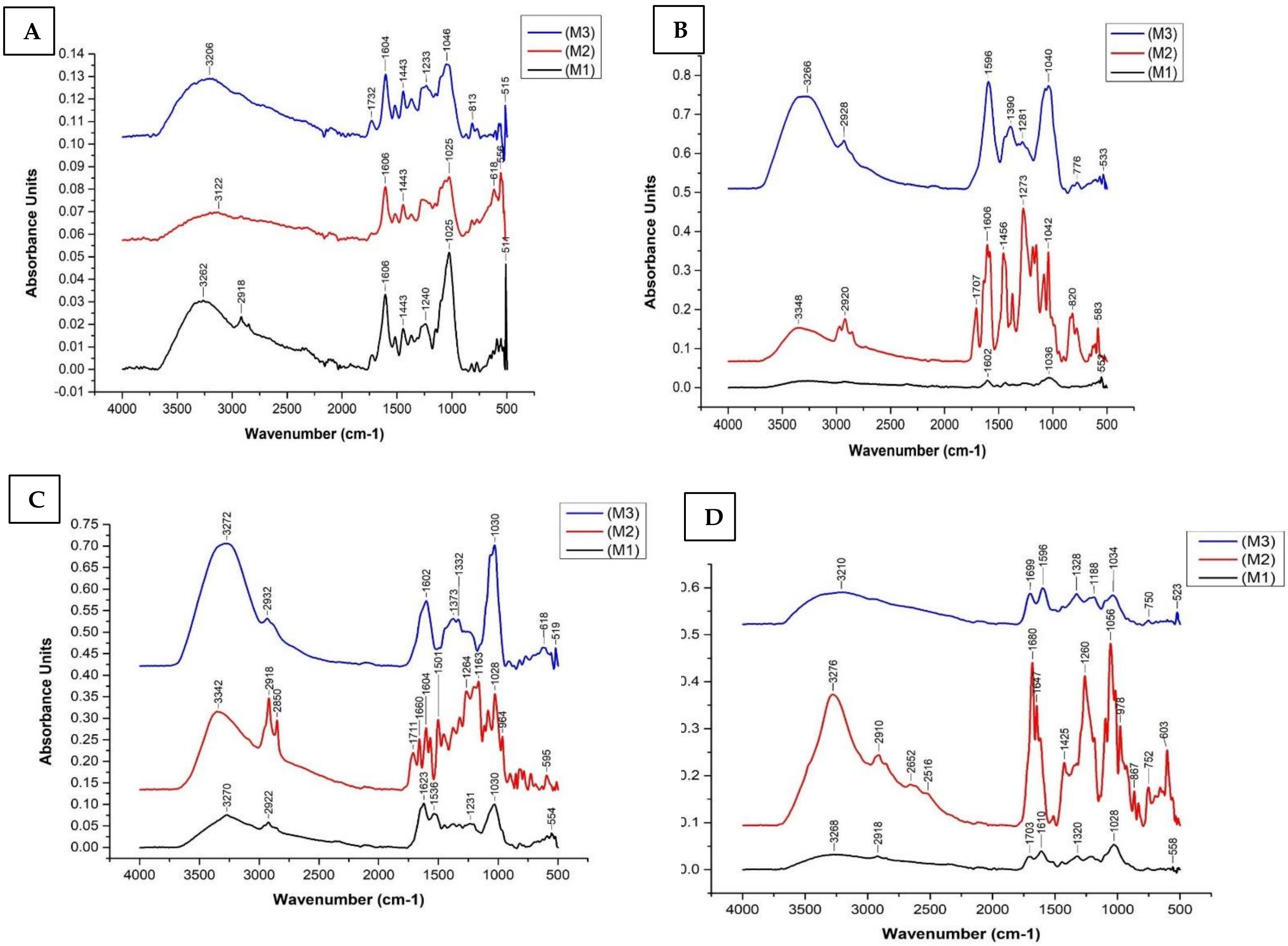
Material	Extraction method	Wavelength / nm	Absorption	Color stability
Mahogany	M1		0.24	High
	M2	280	0.21	Good
	M3		0.20	Low
Onion	M1		0.25	Good
	M2	310	0.30	High
	M3		0.09	Low
Marigold	M1		0.04	Low
	M2	280	0.40	High
	M3		0.11	Good
Rambutan	M1		0.66	High
	M2	220	0.32	Low
	M3		0.38	Good
Mangosteen	M1		0.05	Low
	M2	200	0.10	Good
	M3		0.24	High

pH value

The direct extraction method shows a lower pH value than the Soxhlet extraction methods. The pH values of direct extraction by using distilled water (M1) for mahogany, onion, rambutan, mangosteen and marigold were 5.29, 3.44, 3.19, 3.08 and 4.12 respectively. The pH values of Soxhlet extraction by using ethyl acetate (M2) for mahogany, onion, rambutan, mangosteen and marigold were 6.62, 3.96, 3.25, 5.05, and 4.11 respectively. The pH values of Soxhlet extraction by using distilled water (M3) for mahogany, onion, rambutan, mangosteen and marigold were 5.62, 3.55, 3.23, 4.04, and 4.12 respectively. Results were obtained from the pH meter (MULTI-FUNC.METER MM-42DP).

FTIR analysis

The Fourier Transmission Infrared (FTIR) Spectroscopy (Brand - Alpha) studies were done to confirm the functional group of the extracted dye samples based on the peak value in the region of infrared radiation. The extracted dyes gave the following characteristic absorption peaks as represented in Fig. 2.



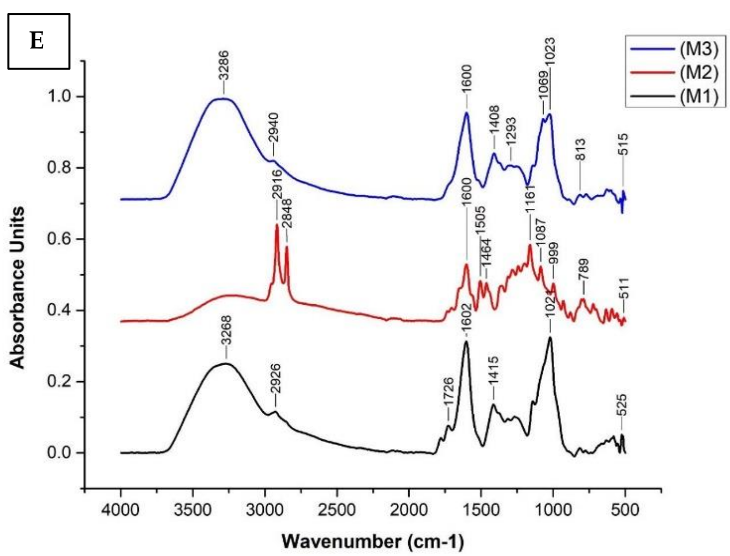


Fig 2. FTIR results of the A) mahogany shale, B) mangosteen peel, C) marigold petal, D) rambutan peel and E) onion peel samples obtained from M1, M2, and M3 extraction methods

The change in intensity and position in the characteristic peaks of different regions of the FTIR spectrum were used to determine the quality and functional groups of the extracted natural colorant powder in direct M1 and Soxhlet M2, M3 extraction methods. The FTIR spectrum of mahogany shale extraction from M1 method shows strong absorption bands at 1606, 1025, 3262 cm^{-1}, M2 method shows strong absorption bands at 1606, 1025, 3122 cm^{-1} and M3 method shows strong absorption bands at 1604, 1046, 3206 cm^{-1} as in fig. 4A. The spectral analysis of the mahogany shale based natural dye powder results in the presence of O-H, C=C, and C-O-C functional groups which are characteristics of the flavonoids. The FTIR spectrum of mangosteen peel extraction from M1 method shows strong absorption bands at 1602, 2921, 3262 cm^{-1}, M2 method shows strong absorption bands at 1042, 1606, 2920, 3348 cm^{-1} and M3 method shows strong absorption bands at 1040, 1596, 2928, 3266 cm^{-1} as in fig. 4B. The spectral analysis of the mangosteen natural dye powder extracts showed the presence of alkane and aromatic, C-C, and O-H functional groups which are characteristics of the anthocyanin and tannin.

The FTIR spectrum of marigold extraction from M1 showed strong absorption bands at 1030, 1623, 2922, 3270 cm^{-1} and M2 showed strong absorption bands at 1028, 1604, 2918, 3342 cm^{-1} while M3 showed strong absorption bands at 1030, 1602, 2932, 3272 cm^{-1} as shown in fig. 4C. The spectral analysis of the marigold natural dye results in the presence of unsaturated polyenoic hydrocarbons, C=C and C-H functional groups which are characteristics of the lutein. The FTIR spectrum of rambutan extraction from M1 showed strong absorption bands at 1028, 1610, 1703, 3268 cm^{-1} and M2 showed strong absorption bands at 1056, 1680, 3276 cm^{-1} while M3 showed strong absorption bands at 1034, 1699, 3210 cm^{-1} as shown in fig. 4D. The spectral analysis of the rambutan natural dye powder extracts showed the presence of C-C, and O-H functional groups which are characteristics of the anthocyanin. The FTIR spectrum of onion extraction from M1 showed strong absorption bands at 1602, 1021, 3268 cm^{-1} and M2 showed strong absorption bands at 1600, 1087 cm^{-1} while M3 showed strong absorption bands at 1600, 1023, 3286 cm^{-1} as shown in fig. 4E. The spectral analysis of the onion natural dye powder extracts showed the presence of O-H, and C=C functional groups which are characteristics of the anthocyanin which result in blue, red or purple colors to batik textile.

Colour stability and Shelf life

During the direct extraction by using distilled water method (M1), the dark maroon color from mahogany shale, dark purple color from onion peel, the dark brown color from rambutan peel, the dark pink color from mangosteen peel, and the dark yellow color from marigold flowers were resulted. During the Soxhlet extraction methods (M2 & M3), light colour ranges were obtained from respective dried plant-based raw materials as shown in table 2.

Fig. 3. Extracted dye powder from plant based raw materials

Table 2: Obtained colors according to the supposed methodologies

Material	Control	Direct	Soxhlet	Batik
Mangosteen Cotton BLP	7/321	1/412	1/578	2/160 & 2/868
Mahogany Cotton BLP	5/934	5/934	5/586	3/170 &3/722*
Rambutan Cotton BLP	12/582	12/582	13/292	1/893
Onion Cotton BLP	3/672	3/672	3/453	12/283
Marigold Cotton BLP	12/615*	12/616	12/615*	1/416*

When comparing the synthetic batik with natural batik, respectively Marigold, Onion, Rambutan, Mahogany and Mangosteen were shown good results to cotton & blue line poplin. The natural batik dyes extracted from Marigold, Onion, Rambutan, Mahogany and Mangosteen were shown good results on cotton & blue line poplin batik textiles. Marigold has shown the best color adjustments after three times washing cycles. The color changes after washing may occur due to dye decomposition, detachment from the substrate, or ionization during alkaline washing, as observed in previous research, Mahala *et al*., 2001. Notably, Detergent mix hot bath method was more effective in removing batik wax than the iron and paper methods in natural dye applied batik textile materials.

The direct extracted dyes used to create cotton and blue line poplin batik textiles. Then final output was given the following appearance/color changes in Fig. 4 after washing three times.

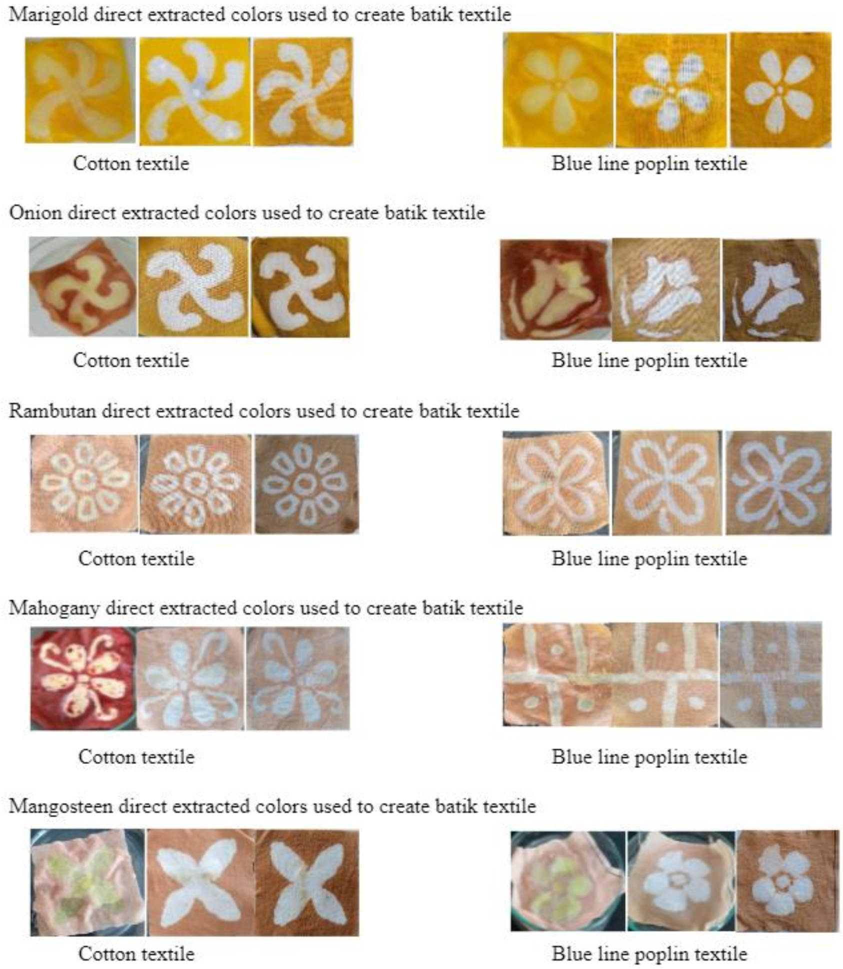


Fig 4. Batik textile appearance and color changes after three times washing

4 CONCLUSION

Natural dyes extracted from raw materials were applied into cotton and blue line poplin successfully. The direct extraction by using distilled water method and Soxhlet extraction by using distilled water method results in a higher yield colorant than the Soxhlet extraction by using ethyl acetate method with high colour stability. Direct extraction by using distilled water method (M1) can be used as the best way to extract natural dye powder with higher percentage (%) yield. Marigold, Onion peel, Rambutan peel, Mangosteen and Mahogany shale extracted colors were suitable for the development of natural colorant for batik textiles on cotton and blue line poplin within alum mordant solution. The exploration of diverse mordant solutions interaction with plant -based colorants is advised to optimize the natural dyeing processes and expand the scope of sustainable dyeing practices instead of synthetic counterparts.

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