

RAUVOLFIA VOMITORIA LEAVES AS A SUSTAINABLE SOURCE OF NATURAL COLOURANTS FOR POLYAMIDE FABRICS

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ABSTRACT

As the global demand for safe products is increasing, the search for new sources of natural dyes need to be intensified. This paper focuses on the extraction procedure and dyeing characteristics of *Raufolesia vomitoria* leaf extract as a new source of natural dyes. It was found that extraction using protic solvent (mixture of C₂H₅OH and 0.5M H₂SO₄) could reach optimal values at a temperature of 70°C after 5 hours. It was also found that the colouring and non-colouring components present in *R. vomitoria* leaf extract were identified as Vanillic acid, Ferulic acid, p-Cumaric acid and Chlorogenic acid. Simultaneous mordanting and dyeing of wool and nylon 6 fabrics were then performed in order to study the dyeability activity on these two textile fabrics. The chelating materials used as mordants were CuSO₄, SnCl₂ and FeSO₄. The colorimetric parameters of the dyed fabric samples were determined using reflectance method. Furthermore, the washing, light and rubbing fastness properties were tested according to standard methods. In particular, the washing and light fastness properties were very good and excellent (6 and 6/7) technically.

Keywords: Natural dyes, Optimised extraction, Simultaneous mordanting, Wool and Nylon 6, Exhaustion percentage

INTRODUCTION

In recent times, consumers of textile products are becoming interested in products that contain safe colours (Wang *et al.*, 2012, Savvidis *et al.*, 2012, de Boer *et al.*, 2019). Due to this reason, the researchers are appreciating natural dye sources due to their environmentally friendly, nontoxic, nonhazardous, and biodegradable nature (Adeel *et al.*, 2023). Thus, natural dyes, more suitable for human use for textile and non-textile purposes are needed. However, the most important issues nowadays for researchers in the production and utilization of natural colourants are sustainability, eco-

friendliness and improved functionality (Saxena *et al.*, 2014, Link *et al.*, 2019). Hence, researchers are now actively directing their research in the field of dye-sensitized solar cells (hao *et al.*, 2005), cosmetics (Beiki *et al.*, 2018, Patil *et al.*, 2016), UV-protective clothing (Ebrahimi *et al.*, 2015), food (Ebrahimi *et al.*, 2016), pharmaceuticals, antimicrobial finishing of textiles and printing of textiles (Haji *et al.*, 2014, Prusty *et al.*, 2010, Ismal *et al.*, 2013, Savvidis *et al.*, 2014, Islam *et al.*, 2016, Basak *et al.*, 2016). Thus, apart from the use of natural dyes in the field of textile industry, the demand for them will definitely rise in the nearest future as

new areas of application emerges. Currently, the amount of natural dyes required by the world's textile industry alone is about 3.5 million tonnes (Ian *et al.*, 2014). This amount, is by far above the current world production level. To this end, the identification of more natural dye-bearing plants and their large scale cultivation and production by interested companies are necessary. This will ultimately bridge up the gap between demand and supply of natural dyes.

Raufolevia vomitoria is a shrub mostly found in tropical Africa including Nigeria. The plant grows up to 40 m tall and about 80 cm in diameter. The plant is a source of many alkaloid compounds such as ajudicine and ajmaline. The leaves of the plant is found to contain the geischshizol alkaloids, whereas the root bark contains reserpine, reserpinine and ajmaline alkaloids. The presence of these alkaloids makes *R. vomitoria* important in the pharmaceutical industry. For instance, reserpinine is known for its anti-hypertensive, antipsychotic and sedative activities [20]. Apart from these uses, *R. vomitoria* leaves are yet to be utilized as a source of natural dyes. The present research, is therefore necessary for introducing *R. vomitoria* leaves as an abundant and cheap source of natural dyes for extraction as well as application on wool and nylon 66 fabrics.

MATERIALS AND METHODS

The leaves of the plant were harvested from the campus of Delta State University, Abraka, Nigeria. The plant was identified by the Botany survey at the University of Calabar, Calabar, Nigeria with a voucher number (2018/CAL/HRB/1733). The wool and nylon fabrics were obtained from a local market. The chemicals such as

C_2H_5OH , CH_2Cl_2 , H_2SO_4 , $FeSO_4$, $SnCl_2$, $CuSO_4$ and silica gel were of laboratory grade (Merck Chemical Co. Ltd., Germany).

Preparation of Stock Solutions of Mordants

The stock solution of $FeSO_4$, $CuSO_4$ and $SnCl_2$ were made by dissolving 10 g of mordant in 1000 mL distilled water respectively.

Preparation of Leaves for Extraction

The leaves of *R. vomitoria* were obtained from matured plants. The dirt and dust on their surfaces were removed carefully and dried under sunlight for two weeks. The Frouton Multipurpose Grinder BIZM, China was used to grind the leaves into powdered form and ready for colour extraction.

Extraction of Dye

The ground leaves (10 g) were placed into a Soxhlet extractor. Extraction solvent (a mixture of C_2H_5OH (80%) and 0.5M $H_2SO_{4(aq)}$ 250 mL) was added and extraction was performed at 20, 30, 40, 50, 60, 70, 80 and 90°C for 1 hour until a negligible colour was obtained in each case. Extraction was repeated using the same solvent system at 70°C and using extraction time of 1, 2, 3, 4, 5, 6 and 7 hours respectively. Extraction was also carried out using ethanol and dichloromethane as the solvent systems respectively. In each case, the absorbance readings were measured and recorded followed by a plot of absorbance against temperature and time respectively (Wang *et al.*, 2016). The crude extract was subjected to column chromatography analysis and the colouring components were eluted using ethanol/chloroform, methanol and ethanol respectively. The various elutes were

monitored by Thin Layer Chromatography (TLC). High Performance Liquid Chromatography (HPLC) analysis was carried out on the elutes respectively (Agilent Hewlett Packard 1050 Series HPLC with a UV-variable wavelength detector, Germany).

Mordanting and Dyeing of Wool Fabrics

The wool fabrics were first pretreated in a bath containing 2.0 gl⁻¹ nonionic detergent and 1.0 gl⁻¹ of Na₂CO₃ for 3 min at 60°C having a liquor ratio of 50:1. The scoured samples were rinsed and treated with 1.0% acetic acid solution to neutralize them. A dye bath containing the dye extract of liquor ratio 1:50 at 2% shade on the weight of fabrics and pH 4.5 was prepared. The scoured wool samples were immersed into the dye bath and dyeing started at 40°C, using the Ahiba laboratory dyeing machine (Data Colour, USA). The temperature of the dye bath was gradually raised over a period of 25 minutes to 100°C. Dyeing was maintained at this temperature for 60 minutes. The unfixed dye molecules were removed by washing the fabrics in a bath which contains 2.0 gl⁻¹ of nonionic detergent for 15 minutes. Another set of scoured wool samples were dyed in a bath containing the following mordants (SnCl₂, CuSO₄ and FeSO₄) respectively. Mordant is commonly used for the dyeing process using natural dyes. Mordant is a substance that binds dyes to the fabric. It's derived from metallic salt including alum, chrome, stannous chloride, copper sulfate, and ferrous sulfate [36]. The fabrics were finally rinsed with cold distilled water and dried in an oven set at 50°C (Zhang *et al.*, 2017, Otutu *et al.*, 2019).

Mordanting and Dyeing of Nylon 6 Fabrics

The nylon fabrics (1.0 g) were scoured in a solution containing sodium carbonate 2.0gl⁻¹ and nonionic detergent for 20 minutes using liquor ratio of 40:1. The nylon samples were then immersed into the dye bath containing a liquor ratio of 1:50 using 2% shade on the weight of fabric and using SnCl₂, CuSO₄ and FeSO₄ as mordants respectively. The dye liquor pH was adjusted to 4.5 by the addition of 10% acetic acid (v/v) and dyeing started at 40°C. The temperature of the dye bath was raised to 100°C at a rate of 2°C min⁻¹. After dyeing at this temperature for 1 hour, the dyed samples were rinsed and dried at room temperature. The dyeing process was performed using Ahiba laboratory dyeing machine (Data Colour, USA). Another set of scoured nylon 6 samples were also dyed without the mordants to serve as a control (Sadeghi *et al.*, 2019, Gasht *et al.*, 2014).

Measurement of Exhaustion Percentage

The exhaustion of dyed fabrics was measured by the method described by Dong (Gasht *et al.*, 2014). The absorbance of the dye solution was measured before and after dyeing, using an Agilent Technologies Cary 300 UV-Visible spectrophotometer. The expression for calculating the exhaustion percentage is as shown;

$$\begin{aligned} \% \text{ exhaustion} &= \frac{A_1 - A_2}{A_1} \\ &\times 100 \dots \dots \dots (1) \end{aligned}$$

Where;
A₁ and A₂ represent the absorbance values before and after dyeing respectively. The absorbance values were measured trice in each case and the average values were recorded.

Colour and Colour Strength Measurements

The dyed fabrics were evaluated for their CIE L*, a*, b* colour space system, which were converted from the tristimulus values of XYZ coordinate under illuminant. The L* represent, the light brightness axis, a* corresponds to the red-green axis and b* represents the blue-yellow component and H which refers to the quantitative parameters of the hue of the colour was also evaluated. These were examined using the reflectance data obtained from a 10 degree observer and an illuminating δ₆₅ source. The colour yield (K/S) values were calculated using the expression [27-29]

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

Where;

K represents the absorption coefficient
 S refers to light scattering coefficient and
 R corresponds to the reflectance of dyed fabric

Colour Fastness Tests

Assessment of colour fastness to washing was performed using ISO-CO3 method [30]. The solution used contains 5 gl⁻¹ soap solution. The dyed samples (5x5 cm) were placed between two adjacent undyed fabrics and treated with the solution for 45 minutes at 60°C using a liquor ratio of 50:1 in an Arora dyeing machine. The combination was then removed, rinsed with

distilled water and air dried at room temperature. The change in colour and staining on adjacent undyed samples were evaluated by comparing with standard scale (1 – 5) where 1 is poor and 5 is excellent.

Colour fastness to light was carried out using ISO-BO1 protocol (ISO 2014). The dyed fabrics were exposed to the effect of sunlight, set alongside standard light sensitive blue wool for a duration of 72 hours. The change in colour after the test was assessed using the blue scale for light fastness. Tests for rubbing fastness were performed using a crockmeter according to ISO 105-X12 2001 method (ISO 2001). The dyed samples were tested following both dry and wet procedures respectively. Staining on the testing cloth was assessed by comparing with the grey scale (1 – 5) where 1 represents poor performance and 5 represents excellent.

RESULTS AND DISCUSSION

Optimization of Dye Extraction

The optimization of natural dye extraction is important in order to maximize dye yield and also reduce the use of energy during processing. The most important parameters in the extraction of natural dyes are time and temperature. In this study, the yield of dye from the leaves of *R. vomitoria* using protic and aprotic solvents is regarded to be proportional in value to the UV-Visible absorbance. The process was monitored using UV-Visible instrument at 213-450nm.

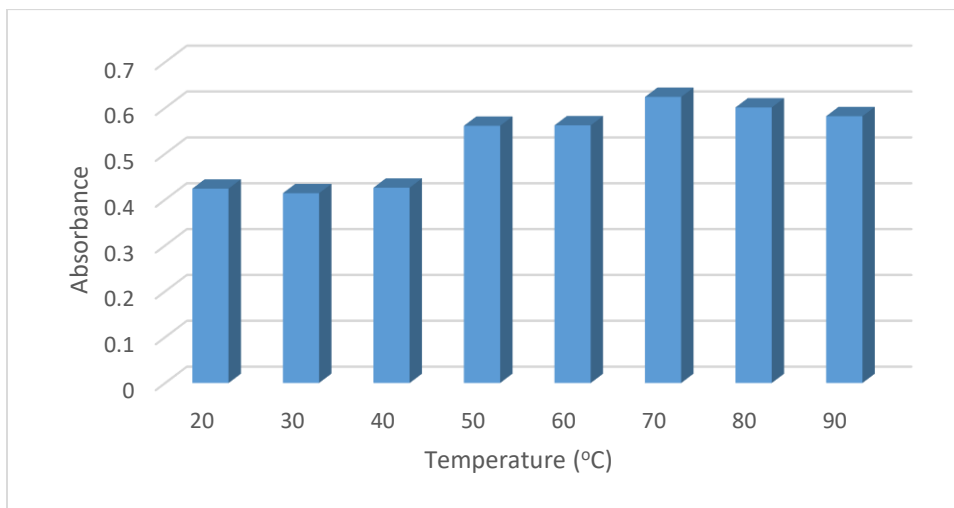


Figure 1: Effect of temperature on dye absorbance using C₂H₅OH (80%) and 0.5M H₂SO₄ mixture as solvent

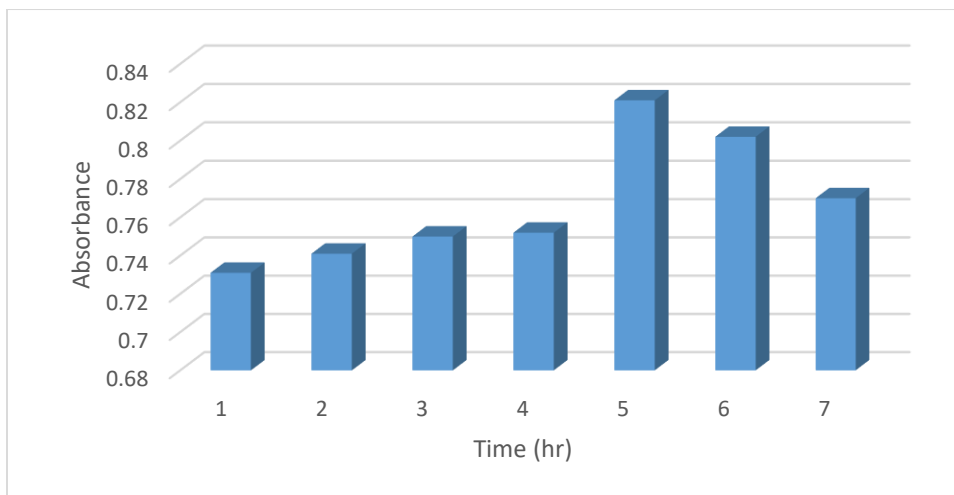


Figure 2: Effect of time on dye absorbance using C₂H₅OH (80%) and 0.5M H₂SO₄ mixture as solvent

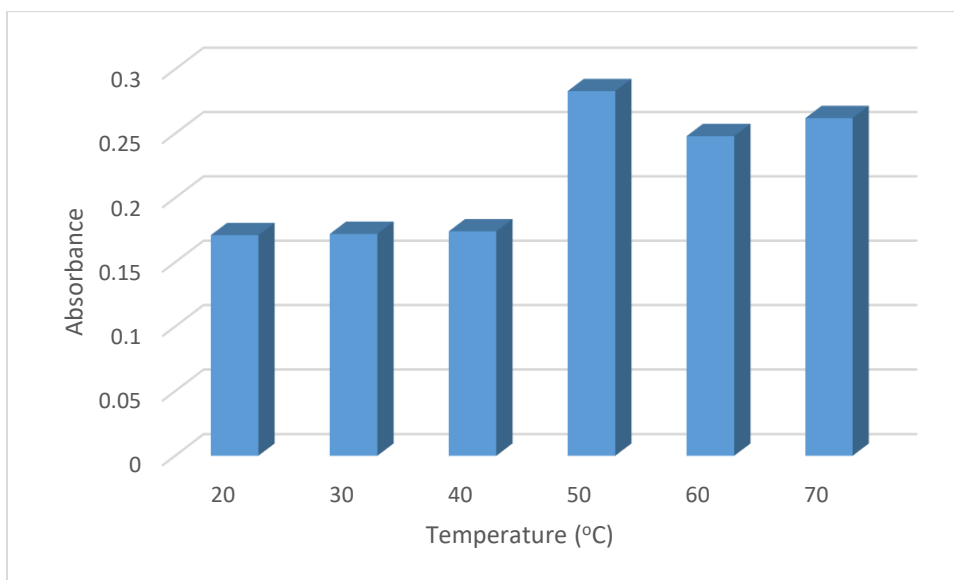


Figure 3: Effect of temperature on dye absorbance using C_2H_5OH (80%) as solvent

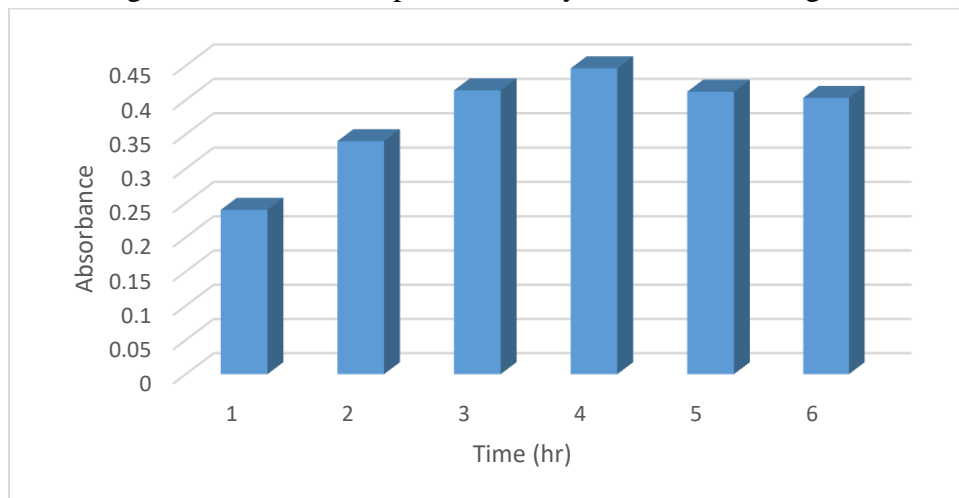


Figure 4: Effect of time on dye absorbance using C_2H_5OH (80%) as solvent

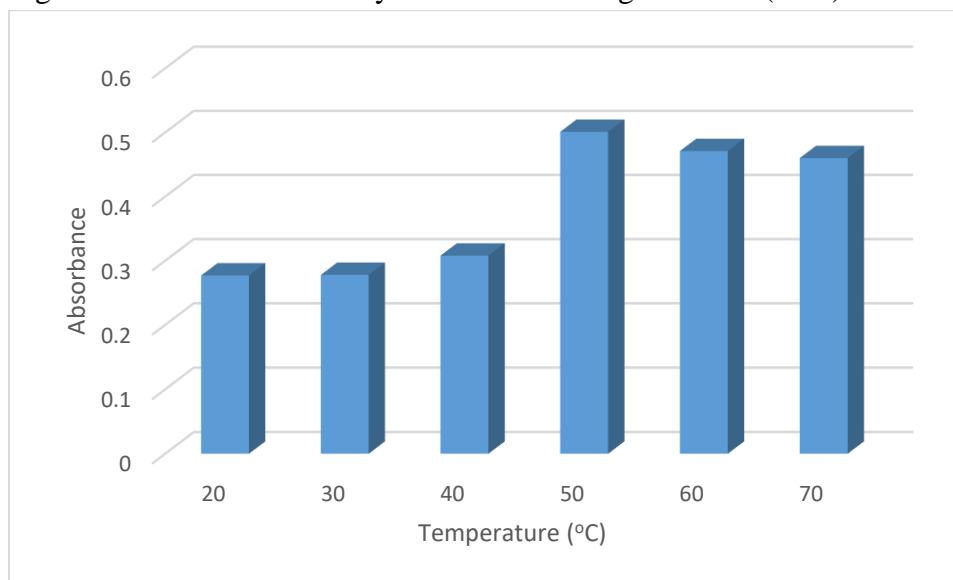


Figure 5: Effect of temperature on dye absorbance using CH_2Cl_2 as solvent

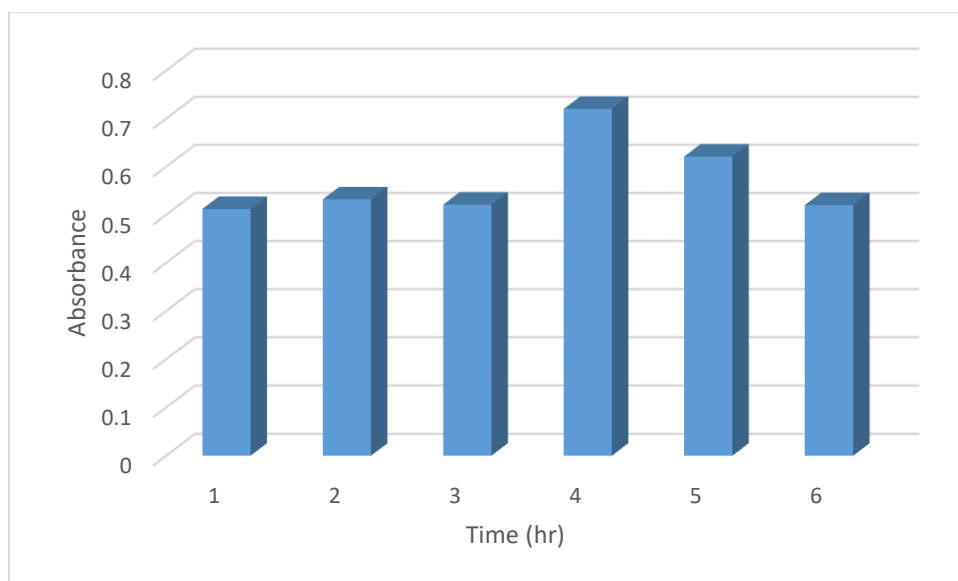


Figure 6: Effect of time on dye absorbance using dichloromethane as solvent

Figures 1 to 6 show the extraction of natural dyes from *R. vomitoria* leaves at various temperatures of 20 - 70°C and it was observed that the extraction was optimal at 70°C after 5 hours and then decreased gradually over time, when a mixture of C₂H₅OH and 0.5M H₂SO₄ was used as a protic solvent. The highest absorbance observed when dichloromethane (aprotic solvent) was used is 0.720 after 4 hours at

50°C. Although, this extraction temperature is lower and preferred, when compared with that of the protic solvent system. The extraction of natural dyes from plant sources at lower temperatures is preferred to that at higher temperatures due to the fact that at higher temperatures, the chemical structure of the dyes could breakup. Results found in this research is, however, still within the allowed temperatures

3.2 Evaluation of the Colouring and non-Colouring Components in the Extract

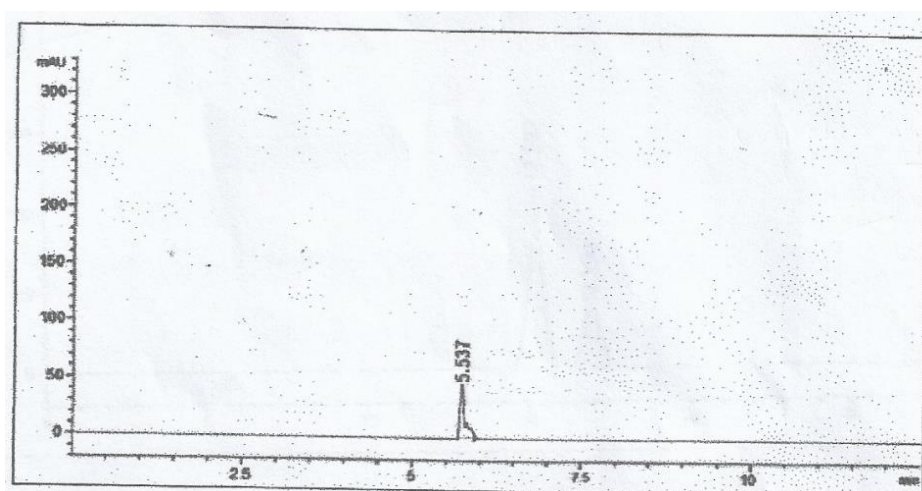


Figure 7: HPLC trace of dye extract at λ_{\max} 270nm

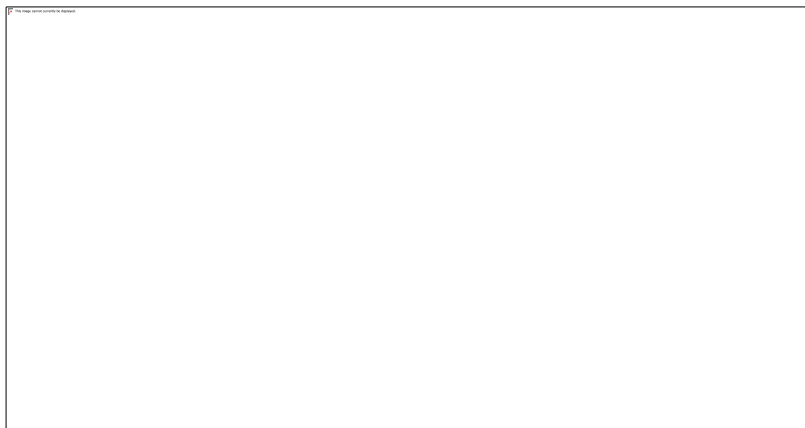


Figure 8: HPLC trace of dye extract at λ_{\max} 280nm

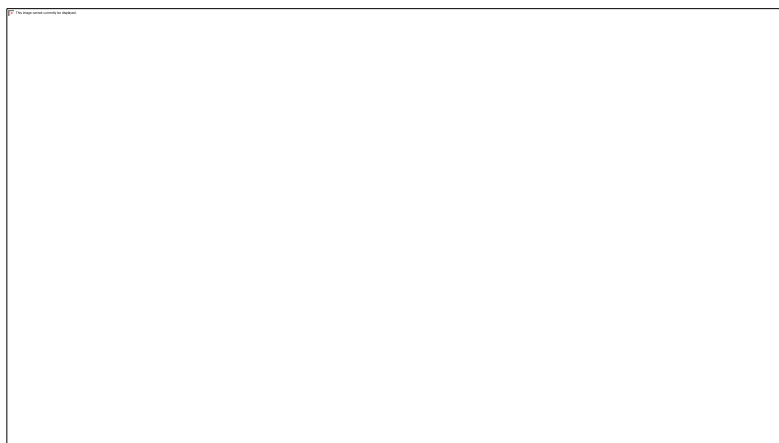


Figure 9: HPLC trace of dye extract at λ_{\max} 320nm

Figure 7, 8 and 9 show the high-performance liquid chromatography (HPLC) traces of the dye extract using Agilent Hewlett Packard 1050 series HPLC with a UV-visible wavelength detector (Germany). The compound whose peak was observed at the retention time of 5.537 min (λ_{\max} 270nm) in Figure 7 was not detected. In figure 8, the peak located at 7.284 min (λ_{\max} 280nm) was detected and identified as Vanillic acid. The peaks observed at 7.351 and 8.573 min (λ_{\max}

280nm) were identified as Ferulic and ρ -Cumaric acid respectively. Finally, the peak found at 14.592 min (λ_{\max} 320nm) in Figure 9, was identified as Chlorogenic acid, while the peak observed at 16.398 min (λ_{\max} 320nm) was not detected. From the HPLC results, the alkaloid components of the leaf extract were not detected. The identified colouring and non-colouring compounds are shown in Figure 10.

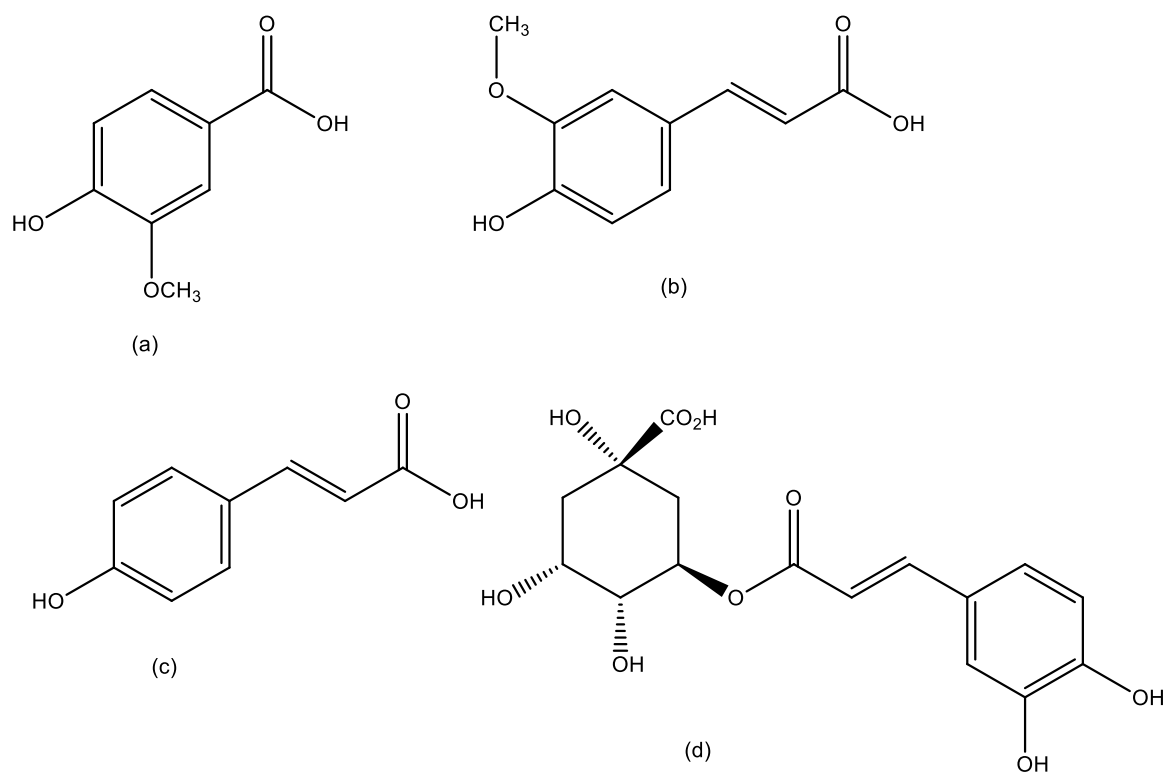


Figure 10: Structures of identified Colour and non-colouring components in *R. vomitoria* leaf extract (a) Vanillic acid, (b) Ferulic acid (c) p-Cumaric acid (d) Chlorogenic acid

Colour Fastness

Table 1: Washing Fastness Properties of *R. vomitoria* Leaf Extract on Wool and Nylon 6 Fabrics

Fabrics	Change in colour				Staining			
	Direct	CuSO ₄	SnCl ₂	FeSO ₄	Direct	CuSO ₄	SnCl ₂	FeSO ₄
Wool	3	5	4	4/5	4	4/5	5	5
Nylon	2/3	4/5	4	5	3/4	4	3/4	4

The dyed wool and nylon samples gave very good washing fastness (Table 1). The shade of the dyed fabrics changed slightly during laundering. For instance, it was observed that dyeing without a mordant had a rating of 2/3 (good) and when mordants were applied the ratings increased to 4/5 and 5 (excellent). This indicates that mordants significantly improved the washing fastness. It was also evident from the staining results, that the removed dye molecules did not attach

to the adjacent fabric, hence, the ratings were very good (3/4 and 4). The dry and wet rubbing fastness values were 3/4 and 4, indicating that slight staining took place during the rubbing process (Table 3). The light fastness of the dyed fabrics were observed to be fair/moderate (3 and 3/4) on wool and nylon fabrics respectively. However, on the introduction of Cu, Sn and Fe salts, an improvement in the light fastness properties was observed (Table 2).

Table 2: Light Fastness Properties of *R. vomitoria* Leaf Extract on Wool and Nylon 6 Fabrics

Fabrics	Direct	CuSO ₄	SnCl ₂	FeSO ₄
Wool	3	6/7	5/6	6/7
Nylon	¾	6	5/6	6/7

Table 3: Rubbing Fastness Properties of *R. vomitoria* Leaf Extract on Wool and Nylon 6 Fabrics

Fabrics	Direct		CuSO ₄		SnCl ₂		FeSO ₄	
	Dry	Wet	Dry	Wet	Dry	Wet	Dry	Wet
Wool	4	¾	4	4/5	4	4	4	4
Nylon	4	¾	4	¾	4	3/4	4	3/4

It is also worthy of note that the essential factors influencing the fastness properties of natural dyes include the bonding type between the dye and the fibre, the concentration of the dye used in dyeing, the chemical structure of the dye, the finishing applied after dyeing as well as the type of fibre(s) involved. Furthermore, it has been reported that Vanillic acid and Ferulic acid (Ismal *et al.*, 2014) can act as UV-absorbers which improve the light fastness of

natural dyed fibres. This could be another explanation for the high photostability of the dye extract on the dyed wool and nylon fabrics. In addition, all the dye components in the dye extract contain at least four hydroxyl groups which contributes to strong complexation between the dye and the textile fibre, leading to high technical performance in terms of photostability (Fitz-Binder *et al.*, 2019).

Table 4: Exhaustion percentage of Dye Extract from *R. vomitoria* Leaf on Wool and Nylon Fabrics

Samples	Exhaustion percentage
Wool (non-mordanted)	55
Wool + CuSO ₄	64
Wool + SnCl ₂	64
Wool + FeSO ₄	58
Nylon (non-mordanted)	61
Nylon + CuSO ₄	68
Nylon + SnCl ₂	60
Nylon + FeSO ₄	69

The exhaustion percentage of the studied fabrics was determined based on the absorbance before and after dyeing of the dyebath. According to Table 4, the exhaustion percentage lies in the range of 55 – 68% for wool fabric and 61 – 69% for nylon fabric. From the results, it is obvious that,

there was improvement in the dye up take as the mordants were introduced into the dye liquor. This indicates also that there was good attachment of the dye molecules to the fibres. In general, acid dyes exhibits a higher affinity on nylon fibres than on wool. However, as a result of the numerous free amine groups of

the two fibres 30 – 50 mmol/kg for nylon, compared to 800 – 900 mmol/kg for wool, used for bonding with acid dyes, the two

fibres showed good exhaustion towards acid dyes

Table 5: Colour Coordinate values and Colour Strength of Wool and Nylon Sample Mordanted with CuSO₄, SnCl₂ and FeSO₄ and Dyed simultaneously with *R. vomitoria* Leaf Extract

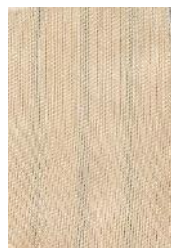
Samples	K/S	L*	a*	b*	H
Wool (non-mordanted)	10.6	42.7	18.3	23.8	65.6
Wool + CuSO ₄	13.8	32.5	19.7	26.5	83.4
Wool + SnCl ₂	12.7	34.8	18.7	23.7	62.6
Wool + FeSO ₄	15.8	30.3	20.8	24.5	86.3
Nylon (non-mordanted)	11.7	41.3	21.9	22.9	67.2
Nylon + CuSO ₄	13.9	39.6	20.8	24.5	69.6
Nylon + SnCl ₂	13.1	36.8	19.5	24.1	63.5
Nylon + FeSO ₄	15.5	30.7	20.7	25.5	73.7

Table 5 shows the colour strength and colour coordinates values of wool and nylon mordanted with CuSO₄, SnCl₂ and FeSO₄ and dyed with *R. vomitoria* leaf extract. From the results, it is observed that K/S values were influenced by the mordant application in comparison with the non-mordanted samples. The improvement in the K/S values is in the order; FeSO₄ > CuSO₄ > SnCl₂ for the wool samples. It also followed the same order in the case of the nylon samples. However, a decrease of L* values was observed after simultaneous mordanting. This could be attributed to a better natural dye diffusion into the wool and nylon fabrics compared with the non-mordanted samples. The L*

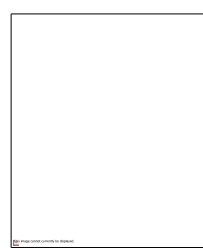
values were found to be higher in the non-mordanted samples indicating a less diffusion and absorption of the dye molecules. The a* values are slightly lower than the b* values indicating that the redness of the samples is little lower than the b* values.



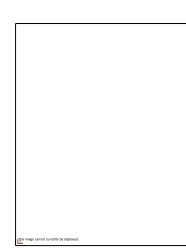
(a)



(b)



(c)



(d)

Figure 11: Scanned images of dyed nylon 6 fabrics using CuSO_4 , SnCl_2 and FeSO_4 as mordants (a) direct dyeing (b) mordanted with CuSO_4 (c) mordanted with SnCl_2 and (d) mordanted with FeSO_4

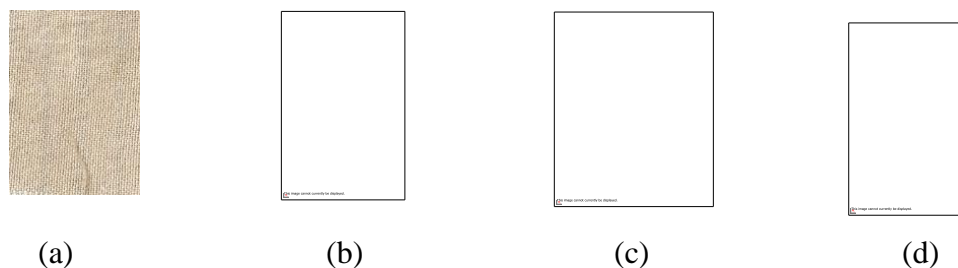


Figure 12: Scanned images of dyed wool fabrics using CuSO_4 , SnCl_2 and FeSO_4 as mordants (a) direct dyeing (b) mordanted with CuSO_4 (c) mordanted with SnCl_2 and (d) mordanted with FeSO_4

The colour hues produced on wool and nylon fabrics by simultaneous mordanting and dyeing with different mordants in comparison with the non-mordanted samples are presented in Figures 11 and 12. It is obvious from the figures that the mordanted samples produced deeper shades compared with the non-mordanted samples. However, among the mordanted samples, it was found that FeSO_4 produced deepest colour shade on both wool and nylon fabrics.

CONCLUSION

In this study, the extraction of natural dyes from *R. vomitoria* leaves was carried out and the dyeability on wool and nylon 6 fabric was investigated. The optimum conditions observed were as follows; extraction temperature of 70°C , extraction time of 5 hours and using a mixture of ethanol and $0.5\text{M H}_2\text{SO}_4$ as solvent. The results of the dyeing properties of the dye extract on nylon and wool, revealed that simultaneous mordanting increased the colour strength and L^* , a^* , b^* values and lowered the K/S values of non-mordanted nylon 6 and wool samples. The light fastness results showed that CuSO_4 , SnCl_2 and FeSO_4 improved the stability of the dye extract on the two dyed textile

fabrics. Interestingly, the results of the HPLC analysis show that the colouring and non-colouring compounds are all acids. In conclusion, it can be established that *R. vomitoria* leaves could be a sustainable source of natural dyes for brown colour shades. This study presents, the extraction, evaluation of colouring and non-colouring components and dyeing characteristics of *R. vomitoria* leaf extract on wool and nylon fabrics. Further research should focus on other non-colouring components such as the alkaloids which also contribute to the final colour and stability of the colour on textile substrates in general, as well as the dyeing properties of this leaf extract on other textile substrates.

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REFERENCES

- Adeel, S., M. Azeem, N. Habib, M. Hussaan, A. Kiran, A. Haji, and W. Haddar. 2023. "Sustainable Application of Microwave-

- Assisted Extracted Tea-Based Tannin Natural Dye for Chemical and Bio-Mordanted Wool Fabric.” *Journal of Natural Fibers* 20 (1): 1–9. <https://doi.org/10.1080/15440478.2022.2136322>. Wang, C, Xu, C, Tian, A, Fu, S
- Adeel, S., S. Ahmad, N. Habib, B. Mia, R. Ahmed, and B. Ahmed. 2022. “Industrial Crops and Products Coloring Efficacy of *Nyctanthes Arborescens* Based Yellow Natural Dye for Surface-Modified Wool.” *Industrial Crops and Products* 188 (PA): 115571. <https://doi.org/10.1016/j.indcrop.2022.115571>.
- Basak, S, Sumanta, KK, Chattopadhyay, SK, Pandit, P, and Malti, S. Green fire retardant finishing and combined dyeing of proteinous wool fabric. *Color Technol*, **2016**; 132: 135-143. doi: 10.1111/cote.12200.
- Beiki, T, Najafpour, GD and Morteza, H. Evaluation of antimicrobial and dyeing properties of Walnut (*Juglans regia* L.) green husk extract for cosmetics. *Color Technol*, **2018**; 134: 71-81. doi: 10.1111/cote.12322.
- Brill, MA. Is CIELAB one space or many! *Color Technol*. **2021**; 83-85. <http://doi.org.10.1111/cote.12486>.
- Commission International de l’Eclairage CIE 15: **2018**. *Colorimetric 4th Edn* Vienna.
- de Boer, PY, Imhot, A and Velikor, K. Encapsulation of colorants by polymers for food applications. *Color Technol*, **2019**; 135: 183-194. doi: 10.1111/cote.12393.
- Dong, C, Lu, Z, Zhang, X, Zhu, P and Li, N. Preparation and dyeing properties of pineapple leaf fiber modified with cationic modifier. *Color Technol*, **2014**; 130: 260-265. doi: 10.1111/cote.12091.
- Ebrahimi, I and Gusti, MP. Extraction of Juglone from *Pterocarya fraximifolia* leaves for dyeing, antifungal finishing and solar UV-protection of wool. *Color Technol*, **2015**; 131: 451-457. doi: 10.1111/cote.12180.
- Ebrahimi, I and Gusti, MP. Extraction of polyphenolic dye from henna, pomegranate rind and *Pterocarya fraximifolia* for nylon dyeing. *Color Technol*, **2016**; 132: 162-176. doi: 10.1111/cote.12204.
- Fitz-Binder, C and Bechtold, T. Extraction of polyphenolic substances from bark as natural colorants for wool dyeing. *Color Technol*, **2019**; 135: 32-39. doi. 10.1111/cote.12378.
- Gasht, MP, Katozian, B, Shaver, M and Kiumarsi, A. Clay nanoadsorbent as an environmentally friendly substitute for mordants in the normal dyeing of carpet piles,

- 2014**; 130: 54-61. doi: 10.1111/cote.12065.
- Grum, F and Bartlesn, J (eds). Optical radiation measurement vol. 2 color measurement New York: Academic Press. **1980/200**.
- Haji, A, Shoushtari, AM and Mirafshar, M. Natural dyeing and antibacterial activity of atmospheric-plasma-treated nylon 6 fabric. *Color Technol*, **2014**; 130: 37-42. doi: 10.1111/cote.12060.
- Hao, S, Wu, J, Huang, Y and Lin J. Natural dyes as photosensitizers for dye-sensitized solar cell. *Solar Energy*, **2005**; 80: 209-214. doi: 10.1016/j.solener.2005-05-009.
- Ian, H. Technical briefing: Natural dyes production and processing advances show potential. *Inter Dyer Finisher Issue 1*: **2014**; 9-11. www.internationaldyer.com
- Islam, MT, Khan, H and Hasan, MM. Aloe vera gel: A new thickening agent for pigment printing. *Color Technol*, **2016**; 132: 255-264. doi: 10.1111/cote.12215.
- Ismal, OE, Ozdogan, E and Yildirim, L. An alternative natural dye, almond shell waste: effects of plasma and mordants on dyeing properties. *Color Technol*, **2013**; 129: 431-437. doi: 10.1111/cote.12047.
- Ismal, OE. A route from olive oil production to natural dyeing: Valorisation dye source. *Color Technol*, **2014**; 130: 147-153. doi: 10.1111/cote.12068.
- ISO 105-B01: 2014. Textile – test for colour fastness part B01: Colour fastness to light Basel: ISO **2014**.
- ISO 105-C10: 2006. Textile – tests for colour fastness part C10. Colour fastness to washing Basel: ISO **2006**.
- ISO 105-X12: 2001. Textile – test for colour fastness part X12. Colour Fastness to rubbing Basel ISO **2001**.
- Link, J. A review: Innovative future for textile finishing. *Inter Dyer & Finisher*, Issue 4, **2019** www.internationaldyer.com
- Manian, AP, Paul R and Bechtold, T. Metal mordanting in dyeing with natural colorants. *Color Technol*, **2016**; 132: 107-103. doi: 10.1111/cote.12199.
- Otutu, JO, Asiagwu, AK, Onyesom, IE, Ebigwai JK and Itoya, PO. Improving the efficiency of Nesogordonia papererifera (Danta) as a natural dye in textile making industry. *Bull Chem Soc Ethiopia* **2019**; 33: 415-424. <http://dxdoi.org/10.4314.bce33i33>.
- Ozomay, M., and M. Akalın. 2020. “Optimization of Fastness Properties with Gray Relational Analysis Method in Dyeing of Hemp Fabric with Natural and Classic Mordant.” *Journal of Natural Fibers* 19 (23): 1–15. <https://doi.org/10.1080/15440478.2020.1837328>.

- Patil, NN and Datar, AG. Application of natural dye from *Ixora coccinea* L. in the field of textiles and cosmetics. *Color Technol*, **2016**; 132: 98-103. doi: 10.1111/cote.12193.
- Prusty, AK, Das, T, Nayak, A and Das, NB. Colorimetric analysis and antimicrobial study of natural dyes and dyed silk. *J. Cleaner Prod*, **2010**; 18: 1750-1758. doi: 10.16/jcpro 2010.06.020.
- Sadeghi-Kiakhani, M, Safapour, S and Mirnezhad, S. Thermodynamic and kinetic studies of the adsorption behavior of the natural dye cochineal on polyamide 6,6,. *Color Technol*, **2019**; 134: 308-314. doi: 10.1111/cote.12342.
- Savvidis, G, Karanikas, E, Nikolaidis, N, Eleftheriadia, I and Satsaroori, E. Ink-Jet printing of cotton with natural dyes. *Color Technol*, **2014**; 130: 200-204. doi: 10.1111/cote. 12067.
- Savvidis, G, Zarkogiann, M, Karanikas, E, Lazavidia, N, Nikoladis, N and Tsatsaroni, E. Digital and conventional printing and dyeing with natural dye annatto: Optimisation and standardization process to meet future demands. *Color Technol*, **2012**; 129: 35-43. doi: 10.1111/cote.12004.
- Saxena, S and Raja, ASM. Natural sources, chemistry, application and sustainability issues. *J Roadmap Sustainable Text and Clothing*, **2014**; 37-79. doi: 10/1007/978-981.065.02.
- Tropical plants database, Ken Fern. The ferns info 2021-02-23. Tropical the ferns in for/viatropical?Id=rouvolfia+v omitoria.
- Wang, C. Extraction of natural dyes from *Alpinia blepharocarlyx* K, Schun for dyeing of silk fabrics. *Color Technol*, **2012**; 129: 32-38. doi:10.1111/1478-4408.20.12.00407.x
- Wang, H, Tang, Z and Zhou, W. A method for dyeing fabric with anthocyanin dyes extracted from mulberry (*Morus rubia*) fruit. *Color Technol*, **2016**; 132: 222-231. doi: 10.1111/cote.12212.
- Zhang, T, Bai, R, Wang, Q, Fan, X, Wang, P, Yuan, J and Yu, Y. A novel strategy to Laccase-mediated coloration of wool fabric. *Color Technol*, **2017**; 132: 65-72. doi: 10.1111/cootr12252.