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Cleaner Production of Bioactive and Coloured Cotton Fabric Using Euclea Divinorum Dye Extract with Bio-Mordants

Čistejša izdelava bioaktivnih in obarvanih bombažnih tkanin z uporabo izvlečka barvila Euclea Divinorum s pomočjo organske čimže

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Abstract
Coloured textile products are more marketable, and are therefore always in higher demand. This has increased the use of synthetic dyes in the textile industry, thus raising environmental pollution associated with synthetic dyes. Natural dyes have been shown to be suitable alternatives. However, the use of metallic mordants during dyeing means the process is not eco-friendly, hence the need to develop bio-mordants that can be used as alternatives to some toxic metallic mordants. In this study, the effects of bio-mordants on the dyeing properties of *Euclea divinorum* Hiern (Ebenaceae) dye extract were assessed using different mordanting methods on cotton fabric. Dyeing characteristics were evaluated in terms of colour fastness and colour strength. Antioxidant textile finishing properties of the natural dye on cotton fabric was determined using the 2,2-diphenyl-1-picrylhydrazyl radical (DPPH) method. The bio-mordants improved the colour strength for dyed cotton fabric from 0.612 to 0.863 and 0.911 for the cotton fabric mordanted with mango and rosemary, respectively. This study identified an important basis of suitable bio-mordants that can be applicable when dyeing cotton fabric with *E. divinorum* natural dye. In addition, the good antioxidant activity of 72.5% indicates that *E. divinorum* dye extract is a promising agent for the future development of bioactive, protective and health textile fabric.
Keywords: natural dye, bio-mordants, antioxidant activity

Izvleček
Obarvani tekstilni izdelki se dobro tržijo in po njih je vedno veliko povpraševanja. Zato so se sintetična barvila v preteklosti veliko uporabljala, to pa je vodilo tudi v povečano onesnaževanje okolja. Naravna barvila so primerna alternativa, vendar je raba kovinskih čimž pri barvanju z naravnimi barvili okolju neprijazna. Iz tega izhaja potreba po razvoju organskih čimž, ki bi lahko zamenjale strupene kovinske. V tej raziskavi so bili ocenjeni učinki organskih čimž na lastnosti obarvane bombažne tkanine z barvilnim ekstraktom iz Euclea divinorum Hiern (Ebenaceae) pri uporabi različnih metod čimžanja. Lastnosti obarvanja so bile ocenjene glede na barvno obstojnost in globino obarvanja.

Antioksidativne lastnosti bombažne tkanine, obarvane z naravnim barvilom, so bile določene z metodo 2,2-difenil-1-pikrilhidrazil radikala (DPPH). Organske čimže so izboljšale globino obarvanja obarvane bombažne tkanine z 0,612 na 0,863 oziroma 0,911 za tkanini, čimžani z mangom oziroma rožmarinom. Ta raziskava je pomembna osnova pri izbiri primernih organskih čimž za barvanje bombažnih tkanin z naravnim barvilom E. divinorum. Poleg tega dobra antioksidativna aktivnost 72,5 % kaže tudi na to, da je ekstrakt barvila E. divinorum obetavno sredstvo za nadaljnji razvoj tekstilij z bioaktivnimi, zaščitnimi in zdravilnimi lastnostmi.
Ključne besede: naravno barvilo, organska čimža, antioksidativno delovanje

1 Introduction

The textile dyeing process is among the leading polluting industrial processes due to use of the toxic synthetic dyes [1]. An environmentally friendly textile dyeing process can be achieved by substituting certain toxic synthetic dyes with natural dyes that have been shown to provide value added to textiles [2–4]. The process of manufacturing synthetic dyes, together with the associated textile application procedures, discharge toxic wastewaters that require costly resources for complete treatment, and thus find their way into the environment [5–7]. Environmental awareness has led to a recent shift to natural dyes, which in turn has stimulated increased research on natural dyes [8, 9].

Most natural dyes exhibit poor to moderate colour fastness on fabric, which has been a major limitation in their use in the textile industries. As a result, natural dyes are used together with mordants that help fix them on textile material. Commonly used mordants include metallic salts that fix dye molecules to the fabric through a combination that involves the dye molecules, the fabric molecules and the metallic ions that form insoluble precipitates [10]. Examples of metallic mordants are potassium aluminium sulphate, stannous chloride, potassium dichromate, copper sulphate, ferrous sulphate, and others [11].

The main purpose of metallic mordants during natural dyeing is to enhance the affinity of the fabric to the dye molecules since the metal ions form coordination complexes that allow the attachment of the dye to the textile fabric [12]. In addition, the use of metallic mordants during natural textile dyeing offers a wider spectrum of shades from a single natural dye extract [13]. The mordanting step in the natural dyeing process is very crucial, especially when dyeing cotton fabric because it lacks functional groups, such as carboxylic and amino acid groups present in other textile fabrics that act as positions where the dye molecules attach [10, 14]. On the other hand, most of these metallic mordants have been found to be harmful and a small amount of the metallic ion takes part in the fixation of the dye to the fibre, hence a huge percentage of it finds its way into the environment [15]. Global restrictions on production industries regarding the use of toxic substance with the aim of curbing increasing environmental pollution have stimulated research on the development of environment-friendly mordants that can be used as alternatives to poisonous metallic mordants [16].

Bio-based mordants are gaining popularity since they are obtained from nature and thus facilitate a suitable approach for making the natural dyeing process a part of green chemistry. Bio-mordants are basically natural substance that are rich in tannins or metal ions, and are mainly obtained from plants. Examples include tartaric acid, tannic acid [17], tamarind seed coat tannins [18], mango bark [19] extracts from myrabolan, pomegranate rinds, banana leave ash, rosemary plant, rhizomes of turmeric, bark of acacia, guava, etc. [7, 20, 21]. In order to lessen the environmental hazards caused by some metallic mordants, there is need to shift to bio-mordants, which will help in realizing the aim of producing ecologically coloured textile materials [22–24].

In addition to colours obtained from different natural dye extracts, functional textile finishing properties have also been achieved. These include antimicrobial [25–29], antioxidant [30, 31], deodorizing [32] and UV-protective properties [33–35]. The discovery of these additional functional properties of textiles brought about by natural dyes is important in the development of healthy and clean textile materials.

Antioxidant activity is one of the most significant properties of bioactive textile since it protects textile material from damage and safeguards the human skin from inflammation and aging due to oxidative stress caused by free radicals [36]. The human skin is continuously exposed to ionizing radicals, which are the primary cause of skin damage and the associated diseases [37]. Ultra-violet radiation is the main source of free radicals in the environment, and accumulate to such a level that the antioxidants in the skin cannot neutralize them, leading to oxidative stress, which may cause skin cancer and other diseases [38].

Antioxidants, also referred to as free radical scavengers, are substances that react with free radicals and neutralize them, thus counteracting their harmful effects. Studies have shown that antioxidant molecules such as phenols and flavonoids have good anti-cancer activity against skin cancer [39]. The antioxidant activity of textile fabric is achieved through the deactivation of very reactive and destructive radicals in the environment, such as oxygen and nitrogen radicals [40]. It has been shown that natural dyes are suitable agents for achieving antioxidants properties in textile material because they are not toxic and do not irritate the skin [41–43]. Clothes are in direct contact with the skin. For this reason, the antioxidant activity of textile materials is important in the development of bioactive, healthy textile fabrics.

The antioxidant activity of *Euclea divinorum* aqueous root extract has been studied and was found to be between 74.5–82.5% DPPH (2,2-diphenyl-1- pic rylhydrazyl) [44]. The objective of this study was to determine the effects of bio-mordants on the dyeing properties of *E. divinorum* natural dye extract, and to explore its potential as an antioxidant finishing agent for cotton fabric. The durability of the antioxidant properties of textile materials after washing was also determined.

2 Material and methods

2.1 Materials

E. divinorum root bark was collected in Nandi County in Kenya (latitude 0° 01' 59.5" S and longitude 35° 3' 17.3" E). Commercially bleached, plain woven cotton fabric with 20 ends/cm, 13 picks/cm, 29.4 tex (Nm 34) warp count, 29.4 tex (Nm 34) weft count and a mass per unit area of 97.1 g/m^2 was purchased from a textile factory in Eldoret, Kenya, while 2,2 -diphenyl-1-picrylhydrazyl radical (DPPH) was used for antioxidant evaluation.

2.2 Extraction

E. divinorum root bark was washed and dried using sunlight, and then ground into powder form using an electric grinder. The natural dye as then extracted using distilled water. The extraction conditions used were: temperature of 84 °C, time of 146 minutes and M:L 7.5:100, as previously determined [45]. Conical flasks were used as extraction containers and a water bath was used to regulate the temperature. After extraction, the extracts were allowed to cool and filtered using filter paper.

2.3 Bio-mordanting

The mango (*Mangifera indica*) bark bio-mordant was extracted using the procedure described by [24], where extraction was performed at 90 °C for one hour using 75g/L of the sample in distilled water. Rosemary (*Rosmarinus officinalis*) was purchased from a local market, cut into small pieces, dried under the sun and then ground into powder. A mixture of 20g/L of rosemary powder in distilled water was used to extract the mordant at 100 °C for one hour [46]. Bio-mordanting was carried out using the pre-, meta- and post-mordanting methods. For the pre-mordanting method, the wet cotton fabric was immersed in the solution of the mordant using a material to-liquor ratio of 1:50 at 60 °C. Continuous stirring was maintained for one hour, followed by dyeing. In meta-mordanting, the wet cotton fabric was immersed in a flask containing the solution of the mordant and the dye extract using a material to-liquor ratio of 1:50 at 60 °C for one hour. For the post-mordanting method, the previously dyed cotton fabric was placed in flask containing the solution of the mordant using a material to-liquor ratio of 1:50 at 60 °C for one hour [24].

2.4 Dyeing

The cotton fabric was cut into equal sizes of 1g. Wetting of the fabric was performed using 5g/L of non-ionic detergent for 30 minutes prior to dyeing. *E. divinorum* aqueous dye extract was used to prepare the dyebath using a material-to-liquor ratio of 1:40 [1]. After dyeing, the dye bath was allowed to cool. The dyed samples were then washed with cold water to remove the unfixed dyestuff and subjected to soaping with a 2 g/L soap solution, followed by rinsing with water and air drying.

2.5 Colorimetric measurements

The colour characteristics of the dyed samples were measured with a Spectro -Flash X-rite SP62 spectrophotometer, using a D65 source of light and 10° standard observer. The CIELAB coordinates were measured. The un-dyed cotton fabric was used as the blank. The relative colour strength (K/S) values were determined using the Kubelka–Munk equation (equation 1).

$$K/S = \frac{(1 - 0.01\,R)^2}{2 \times 0.01\,R} \quad (1)$$

where K represents the absorption coefficient, S represents the scattering coefficient and R represents the minimum reflectance of dyed substrate samples.

2.6 Colour fastness

The ability of cotton fabric to retain dye during washing and rubbing, and when exposed to light and perspiration was determined using the relevant standard colour fastness tests. These were conducted according to ISO 105-C02:1989, ISO 105 A02:1993, ISO 105-X12:2000 and ISO AATCC-2009 for washing, exposure to light, rubbing and perspiration fastness, respectively, with some changes, where the grey scale used to rate the colour fastness was between one and five, with five representing the best fastness [25].

2.7 Antioxidant activity

The antioxidant activity of the pure and dyed cotton fabric was evaluated using a DPPH radical scavenging assay [41]. A total of 2.54 cm^2 of the pure and dyed cotton fabric were separately immersed in a test tube containing 50mL solution of 2,2-diphenyl-1-picrylhydrazyl radical (DPPH) in methanol (0.15 mM) and mixed thoroughly. The samples were incubated in the dark at room temperature for 30 minutes. The absorbance of the solution was measured at 517 nm using a UV-Vis spectrophotometer. The percentage antioxidant activity was calculated according to equation 2.

$$A = \frac{A_{control} - A_{sample}}{A_{control}} \times 100\ (\%) \quad (2)$$

where A represents antioxidant activity, $A_{control}$ represents the initial absorbance of the DPPH solution and A_{sample} represents the absorbance of the remaining DPPH solution after incubation with the sample.

The durability of the antioxidant activity of the dyed samples was assessed by subjecting the dyed fabric to washing cycles, with the antioxidant activity determined after every washing. The washing tests were performed by putting the samples into a washing solution comprised of commercial detergent (2 g/L) with a material-to-liquor ratio of 1 : 50. The antioxidant activity was determined after the 1^{st}, 5^{th} and 10^{th} washing cycles [41].

3 Results and discussion

3.1 Colorimetric analysis

The colour characteristics of the dyed samples were measured and are presented in Table 1. In terms of lightness (L^*), the mango and rosemary bio-mordants increased lightness from 63 to 66.33 and 67.27, respectively, providing lighter shades of brown. Similarly, the intensity of the colour increased as indicated by an increase in the value of C^*. The ability of mango and rosemary bio-mordants to form lighter shades indicates that they can be used as substitutes for metallic mordants that are used to brighten the colour of the dye on the fabric, such as alum and tin, an observation that has been reported previously in literature [46]. The polyphenols in the rosemary bio-mordants are responsible for the increased dye absorption by the cotton fabric, which leads to increased colour strength [20]. Pre-mordanting method for both mordants showed the best colour strength, as has been observed in other studies [19].

3.2 Colour fastness

Colour change in terms of washing and perspiration fastness was in the range of 4–5, except for the pre-mordanted fabric using mango and rosemary bio-mordants, which was 5 (Table 2). The pre-mordanting method allows the poly-phenols to attach to the cellulosic fibre then act as bridge between the fibre and the dye molecules, thus improving the ability of the dye to fix to the fabric, which in turn enhances the fastness properties of the dye [20]. The excellent light fastness throughout the dyeing process indicates that the dye-fabric complex formed is resistant to fading during the exposure to ultraviolet radiation [1]. Generally, the colour fastness of the cotton fabric dyed with *E. divinorum* dye extract was very good and is suitable for application in the textile dyeing industry.

3.3 Antioxidant activity

Antioxidant activity as a percentage is measured by the reduction of the absorbance of DPPH. When the phenolic hydroxyl donates a proton to the DPPH radical, the solution is decolorized and its absorbance is reduced [49]. The antioxidant activity of the assayed

Table 1: Colorimetric values and colour strength of the dyed and mordanted cotton fabric

Method	Mordant	L^*	a^*	b^*	C^*	$H°$	K/S	Shade
-	-	63.47	+4.63	+16.86	17.53	74.53	0.612	
Pre	Mango bark	67.27	+9.66	+21.01	23.13	65.32	0.863	
	Rosemary	66.33	+8.65	+18.80	20.72	65.19	0.911	
Meta	Mango bark	64.59	+11.82	+19.55	22.85	58.84	0.708	
	Rosemary	64.74	+10.65	+17.27	20.31	58.30	0.720	
Post	Mango bark	66.14	+9.97	+19.18	21.64	62.51	0.691	
	Rosemary	65.76	+7.26	+16.72	18.22	66.58	0.724	

Table 2: Colour fastness of the dyed fabric using different methods of mordanting

Method	Mordant	Washing fastness		Rubbing fastness		Perspiration fastness		Light fastness
		C.C [a)]	C.S [b)]	Dry	Wet	C.C	C.S	
	Without	4–5	5	5	5	4–5	4–5	5
Pre	Mango bark	5	5	5	5	5	5	5
	Rosemary	5	5	5	5	5	5	5
Meta	Mango bark	4–5	5	5	5	4–5	5	5
	Rosemary	4–5	5	5	5	4–5	5	5
Post	Mango bark	4–5	5	5	5	5	5	5
	Rosemary	4–5	5	5	5	5	5	5

[a)] colour change, [b)] colour staining

cotton fabric samples is shown in Table 3. Dyeing with the *E. divinorum* dye extract increased the antioxidant activity from 26.9% (undyed cotton) to 72.5% (dyed cotton), which can be attributed to the molecules adsorbed by the cotton fabric from the dye extract, which consequently imparts the radical scavenging activity into the fabric. The antioxidant activity of aqueous root extracts of *E. divinorum* was found to be between 74.5–82.5% DPPH [46]. It was also noted that the antioxidant activity of the fabric samples bio-mordanted with mango (82.4%) and rosemary (85.3%) was higher than that of the un-mordanted (72.5%) fabric, which could be due to the additional activity from the bio-mordant extracts that have been shown to have good antioxidant activity [50]. The mango bark extract comprises polyphenols that are responsible for the radical scavenging ability of the extract [48], [51].

Table 3: Antioxidant activity of the sample fabric

Sample	Antioxidant activity (%)
Undyed cotton	26.9
Dyed	72.5
Mango mordanted	82.4
Rosemary mordanted	85.3

The durability of the antioxidant activity after washing cycles was as indicated in Figure 1. A subsequent

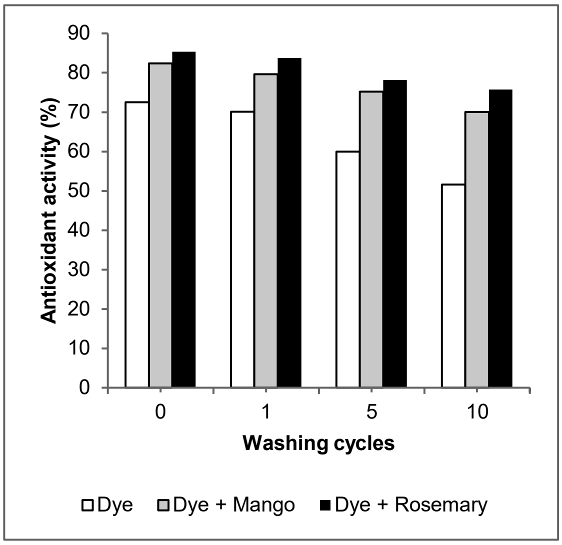

Figure 1: Antioxidant activity of dyed and mordanted cotton fabric after washing cycles

reduction in the antioxidant activity of the dyed samples was observed, and was found to be similar to what has been reported in other studies. However, the reduction in antioxidant activity became minimal after the 5th washing cycle, which is a significant attribute for this extract and can be ascribed to its good fastness properties [36]. The rate of reduction was lower in the mango and rosemary mordanted fabric than in the unmordanted fabric due to the enhanced fastness of the dye to the fabric [41]. The rosemary mordanted fabric showed good antioxidant durability (above 80%) after the 10th washing cycle), indicating that the elements responsible for bioactivity were not affected by the washing process and thus remained very active [36].

4 Conclusion

The bio-mordants improved the colour strength from 0.612 to 0.863 for the dyed cotton and to 0.911 for the cotton fabric mordanted with mango and rosemary, respectively. In addition, the bio-mordants modified the colorimetric values, resulting in different shades of brown colour on the cotton fabric. All the fastness properties in this case showed good results of 4–5 and higher. This study thus shows that bio-mordants provide the opportunity to make natural dyeing an eco-friendly process as suitable alternatives to toxic metallic mordants, and thus need to be exploited in the textile industry. The dye extract imparted the anti-oxidant activity onto the cotton fabric, which showed a good radical scavenging activity of between 72.5% and 85.3%. The antioxidant activities were found to be durable, even after several washing cycles. As a result, the dye extract is proposed as an agent for the future development of bioactive, protective and healthy textile fabrics.

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