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Application of Eco-friendly Natural Dye from *Aerva sanguinolenta* on Cotton Yarn Using Biomordant

Sohini Mukherjee^{1*} and Shyamala Kanakarajan²

¹Research Scholar, Department of Plant Biology and Plant Biotechnology, Ethiraj College for Women, Chennai-600008, Tamil Nadu.

²Associate Professor (Retired), Department of Plant Biology and Plant Biotechnology, Ethiraj College for Women, Chennai-600008, Tamil Nadu.

ABSTRACT

Dyeing clothes with natural dyes, started many years ago. The recent prohibition on use of synthetic dyes in several countries has again increased the interest for non toxic, biodegradable natural dyes. In the present study, an environment friendly approach was developed to impart colour to yarn. The natural dye was extracted from *Aerva sanguinolenta* leaves. Effect of time, temperature and pH on extraction has been studied to optimise the extraction method. Alum, less toxic metal mordant and bio mordant of *Peltophorum pterocarpum* bark were used to bind the dye to the yarn. Extraction of biomordant from the bark and mordanting process has been standardised using various standard parameters. Yarn was dyed with mordants using three different mordanting techniques, i.e., pre mordanting, meta mordanting and post mordanting. The mordanting technique was found to influence the result of dyeing process. Effect of dyeing time, dyeing temperature, pH, and concentration were studied to standardise the dyeing method. As mordants have significant effect on colours of dyed yarn, effect of mordant on fastness properties of dye has been analysed. Investigations have been done to know pigments present in it. Results showed presence of betalain as pigment that gives reddish colour. It is concluded that the dye extracted from the *Aerva sanguinolenta* leaf can be successfully used as an alternative source in place of harmful synthetic dyes.

Keywords: Natural dye, *Aerva sanguinolenta*, biomordant, fastness property, Betalain.

*Corresponding author

INTRODUCTION

Dyeing is an ancient art. Evidence of practice of dyeing was found during Bronze age in Europe. Primitive dyeing techniques include sticking crushed plant to fabric or rubbing pigment to fabric. With time these method of dyeing became more sophisticated by using natural dyes extracted from plant materials [1,2].

In 1856, a basic synthetic dye 'mauve' was accidentally discovered by William Perkin. This discovery declined the use of natural dye because of its drawbacks like inconsistency of colours, less availability, lack of fastness etc [3].

However, recently there has been increase in use of natural dyes in textile industry due to hazardous, toxic and carcinogenic effect of synthetic dyes and also because of a recent ban imposed by Germany, USA, India and some other European countries on the use of some synthetic dyes [4]. Demand for natural dye is increasing in textile industries for it's eco-friendly, nontoxic nature.

Natural colorant from different plant parts can be extracted through various methods including aqueous extract, alcoholic or different solvent extract and enzyme assisted extraction [5].

Using natural dyes in textiles has certain limitations like low dye uptake and inadequate fastness properties which can be solved using chemicals called mordant [6,7]. Mordants are metal salts that can increase the affinity between yarn and dye. Generally copper sulphate, ferrous sulphate, chromium sulphate, zinc sulphate and alum are used to fix the dye to yarn. Alum is environment friendly in comparison to other metal salts.

In recent past, biomordants can be another alternative to chemical mordants to solve the fastness problem of natural dyes. The use of biomordant also can be a possible eco-friendly approach to improve the dye uptake and colour fastness [8,9,10].

To colour any substrate it is important to remove all dirt, either natural or acquired, so that colour can stick or penetrate to the substrate efficiently. Therefore, yarn should be pre-treated before colouring [11].

Aerva sanguinolenta belongs to genus *Aerva*, family *Amaratheceae* and Order *Caryophyllales*. Its habitat ranges from eastern Asia to Southern China, Palisthan, India, Bhutan, Nepal, Maynamar, Malaysia, Indonesia etc. It grows mainly as perennial herbs.

Betalain is red - violet in colour, water soluble, nitrogen containing pigment. Presence of betalain is characteristic feature of order *Caryophyllales*. They divided into two groups, i.e., betacyanin (red- violet in colour) and betaxanthin (yellow in colour). Betalain gives bright colour to flowers like *Mirabilis* sp [12,13] *Portulaca* sp [14], food items such as *Beta vulgaris* (beetroot) [15], *Opuntia* (Cactus pear)[16]. The main chromophore of betalain is betalamic acid [17].

The objective of the present study is to extract natural dye from *Aerva sanguinolenta* leaves and its application on pre treated cotton yarn in combination with biomordant and chemical mordant alum. Light, wash and perspiration fastness have been analysed to check the colour's stability. Further, identification of the pigments responsible to dye the yarn was carried out.

MATERIALS AND METHOD

Collection of plant material:

The leaves of *Aerva sanguinolenta* was used to extract the dye. The leaves were collected from Chennai.

Collection of yarn:

Cotton yarn was purchased from TANSPIN, Erode, Tamil Nadu.

Optimisation of extraction of dye:

In the present study, extraction has been done using water to avoid solvent toxicity. To optimise the colorant extraction, different parameters have been maintained. Fresh leaves of *Aerva sanguinolenta* were collected, washed and cut into small pieces. Further, it was soaked in distilled water for 30min at room temperature. Sample to Liquor ratio of 1:10 was maintained. Different parameters have been taken into account to optimise extraction of dye such as pH (5,7,9), Temperature (40°C - 100°C) and time (15 min, 30 min, 45min, 60min). On comparison with standard calibration curve optimum pH, temperature and time were fixed for further analysis.

Pre-treatment of Cotton Yarn:

Dirts, impurities and natural colours of cotton yarn should be removed to increase the dye uptake in cotton for making it brighter. Initially the yarn was washed in distilled water, after which it was boiled in sodium hydroxide solution. Yarn to NaOH ratio at varied range, i.e., 1:1, 1:2, 1:4 and 1:6 were screened to optimise the concentration of sodium hydroxide solution for pretreatment.

Optimisation of Dyeing:

The pretreated cotton was dyed using *Aerva sanguinolenta* leaf extract. The dyeing process has been standardised using different parameters like dyeing time (15min, 30min, 45min, 60min), Temperature (30°C, 50°C, 70°C, 90°C), pH (5, 6, 7, 8, 9), and yarn to dye ratio (1:20, 1:40, 1:60). On the basis of the percentage of dye exhaustion, optimum time, temperature, pH, and yarn to dye ratio were selected.

Measurement of dye exhaustion:

Percentage of dye exhaustion,

$$\% E = \frac{C_1 - C_2}{C_1} \times 100$$

Where, C1 and C2 are concentration of dye bath before and after dyeing process respectively.

Dye concentration was checked at specific wavelength (λ_{max}) in UV Vis Spectrophotometer. Dye exhaustion is the amount of dye which diffused to yarn (Jigar P R *et al.*, 2013).

Mordanting:

Chemical and biomordant have been used to improve fastness property of the dye. In this study, chemical mordant alum and in case of biomordant, tannin extracted from bark of *Peltophorum pterocarpum* was used. Three different mordanting processes have been carried out with both chemical and biomordant, i.e., pre mordanting, meta mordanting and post mordanting.

Standardisation of chemical mordanting:

Different concentration of alum ranging from 1-5% o/w (on weight of yarn) has been screened. Wet yarn sample was brought into mordant solution and the temperature was maintained at 80°C for 1 hour. Further, the yarn was squeezed and soaked in dye bath. As few mordants are sensitive to light, therefore yarn has to be immediately dyed after mordanting. Similar process has been followed for pre, meta and post mordanting.

Biomordant:

Different extraction methods were screened to extract tannin efficiently.

Extraction of tannin:

Plant sample to liquor ratio was fixed at 1:15 ratio. Extraction was carried out with distilled water, distilled water with 1% and 5% sodium carbonate solution. Distilled water to sodium carbonate was maintained at 2:1 ratio. The setup was kept for boiling for 1 hour. Further, the extract was filtered and was used as biomordant. Presence of tannin was found through phytochemical analysis.

Standardisation of biomordanting:

Different concentrations (50% to 100%), time (30min to 60min), temperatures (40°C to 100°C) were maintained for biomordanting process of cotton yarn.

Colour fastness:

Dyed cotton yarn was analysed for colour fastness to light (ISO 105 B02-2013), wash (ISO:105 C-10:2006) and perspiration(ISO 105 E04-2013) using standard ISO method.

Identification of Pigment:**Phytochemical analysis:**

Initial analysis of chemical constituents of dye solution was carried out by phytochemical analysis [18, 19, 20] following standard procedure.

Thin layer chromatography:

Silica gel coated plate was used to separate pigments. Different Solvent systems such as methanol: chloroform, ethanol: water and 1% aqueous HCl were used. Finally Rf value has been calculated.

UV Visible Spectroscopy:

The extract was analysed in UV Visible Spectrophotometer SHIMADZU 1650PC at wavelength of 400-800nm and the corresponding peaks were recorded.

FTIR Spectroscopy:

The extract was mixed with KBr (1:100) using a mortar pestle and compressed into a thin pellet and the spectrum was recorded on SHIMADZU IR Prestige - 21FT-IR with a diffused reflectance mode (DRS-8000) attachment. All measurements were carried out in the range of 500-4000 cm^{-1} at a resolution of 4cm^{-1} . This range was used to study the fundamental vibration and associated rational vibration structure.

RESULTS AND DISCUSSION

The initial screening showed the leaves of *Aerva sanguinolenta* (Figure: 1) yield reddish colour (Figure: 2) which can be used as textile dye. Therefore, detailed study was carried out to standardise the extraction of dye and also dyeing of cotton yarn with the help of mordants.



Fig 1: *Aerva sanguinolenta*



Fig 2: *Aerva sanguinolenta* leaf extract

Optimisation of extraction:

Dye extracted from leaves majorly contain polyphenol chromophore and the dye extraction is better in aqueous medium [21, 22]. Hence, in the present study, dye has been extracted from *Aerva sanguinolenta* leaves using aqueous solvent.

Optimisation of aqueous extraction of dye was conducted with the help of different parameters i.e. time, temperature and pH. On comparing the absorbance of the sample with the calibration curve it can be concluded that pH 7 is optimum pH for dye extraction. However, pH has less effect on shades of extracted dye on comparison to time and temperature. No significant change in dye colour was observed with the change of pH. Jackman and Smith (1996), Stintzing and Carle (2004)[23,24] also reported that colour of betalain shows stability at pH 3-7. Hence, *Aerva sanguinolenta* leaf extract may contain betalain pigment.

Furthermore, the optimum extraction was standardized at 70° C for 30min. The material to liquor ratio was maintained at 1:10 ratio. Temperature and time are more crucial in the process of dye extraction than any other parameters.

Mordanting of Cotton Yarn:

Yarn to NaOH ratio was optimised at 1:4 concentration of sodium hydroxide solution for pretreatment of cotton yarn. Tannin helps to retain natural dye to cotton [25]. So, extraction of biomordant tannin from *Peltophorum pterocarpum* has been standardised. Temperature and time for biomordanting was optimised at 60°C for 60 min (1 hr) and concentration of biomordant has been selected as 50% on weight of yarn (owy). Alum as non toxic metal salt plays an important role to attach dye to yarn. Further, mordanting of yarn with alum has been optimised at 5% on weight of yarn (owy) and mordanting was done at 80°C for 1 hour. Alum can form coordination complex with dye and chelate successfully [26]. As dye exhaustion is more in mordanted fabric than unmordanted [26], mordanting plays a significant role on dyeing and dyed yarn. In the present study, pre, meta and post mordanting was carried out. Pre and meta mordanting both showed good ability to dye cotton, however, premordanting gave better result than meta and post mordanting. Similar result was found by M Mahabub Hasan *et al.*(2015)[26]. Therefore, pre mordanting can be recommended for its efficiency to produce more active sites on yarn, where dye molecule can be attached successfully and form yarn-mordant-dye complex [27].

Effect of time on dyeing:

The effect of time on dyeing is shown in figure 3 & 4. The dye uptake increased with increase of time. There was a sharp increase of dye uptake from the time period of 15min to 30min, following which, amount of dye uptake was reduced. Therefore, 45min was considered as optimum time for alum and 60min for biomordant. Similar result was supported by M Mahabub Hasan, 2015[26]. After 1 hour, desorption of dye could decrease the dye uptake due to overheating [26].

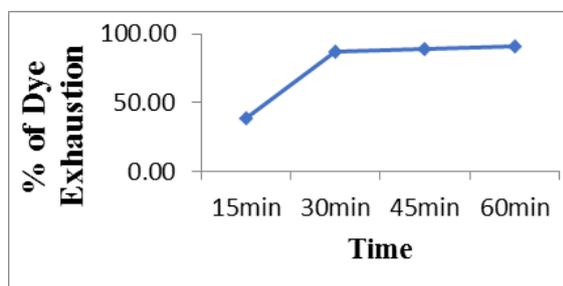


Fig 3: Effect of Time on dye exhaustion using alum

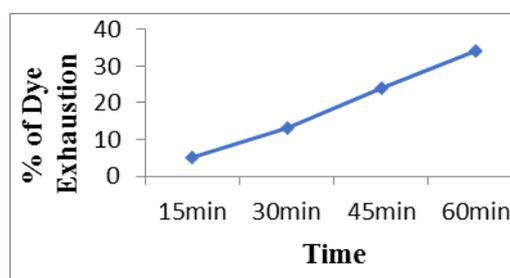


Fig 4: Effect of time on dye exhaustion using biomordant

Effect of temperature on dyeing:

Figure 5 & 6 shows effect of temperature on dye exhaustion of cotton yarn. Maximum dye uptake was found at 70°C for both alum and biomordant. Similar result was found by S.Ali *et al.*,(2009) [28]. There is

increase of dye uptake from range 30°C to 70°C, however, after 70°C, the dye uptake found to be decreasing. The loss of thermal stability of the pigment at high temperature might be the reason for the decreased level in dye uptake [29].

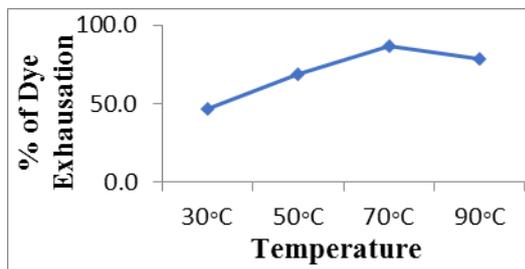


Fig 5: Effect of Temperature on dye exhaustion using alum

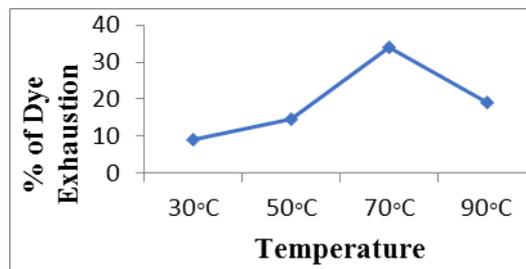


Fig 6: Effect of Temperature on dye exhaustion using biomordant

Effect of pH on dyeing:

pH has a significant effect on dye adsorption process. Effect of pH has shown in figure 7 & 8. pH 6 showed best result for biomordant. Dyeing cotton yarn in acidic pH shows better result than alkaline pH. Same result has found by Chunxia Wang *et al.*, 2012 [27]. According to him higher pH oxidise the conjugate structure of natural dye which decrease the dye ability of natural dye. In case of alum, pH was standardised at 7, where dye exhaustion was highest by cotton yarn. Similar result was reported by S. Ali *et al*, 2009[28].

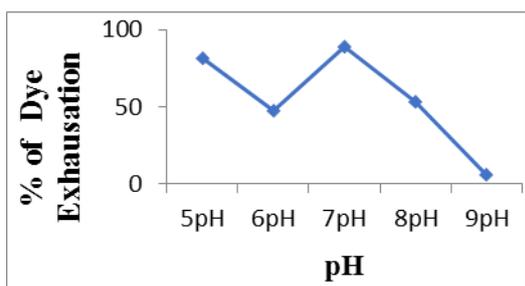


Fig 7: Effect of pH on dye exhaustion using alum

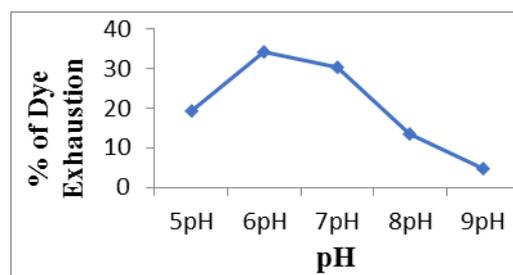


Fig 8: Effect of pH on dye exhaustion using biomordant

Effect of concentration on dyeing:

Figure 9 & 10 showed effect of material to liquor ratio on dyeing of cotton yarn. 1:20, 1:40 and 1:60 are different material to liquor ratios selected for optimisation. Both 1:40 and 1:60 showed almost equal result and 1:40 ratio was selected as optimum in case of alum. However, for biomordant 1:60 was considered as standardised concentration for dyeing as dye exhaustion was maximum in case of 1:60. Percentage of dye uptake was similar in case of 1:20 and 1:40. With increase of dye concentration, dye uptake increase. Because if dye concentration increases concentration gradient increases, which help to uptake more dye in dye bath. Similarly, less concentration results less dye uptake [30].

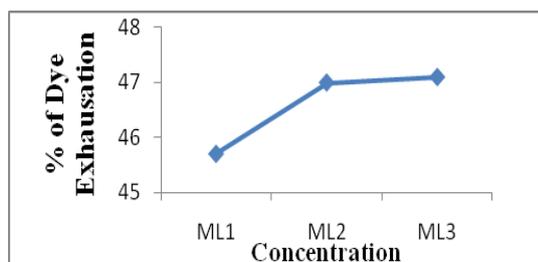


Fig 9: Effect of concentration on dye exhaustion using alum

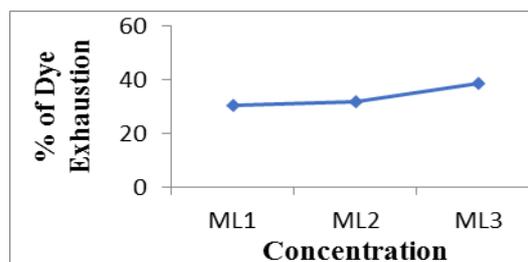


Fig10: Effect of concentration on dye exhaustion using biomordant

Fastness Properties:

Fastness properties of dyed yarn with both alum (Figure: 11) and biomordant (Figure: 12) have been tested through standard methods in laboratory. Further, depending on best dye uptake and fastness properties, dyed cotton yarn treated with biomordant was tested using standard ISO method.



Fig 11: Dyeing with *Aerva sanguinolenta* leaves with alum



Fig 12: Dyeing with *Aerva sanguinolenta* leaves with biomordant

There is a relationship between complex formation of dye and mordant with dye removal under washing solution, perspiration solution and also under light [31]. This complex forms mainly through hydrogen bond. Stronger the bond, better fastness properties of dyed yarn is obtained.

Colour fastness to wash showed fair to good result however the light fastness and perspiration fastness of treated cotton yarn in both acidic and alkaline treatment showed good result (Grey Scale: 3-4). Change in colour is less in case of light and perspiration than wash fastness.

Characterization of Dye:

Phytochemical screening of plant extract has been done to simplify the characterization of dye [20]. Phytochemical analysis showed presence of alkaloids, carbohydrates, terpenoids, phenols, flavonoids, tannins, proteins and amino acids in the extract. Presence of flavonoids, tannins, terpenoids, reducing sugar in the extract considered as significant characters for textile dyes [20].



Fig 13: TLC

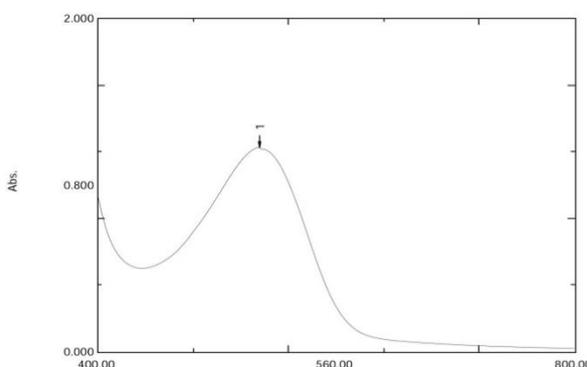


Fig 14: UV Vis Spectroscopic analysis of Dye extract

Thin layer chromatography (TLC) generally use to identify colour components in textile dye[32].Figure 13 showing TLC plate of *Aerva sanguinolenta* leaf extract, where mobile phase is 1% aqueous HCl. The reddish purple colour is the separated pigment. Rf value of the leaf extract is 0.914 .The Rf value of TLC in 1% aqueous HCl 0.916 proves the presence of betacyanin [33]. Thus the result is indicating the presence of betalain in the extract.

Any dyes absorb visible light in different specific wavelength to show the presence of hue [7]. In this study, figure 14 showing the visible spectrum of leaf extract ranging from 400-800nm, where λ_{max} is 536 nm.This result can be supported by K.K.Woo, 2011[34] and A.Guesmi *et al.*, 2012[29].Harborne (2007)[33] reported visible spectrum of betalain gives maximum absorbance in the range of 532-554 nm.

Further analysis was done to identify the major functional groups present in the dye solution by FTIR analysis [35].Peaks between 1653-918 cm^{-1} supports the presence of betacyanin in leaf extract [36].Figure 15 shows five distinct peak at 1075.36, 1079.22, 1132.26, 1158.3 and 1626.06 which comes in between the above mentioned range. Stretches around 1651-1667 cm^{-1} implies the presence of carbonyl (C=O) and stretches at 3406 cm^{-1} represents presence of hydroxyl group (-OH) which corresponds to CO=OH group of betalains pigments. Similar result was found by Shiv Narayan Amit Kumar; 2013[37] and Siyafinar *et al.*, 2015 [38].

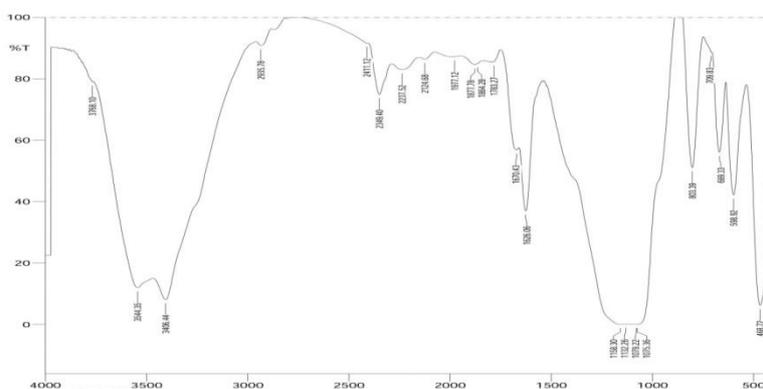


Fig15: FTIR analysis of dye extract

CONCLUSION

From the present work, it can be concluded that Red-violet natural dye can be efficiently extracted from *Aerva sanguinolenta* leaves. The extracted dye could be successfully attached to cotton yarn with the combination of biomordant and chemical mordant (alum).However, Biomordant showed better result than alum in case of light, wash and perspiration fastness properties and premordanting showed maximum dye exhaustion in dye bath. Further, the extracted dye was analysed for its chemical constituent and found the presence of betalain as red pigment. Hence, dyeing of cotton yarn with *Aerva sanguinolenta* in combination with biomordant extract at pH 6, 70°C for 60min can be recommended as successful alternative to harmful synthetic dye.

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