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A Potential Dye From *Acalypha Wilkesiana* Muell Arg. For Conventional and Sonicator Dyeing of Cotton And Silk Yarn.

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ABSTRACT

Synthetic dyes used for colouration has been causing environmental pollution and affecting aquatic life forms and human beings adversely. In the recent past, demand for natural dyes gained importance same as in ancient time because of its safe and eco-friendly nature. In the present study, dye from *Acalypha wilkesiana* leaves was extracted only with water to avoid solvent toxicity. The conditions optimal for the extraction of dye such as pH, temperature and time were carried out. Pretreated cotton and silk yarn were mordanted using chemical mordant alum for better fixation of dye. Both conventional and sonicator dyeing methods were followed. The conditions optimal for dyeing such as pH, temperature, time and dye concentration were also standardized. Dyed yarn has been evaluated for its colour fastness to light, wash and perspiration using standard ISO methods. The results showed good fastness properties. To study the pigments present in the dye, Phytochemical, TLC, UV-Visible, and FTIR analysis were done. Results showed the presence of various pigments of which Anthocyanin was found to be the principal pigment which is responsible for the red colour of the dye. Hence, it can be efficiently used for dyeing of textile materials.

Keywords: Natural dye, *Acalypha wilkesiana*, Alum, Sonicator, Colour fastness, Anthocyanin.

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INTRODUCTION

Colour is one of the elements of nature which made human life artistic and ravishing in the world. Dye is derived from the English word 'Daeg' or 'Daeh' means colour [1]. A basic dye was synthesized accidentally by William Perkins in 1856 which declined the use of natural dyes tremendously because the natural dyes failed to accomplish the demand of the market [2]. But synthetic dyes pose a threat to the environment due to their harmful effects on both the land and water bodies and also on human health. The synthetic dye residues affect the photosynthetic activity in aquatic life by preventing the light penetration and the toxic chemicals produced have a deleterious effect on flora, fauna and human beings [3,4]. So once again the natural colourants from renewable resources has been reintroduced into the dyeing industry by the researchers [5]. The European Union has recently banned the use of azo dyes which is a highly pollutant and toxic dye which also increased the scope of natural dyes [6]. 100,00 dyes are commercially available with a production rate of over $7x10^5$ to $1x\ 10^6$ per year and 10-15% of those used dyes enters the environment through effluents [7,8].

Natural dyes are obtained from sources like plants, animals, insects and minerals [9]. They not only provide unique colours but also give antimicrobial, deodorizing and UV- protective functions to the textile materials. Natural dyes global demand per year is around 10,000 tons which equals to 1% of the world's synthetic dye consumption [6]. Exports of natural dyes have grown to an impressive annual rate of 181.0% between January and September 2010 [2]. The use of natural dyeing materials since ancient times in India is very well evident from the wall paintings of Ajanta, Ellora and Sithannavasal [1]. A limitation in the use of natural dyes in textile dyeing is inadequate fastness properties. To overcome this problem chemical called mordants were used. Mordants are metal salts that produce affinity between the textile material and the dye [10,11]. Mordants are essential substances which are used not only to fix dye to the fibres but also help in improving the colour and fastness properties [12].

Non-conventional methods such as Sonicator (Ultrasound), Microwave and Supercritical fluid extraction methods are gaining importance for the better extraction of dye and dyeing of textile materials. Ultrasound is a sound wave with frequency of 16Hz-16KHz which is above the human audible range [13]. Ultrasound is efficient and energy conservative which makes the extraction process and also dyeing much faster [14]. It is less time consuming, cost effective and also provides better fastness properties. A number of dyeing can be carried out in good colour strength, lesser dyeing time under mild conditions using Sonicator [15,16].

Acalypha wilkesiana Muell Arg. (copper red) is an ornamental plant from the family Euphorbiacaece. They are distributed all-over the world particularly in the tropics of America, Africa and Asia. It is a fast growing shrub and the heart shaped leaves gives a splash of colour with bronze red to muted red [17]. A study was reported on dyeing of silk with various mordants and its fastness by *Acalypha wilkesiana* [18].

Anthocyanins are the most important and largest group of water-soluble vacuolar pigments which are flavonoids responsible for pink, scarlet, red, mauve, violet and blue colours in the petals, leaves and fruits of higher plants [19].

In the present study natural dye was extracted from *Acalypha wilkesiana* leaves by optimizing the extraction process with different parameters and dyed with pretreated cotton and silk yarn under different conditions. Conventional and Sonicator dyeing were compared for better dyeing process. Chemical mordant alum (Aluminium potassium sulphate dodecahydrate) was used for mordanting and the colour fastness of the yarns was evaluated. The plant pigments were also identified using various analyses.

MATERIALS AND METHODS

Collection of Plant Material:

The plant used in this study for extraction of dye is *Acalypha wilkesiana* which was collected from Society of Horticulture and maintained. The part used for the dye extraction was only leaves.

Extraction of Dye:

Acalypha wilkesiana leaves were collected, washed and dried. The leaves were weighed 10gm, cut into small pieces and soaked in 100 ml of distilled water for 30 minutes at room temperature and then extracted. Temperature (60˚C, 80˚C, 100˚C), pH (5,7,9) and time (15, 30, 45 minutes and 1hour) were optimized to study the efficient extraction of dye. The extracted dye was filtered and cooled.

Pretreatment of Cotton and Silk Yarn:

Cotton yarn was purchased from TANSPIN, Erode. To remove the natural cellulosic content (Starch) and the colouring matter of cotton yarn, scouring was done using Sodium hydroxide (NaOH) with 1:4 ratio. Yarn was boiled with NaOH solution for 2 hours and then washed thoroughly in running tap water. Effective scouring were tested using standard test methods such as Weight loss method, Drop test method and Tegewa scale method.

Silk yarn was purchased from TANSILK, Salem. The degummed silk yarn was soaked in sterile distilled water with mild detergent for 1hour at room temperature and then washed to remove the natural impurities and improve the texture of silk yarn for dyeing.

Mordanting:

Chemical mordant alum was used in the study to avoid effluent toxicity caused by other chemical mordants. Different concentrations of alum (1-10%) were optimized for both cotton and silk yarn. Premordanting of cotton and silk yarn was done by dipping 1gm of yarn in different concentration of alum solution (1-10%) and maintained at 90˚C, 1hour for cotton and 60˚C, 1hour for silk yarn. After mordanting the yarns were squeezed and used for dyeing. Metamordanting was done by adding alum (1-10%) to the dye bath containing cotton and silk yarn separately and dyeing was carried out. Post mordanting was done after dyeing of cotton and silk yarn and mordanted with alum solution (1-10%) and maintained at 90˚C, 1 hour for cotton and 60˚C, 1hour for silk yarn.

Dyeing:

Conventional dyeing:

The pretreated cotton and silk yarn was dyed using dye extracted from *Acalypha wilkesiana*. Optimization of dyeing of cotton yarn was studied using different parameters such as pH (5,6,7,8,9), temperature (30˚C, 50˚C, 70˚C, 90˚C), dyeing time (15, 30, 45 minutes and 1 hour) and material to liquor ratio (1:20, 1:40, 1:60). Optimization of dyeing of silk yarn were also studied at different pH (5,6,7,8), temperature (30˚C, 40˚C, 50˚C, 60˚C), time (15, 30, 45 minutes and 1 hour) and material to liquor ratio (1:20, 1:40, 1:60). The dyed yarns were air dried and checked for colour fastness.

Sonicator dyeing:

Dyeing of cotton and silk yarn was carried out in ultrasound for 15 minutes at 60˚C in pH 6 for cotton and pH 5 for silk. Pre mordanting, Meta mordanting and Post mordanting with alum was done same as conventional dyeing.

Dye Exhaustion:

The percentage of dye bath exhaustion was calculated using an equation on the basis of absorbance values recorded before and after dyeing with a UV-Visible spectrophotometer.

$$\%E = \frac{A_0 - A_1}{A_0} \times 100$$

where, %E is dye exhaustion, A_0 and A_1 are the absorbance of the dye bath before and after dyeing, respectively.

Colour fastness:

The dyed cotton and silk yarn was evaluated for colour fastness to light (ISO 105 B02-2013), washing (ISO: 105C-10:2006) and perspiration (ISO 105 E04-2013) using standard ISO methods.

Identification and Characterization of dye:

Phytochemical analysis:

The aqueous dye extract was checked for the presence of secondary metabolites such as alkaloids, flavanoids, terpenoids carbohydrates, phenols, tannins, proteins and amino acids, gum/mucilage using standard procedure [20].

Thin Layer Chromatography:

Silica gel coated plates were used for the separation of plant pigments. The solvent system used were Methanol: HCl (85:15), Methanol: HCl (99:1), Conc. HCl: Formic acid: H_2O (19:39.6:41.4) and HCl: H_2O (3:97). After separation the Rf values were calculated.

UV-Visible Spectroscopy:

The dye extract was analyzed in UV-Visible spectrophotometer Shimadzu-1650 PC at a wavelength 200-800nm to study the maximum absorption spectrum of the dye extract.

FT-IR Spectroscopy:

The dye extract was mixed with Potassium bromide (Kbr) salt and compressed into a thin pellet and the spectrum of sample was recorded on FT-IR instrument (Shimadzu, IR Affinity 1, Japan),equipped with a DLATGS detector with a mirror speed of 2.8mm/sec. All measurements were carried out in the range of 500-4000 cm-1 at a resolution of 4cm-1.

RESULTS AND DISCUSSION

Optimization of dye extraction:

Figure 1: *Acalypha wilkesiana* Muell Arg. – Habit

Figure 2: Dye extract from *Acalypha wilkesiana*

Different parameters such as pH, temperature and time were optimized for the aqueous extraction of dye from *Acalypha wilkesiana* (Fig.1). The dyes from leaves will have a polyphenolic chromophoric structure which helps for better extraction in aqueous solution [21,22]. Best extraction was obtained at pH 5, 60˚C for

1hour (Fig.2). At neutral and alkaline pH 9 the colour of the dye changed from red to brownish black, while in acidic pH 5 the red colour of the dye was maintained. This clearly shows that pH is having a major effect on the dye extraction thereby degrading the plant pigment at higher pH. In case of temperature, 60˚C was found to maintain the stable red colour of the dye, while in higher temperatures the colour changed to reddish brown.

So this temperature and pH sensitive red colour pigment resembled the features of the pigment Anthocyanin with reference to the literatures cited. According to [23] the anthocyanin colour depends on the acidity of the medium, where they exist chiefly in the form of the red flavylium cation which due to hydration to produce the colourless carbinol pseudobase. When the pH shifts higher from neutral to alkaline, a rapid proton loss of the flavylium cation happens. [24] stated that higher temperatures leaded to anthocyanin degradation. Similar results were seen in the present study confirming it as Anthocyanin.

Mordanting:

According to [25] adding excess amount of metallic salts leads to a reduction in the extract solubility which makes them to precipitate and hence finding difficult to penetrate during dyeing. In the present study, only alum has been used to avoid toxicity caused by other metallic mordants. 1-10% of alum was optimized for the mordanting of cotton and silk yarn, of which 3% of alum gave best results for cotton and 1% of alum gave best results for silk yarn. The colour of the dye was slightly changed from red to reddish brown when alum was used for mordanting thereby developing a new shade. There is a sudden, dramatic change in colour when a mordant is added to the dye bath solution because of the incorporation of the metal atom to the delocalized electron system of the dye [26], which supports the results of the present study.

Hydroxyl and Carbonyl groups present in the dye structure has the ability in forming a metal complex with the positively charged metals. Generally, one dye molecule will form a bond with one active site present in the fibre molecule, but one molecule of mordant is capable of forming bonds with two or more molecules of dye. Hence use of mordant increases the colour uptake and strength. In case of silk an ionic bonding is formed between dye and fibre, metal and fibre and then dye and metal ions because of the strong attraction of dye anions and metal cations towards positively charged amino and negatively charged groups. Coordinate bonds were also formed by the dye- metal complex with the uncharged amine ($-NH_2$) groups of silk [27].

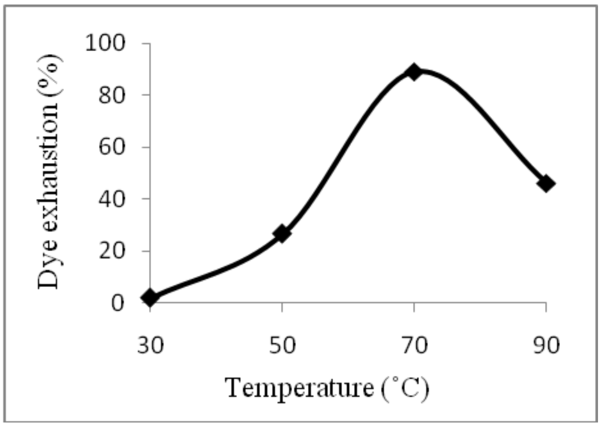
[28] states that, the natural dye has less affinity for fibres, so to improve the fastness metal mordants are used which forms an insoluble complex with dye molecules. The keto hydroxyl structure in the flavones dye molecules improves the chelation of Al ions, thereby forming five-member chelate ring with aluminium ions. Due to these reasons the alum used in present study helped the yarns in better dye uptake and strength. According to [29] alum increased the number of bonds between dye and fibre.

In different methods of mordanting , pre mordanting gave best and meta mordanting gave moderate results. During pre mordanting and meta mordanting the mordant interacts with the yarn and increase the active sites for more dye molecules to get attached, whereas in case of post mordanting the dye uptake was less due to the lack of mordant during dyeing. Similar results were supported by [30].

Conventional Dyeing:

Effect of temperature on dyeing:

The effect of temperature on the dye ability of cotton and silk yarn were studied at different temperatures such as 30˚C, 50˚C, 70˚C, 90˚C and 30˚C, 40˚C, 50˚C, 60˚C respectively. As shown in the Fig.3and 4 the maximum dye exhaustion was obtained at 70˚C for cotton and 60˚C for silk yarn. Similar results for cotton were reported [31]. At higher temperature 90˚C for cotton, the dye colour found to be unstable and started degrading. The reason behind this is two processes occurs during dyeing of cellulosic fibre, sorption and stripping of dye from fibre to dye bath. First the sorption rises by heating but at higher temperature there might be a stripping and hydrolytic degradation of dye which lowered the colour strength [32]. According to [33] the colour strength remained constant between 60-80˚C during dyeing with anthocyanin- based mangosteen dye.



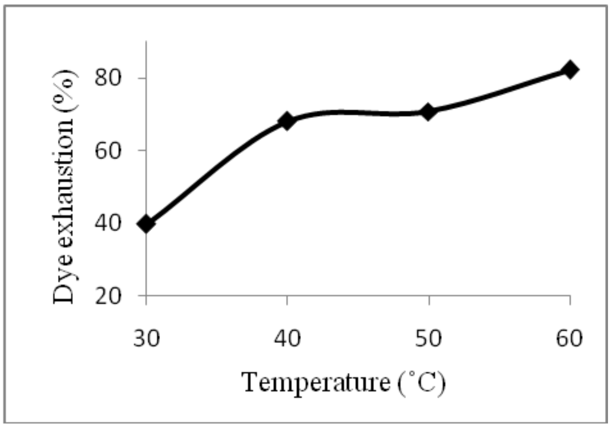
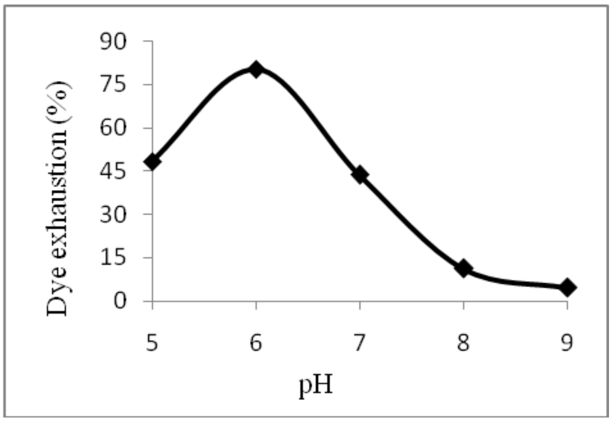


Figure 3: Effect of temperature on dyeing of cotton yarn **Figure 4: Effect of temperature on dyeing of silk yarn**

Effect of pH on dyeing:

The effect of pH on the dye ability of cotton and silk yarn were studied at different pH such as 5,6,7,8,9 and 5,6,7,8 respectively . As shown in the Fig.5 and 6 the maximum dye exhaustion was obtained at pH 6 for cotton and pH 5 for silk. As discussed earlier, the dye is unstable in neutral and alkaline pH and stable in acidic pH, the dyeing was efficient in acidic pH only. Similar results were reported by [34].



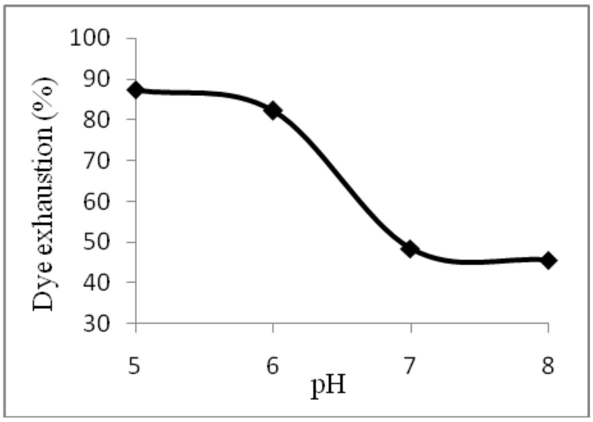


Figure 5: Effect of pH on dyeing of cotton yarn **Figure 6: Effect of pH on dyeing of silk yarn**

In case of silk the ammonium ion (NH_3^+) is converted to amino groups (NH_2) by the hydroxide ions (OH^-) in the alkaline pH [35,36]. Hence there is a decrease in the dye uptake due to the electrostatic repulsion between anionic colourants and the protein fibres [33]. Similary, at a higher pH than 6 because of oxidation the conjugate structure of the natural dyes could be destroyed; decreasing the colour strength [30].

Effect of time on dyeing:

The effect of time on dye ability on cotton and silk yarn were studied at different time such as 15, 30, 45 minutes and 1hour for both yarns. As shown in the Fig.7 and 8 the maximum dye exhaustion was obtained in 45 minutes for cotton and 1 hour for silk yarn. Similar results were supported by [9,25,34].

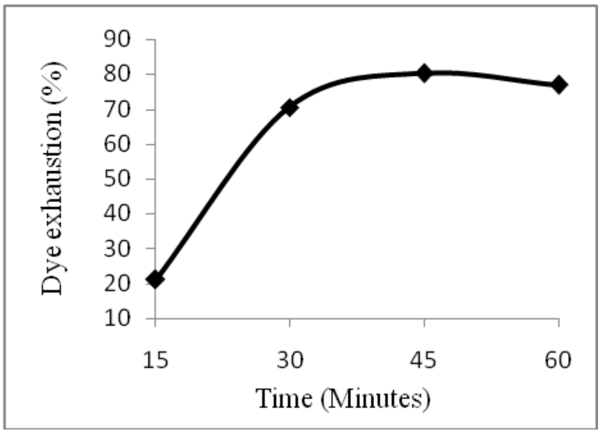


Figure 7: Effect of time on dyeing of cotton yarn

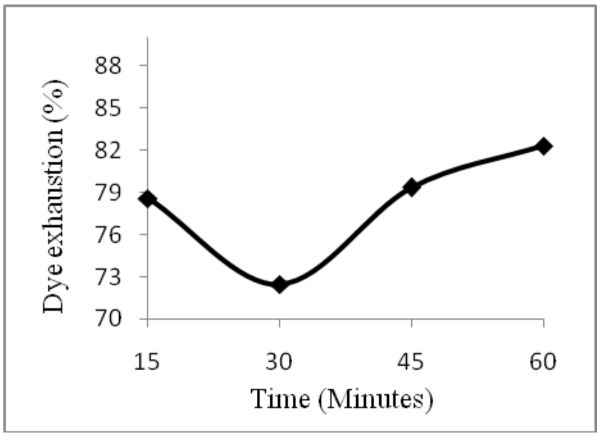


Figure 8: Effect of time on dyeing of silk yarn

Effect of dye concentration:

The effect of dye concentration (Material to liquor ratio) such as 1:20, 1:40 and 1:60 for both cotton and silk yarn were studied. As shown in Fig.9 and Fig.10, maximum dye exhaustion was obtained at 1:20 concentration for cotton and silk yarn but the best shade and equally better exhaustion was obtained at 1:40 concentration. In case of 1:20 concentration the dye exhaustion was maximum because the dye concentration was found less and not enough for the amount of yarn used, hence a smooth dyeing was not achieved. But in case of 1:40 concentration the dye exhaustion was also better and an even dyeing was achieved. Hence 1:40 concentration was preferred for the dyeing process. Concentration gradient increases when the concentration of dye increases in the dye bath which ultimately increases the rate of diffusion of dye to the surface of the yarn [5]. So the 1:40 concentration will increase the rate of diffusion of dye to the yarn.

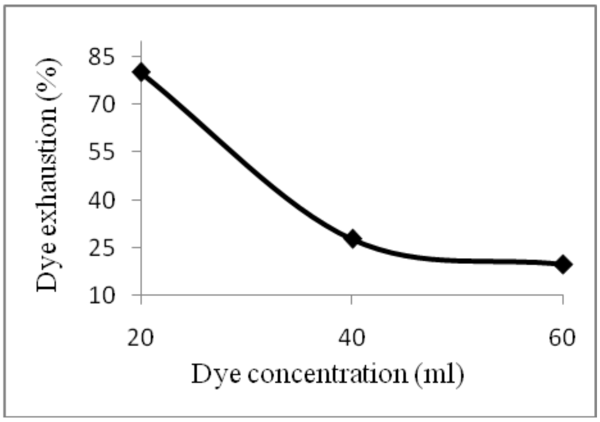


Figure 9: Effect of dye concentration on cotton yarn

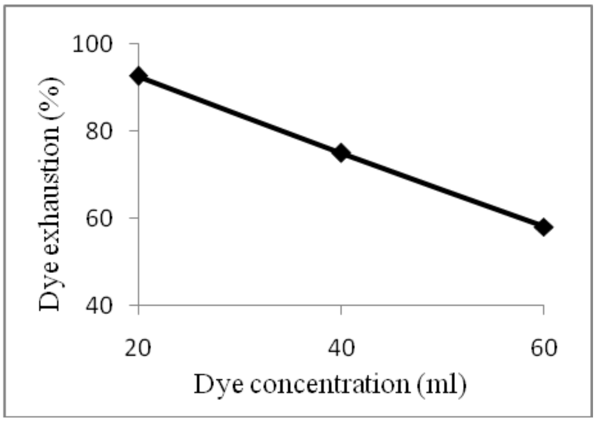


Figure 10: Effect of dye concentration on silk yarn

From the optimization of conventional dyeing of premordanted cotton and silk yarn with *Acalypha wilkesiana* dye extract using different parameters like pH, temperature, time and material to liquor ratio, the best parameter selected based on the dye exhaustion and colour fastness for dyeing of cotton yarn was 70˚C, pH 6, 45 min, ML-1:40 (Fig.11) and for silk yarn 60˚C, pH 5, 1 hr, ML-1:40 (Fig.12).

Figure11: Dyeing of cotton yarn with *A.wilkesiana* (70ºC,pH6,45min,ML-1:40)

Figure 12: Dyeing of silk yarn with *A.wilkesiana* (60ºC,pH5,1hr,ML-1:40)

Sonicator dyeing:

Sonicator dyeing of cotton (Fig.13) and silk yarn (Fig.14) was carried out at 60˚C for 15minutes with a dye concentration of 1:40 in pH 6 for cotton and pH 5 for silk. Pre mordanting and meta mordanting gave best results. The results showed (Fig.15) dye exhaustion maximum in sonicator method when compared to the conventional method in short duration of time (15minutes). In comparison with premordanted cotton yarn, dye uptake on premordanted silk yarn was found to be very efficient. Hence silk yarn was checked for colour fastness. Similarly [29] maintained the dyeing time for 15 minutes. The principle behind the sonicator dyeing is that the acoustic cavitation occurs near the surface of the yarn which generates microjets that facilitate the liquid to move with a greater velocity and results in efficient diffusion of dye molecules inside the fibre pores [28]. Hence the dyeing efficiency was increased by sonicator method when compared to conventional dyeing.

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Figure13: Dyeing of cotton yarn with *A.wilkesiana* in sonicator (60ºC,pH6,15min,ML-1:40)

Figure14: Dyeing of silk yarn with *A.wilkesiana* in sonicator (60ºC,pH5,15min,ML-1:40)

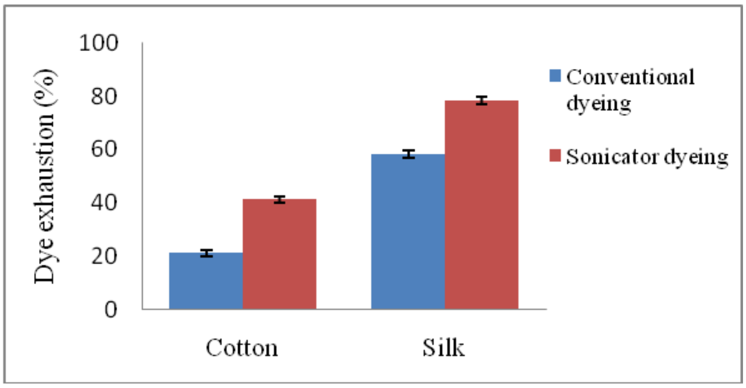

Figure15: Comparison of Conventional and Sonicator dyeing for 15 minutes

Colour fastness:

Fastness properties of dyed yarns were evaluated. Premordanted cotton and silk yarn dyed using conventional dyeing method and premordanted silk yarn dyed using sonicator were checked for colour fastness. In conventional dyeing method the cotton and silk yarn showed fair to good fastness to light and perspiration but showed poor wash fastness. But in sonicator method of dyeing, the silk yarn showed fair to good fastness to light, wash and perspiration. Similar results were supported by [28]. When metallic mordants were used the complex formed with transition metal protects the chromophore from photolytic degradation. The chromophoric group absorb the photons which by resonating within six- member ring dissipate their energy thereby protects the dyes [27]. Hence the metal mordant alum used in the present study protects the dye from photolytic degradation, thereby giving good light fastness.

The washing and perspiration solution influences the relation between dye removal and dye mordant nature. If the number of groups which is capable of forming hydrogen bonding and metal complex is higher, the magnitude of dye removal will be lower [25]. Similarly the yarns premordanted with alum have given relatively good wash and perspiration fastness.

Identification and Characterization of dye:

Phytochemical analysis:

Phytochemical analysis of aqueous dye extract from *Acalypha wilkesiana* showed the presence of Alkaloids, Glycosides, Carbohydrates, Proteins and Aminoacids, Phenols,Flavonoids,Tannins and Terpenoids . Similar phytochemical results of *Acalypha wilkesiana* was reported [17, 37].

Thin Layer Chromatography (TLC):

The Thin layer Chromatography with a solvent system Methanol: HCl (99.9:0.1) showed (Fig.16) the presence of anthocyanin after the separation of pigments with reference to the Rf value calculated. The Rf value calculated was 0.53 which corresponds to the monoglycosylated anthocyanins.

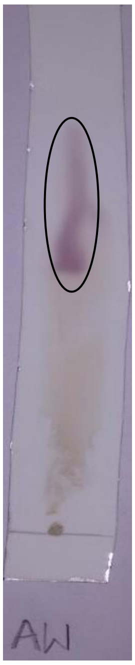

Figure 16: TLC of dye extract

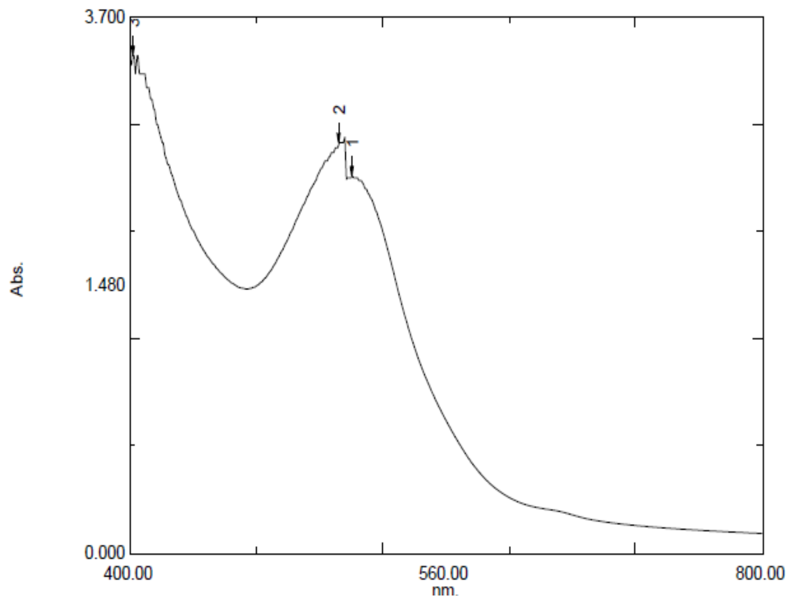

Figure 17: UV-Visible spectrum of *Acalypha wilkesiana*

UV-Visible Spectroscopy:

The dye extract scanned from 400-800nm showed a maximum absorbance at 540 nm in visible range (Fig.17) which is a characteristic of aglycone of anthocyanin molecule. Similar results were obtained by [38]. All flavonoids show maximum absorbance in the range of 250-270nm and especially anthocyanin will have an intense absorption in the range of 520-560nm [39]. According to [40] the maximum absorption of anthocyanin falls in the range of 530- 550nm. Anthocyanins have maximum absorption between 475-550nm [19]. With the literatures cited it confirms that the red coloured dye pigment is anthocyanin which is a flavonoid.

FT-IR Spectroscopy:

FT-IR analysis has been used to identify the auxochromes which are responsible for the substantivity of dye [5]. The FT-IR analysis (Fig.18) of the dye extract showed the corresponding peaks at 3429 cm^{-1} assigned to fundamental stretching vibration of O-H hydroxyl groups. The aromatic C=C bands are present at 1527 and 1447 cm^{-1}. The peak at 2934 cm^{-1} corresponds to the aliphatic CH vibrations and 1632 cm^{-1} to the C=O conjugated with aromatic ring. Very much similar results have been reported by [40,41]. These peaks therefore are the characteristic feature of anthocyanin. The regions below 1200 cm-1 showing peaks are due to the vibrations of the whole molecule and are called as 'fingerprint region'.

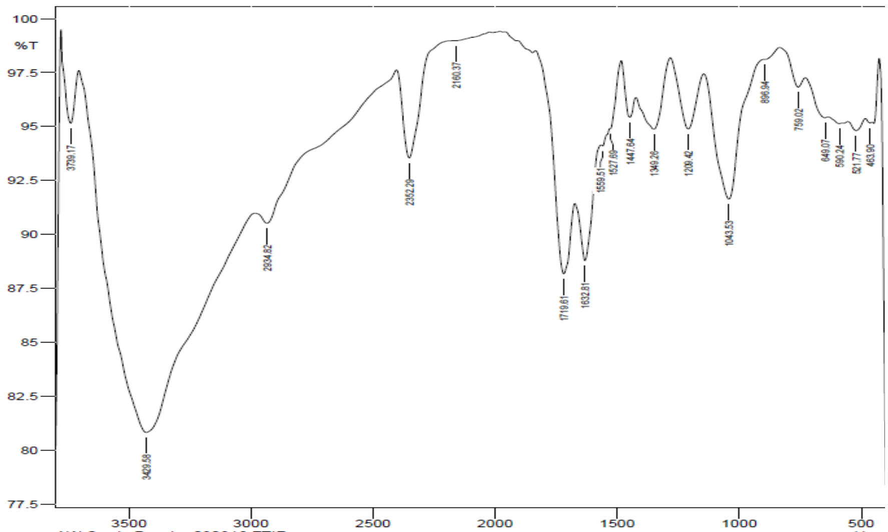

Figure18: FT-IR spectrum of *Acalypha wilkesiana*

CONCLUSION

In the present study, *Acalypha wilkesiana* was found to have good agronomic potential as a dye plant. Best extraction of dye can be obtained at pH5, 60˚C for 1hour. Metal mordant alum has found to increase the dyeability and fastness properties when samples (yarn) were premordanted. In conventional dyeing method, the best dyeing was obtained at 70˚C, pH 6 for 45mins with 1:40 dye concentration for cotton and 60˚C, pH 5 for 1hour with 1:40 concentration for silk yarn. The dye uptake and fastness of was found to be increased by the sonicator dyeing when compared with the conventional dyeing, thereby made the dyeing process energy saving and economical. The results showed fair to good fastness properties to light, washing and perspiration. The dye pigment was identified as Anthocyanin which gives red colour to the dye. Hence it is recommended to be used commercially in textile industries without any toxicity caused by synthetic dyes.

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