# Natural Dyeing of Leathers Using Natural Materials

by
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## Abstract

Environmental constraints on discharge of dye containing wastewater has forced us to look for natural dyes, which have several advantages like low toxicity, cheap and easy availability etc. Natural dyes are obtained from plants (e.g., Alizarin, Catechu, Indigo and Logwood), from animals (e.g., cochineal, kermes and tyrian purple) and from certain naturally occurring minerals (e.g., Ocher and Prussian blue). In this study, an attempt has been made to develop leathers, colored using natural dyes such as Rhine, Rhine M, Indus, Pacific, Caspian, Henna and modified Logwood. This study would be more beneficial to the leather industry. Also, an effort has been made to produce wide range of shades through mordanting with eco-benign metals such as aluminum, copper and tin. Twenty four shades were developed using combination of seven natural colorants by mordanting with three metal ions. The reflectance spectra for all colors have been studied. The visual appearance and their fastness properties have also been evaluated and the test results are satisfactory. Developed colors have potential value in the global leather market in the context of environmentally benign leather processing.

## Resumen

Limitaciones en relación al medio ambiente sobre las descargas líquidas que contengan colorantes nos han obligado a buscar colorantes naturales, los cuales tienen ciertas ventajas como baja toxicidad, bajo costo y fácil disponibilidad, etc. Colorantes naturales son obtenibles de plantas (Vg. Alizarina, [extracto de] Mimosa, Índigo, Haematoxylum campechianum), de animales (Vg. Cochinilla, Kermés Ilicis y/oVemilio, Bolinus brandaris [púrpura]) y de minerales de natural ocurrencia (Vg. Ocre y Azul de Prusia). En este estudio se trató de desarrollar cueros, el coloreado utilizado por medio de colorantes naturales tales como [El río] Rin, Rin M, Indus, Pacifico, Caspio, Henna y Púrpura modificado. Este estudio sería más benéfico a la industria de cuero. Un esfuerzo también se ha hecho para producir una extensa gama de tonos por medio de mordientes metálicos eco-benignos tales como el aluminio, cobre y estaño. Veinticuatro tonos fueron desarrollados utilizando combinaciones de siete colorantes naturales amordantados con tres iones metálicos. Los espectros de reflexión de todos los colores fueron estudiados. El aspecto y propiedades de firmeza también fueron evaluados y los resultados son satisfactorios. Los colores desarrollados tienen potencial valor en el mercado mundial debido al contexto benigno del procesamiento en el medio-ambiente.

## Introduction

Global leather industry is currently undergoing radical transformation due to pollution and discharge legislations. Thus, the leather industry is pressurized to look for natural materials for processing the raw hides and skins. Conventional method of dyeing results in the generation of colored effluent with toxic substances.[1] Nowadays, there is a growing interest in the revival of natural dyes in leather dyeing; arguments based around keywords such as sustainability, green chemistry and improved eco-balances and thereby leading to niche products for special markets.[2,3] Therefore the tanners are looking for eco-dyeing method for making leathers.

Natural dyes are a class of colorants extracted from vegetative matter, animal residues and certain naturally occurring minerals. These dyes in vogue during ancient days were indigo for dark blue/ light blue, pomegranate rind for yellow/ brown/ green, lac for scarlet/crimson/purple, jackfruit heartwood for yellow/green, manjistha root for rust red, myrobalan for khaki/green/black. These are considered as mordant dyes as they require the inclusion of one or more metallic salts of aluminum, iron, chromium, copper and others for ensuring reasonable fastness of the colors to sunlight and washing.[4-8] These metallic salts combine with the dyestuff to produce dye

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**TABLE I**
**Process for Eco-dyeing of Leathers Using Natural Dyes With and Without Mordants**

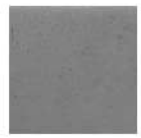
| Process | Chemical | % | Duration (min) | Remarks |
|---|---|---|---|---|
| Acid washing | Water | 100 | | |
| | Acetic acid | 0.5 | 45 | Drain |
| Rechroming | Water | 100 | | |
| | BCS | 5 | | |
| Basification | Sodium formate | 1 | 10 | |
| | Sodium bicarbonate | 1 | 3X15+45 | PH 3.8-4.0 drain |
| Wash | Water | 300 | 10 | Drain |
| Neutralization | Water | 100 | | |
| | Sodium formate | 1 | 10 | |
| | Sodium bicarbonate | 1 | 3X15+45 | pH 5.0-5.2 drain |
| Wash | Water | 300 | 10 | Drain |
| Dyeing, fat liquoring and retanning | Water | 100 | | |
| | Natural coloring Material[a] | 2 | 60 | Check for penetration |
| Mordanting | Metal[b] | X[c] | 45 | |
| | Densodrin ENS | 1.5 | 20 | |
| | Lipoderm Liquor FB-FP | 2 | | |
| | Lipoderm Liquor FB-W | 2 | | |
| | Balmol SXE | 2 | 40 | |
| | Relugan RE | 2 | 20 | |
| | Basyntan DI | 4 | | |
| | Basyntan AN | 4 | | |
| | Basyntan FB6 | 4 | 45 | |
| | Formic acid | 1 | 3X15+45 | Drain |
| Washing | Water | 300 | 10 | Drain pile for 18 h. Next day, leathers were dried, staked and trimmed. |

[a] Natural coloring materials such as Rhine, Rhine-M, Indus and Pacific,
[b] Metals such as $Al^{3+}$, $Cu^{2+}$ and $Sn^{2+}$.
[c] X was 0 and 1%

aggregates, which cannot be removed from the fabric easily. Compound shades were also obtained by over dyeing of fabric with two colors or by cross weaving of fabric. These natural dyes were eco-friendly and non-carcinogenic.[9-11]

Synthetic dyes (acid, basic, reactive and metal complex) are widely used in dye industries like textile, paper and leather. Environmental challenges on account of synthetic dyes have resulted in a revisit to natural dyes. The pollution caused by the synthetic dyes provides an opportunity for the reintroduction of natural dyes. It has reported that natural colorant, eco-yellow extracted from an indigenous material for leather dyeing. Development of black and brown using vegetable materials on chrome-iron tanned leathers is gaining importance.[12] Although reports on the use of natural materials for dyeing of fabric are available, the application for leather dyeing is limited.[13] Most of the natural dyes produce comparatively dull shades with poor fastness properties. The fashion trends around the globe are looking forward for a shift in leather dyeing towards eco-benign leathers. Moving from synthetic to natural dyes is an immediate possibility based on forecasts for natural colors in the global market for fashion trends. The scope of work is derived from this need, for which a revisit of natural materials and mordants is the objective forward. The objective of this work is to develop different shades from natural materials such as Rhine, Rhine M, Indus, Pacific, Caspian, Henna and modified logwood by striking with non-toxic metal ions such as aluminum, copper and tin. Color and reflectance measurements have been carried out. Fastness characteristics of the naturally colored leathers have been tested. Further, the spectral characteristics of natural dyes with presence of mordants have been analyzed.

**TABLE II**
**Different Shades Developed from the Natural Dyes With and Without Mordants**

| Natural Dyes | Without Mordant | $Al^{3+}$ Mordanted | $Cu^{2+}$ Mordanted | $Sn^{2+}$ Mordanted |
|---|---|---|---|---|
| Rhine |  |  |  |  |
| Rhine M |  |  |  |  |
| Indus |  |  |  |  |
| Pacific |  |  |  |  |
| Caspian |  |  | | |
| Henna |  |  | | |
| Vegdye BRE |  |  |  |  |
| | Vegdye VO | Hematine CFMK | Vegdye MUS | Logwood |

## Experimental

### Materials

Compact wet blue goatskins of 3 - 4 sq. ft. were chosen as the raw material for this study. Natural dyes such as Rhine, Rhine M, Indus, Pacific, Caspian and Henna were sourced from Alps Industries Ltd., India. Modified logwood was sourced from M/s SCRD, France. Chemicals for mordanting were of laboratory grade. The chemicals used for the leather processing were of commercial grade.

### Dyeing Trials

Wet blue goatskins were sammed, shaved to uniform thickness of 1.1±0.1 mm. The post tanning process for the dyeing trial is as given in Table I. Dyeing trials were carried out using three wet blue goatskins for each experiment.

### Reflectance Measurements

The principle involves measuring the amount of light reflected from the surface of opaque specimen at wavelengths throughout the visible spectrum as a fraction of that reflected by a white standard identically illuminated. This is known as the reflectance factor. The white standard used should be an absolute one i.e., it should be a perfect reflecting diffuser whose reflectance at every wavelength is 100%. The crust leathers using natural dyes made in this study were subjected to reflectance measurements using a Mitton Roy Color Mate HDS instrument.

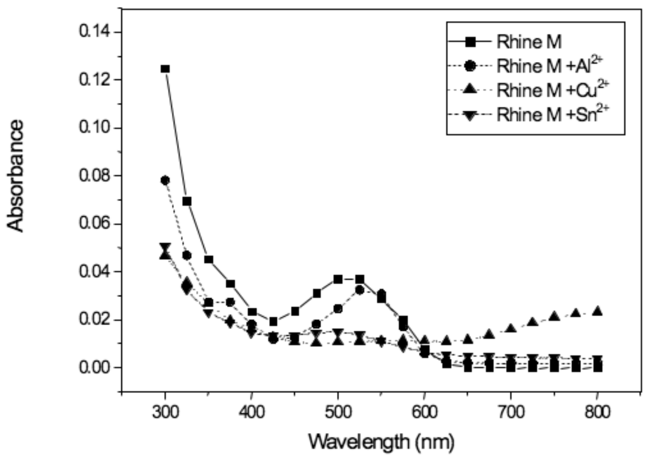



Figure 1: Spectral characteristics of Rhine M dye with and without mordants

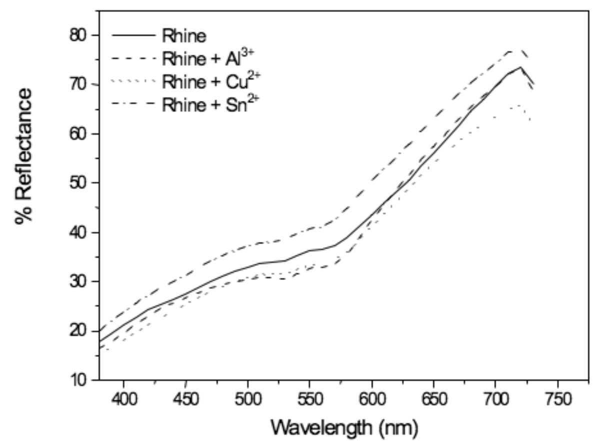



Figure 2: Reflectance spectra for leather dyed using Rhine with and without mordants

**TABLE III**
**Color Measurement Values for Eco-dyed Leathers Using Natural Dyes With and Without Mordants**

| Natural dyes/mordants | L | a* | b* |
|---|---|---|---|
| Rhine | 62.84 | 12.11 | 2.68 |
| Rhine + $Al^{3+}$ | 71.84 | 10.28 | 2.54 |
| Rhine + $Cu^{2+}$ | 70.39 | 8.16 | 0.9 |
| Rhine + $Sn^{2+}$ | 69.16 | 11.18 | 3.7 |
| Rhine-M | 43.49 | 36.77 | 3.76 |
| Rhine-M + $Al^{3+}$ | 47.57 | 26.96 | -0.10 |
| Rhine-M + $Cu^{2+}$ | 41.44 | 25.09 | -0.33 |
| Rhine-M + $Sn^{2+}$ | 40.92 | 31.18 | 7.68 |
| Indus | 51.48 | 20.73 | 15.51 |
| Indus + $Al^{3+}$ | 54.67 | 22.34 | 22.19 |
| Indus + $Cu^{2+}$ | 58.73 | 15.55 | 19.55 |
| Indus + $Sn^{2+}$ | 57.21 | 20.14 | 15.55 |
| Pacific | 67.23 | 3.59 | 14.81 |
| Pacific + $Al^{3+}$ | 70.55 | 1.85 | 15.83 |
| Pacific + $Cu^{2+}$ | 63.38 | 2.24 | 16.77 |
| Pacific + $Sn^{2+}$ | 70.15 | 2.29 | 15.85 |
| Caspian | 63.54 | 9.71 | 13.95 |
| Caspian + $Al^{3+}$ | 68.67 | 8.43 | 13.61 |
| Henna | 73.66 | 3.76 | 2.37 |
| Henna + $Al^{3+}$ | 74.67 | 3.85 | 1.18 |
| Vegdye BRE | 59.37 | 21.77 | 8.47 |
| Vegdye VO | 45.81 | 0.67 | -7.70 |
| Hematine CFMK | 42.76 | 4.62 | -7.75 |
| Vegdye MUS | 60.00 | 5.55 | 35.86 |

**TABLE IV**
**Wet and Dry Rub Fastness of Leathers from Eco-dyeing Using Natural Dyes With and Without Mordants**

| Dye | Felt | | Leather | |
|---|---|---|---|---|
| | Dry rubbing | Wet rubbing | Dry rubbing | Wet rubbing |
| Rhine | 5 | 4 | 4-5 | 4-5 |
| Rhine + $Al^{3+}$ | 5 | 4-5 | 4-5 | 4-5 |
| Rhine + $Cu^{2+}$ | 5 | 4 | 4-5 | 4-5 |
| Rhine + $Sn^{2+}$ | 5 | 4 | 4-5 | 4-5 |
| Rhine-M | 4-5 | 4-5 | 4-5 | 4 |
| Rhine-M + $Al^{3+}$ | 4-5 | 4-5 | 4-5 | 4-5 |
| Rhine-M + $Cu^{2+}$ | 4-5 | 4-5 | 4-5 | 4 |
| Rhine-M + $Sn^{2+}$ | 4-5 | 4-5 | 4-5 | 4 |
| Indus | 4 | 4 | 4-5 | 3-4 |
| Indus + $Al^{3+}$ | 4-5 | 4-5 | 4 | 3-4 |
| Indus + $Cu^{2+}$ | 4 | 4 | 4 | 4 |
| Indus + $Sn^{2+}$ | 4 | 3-4 | 4 | 3-4 |
| Pacific | 5 | 4-5 | 4-5 | 4-5 |
| Pacific + $Al^{3+}$ | 5 | 4-5 | 4-5 | 4-5 |
| Pacific + $Cu^{2+}$ | 5 | 4-5 | 4-5 | 4-5 |
| Pacific + $Sn^{2+}$ | 5 | 4-5 | 4-5 | 4-5 |
| Caspian | 5 | 4-5 | 4-5 | 4-5 |
| Caspian + $Al^{3+}$ | 5 | 4-5 | 4-5 | 4-5 |
| Henna | 5 | 4-5 | 4-5 | 4 |
| Henna + $Al^{3+}$ | 5 | 4-5 | 4-5 | 4-5 |
| Vegdye BRE | 5 | 4-5 | 4-5 | 4-5 |
| Vegdye VO | 5 | 4-5 | 4-5 | 4-5 |
| Hematine CFMK | 5 | 4-5 | 4-5 | 4-5 |
| Vegdye MUS | 5 | 4-5 | 4-5 | 4-5 |

**Color Measurements**

Color measurement parameters viz., L, a* and b* were recorded using a Milton Roy Color Mate HDS instrument for all the crust leathers made using natural colorants.[14]

Characterization of Dyeing of Leathers

Three dyeing experts were assessed all the dyed crust leathers for uniformity in color, dye affinity and penetration of dye. The leathers were rated on a scale of 0-10 points for each property, where higher points indicate better property.

**Light, Wet and Dry Rub Fastness**

Crust leather samples were cut from the official butt portion as per IUP method.[15] Samples were conditioned at 80±4°F and 65±2% R.H. over a period of 48 hrs. Wet and dry rub and Light fastnesses were carried out using standard procedures.[16,17]

## Results and Discussion

In this study, an attempt has been made using seven natural dyes with and without mordanting with three metal ions for developing natural shades for leathers. Natural dyes such as Rhine, Rhine M, Indus, Pacific, Caspian, Henna and Modified logwood and metal ions aluminum, copper and tin have been

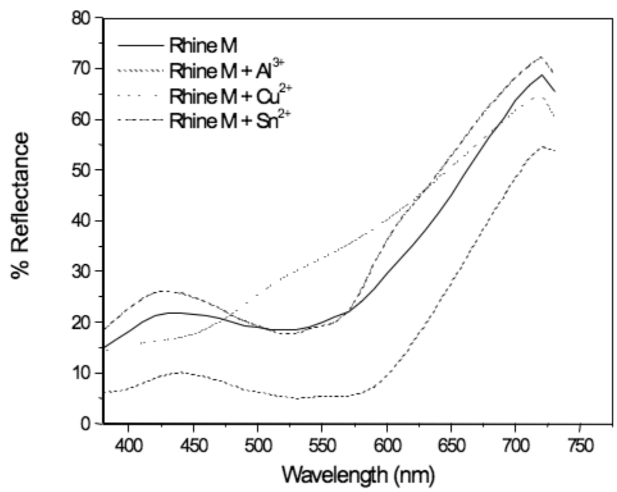



Figure 3: Reflectance spectra for leather dyed using Rhine M with and without mordants

chosen for this study. The offer of natural dye was fixed as 2% and for metal mordanting the metal offer was fixed as 1%. From this combination, twenty four shades have been developed and presented in Table II. The developed colors have been tested for various physico-chemical measurements.

**Spectral Characteristics of Dye with and without Mordant**

The spectral characteristics of the Rhine M dye have been studied. The behavior of Rhine M dye with and without mordants is shown in Fig 1. From the spectrum, it is evident that the addition of mordant shifts the absorbance maxima wavelength of the dye when complexed with the metal ions, thus resulting in different shades (as seen from Table II and III). This is attributed to a charge transfer interactions between the metal and the phenolic moieties in the dye. Similar trend has been observed for other dyes as well.

**Color Measurement of Dyed Leathers**

The color measurement values of leathers dyed using natural dyes with and without mordants are presented in Table III. The color of the leathers is sandal for Rhine treated leathers. It is observed that change in shade is towards lighter shade on mordanting. L, a* and b* values indicate no significant change in base color. The reflectance spectra (Fig 2) indicate that mordanting with aluminum results in lighter shade as compared to no mordanting leathers. This is in accordance with the L, a* and b* values.

The color of the leathers is reddish brown for Rhine M treated leathers. It is observed from Table III that the color shade shifts from maroon to reddish pink and deep brown on mordanting with metal ions. The shade variation is also observed from the reflectance spectra given Fig 3. The change in a* values indicate the reddish tone. As seen from the reflectance spectra and color measurements, aluminum mordanting results in lighter shade whereas tin mordanting provides darker shade. The shade variation can be attributed to the complexation of dye with metal ions.

Indus dye produces brown as the base color. The color shade shifts from carmine to rose brown, brown and dark brown. Reflectance and color measurements (Fig 4 and Table III) reveal that no significant change in base color is observed on mordanting.

The color obtained from pacific is apple green. There is a slight in shade of apple green is observed on mordanting. Mrodanting with copper results in darker shade as compared to aluminum and tin. This observation is in accordance with the reflectance spectra (Fig 5) and color measurements (Table III). Caspian produces brown to light brown color on mordanting with aluminum. No change is observed with copper and tin and hence the shades are not shown. Dyeing of leathers with henna powder provides beige color. No change in color shade on mordanting. Color measurements (Table III) indicates no significant change in lightness factor for henna on mordanting whereas the shade became lighter on mordanting with aluminum for caspian.

Four commercially available modified logwood dyes have been employed for the study. The colors obtained are golden yellow, grayish black, grayish blue and dark pink color on treatment with modified logwood (Vegdye BRE, Vegdye VO, Hematine CFMK

**TABLE V**
**Light Fastness of Eco-dyed Leathers Using Natural Dyes With and Without Mordants**

| Dye | Leather |
|---|---|
| Rhine | 4-5 |
| Rhine + $Al^{3+}$ | 4-5 |
| Rhine + $Cu^{2+}$ | 4-5 |
| Rhine + $Sn^{2+}$ | 4-5 |
| Rhine-M | 4-5 |
| Rhine-M + $Al^{3+}$ | 4-5 |
| Rhine-M + $Cu^{2+}$ | 4-5 |
| Rhine-M + $Sn^{2+}$ | 4-5 |
| Indus | 3-4 |
| Indus + $Al^{3+}$ | 3-4 |
| Indus + $Cu^{2+}$ | 3-4 |
| Indus + $Sn^{2+}$ | 3-4 |
| Pacific | 4-5 |
| Pacific + $Al^{3+}$ | 4-5 |
| Pacific + $Cu^{2+}$ | 4-5 |
| Pacific + $Sn^{2+}$ | 4-5 |
| Caspian | 4-5 |
| Caspian + $Al^{3+}$ | 4-5 |
| Henna | 4-5 |
| Henna + $Al^{3+}$ | 4-5 |
| Vegdye BRE | 4-5 |
| Vegdye VO | 4-5 |
| Hematine CFMK | 4-5 |
| Vegdye MUS | 4-5 |

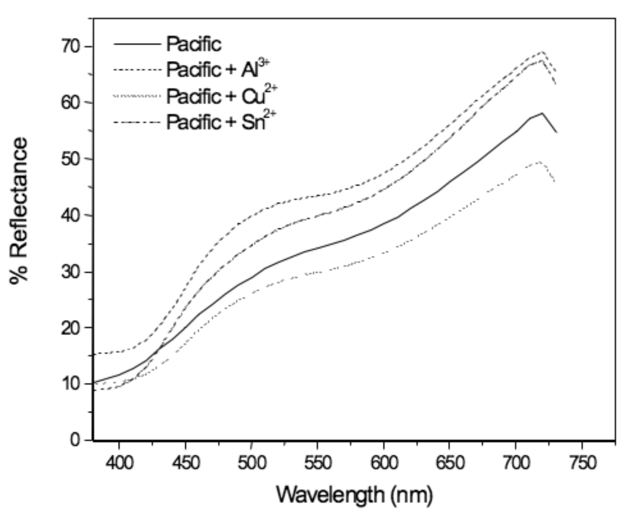



Figure 5: Reflectance spectra for leather dyed using Pacific with and without mordants

**TABLE VI**
**Dyeing Characteristics of Natrual Dyes With and Without Mordants**

| Trials | Dye uniformity | Dye affinity | Dye penetration |
|---|---|---|---|
| Rhine | 7±1 | 7±1 | 8±1 |
| Rhine + $Al^{3+}$ | 7±0.5 | 8±0.5 | 7±1 |
| Rhine + $Cu^{2+}$ | 8±0.5 | 7±1 | 8±0.5 |
| Rhine + $Sn^{2+}$ | 7±0.5 | 7±0.5 | 7±1 |
| Rhine-M | 8±0.5 | 9±1 | 8±0.5 |
| Rhine-M + $Al^{3+}$ | 8±1 | 8±0.5 | 9±1 |
| Rhine-M + $Cu^{2+}$ | 8±0.5 | 8±1 | 8±0.5 |
| Rhine-M + $Sn^{2+}$ | 8±1 | 9±0.5 | 9±0.5 |
| Indus | 8±0.5 | 8±0.5 | 8±1 |
| Indus + $Al^{3+}$ | 8±0.5 | 8±1 | 9±0.5 |
| Indus + $Cu^{2+}$ | 8±1 | 8±0.5 | 7±1 |
| Indus + $Sn^{2+}$ | 7±1 | 7±0.5 | 7±0.5 |
| Pacific | 8±0.5 | 8±0.5 | 8±0.5 |
| Pacific + $Al^{3+}$ | 8±1 | 8±1 | 8±0.5 |
| Pacific + $Cu^{2+}$ | 8±1 | 8±0.5 | 7±1 |
| Pacific + $Sn^{2+}$ | 7±0.5 | 8±1 | 7±0.5 |
| Caspian | 8±0.5 | 8±1 | 9±0.5 |
| Caspian + $Al^{3+}$ | 8±0.5 | 8±0.5 | 8±0.5 |
| Henna | 7±1 | 7±0.5 | 7±0.5 |
| Henna + $Al^{3+}$ | 7±0.5 | 8±1 | 7±0.5 |
| Vegdye BRE | 8±1 | 8±1 | 8±0.5 |
| Vegdye VO | 8±1 | 8±0.5 | 8±1 |
| Hematine CFMK | 8±0.5 | 8±1 | 8±0.5 |
| Vegdye MUS | 8±0.5 | 8±1 | 8±0.5 |

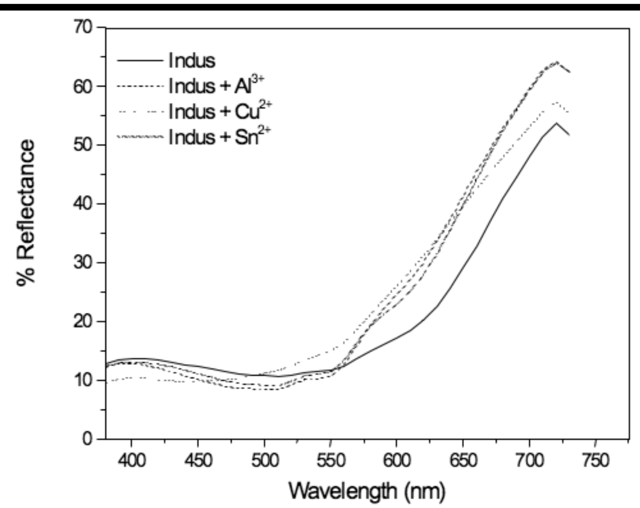



Figure 4: Reflectance spectra for leather dyed using Indus with and without mordants

and Vegdye MUS). The shade difference is seen from Table II.

### Wet, Dry and Light Fastness

The results for the wet and rub fastness for all the crust leathers is presented in Table IV. It can be seen that the all the leather possess relatively high fastness to wet and dry rub fastness. This is mainly due to the high affinity and reactive nature of natural dyes. It is known that the fabric dyed using natural dyes has poor fastness to light. Hence, light fastness of the leather dyed using natural dyes with and without mordants was carried out. Light fastness data for the crust leathers dyes using natural dyes are presented in Table V. It is interesting to note that the crust leather possess high fastness to light. This could be due to the interaction of natural dyes with skin protein, which prevents the oxidation of natural dyes.[18] However, the leathers treated with Indus exhibited slightly lower fastness properties.

### Visual Assessment

The visual assessment of leathers was carried out to assess the dye uniformity, dye affinity and penetration of dye. The visual assessment data is given in Table VI. The data indicates that the natural dye provides a uniform and level color to the entire surface of the leather. Generally, natural dyes have good affinity towards skin proteins. Hence, the natural dyes employed in this study possess good dye affinity behavior. Further, the penetration of dye in to the matrix is vital since the natural dyes are plant polyphenols with higher molecular weight. However, the dye penetration is good for all the crust leathers dyed naturally.

## Conclusions

Leather industry is poised for an upward surge. In order to realize better value for our raw material, it is essential for the industry to couple fashion with utility alongside an eco-benign character. In this direction, the role of dyeing is extremely important. A revisit to the use of natural dyes for coloring of leather has been made. It has been possible to develop a range of colors by using commercially available natural dyes either using them directly or through mordanting with metal ions commonly used in households like aluminum, copper and tin. The color developed from this work highlights the significance of the work in generating colors to meet the demands of the fashion world. Some highlighting features of this work are

- o 24 natural shades have been developed
- o Mordants play an important role with natural dyes
- o Colors have good affinity and light fastness
- o Provides better dye affinity and penetration

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