THE ISOLATION AND WASHED FASTNESS TESTING OF CRUDE NATURAL DYES OF BAGBAGOTOT {Phyllantus reticulatus) BERRIES FROM VIGAN ILOCOS SUR

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ABSTRACT

Bagbagotot (Phyllantug_reticulatys) berries were collected from ligan, 1locos Sur; then soaked in distilled water and concentrated in rotavapor at 4OC. Water- soluble crude natural dyes were obtained through freeze-drying and yielded 9.20%.

The fastness of the natural dye was tested. Ferrous sulfate, copper sulfate, alum and potassium dichromate were used as mordants applying in 2 different yarns, i.e., cotton and silk.

1mong all mordants in the 3 techniques namely post mordanting, pre-mordanting and simultaneous mordanting, Jerrous sulfate is the darkest, with copper sulfate, alum and potassium dichromate as the lightest.

Dyeing a yarn with this extract with mordants shows darker washed fastness in silk than in cotton, mordant generally helps the fixation of the natural dye on fibers.

Dyeing without mordant is similar to dyeing with alum mordant at 5% concetration.

Increasing the concentration of mordant to 10% potassium dichromate manifests a significant change which can not be seen in all the treatments and strategies.

INTRODUCTION

Many plants and trees are just growing without being given importance. Some are even poisonous and arc wastes to the environment especially when thcy bcar fruit. People can't find ways to utilize them so that their existence will not be useless.

With this situation. people in contact with these plants don't have intcrest and dare not to identify their actual common names more so with the scientific names. Some plants are endemic to a place and no matter how much we destroy and eradicate such species, they continue to grow. But there will come a time, that when they had already disappeared, we come to realize that they have significant uses, and are very important renewable sources of some potentials.

At the start of the coming century. we have scen the shift of most scientists and industrialists from the vcry destructivc technology which is synthetics to an cnvironment friendly world of natural resources.

In thc world of dyes, many entrepreneurs have sccen thc potentialitics of natural dycs. Thesc are isolated from plants mainly trees and shrubs.

The llocos region is the endemic habitat of many tropical plants, trccs and shrubs which are not so familiar because ofunkown and undiscovered potentials. Otce of these plants is a shrub locally known as "bagbagotot". Referred to a botanist at the University of the Philippines at Los Banos, shc identified thc plant as Phyllantus reticulatus. In the local Tagalog dialect. it is called "tinta- tintahan".

Bagbagotot is a shrub with broad branches arising from the base with heights up to 3 meters from the ground. It has minuted leaves. entire margin, reticulate in shape and ever green all throughout. It bears pink flowers at the branches where the lcaves are and fruits comc in bcrrics. Berries are colored rank or deep blue and turn indigo when matured. Berries range from I.5 to 2.0 cm in diameter and juices range from maroon to violet in color. They thrive in both wct and dry soil. Usually growing endemically without being propagated, they bear fruit throughout thc ycar and can survive in both cxtremc seasons.

Bagbagotot is believed to be a potential natural source of dyes bccause when accidentally stained on clothes, one finds it hardly removed. It has bcen observed to stay in color for ycars.

It is the main objcctivc of this rcscarch to isolate the natural dye cxtracts from the fruit berries and test their dyc properties on textiles. after which future identification and characterization will be donc.

When fully identificd or *tested,* the bright future of culturing. growing and harvesting fruits and putting them into industrial scale will be done.

REVIEW OF RELATED LITERATURE

Dyes come in many forms. They are found to be useful and they havc wide range of uses as cited Zajac (1992) natural dyes are important in food industry. Dyes such as anthocyancs from black current pomace are tried for industrial produciion stability of dried dye concentrates was estimatcd. The anthocyane losses were estinated after one year storage for I0-14 percent and thus a highly stable product was obtained. Recipes were prepared with practical guide on edible and useful plants were (Jull, 1987). Dyes are important in microbiology for they are used as substrats in arting Bruella mglintusis biovar I atyphical strains in indicating the evolution of a new variant (Banai, et. al, 1990). Scott and Jons performed an experiment by evaluating Lycopcrsicon sp. accessories for resistance to bacterial spot. (Xanthomonas computric pr. vcsicatoria (Dridge) dye. They are used for labclling and marketing. Retention time and toxicity of dye marker Sudan' red TB on Formson and eastern subterranean termites (Su, Scheffrahn & Bon, 1988). They are utilized in detecting of virus. Sibsen (1988) determined slot immunobinding assays for virus detections in potatoes which manifest advantages of soluble dyes. Gracc and Abdally (1990) labelled eastern subterranean termites, Reticuliterms flayipes with dyes using Sudan Blue 35 in foraging and behavioral studies. Termite workers were fed filter paper containing dye concentrations of 0.1, I, 2, 4, 6 and 8 percent for period of 5-15 days. Concentrations greater than 2% deterred feeding, which resulted in **less effective** labelling. and elicited significant mortality.

As an alternative to other dye makers. Sudan Blue 35 would be suitable for field studies with R. Flayipes employing mark-

release-capture cycles **of** 5- 6 days, or for labellings termites in laboratory studics of behavioral interations for fceding preferences.

Sources and Classifications

Triphcnylmethane dyes wcrc cxtractcd from macrophytzcs (Timofccva and Cheromnyth). 1989). The symplastic transfer of fluorescent dyes from mcsophyll **to** sieve tubc in stripped leaf tissuc and partly isolatcd minor veins of Commclincr bcnqhalgnsis Tccton grandis L. and Vilgy payiflora Juss revcalcd thc localization of tannins and pigments in the parcngyma cells of the lcavcs and stems. Both colorants from the test plants occurcd at varying concentrations and color intensities. Aqucous cnudc dye cxtracts from the plant samples scanned from 360-780 um showed that the colorants wcrc water soluble and cxistcd in mixture as exhibitcd by broad pcaks. (Quinto, 1992). Turmeric (Curcuma longa) rhizome finds usc as colouring matter and also as condiment. The yellow matter distributed throughout thc plant is mainly concentrated in the rhizome. In Indian systems of mcdicinc it is also uscd to some extent asa tonic acid (blood purifier Agrawal, Goel and Gupta. 1992). Curcumin is the main coloring matter which dcvclops ycllowrcd color with concentrated sulfuric acid (Wealth of India, 1950).

Natural dyes can broadly bc classificd as natural organic dyestuffs of vcgetablc and animal origin and mineral dyestuffs an inorganic pigments. Thc organic dyestuffs arc obtained from roots, stems. lcavcs, berries and flower of various plants and from certain insccts and shellfish with a very claboratc scrics of processes. Thcsc dyes, with a vcry few to be used with a mordant for fixation. Thc inorganic pigments arc insoluble salts precipitatcd on fiberby suitablc double decomposition. Natural organic dyes arc subdividcd into indigo and related compounds, logwood,' dyes producing red shades and dyes producing other than red shades.

Indigo is extracted from Indigofera Tinctoria a bush of the pepper family. It is a maximum dye content of 0.4% (Kamat & Alat, 199). Wood is similar to indigo from fleshy leaves of the plant "Isatis tinctoria". The extraction is similar to indigo, Logwood produces a blue-black shade on wool and black shade on silk containing Haematoxylin which is colorless and having the following formula $C_{,}H_{,,}O_{,}$. Which on oxidation is converted to Haematein $C_{,,}H_{,,}O_{,}$ the blue-black dye. Natural indigo is manufactured by a process that has remained essentially unchanged throughout the ages. It consists of two distinct operations, the fermentation of thc plant and arial oxidation of the liquors. It is now extensivcly used in dyeing denims (Aberydira, 1979). Catechu apart from being a dye has also a prescrvativc action and it is used in printing. Il is used for Khaki dyeing. Annatto dye is also extracted from Bia orgllana, a tropical shrub growing in South America and South Asia. lts dye is bright crimson taken from seed. Sappan wood from Sri Lanka denim its color from Brazilin. $(C_{,}H_{,,}O_{,})$ that is soluble in water and alcohol. It is converted into Brazilein $(C_{16}H_{12}O_{5})$. When exposed to atmospheric oygen. The heartwood or sawdust of Arthocarpus inteqrifolia yield a yellow dye employed in colour monks robes. It is fixed with alum and intensified with turmeric. (Abeyrida. 1979).

MATERIALS AND METHODS

The specimen, Phyllantus reticulatus fruits were collected in Rugsuanan, Vigan, llocos Sur and transported to WST, UPLB where they were processed. Fresh **berries** were weighed and boiled with distilled water. Extracts containing water were

concentratcd in a rotavapor at vacuum and 80C-90C water bath. The mixed distilled watcr were scparatcd leaving the extract natural dyes.

Isolation

Dyes arc now frozen dricd using vacuum and temperature of 50C for three days after samples had turned powdery and dry at the Forest Product Rescarch and Development Institute (FPRDI). Testing of Textile Research Institute at Bicutan. Taguig, Metro Manila.

APPLICATION OF MORDANTS (PRE-MORDANTING)

The fastness of the natural dye was tested. It is done initially by thc application of four different mordants (ferrous sulaftc. copper sulfate, alum and potassium dichromatc).

Thie four mordants were applicd scparatcly in two types of yarns. Thcsc arc cotton and silk. Wcigh the yarns into four samples. Determinc the amount by setting it at a spccificd concclrations both on silk and cotton. Similarly. the volume of water uscd is based on the liquor ratio. Mix the mordant and the water (solvent) and hcat for IO minutcs at 6rC water bath. Squcezc it until it is totally draincd.

Dyeing

Prepare now the dye to be used. The amount of dyes is 50% of the wcight of the yarn. The dye is mixed with thc water which is bascd on thc liquor ratio. Cotton yarn requires L:3() while silk is 1:2() which is computed at 30% of the weight of yarn. Placc the yarn in thc mixture. Heat in water and drain for an hour. Nowcompletely rinse it to clear.

Washing

Prepare a washing solution of detergent with water at 20% concentration, Wash the cooked and dyed yarns up to "bleedfree". Dry it in a shady area. Fastness is determined.

APPLICATION OF MORDANTS (POST-MORDANTING)

The fastness of the natural dye was tested. It is done initially by the application of four different mordants (ferrous sulfate, copper sulfate, alum and potassium dichromate) after the dye has bcen mixed with the fibers. The same specifications were followcd only that dyeing comcs ahcad of mordanting.

APPLICATION OF MORDANTS (SIMULTANEOUS-MORDANTING)

Theeffectiveness of the dye was tested via this process. The natural dye together with the mordants were mixed in specific volume of water at the same time and allowed to color the fibers (cotton or silk) altogether or they are stirred occasionally in a hot water bath at IO0C [or one () hour.

VARYING THE CONCENTRATION OF MORDANTS

Thc effectiveness of the dye is tested by varying thc mordant concentration. After 5% initial application, it was followed by 10%. The determination of the amount of mordant depends on the concentration being sct.

Figure I
Size of Baghagotot shrubs (Phyllanthuy rgticnlatus) grown endemically in drylands of 1Ilocos

Figure 2
PIlanths reticulatus branch and leaves

Figure 3
Phy/lanthus reticulatus bearing pink flowers in bloon

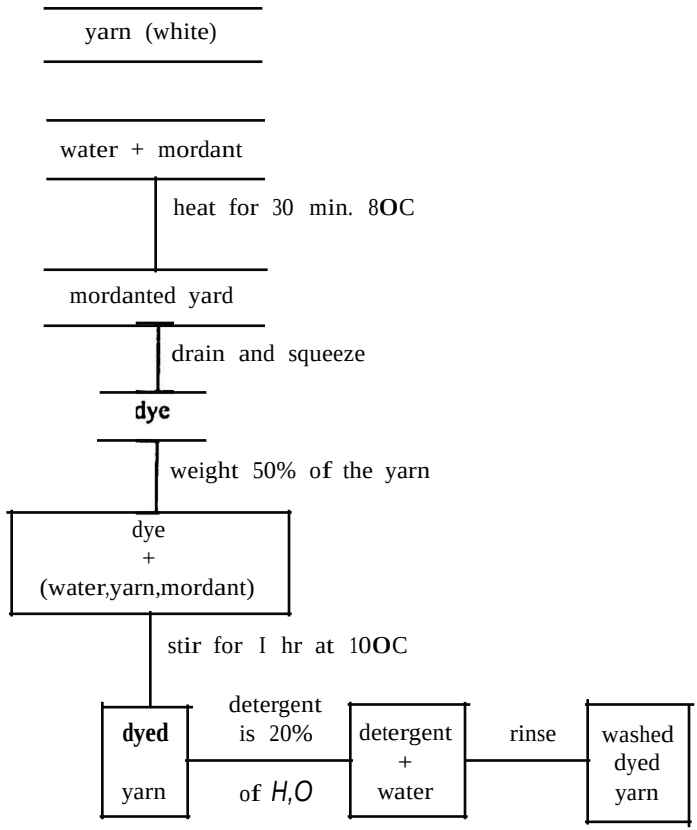

Figure 5. Pre-Mordanting Followed by Dyeing and Washing

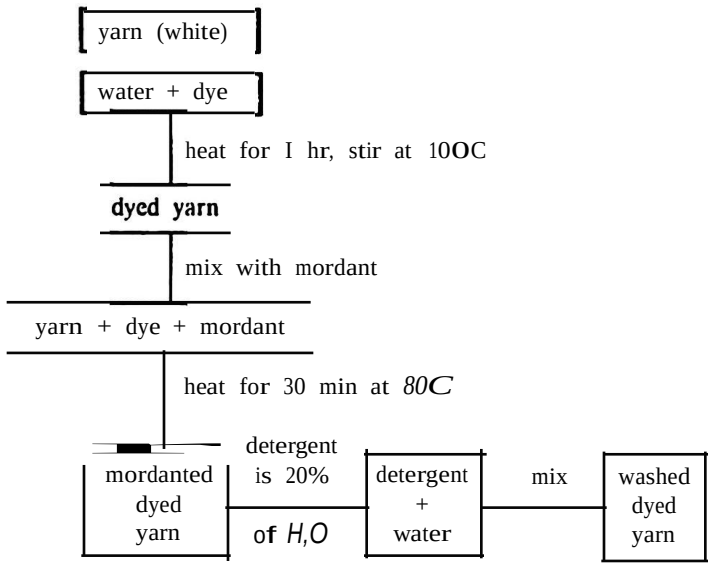

Figure 6. Dyeing Followed by Mordanting and Washing (Post Mordanting)

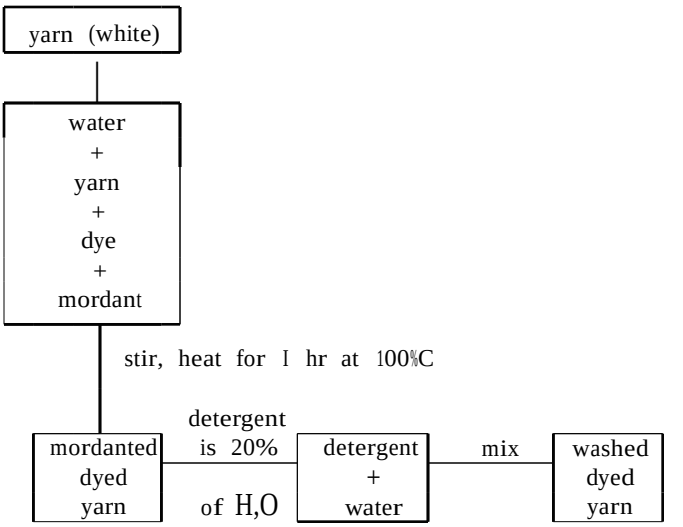

Figure 7. Dyeing and Mordanting Simultaneously Followed by Washing

GENERAL SPECIFICATIONS

Crude dye: Phyllantus reticulatus dye

Sources of dye: water-soluble; freeze dried berries (blue violet)

Mordants:
- Ferrous sulfate (white)
- Copper sulfate (blue)
- Alum (white)
- Potassium dichromate (orange)

Yarns:
- silk
- cotton

Solvent:
- water

Detergent:
- sodium sulfactants

Concentration used:
- 50%. 10%

Technology applied:
- pre-mordanting
- post-mordanting
- simultaneous dyeing mordanting

RESULTS AND OBSERVATIONS

Phyllanthus reticulatus berries contain an indigo to black color juices which are soluble in water. These substances cause staining of cloth fibers. The chemicals were isolated in Soxhlet extractors using water as solvent, concentrated in a rotavapor and solidified by a freeze dryer at -50C in 24 hours.

The percentage yield is 9.205%.

The effectivity of the natural dye is tested using the washed fastness as the basis where the dye is mixed with a solvent water and a fixing chemical - a mordant. There are four (4) mordants: namely ferrous sulfate, copper sulfate, alum and potassium dichromate.

The dye is tested into two (2) varying concentration, 5% and 19% of mordant mixed. In four different strategies: without mordant, pre- mordanting, post-mordanting and simultaneous deying and mordanting. The dye is tested on 2 types of fabrics or yarns, the cotton and the silk.

The range of the colour are from dull white up to decp black. Usually dull white, dank white, yellowish brown, light brown, dark brown gray, dark gray. light black, black and deep black. For the first treatment, 5% concentration of the mordants added. Only pre-mordanting was performed while 10% concentration of the mordant added to all the three were performed. (Pre-Post-Simultaneous). For 5% concentration of mordant the following were computed and manifested the result in Figure 9.

A. Silk Pre-Mordanting

weight of the silk = .25g
at 5%; weight of mordant
= .05x.25g=0.0125g
volume of water
at 1:30 liquor ratio= 30 x .25
= 7.5 ml
heatcd at 60C for 30 min

Dyeing

weight of dye
(50% of the yam)...=.50x.25=.125 g
volume of water = .50 x .25 = 7.5 ml
heat at 100C for I hr, stir occassionally

Washing

washed with detergent (quickwash)
at 20% concentration with water,
300 ml water
weight of detergent=30 ml x.20 = 60g
totally washed and rinsed until there
was no bleed

B. Cotton

Pre-Mordanting

weight of the cotton = .75g
at 5% weight of mordant =.05x.75g
= 0.0375g
volume of water
at 1:20 liquor ratio = 20 x .75
= 15 ml
heat at 60°C for 30 min.

Dyeing

weight of dye
(50% of the yarn) = .50 x .75
= .375 g
volume of water = .20 x .75
= I5 ml
heat at 100C for I hr,
stir occassionally

Washing

washed weight quickwash detergent
at 20% concentration with water,
300ml water
weight of detergent = 300 ml x .20
= 60g
washed and rinsed until there was
no bleeding

Post Mordanting

A. SILK - dyeing, mordanting, washing
B. COTTON - dyeing, mordanting, washing

Note: The computations were bascd on the same formula. Only the sequence is changed.

Simultaneous **Dyeing and Mordanting**

A. SILK - (dyeing/mordanting), washing
B. COTTON - (dyeing/mordanting), washing

Note: Total time for mixing is I hr at water bath at 1I0C

Dyeing without mordant was performed. There was just a slight alteration of the color of the white yarn which is specified by the PTRI.

DISCUSSION

Dyeing cotton without mordant is similar to dyeing acotton having 5%pre-mordanting using copper sulfate or alum. Comparing 5% with 10% pre- mordanting, there was no change in the color of cotton but for 5%and no mordant, dyeing without mordant matches with the result of $CuSO'$ mordanted and slightly with alum mordantcd. Among the results in 10% concentration, post mordanting is the darkest, followcd by simultaneous and lastly thc prc-mordanting. But there is a similar color or washed fastness degree at 10% in the copper sulfate (simultaneous) and potassium dichromatc (post- mordanting).

Among all mordants in 3 strategies, ferrous sulfate is thc darkest. copper sulfate, alum and potassium dichromate being the lightest except in 10% post-mordanting that alum is the lightest where potassium dichromate outcolored alum.

Washed fastness of silk **are far** better than cotton with a shinier physical **appear**ance manifested. Among the 10% concentration treatments, all the results are the same for post, pre and simultaneous except for a similar outcome in 10% between copper sulfate (simultaneous) and potassium dichromate (post-mordanting). Comparing 5% and 10% (pre-mordanting), there is a darkening of the silk. Dyeing without mordant is similar *to* 5% (Pre) using alum.

Among all mordants in 3 strategies, ferrous sulfate, is the darkest, copper sulfate alum and potassium dichromate, respectivcly. Except in 10% post mordanting that alum is the lightest which is outcolored by potassium dichromate.

CONCLUSION

The yield is higher than most renewable sources. The chemicals found in bagbagotot (Phyllanthus reticulatus) is water-soluble which is a potential dyestuff.

Increasing the concentration of mordant results to darker washed fastness in both silk and cotton.

In a uniform mordant concentration, of the three (3) strategies, post mordanting is the most effective.

Generally, dyeing a yarn (fiber) with Phyllanthus reticulatus dye with mordants shows darker washed fastness in silk than in cotton. Without a mordant, the dye is slightly fixed on both cotton and **silk.** Mordant generally helps the fixation of the natural dye on fibers.

Dyeing without mordant is similar to dyeing with alum mordant at 5% concen(ration.

Increasing the concentration of mordant to 10%; potassium dichromate manifests a significant change which can not be seen in all the treatments and strategies.

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