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Natural Red Lake Pigments Extraction from Selected Plants in Agusan Del Norte, Philippines: Characterization using Fourier Transform Infrared Spectroscopy

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ABSTRACT

Vibrant colors are all around us, but natural pigments are gaining importance as we become more aware of environmental and health risks. In particular, while yellow is abundant in nature, red is limited. Thus, this study aims to determine the plant sources of red pigments that could help increase the production of natural dyes. Five plant samples, namely Achiote (*Bixa orellana* Linn.), Vine spinach or Alugbati (*Basella alba* Linn.), Coleus or Mayana (*Plectranthus scutellarioides* (L.) *R. Br.*), Scarlet jungle flame or Santan (*Ixora coccinea* Linn.), and Teak (*Tectona grandis L.f.*), were extracted using distilled water as a solvent with alum (aluminum potassium sulfate, $KAl(SO_4)_2 \cdot 12H_2O$) as a mordant at a ratio of 1:50. The resulting dye extracts were then treated with 25 g of sodium carbonate Na_2CO_3 to extract red lake pigments. The results showed that the extracted lake pigments varied in color, with orange, white, green, reddish-brown, and brown, respectively. While red lake pigments were not produced, the findings suggest the potential for increased natural dye production and provide a promising source for further investigation to support research on natural pigments.

Keywords: *Alum, Natural dyes, Red lake pigments*

1 Introduction

Environmental pollution is a critical problem today (Sobhanie et al. 2022). The textile industry is one of the major contributors to pollution, generating high liquid effluent pollutants through synthetic dyes and large quantities of water during textile processing (Periera and Alves 2012). The industry utilizes around 8,000 chemicals during different processes, including dyeing and printing, and many of these chemicals have long-term hazardous and damaging impacts on the environment and human health (Kant 2012). The chemicals can cause skin irritation, itchy or blocked noses, sneezing, sore eyes, and respiratory problems due to inhalation of dye particles, especially when exposed to reactive or synthetic dyes (Hassaan and Nemr 2017). Thus, there has been a growing demand for natural colorants that do not have these harmful effects (Pervaiz et al. 2016).

Before synthetic dyes, traditional dyes were made from natural sources such as plants, fungi, insects, animals, and minerals for coloring silk, wool, and cotton fabrics (Janani et al. 2014). Natural dyes are characterized by their wide variety and economic advantage over synthetic dyes due to their non-carcinogenic, non-toxic, non-poisonous, non-hazardous, biodegradable, and renewable properties (Pervaiz et al. 2016). Additionally, natural dyes generate unusual, soothing, and gentle shades compared to synthetic dyes (Arora et al. 2017). Therefore, the need for natural dyes has been increasing (Talib et al. 2023).

While many plants can produce various colors, the production of red color is limited in nature (Vancar 2000), making it highly sought after.

An example of a plant that produces red color is provided by Sappan Wood or *Caesalpinia sappan* L. Light yellow is produced by its heartwood when freshly cut but quickly changes to red. The extract from this plant has a strong antibacterial action that could preserve the purity of formulations in cosmetic creams, gels, and other products. It also gives a bright color and artistic use to bath soaps and color cosmetics. The property of color change with pH from yellow to red can be advantageously used to produce color-changing lipsticks (Suganya et al. 2016). According to Barhanpurkar et al. (2015), natural dyes are becoming essential for people due to increasing environmental awareness. While not all natural dyes are non-toxic, they are generally eco-friendly.

Thus, this study aims to determine plant sources of red lake pigments to increase the production of natural dyes. Specifically, it seeks to extract natural dye solutions from Achiote (*Bixa orellana* Linn.), Vine spinach or Alugbati (*Basella alba* Linn.), Coleus or Mayana (*Plectranthus scutellarioides* (L.) R. Br.), Scarlet jungle flame or Santan (*Ixora coccinea* Linn.), and Teak (*Tectona grandis L.f.*); produce natural red lake pigment from the extracted natural dye; and partially characterize the structure of the red lake pigment using Fourier transform infrared spectroscopy.

2 Materials and Methods

Selection of Natural Dye Sources

The study utilized various samples and their corresponding common names, local names, scientific names, and the plant parts selected to extract red lake pigments (Table 1). The five samples used in the study were selected based on their known color properties and pigment content. The *B. orellana* seeds and *I. coccinea* flowers were chosen for their strong red dye (Kala et al. 2015, Ghurde et al. 2016), while *B. alba* fruit was selected for its red pigment content (Lin et al. 2010). Also, *P. scutellarioides* leaves were used because of their high anthocyanin content, which is responsible for producing red color (Borek et al. 2016), and *T. grandis* young leaves were selected for their yellowish-brown or reddish dye (Asiong et al. 2017).

Collection and Preparation of Plant Samples

The methods used in all steps, except for mass content analysis, were adopted from Verenkar and Krishnan (2017) and an unpublished manual on the preparation and application of colorants from natural dyes (Faculty of Science, Silpakorn University, Thailand, 2018).

Five different plant samples, namely *B. orellana, B. alba, P. scutellarioides, I. coccinea,* and *T. grandis* were collected for this study. A taxonomist from the Biology Department of Caraga State University identified these plant samples. After collection, the plant samples were washed with tap water, rinsed with distilled water, and air-dried in the laboratory room for about 4-5 days or until thoroughly dried before the extraction analysis. A portion of each plant sample was set aside for moisture content analysis.

Moisture Content Analysis

Fifteen porcelain crucibles were weighed and dried at 110 ± 5 °C in an oven. Once the desired mass was reached, 1 g of each plant sample was added to each crucible (performed in triplicate) and dried in the oven to constant mass. (Adopted from the Department of Transportation 2010)

Extraction of Dye

Dye extraction was performed by dissolving 10% alum ($KAl(SO_4)_2 \cdot 12H_2O$) in 500 mL of simmered distilled water. A 100 g plant sample was mixed in the solution and heated for 4 hours on a hot plate set at 80 °C. The resulting solution was filtered using a cloth, and the filtrate was

Table 1. Plant sources used for red lake pigments

Common name	Local name	Scientific name	Parts of plants extracted with red pigment
Annato	Achiote	*Bixa orellana*	Seed
Malabar spinach	Alugbati	*Basella alba*	Fruit
Mayana	Mayana	*Plectranthus scutellarioides* (L.) R. Br.	Red leaves
Scarlet jungle flame	Santan	*Ixora coccinea* (Linn.)	Read flower
Teak tree	Hadlayati	*Tectona grandis*	Leaf bud

used for pigment extraction. The same process was repeated without using alum as a mordant. It served as a negative control to determine if any observed effects were due to alum or simply a result of the extraction process.

Preparation of Red Lake Pigment

Each dye solution was filtered using cotton fabric, and the filtrate was slowly added with 25 g of sodium carbonate while stirring. It was then filtered again using ordinary filter paper. The residue was air-dried and weighed when thoroughly dried.

Calculation of Crude Lake Pigment Yield

The percentage yield of red lake pigment was calculated using the following formula:

$$\%\ yield\ of\ dry\ sample = \frac{W_{pigment}}{W_{dry\ sample}}$$

Where,

$W_{pigment}$ = weight pigment (g)

$W_{dry\ sample}$ = weight of the dry sample (g)

Partial Characterization of the Structure of the Pigment

A total of 2 mL of dye extract and 0.1 g of lake pigment, taken from each five extracted samples, were subjected to characterization using Fourier-transform infrared spectroscopy.

3 Results and Discussion

Extraction of Natural Dye

The natural dye solutions were extracted from *B. orellana, B. alba, P. scutellarioides, I. coccinea*, and *T. grandis* using distilled water as a solvent. The hues of the extracted dyes were recorded and confirmed using a color wheel chart (Table 2). Also, further investigation was conducted to examine the effect of a mordant on the dye. Specifically, the Achiote sample was used for extraction with and without mordant (alum) to compare the resulting colors. The dye extract without alum resulted in a darker red color (Figure 1A & C) than the extract with alum (Figure 1B & D). These findings align with the study of Prabhu and Bhute

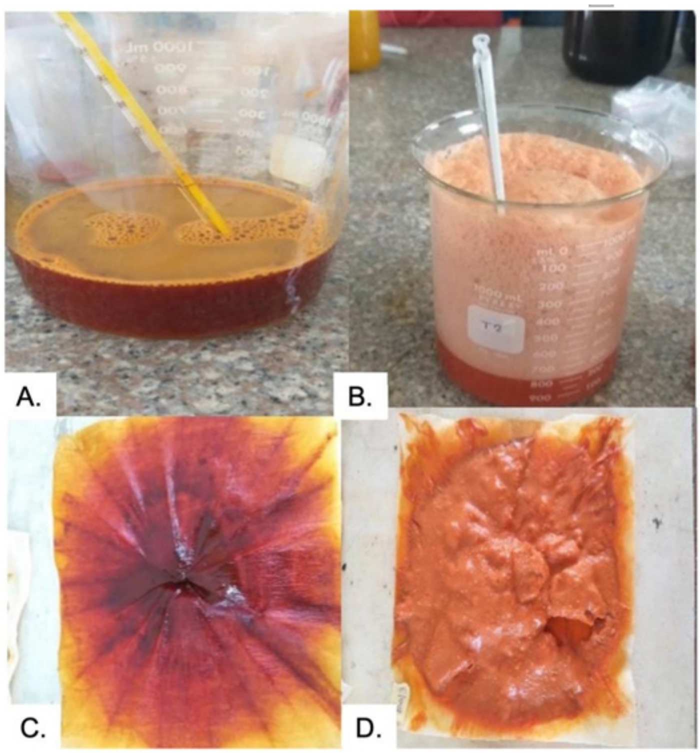

Figure 1. Water extraction of Achiote: Dye A) without Alum, B) with Alum, C) without Alum, D) with Alum

(2012) that mordants produce bright colors, thus enhancing the brightness of the dyes.

In addition, FT-IR characterization was conducted to compare the functional groups present in the dye extracts and their corresponding lake pigments that are proposed to be the primary contributors to enhance chemical interactions.

Figure 2 displays the FT-IR spectra of the dye solutions extracted from the five plant samples (Table 1). Figures 3 and 4 present the FT-IR spectra of alum aqueous solution and distilled water, respectively. The results revealed that the extracts from five plant samples (Figure 2) exhibited similar peaks in their spectra, including the broad and strong O-H stretching at 3200-3550 cm^{-1}, and OH bending at 1630-1780 cm^{-1}. These peaks are due to the water solvent used in the study, as indicated by Figure 4 and supported by prior research (Vrancken et al. 2017).

Also, –C-O stretching peak at 1020-1275 cm^{-1} in both the dye extracts (Figure 2) and the alum solution (Figure 3) was also found. This peak indicates high amounts of dye in the solution for the dye extracts, as noted by Al-Alwani (2017). This peak for the alum solution is evident in a pigment derived from the dye extract added with alum, as reported by Deveoglu et al. (2012).

Extraction of Red Lake Pigments

The expected color of the lake pigments produced was red, but the actual pigments obtained were orange, white, green, reddish brown, and brown (Table 3). According to Asiong et al. (2017), several factors can significantly affect the final color of the pigment. These factors include the solvents used, temperature, pH of the solution, and the mordant used.

Additionally, the color of the pigment can

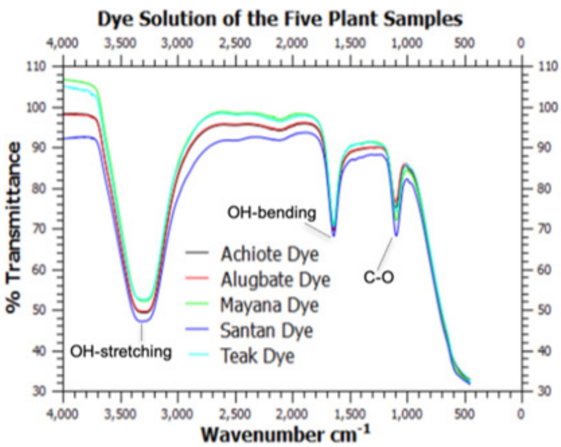

Figure 2. IR spectra of the dye solution extracted from the five plant samples

Table 2. Color of dyes

Plant samples	Color produced
Achiote	Red orange
Alugbate	Red orange
Mayana	Purple
Santan	Red
Teak	Dark red

Table 3. Color of pigments

Plant samples	Color produced
Achiote	Orange
Alugbate	White
Mayana	Green
Santan	Reddish brown
Teak	Brown

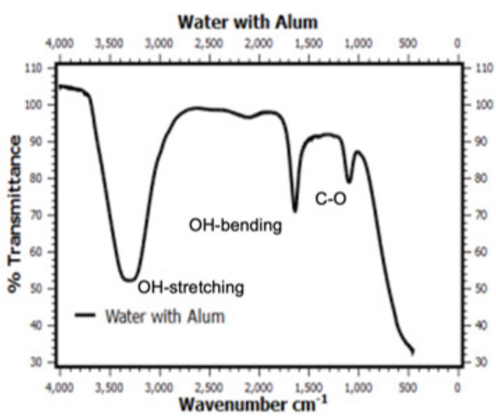

Figure 3. IR spectra of alum aqueous solution

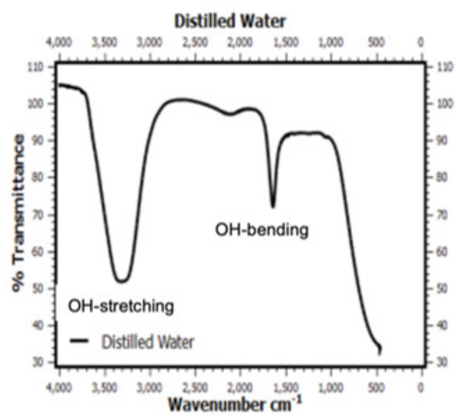

Figure 4. IR spectra of distilled water

vary depending on various factors, such as when the plant is picked, how it was grown, soil conditions, and minerals in the water used in a dye bath (Wahyuningsih et al. 2017). Therefore, the color of the pigment obtained may sometimes be different from the expected color and can vary depending on several factors involved in the production process.

Several methods are available for extracting colorants, with water extraction being one of the oldest and most traditional methods (Sanku et al. 2021; Akogou et al. 2017). The efficiency of pigment extraction depends on the chosen method (Raddatz-Mota et al., 2016). Table 4 presents the average amounts of lake pigments obtained from the five plant samples. Since this study utilized distilled water for pigment extraction due to its eco-friendly process (Sanku et al. 2021), the results indicated that Mayana exhibited the highest percent yield at 27.62 g, while Achiote had the lowest at 22.32 g. Also, statistical analysis revealed no significant difference (p>0.05) among the five plant samples, suggesting a similar overall pigment content when comparing the dry samples.

Table 4. Average amounts of lake pigments

Plant sample	Ave. wt. of wet sample	Ave. % moisture	Wt. of dry sample	Ave. wt. of pigment	% yield of dry sample
Achiote	100.06	63.03	112.10	25.02	22.32
Alugbate	100.13	72.28	102.90	24.17	23.48
Mayana	100.16	88.75	86.41	23.86	27.62
Santan	100.15	80.24	94.86	24.06	25.37
Teak	100.75	73.78	101.60	26.69	26.28

Characterization of Red Lake Pigments

The characterization of the lake pigments from five plant samples is shown in Figure 5. The absorption band at 3200-3550 cm^{-1} indicates OH stretching, while the absorption band at 1420 cm^{-1} (C-O) is attributed to the carbonate (CO23) group. This can be inferred that carbonate co-precipitates with hydrated alumina, as Deveoglu et al. (2012) reported.

Also, the shifts observed in the peaks at 1450-1600 cm^{-1}, which correspond to C=O (carbonyl) and hydroxyl absorptions observed in 3200-3550 cm^{-1}, can be attributed to the strong chelation between the dye molecules and the alumina surface (Deveoglu et al. 2012). As mentioned previously, the peaks observed at 1020-1275 cm^{-1} can be attributed to C-O stretchings, which indicate a high amount of dyes.

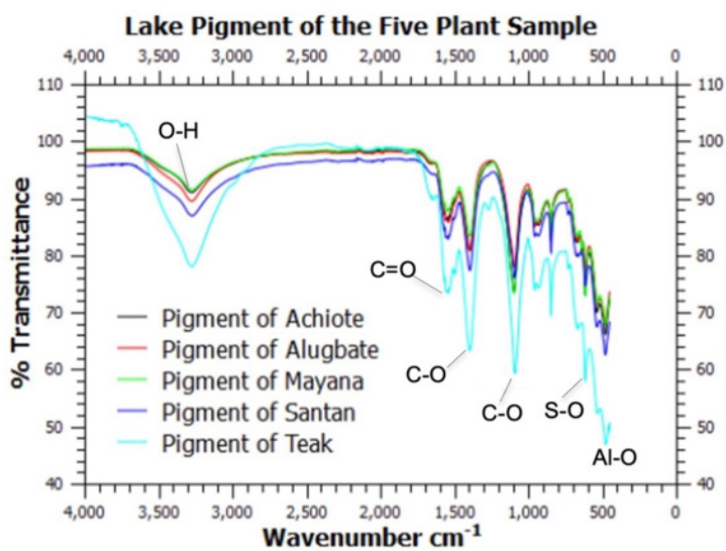

Figure 5. IR spectra of the pigments extracted from the five plant sample

Lastly, the peak observed in the region between 500-750 cm^{-1} is associated with Al-O vibrations (Girdthep et al. 2018), while the peaks at 1125-985 cm^{-1} indicate S-O (sulfate) bands which are of similar band size to Al-O (Kirby et al. 2005).

The presence of alum in the dye solution (Figure 2) was not detected in the FTIR analysis. Still, it was observed in the peaks of the lake pigments (Figure 5), indicating that the alum may be potentially converted to the lake pigment form. Furthermore, the carbonate group was only detected in the lake pigments, as it was added during the pigment extraction process and not during the dye extraction process. These findings suggest that dye solutions (Figure 2) and lake pigments (Figure 5) exhibit similarities and distinct differences in their functional groups.

4 Conclusion and Recommendations

In summary, the five plant samples showed potential for natural dye extraction, with the conversion to pigments resulting in color changes. While red lake pigments were not produced, the study suggests the potential for increased natural dye production and provides a potential source for further investigation. Future studies could explore different solvents, temperatures, and time to improve color quality and assess pigment toxicity. Lastly, comparison with synthetic pigments could determine the viability of natural pigments as an alternative.

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Statement of Conflict of Interest

The authors declare that there are no conflict of interest regarding the publication of this paper.

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