

Preparation of Organic Ink from Natural Pigments Present in *Beta Vulgaris L.* (Beetroot) and *Curcuma Longa L.* (Turmeric) for a Sustainable Environment

Dr. Rupa Pegu

Assistant Professor, Department of Chemistry, Biswanath College

Abstract:

Organic inks containing plant-based pigments provide a sustainable shift in reducing environmental impact and toxicity compared to synthetic inks. Therefore, in this work, organic ink formulations from beetroot and turmeric extracts were explored, emphasizing the natural pigments present in them. Four different ink formulations for each extract were prepared by mixing with different proportions of extra virgin olive oil, drumstick resin, and lemon juice, using water or ethanol or both as a solvent. Out of all formulations, a remarkable result has been achieved using one formulation of each pigment (formulation 3 of beetroot extract, which is composed of 65% extracted pigment (v/v), 15% ethanol (v/v), 15% water (v/v), 5% lemon juice (v/v), three drops of extra virgin oil, and three drops of drumstick resin, and formulation 4 of turmeric extract, which is composed of 55% extracted pigment (v/v), 40% ethanol (v/v), 5% lemon juice (v/v), four drops of extra virgin oil, and four drops of drumstick resin). The two best ink formulations retained their vibrant colour till the 28th day while applied on paper using an ink pen and paintbrush, and the ink solutions can be stored for 28 days in a refrigerator without developing any foul smells.

Keywords: organic ink, natural pigments, light fastness, painting, writing.

1. Introduction

The preparation of natural inks is an ancient practice that has been part of human civilization for thousands of years. China and Ancient Egypt were among the first known users of natural ink, dating back to 2500 BC.¹⁻² The use of natural colors has deep historical roots and still continues in some parts of India, though on a smaller scale today.³⁻⁴ However, this tradition faces challenges due to the widespread availability of synthetic dyes, which are more durable and cost-effective. While not all natural dyes are completely safe, they are generally less toxic than synthetic ones.

Organic inks made from natural sources have become popular because they are eco-friendly and sustainable. Natural dyes are biodegradable and do not cause disposal problems. These inks are visually appealing and free from harmful chemicals often found in synthetic inks.⁵ The preparation of organic inks usually involves only mild reactions. Different colors can be obtained by using various fixatives during the dyeing process. However, making stable and

*Corresponding Author Email: rupa@biswanathcollege.in

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durable natural inks has been difficult because they tend to degrade and break down under UV light.⁶⁻⁷

Plants such as red beetroot, cabbage, onion, and turmeric are known to be used in natural ink production due to the presence of vibrant natural pigments, which are non-toxic and edible. These natural inks have versatile applications in textiles, tissue engineering, food packaging, etc.⁸⁻¹² However, preparation of stable and durable organic inks for writing, painting and printing purposes from such natural sources has always been challenging due to their biodegradation and decomposition under heat and UV light.

The present study aims at preparation of a durable and eco-friendly organic ink from beetroot (*Beta vulgaris* L.) and turmeric (*Curcuma longa* L.), as they are naturally abundant, cheap, and easily available. The red beetroot (*Beta vulgaris* L.) is a valuable source of red and yellow pigments known as betacyanins (red) and betaxanthins (yellow), respectively. The major betacyanin is betanin and accounts for 75–95% of the red pigment.¹³ It is a water-soluble pigment and is known for its potential antioxidant and anti-inflammatory properties. On the other hand, curcuminoids are important natural pigments that are responsible for the bright yellow colour of turmeric. They are composed of three components, viz., curcumin [1, 7-bis(4-hydroxy-3-methoxyphenyl)-1, 6-heptadiene-3, 5-dione], demethoxycurcumin [1-(4-hydroxy-3-methoxyphenyl)-7-(4-hydroxyphenyl)-1, 6-heptadiene-3, 5-dione], and bisdemethoxycurcumin [1, 7-bis-(4-hydroxyphenyl)-1, 6-heptadiene-3, 5-dione].¹⁴ The major component responsible for its yellow colour is curcumin. It is a fat-soluble pigment but insoluble in aqueous medium. Betanin possesses maximum stability at pH ~4-5, while curcumin at pH < 7.¹⁵⁻¹⁶

Chemically, inks are a mixture and are composed of colourant, a vehicle, and additives. The colourant is either dyes or pigments in a vehicle, either water or an organic solvent containing additional ingredients, including pH modifiers; humectants to retard premature drying; polymeric resin to impart binding and allied properties; defoamers to regulate foam efficiency; wetting agents such as surfactants to control thickness and rheology; modifiers to control ink application; and biocides and bacteriostatic agents to preserve ink from bacterial and fungal growth and impart foul smells.¹⁷ Therefore, in the present work, some naturally abundant and easily accessible organic ingredients are chosen as pH regulators, binders, lubricants, and antioxidants to meet requirements for obtaining an effective and desirable ink.

Different natural plant species require various extraction and purification processes to produce a pigment. There have been very simple-to-complex methods used for obtaining the colour pigment from the plants, such as solvent extraction, boiling, pressing, crushing, microwave extraction, molecular distillation, enzyme reaction, supercritical extraction, etc.¹⁸ As this work is just focused on obtaining the colour pigment-containing liquid from the plants, a traditional method, the grinding of the plant's specific part with a mortar and pestle and finally sieving the solution to obtain the colour solution, is employed. The trial-and-error method is applied to obtain the best-formulated inks. Sensible methods, the use of normal ink pens, brushes, and cotton sticks, which are readily available, are used for the application of the ink on paper.

Although the natural pigments present in beetroot and turmeric have been widely explored as colouring agents in traditional medicines, textiles, and the food industries, the deficiency of colour stability limited their applications. A few works have been reported on the preparation of organic inks from these natural pigments for day-to-day writing, printing, and painting purposes.¹⁹ Various ink compositions and complex methods of preparation and standardisation have also been explored by many researchers to get improved colour stability. However, organic inks using these natural pigments with remarkable colour stability have not been achieved yet. The creation of inks with promising colour stability from these natural pigments in combination with other environmentally friendly materials using a simplified method will provide a sustainable shift in reducing adverse environmental impact.

2. Experimental Section

2.1. Materials

Beetroot (*Beta vulgaris* L.), turmeric (*Curcuma longa* L.), and lemon fruits were purchased from a local vegetable market (Assam, India). Drumstick resin was collected freshly from the drumstick tree (*Moringa oleifera*). Extra virgin olive oil was purchased from GAIA, available in the market, and ethanol was purchased from Sigma-Aldrich. Distilled water was used throughout the study.

2.2. Extraction of Beetroot, Turmeric, and Lemon Juice

About 250g of each of raw turmeric and beetroot was washed thoroughly to remove the dirt, peeled off, and cut into small pieces. Then it was pounded in a large-sized mortar and pestle to obtain the paste. The resulting pastes were filtered and squeezed using clean cheesecloth to separate the pigments from the pulp. Similarly, about 50 ml of lemon juice was extracted from green lemon fruit by simply squeezing the fruit, followed by filtration using cheesecloth.

2.3. Preparation of Drumstick Resin Solution

The solution of drumstick resin was prepared by dispersing 2 g of the dry resin in 50 ml of distilled water and boiling it till the solution became lightly sticky. After cooling, 1 ml of ethanol was added to the solution.

2.4. Preparation of Ink Formulations using Beetroot and Turmeric Pigments

Four different ink formulations for each pigment were prepared by mixing with different proportions of extra virgin olive oil, drumstick resin, and lemon juice using water, ethanol or both as a solvent. The compositions of each formulation are as given in the following table (Table 1).

Table 1. Compositions of various ink formulations using beetroot and turmeric.

Pigments	Ink Formulations ^a	Ink Composition
	1	80% extracted pigment (v/v) + 10% ethanol (v/v)+ 10% lemon juice (v/v) + 1 drop drumstick resin + 1 drop extra virgin olive

Beetroot		oil
	2	75% extracted pigment (v/v) + 10% ethanol (v/v) +10% water (v/v)+ + 5% lemon juice (v/v) + 2 drop drumstick resin + 2 drop extra virgin olive oil
	3	65% extracted pigment (v/v) + 15% ethanol (v/v) +15% water (v/v) + 5% lemon juice (v/v) + 3 drop drumstick resin + 3 drop extra virgin olive oil
	4	55% extracted pigment (v/v) + 20% ethanol (v/v) + 20% water (v/v) + 5% lemon juice (v/v) + 4 drop drumstick resin + 4 drop extra virgin olive oil
Turmeric	1	80% extracted pigment (v/v) + 10% ethanol (v/v)+ 10% lemon juice (v/v) + 1 drop drumstick resin + 1 drop extra virgin olive oil
	2	75% extracted pigment (v/v) + 20% ethanol (v/v) + 5% lemon juice (v/v) + 2 drop drumstick resin + 2 drop extra virgin olive oil
	3	65% extracted pigment (v/v) + 30% ethanol (v/v) + 5% lemon juice (v/v) + 3 drop drumstick resin + 3 drop extra virgin olive oil
	4	55% extracted pigment (v/v) + 40% ethanol (v/v) + 5% lemon juice (v/v) + 4 drop drumstick resin + 4 drop extra virgin olive oil
^a The pH of ink formulations were 4-5, as beetroot and curcumin pigments possesses maximum stability in this range.		

In the above formulations, lemon juice is used to maintain the acidic conditions of the ink formulations, which can impart stability to the respective pigments. Since curcumin is insoluble in an aqueous medium, only ethanol (as turmeric is most soluble in ethanol) is used as a solvent for its ink formulation. Fresh drumstick resin is added in a small amount for consistency and as a binder, and extra virgin oil is used as an antioxidant. The resulting ink formulations were then water-bathed to form an emulsion with the oil.²⁰

2.5. Methods Employed to Study the Stability of the Ink Formulations

The stability of the resulting ink formulations for painting and writing purposes is studied in terms of light fastness, colour consistency, odour, and drying time. In order to study the light fastness property, paper strips were prepared for all the ink formulations. The resulting paper strips were allowed to stand for 28 days indoors. The ink solutions of each formulation were divided equally into two halves. The first half of each formulation was kept at room temperature in the dark for 28 days under regular observation, while the second half was kept in a refrigerator for the same period of time. The colour stability of the resulting formulations

was also monitored visually on the 28th day. The drying time was observed in minutes and seconds using a stopwatch just after writing or painting on paper. The assessment of odour and colour consistency of the resulting formulations was performed through visual observations.

3. Result and Discussion

The study of the lightfastness property of all ink formulations has shown that the dark pink colour of beetroot pigments (**Figure 1**) and the intense yellow colour of pigments in the ink strips (**Figure 2**) gradually change their vibrant colour over time, except for formulation 3 of beetroot and formulation 4 of turmeric. All other ink formulations of beetroot and turmeric turned into a light brown colour, while the two best formulations had almost no colour change over the days.



Figure 1. Ink strips of the four beetroot formulations on a) day 1, b) day 7, c) day 14, and d) day 28.

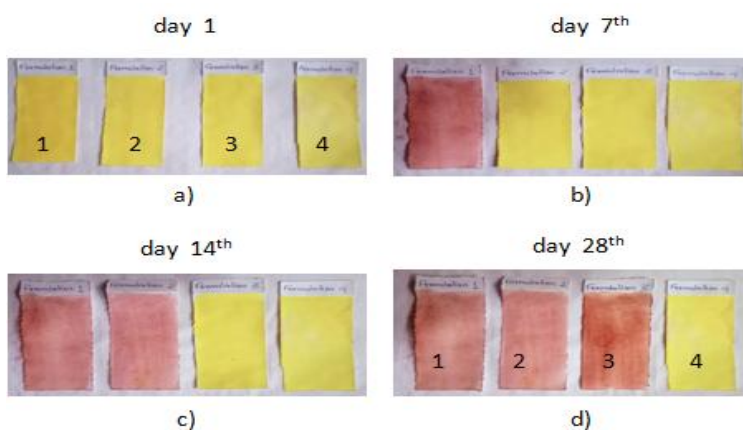


Figure 2. Ink strips of the four turmeric formulations on a) day 1, b) day 7th, c) day 14th, and d) day 28th.

The pigments present in both beetroot and curcumin undergo degradation in the presence of light and excess oxygen.²¹⁻²² However, the presence of a calculated amount of antioxidants, preservatives, and solvents and a balanced pH can slow down the degradation process. In this study, the best formulation was achieved from both pigments (i.e., Formulation-3 of beetroot and Formulation-4 of curcumin) by adding higher amounts of extra virgin olive oil, resin, and

ethanol as a solvent compared to the other formulations. It is known that extra virgin olive oil possesses antioxidant and lubricating properties, and ethanol itself works as a preservative. Since all the ink formulations consist of equal volumes of lemon juice as a pH modifier, the better light fastness property of formulation 3 of beetroot and formulation 4 of curcumin compared to other formulations may be ascribed to the presence of a higher quantity of antioxidants and preservatives. Thus, the fast colour changes observed in the case of other ink formulations containing a lower quantity of antioxidants and preservatives are reasonable. Though beetroot contains a lower amount of antioxidant in Formulation-3 compared to Formulation-4, the former has shown more persistent colour during the course of the study, which may be due to the presence of a higher percentage of water as a solvent than in Formulation-4, which initiates easier degradation.

The various ink formulations have also shown different drying times when applied for writing and painting. They also exhibit different odour stabilities as the days pass by. The variable drying times taken by the ink formulations after being applied for writing and painting are shown in the following diagram (**Figure 3**).

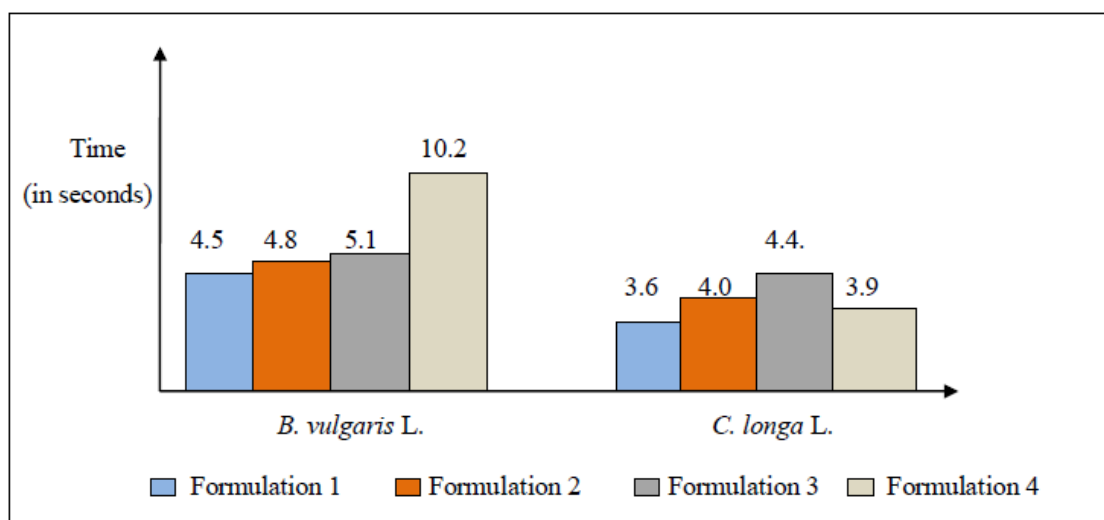


Figure 3. Drying time of the various ink formulations.

It was generally observed that the drying times of the ink formulations were directly proportional to the amount of solvent used. Inks dry primarily by the process of adsorption of the ink onto the surface of a paper, followed by evaporation of the solvent from the ink film. 21 inks are composed of solids dispersed in a liquid. Due to vapour pressure, the liquid evaporates, and the solid remains on the paper. Therefore, the more liquid ink there is, the longer it will take to dry. The drying time is completely reasonable for beetroot formulations, as the volume of solvents was gradually increased from Formulation-1 to Formulation-4; the drying times of the ink formulations were also gradually increased, as shown in the above graph (**Figure 3**). The ink formulations of curcumin had a smaller drying time as compared to the ink formulations of beetroot because curcumin inks were formulated in ethanol, which evaporates more quickly and took comparatively less time to dry.

As the days passed, the solutions of all ink formulations of both pigments, except Formulation 3 of beetroot and Formulation 4 of curcumin, developed some odour over the

week and precipitated when stored at room temperature. A moderate odour was detected for the formulations of beetroot, and a strong odour was found in the case of the curcumin formulations during the course of studies. However, the ink strips were found to maintain a low odour throughout the study. The beetroot formulation 3 has maintained low odour throughout the study, while in the case of formulation 4 of curcumin, the odour was low from the 1st to the 14th day and moderate on the 28th day at room temperature. When these two formulations were stored in a refrigerator, a little odour was detected till the 28th day. In summary, Formulation 3 of beetroot and Formulation 4 of curcumin were found to be the best among the other formulations because these two formulations have shown excellent colour stability, low odour, and lower drying time. Therefore, the two formulations can be used for writing and painting (**Figure 4**) using an ink pen and paintbrush effectively, while desired results were not achieved using other formulations due to low colour stability.



Figure 4. Painting with both beetroot and curcumin ink using a paintbrush.

4. Conclusion

In the present study, two organic ink formulations were successfully prepared by using natural pigments present in the two root vegetables, viz., beetroot and turmeric, by the trial-and-error method. Some other naturally abundant and easily accessible organic ingredients were added to the inks as pH regulators, binders, lubricants, and antioxidants. A simple and sustainable approach was proposed for the preparation of all the ink formulations. The real-time applicability of the formulated inks was evaluated in terms of lightfastness, colour consistency, odour, and drying time. Studies have shown that the two best formulations of both pigments in the ink strips have displayed very low drying time with good colour consistency, retained excellent colour quality, and maintained low odour till the 28th day at room temperature. Storage of both ink solutions was also possible up to the 28th day under refrigeration. It was also observed that the quantity of binder, the antioxidant, and the nature and amount of solvent used play a crucial role in the colour stability of the ink. Generally, the presence of a higher amount of antioxidant, binder and 30% solvent (15% ethanol and 15% water) provided better colour stability to the beetroot ink. Similarly, a higher amount of antioxidant and binder and 40% of ethanol gave rise to the most stable turmeric ink. Hence, this study demonstrates the feasibility of sustainable organic ink for writing and painting purposes using natural additives in the pigments of beetroot and turmeric through a simple method of preparation.

Author Information

Corresponding Author

Dr Rupa Pegu, Department of Chemistry, Biswanath College, Biswanath, Assam-784176, India; E-mail: rupa@biswanathcollege.in

Notes

The authors declare no conflicts of interest to disclose.

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