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NATURAL INK SYNTHESIS FROM FLOWERS OF FLAME OF FOREST (*Butea monosperma*), COMMON LANTANA (*Lantana camara*), AND BERRIES OF MALABAR SPINACH (*Basella alba*)

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Abstract

The art of writing and printing requires ink which can be of natural or synthetic origin. Synthetic inks owing to their merits have an edge over natural inks. However, Volatile Organic Compounds and the non-degradable nature of synthetic ink pose a threat to the individual engaged with it and the environment. Thus to overcome these issues this study was carried out with the objective to synthesize natural inks from the flowers of Flame of forest (Butea monosperma, Papilonaceae*), Common lantana (Lantana camara,* Verbenaceae*), and berries of Malabar spinach (Basella alba,* Basellaceae*). The natural ink was synthesized by using water as a solvent, with vinegar as an additive. The synthesized natural inks' physical properties revealed free-flowing, non-clogging nature and drying time in the range of 5-7 sec. to 2-4 sec. for cardboard and paper respectively. The exposure of these inks to solar radiation for two weeks revealed no colour change in Flame of forest and, Common lantana however faint colour change in Malabar spinach. No ink residue falling off on the sticky transparent tape indicates good adhesion by these inks and no ink residue falling off at all either after various rubbing times indicates the inks does not worn off. The results obtained in this study indicate that the flowers and berries of these plants have the properties of the ink and the ink synthesized from them can be used for commercial purposes. The synthesized inks are eco-friendly, devoid of chemicals, and degradable. The inks synthesized from the Flame of forest, and Common lantana perhaps may be reported for the first time.*

Key Words: *Ink, Natural ink, Flame of forest, Common lantana, Malabar spinach*

Introduction

The art of writing developed in growing civilization with reformation. It is a skill by which a person expresses his thoughts, idea, feelings, and emotions. The writing ability makes the human being a pioneer of the ecosystem and sets him apart from this world among the animal kingdom. Writing is an advanced mode to make the imagination come true that one would arise, preserved, secure, and stable documentation. To achieve this

target magnetic, electric, and electronic inks are arising which will provide certainly a step toward a conservative society Sharma *et al*., 2014).

The art of using natural colour is very old and in China dated 2600 BC which was the first country to use natural dyes (Singh and Sharma, 2017). The history of writing ink is more than five thousand years old in India. In those days carbon inks were in use for writing on palm leaves before the invention of papers (Daumas, 1969). Natural ink is still used in some parts of India, however on a small scale, due to the easy availability of synthetic dyes, long-lasting colour, and low cost. Organic ink is a natural product prepared from a natural colourant. It is very useful for printing and writing material and writing work. Organic inks are considered eco-friendly because they are derived from natural resources like plant leaves, roots, flowers, fruits, and minerals sources. There are more than 500 dye-yielding plants in nature giving different shades of colour. Natural ink overcomes a few of the problems lead by synthetic ink via a less expensive and not tedious process, non-toxic and safe. The natural pigments, although they have been used for centuries, have been replaced by synthetic dyes from the oil industry at the end of the 19th century. Indeed, they have the advantages of reproducibility of the properties, unlike natural pigments. However, they do have mired adverse effects on the environment and workers engage with them (Singh and Sharma, 2017).

The printing ink consumption by volume is expected to increase by 9% from 3.3 million tonnes in 2013 to 3.6 million tonnes in 2023 (Future of Global Ink Markets, 2023). In the case of India, it was estimated at 0.36 million tons for 2019-2020 (Printing Ink). The colour changes, press cleaning, and poor ink management practices generate waste ink. The spent inks have the potential to cause serious water pollution to drinking water supplies if illegally disposed of in a septic system or can if discharged to surface water, reduce the dissolved oxygen resulting in mortality of fish and other aquatic life (Davis, 2001). The hazardous health effects of synthetic ink on human beings include headaches, skin irritation and nervous system, etc. These harmful effects can be caused by solvents, and pigment/dye which help to create the colour of ink (Noah *et al*., 2014). According to the American Lungs Association breathing Volatile Organic Compounds (VOCs) can irritate the eyes, nose, and throat, can cause difficulty in breathing, damage the central nervous system as well as other organs, and nausea whereas some VOCs can cause cancer. In 1996 Germany was the first country which bans azo dye (synthetic dye) from the printing industry which causes harmful effects on the environment with emission of the VOCs (Ghosh and Malik, 2019). Synthetic inks owing to their disadvantages cause a great boost to the natural inks research work. Literature review revealed the paucity of studies on the natural inks synthesized from the flowers of Flame of forest, Common lantana, and berries of Malabar spinach. Thus, this is the identified gap in the subject domain. To fill this knowledge gap this study was carried out with the objective of synthesis of natural ink from these plant materials and testing them for discolouration, best penetration time, and ink residue falling off.

Materials and Method

For the synthesis of natural ink in this study three plants viz. Flame of forest (*Butea monosperma*, Papilonaceae), Common lantana (*Lantana camara*, Verbenaceae), and Malabar spinach (*Basella alba*, Basellaceae) were selected. These plants are native to the region and are easily available. The collected plants were identified by a taxonomist and the specimen of these plants is preserved in the Department of Environmental Science, Sardar Patel College, Chandrapur, India. The flowers and berries of these plants were collected and shade dried and used for the synthesis of ink. The solvent extraction methodology was adopted for the synthesis of the inks from these plant products. The synthesized inks were tested for discolouration by exposure to solar radiation, drying time for paper and cardboard, adhesion test by using standard sticky transparent tape number 600 (ASTM D3359) and worn off test by using a rubbing tester (ASTM D5264-98).

Ink from Flame of forest (Butea monosperma,* Papilonaceae*)

The dried flowers of Flame of forest (200 g) were taken in a beaker and 100 mL of double distilled water was added and boiled at 70ºC for 15-20 minutes on a flame. After boiling the solution was cooled for 5 minutes. This solution was again boiled at 70ºC for 10 to 15 minutes and after cooling 2-3 drops of vinegar was added. The synthesized ink was filtered with the help of Whatman filter paper (No. 1).

Ink from Common lantana (Lantana camara,* Verbenaceae*)

The dried flower petals of Common lantana (200 g) were taken in a beaker and 100 mL of double distilled water was added and boiled at 70ºC for 15-20 minutes on a flame. After boiling the solution was cooled for 5 minutes. This solution was again boiled at 70ºC for 5 to 10 minutes and 2-3 drops of vinegar was added. This synthesized ink was flittered with the help of Whatman filter paper (No. 1).

Ink from Malabar spinach (Basella alba,* Basellaceae*)

The wet Malabar spinach berries (200 g) were crushed with the help of a mortar and pestle. The half quantity (100 g) of these crushed berries was taken in a conical flask and 100 mL of ethyl alcohol (95%) was added and this solution was boiled at 70ºC for 10-12 minutes in a water bath. After boiling, this solution was cooled for 5-10 minutes and again boiled at 70ºC for 8-10 minutes and 2-3 drops of vinegar was added. This synthesized ink was filtered with the help of Whatman filter paper (No. 1).

Results and Discussion

The physical properties of the natural inks synthesized from the Flame of forest, Common lantana, and Malabar spinach are presented in Table 1. The synthesized inks were free-flowing, non-clogging, and bright. The permanency of colour and stability was stable. The ink colour was yellow, green, and maroon for Flame of forest, Common lantana, and Malabar spinach respectively. The drying time on paper and cardboard was in the order of Flame of forest > Common lantana > Malabar spinach. The drying time of all the synthesized inks was comparable with synthetic ink. Common lantana and Flame of forest were never used previously for ink preparation and the prepared inks have the prerequisite characteristics of the ink. The drying time of the ink was noted to be 2-4 and 5-7 seconds for paper and cardboard respectively which is within the range of

standard synthetic ink. The synthesized inks do not erase easily. This may be because the flowers and berries may have the properties of ink. The physical properties of the synthesized ink indicate that the inks have good quality.

Table 1: Physical properties of natural inks synthesized from plant origin

Property	Flame of forest	Common lantana	Malabar spinach
Flow-ability	Free-flowing	Free-flowing	Free-flowing
Clogging nature	Non-clogging	Non-clogging	Non-clogging
Colour	Yellow	Greenish	Maroon
Brightness	Dark	Dark	Dark
Permanency of colour	Stable	Stable	Stable
Stability	Stable	Stable	Stable
Drying time (on paper)	3-4 seconds	2-3 seconds	2-3 seconds
Drying time (on cardboard)	6-7 seconds	5-6 seconds	5-6 seconds

The comparison between synthetic ink and synthesized natural inks was attempted (Table 2) for various components. In synthetic ink preparation, the solvent used includes toluene, acetone, and methanol; whereas, in the case of synthesized natural inks water was used. The pigment used in natural ink synthesis is of plant origin; whereas, titanium dioxide, and calcium carbonate are in synthetic ink. The additive added in synthetic ink was dibutyl phthalate monoethanolamine; whereas, vinegar in a synthesized in natural ink. From thetable, it can be arrived at synthesized natural inks contain fewer chemicals as compared to synthetic ink.

Table 2: Comparison between synthetic ink and synthesized natural inks

Component	Synthetic ink	Synthesized natural inks from		
		Flame of forest	Common lantana	Malabar spinach
Solvent	Toluene, acetone, methanol	Water	Water	Alcohol
Pigment	Titanium dioxide, calcium carbonate	Pigment from flowers	Pigment from flowers	Pigment from berries
Additive	Dibutyl phthalate monoethanolamine	Vinegar	Vinegar	Vinegar

The natural inks synthesized from these plant products from the aforementioned procedure were filled in an ink pen and tested for discolouration after writing on paper. After writing, these written papers were exposed to solar radiation for two weeks in February and March to test for discolouration. The climatic conditions during the exposure period were Temperature: February 20.8^{o}C min. temp., 31.5^{o}C max. temp., March 26.0^{o}C min. temp., 35.5^{o}C max. temp., Cloud amount: February 1.5 min., 1.9 max., March 1.7 min., 2.3 max., Humidity: February 27-57%, March 21- 44%, Wind: February 11 m/s min., 15 m/s max., March 9 m/s min., 14 m/s max.). The exposure time of these papers was from 9.00 am to 5.00 pm daily for two weeks. The results obtained for discolouration after exposure to the solar radiation for 1st and 2nd week are presented in Tables 3 and 4 respectively. From Table

3, it was observed that the first week of exposure of pages to the solar radiation resulted in no colour change. In the second week of exposure of written pages to the solar radiation (Table 4) revealed no colour change in Flame of forest, and Common lantana; however, a slight colour change in Malabar spinach. The synthesized natural inks gave the good colour strength of yellow, greenish, and maroon for Flame of forest, Common lantana, and Malabar spinach respectively after writing on a paper.

Table 3: Synthesized natural ink exposed to the solar radiation (First week)

Exposure to solar radiation	Flame of forest	Common lantana	Malabar spinach
1st day of exposure	No colour change	No colour change	No colour change
2nd day of exposure	No colour change	No colour change	No colour change
3rd day of exposure	No colour change	No colour change	No colour change
4th day of exposure	No colour change	No colour change	No colour change
5th day of exposure	No colour change	No colour change	No colour change
6th day of exposure	No colour change	No colour change	No colour change
7th day of exposure	No colour change	No colour change	No colour change

Table 4: Synthesized natural ink exposed to the solar radiation (Second week)

Exposure to solar radiation	Flame of forest	Common lantana	Malabar spinach
8th day of exposure	No colour change	No colour change	Slight colour change
9th day of exposure	No colour change	No colour change	Slight colour change
10th day of exposure	No colour change	No colour change	Slight colour change
11th day of exposure	No colour change	No colour change	Slight colour change
12th day of exposure	No colour change	No colour change	Slight colour change
13thday of exposure	No colour change	No colour change	Slight colour change
14th day of exposure	No colour change	No colour change	Slight colour change

It was observed that 40% ink concentration in all the inks prepared in this study has optimal with best penetration time of 2-4 seconds and 5-7 seconds for paper and cardboard respectively. These values compare favourably with the penetration time of 1.50±0.42 sec and 5.70±0.71 sec for the commercial ink on writing paper and cardboard respectively. As per the ASTM D3359 standard sticky transparent tape number 600 revealed no ink residue falling off on the tape at all. Moreover, as per ASTM D5264-98 standard, rubbing times were 50, and 100 revealed there was no ink residue falling off at all either.

Singh and Sharma (2017) prepared a natural ink from black current (*Syzygium cumini*). It gave good colour strength of purple colour when printed on a paper, eco-friendly and easy to decompose. These results corroborate the findings of the study. Basri *et al*., (2021) reported low amount of glycerol used was found to increase the value of colour lightness of mangosteen (*Garcinia mangostana*) leaves-based marker ink. A decrease in carboxymethyl cellulose amount resulted in the low viscosity of marker ink. The additive vinegar addition in natural ink synthesized in this study preserves the ink, and enhances its stability and permanence on paper once it has dried.

Brown ink was synthesized from fruit extract of the candahar tree (*Gmelina arborea*). The 40% concentration level of the prepared ink has the best penetration time of 5.29±0.43 sec and 1.55±0.46 sec for cardboard and writing paper respectively (Noah *et al*., 2014). These results are in agreement with the results reported in this study. The extract of the flowers of the butterfly pea (*Clitoria ternatea*) can serve as a natural blue colourant, tend to be convenient to use and possesses a longer shelf life than comparable plant-based colourants (Siti Azima *et al*., 2017).

Printing ink prepared from corn flour and coffee powder showed that the ink owned exceptional adhesion properties well resistance to rubbings, and the delta-E value variation was less than 1.0 which indicated that the colour change was not visible to naked eyes. There was no ink residue falling off on the tape and no ink residue falling off at all either after using a rubbing tester (Chotithammaporn, 2017). These results corroborate with the findings of the study. Nwafulugo *et al*., (2019) produced a marker ink from the berry seed extract of huckleberry (*Gaylussacia baccata*). The ink was of high quality with alkaline pH (9.3), drying time of 2.3 seconds and viscosity of 9.5x 10^{-4} N s/m^2. These findings corroborate the results reported in this study.

Conclusion

In this study, natural inks of yellow, green, and maroon colour were successfully synthesized by solvent extraction method from the flowers of Flame of forest, Common lantana, and berries of Malabar spinach respectively. The yield of synthesized inks was satisfactory. From the study conducted, it was discovered that these plant materials have the prerequisite properties which can be used for the synthesis of natural inks. The physical properties of the synthesized inks are in agreement with synthetic ink. The inks produced in this study were found to be very effective on writing a paper. These inks have a higher penetration time for cardboard as compared with paper. The ink's colour does discolour even after exposure to solar radiation for a sufficient period of time indicates the discolouration was unable to notice with necked eyes. The rubbing test revealed no ink residue falling off at all which indicates these inks were resistant to rubbings and does not worn off. As pointed out by the test results, there was no part of the inks falling off on the transparent tape indicates the inks have exceptionally adhered to the printing material and has good adhesion properties.

The use of water as a solvent and vinegar as an additive makes these inks environment-friendly and bio-degradable. As a result of these characteristics, the limitation of the synthetic ink regarding the presence of VOCs, solvents, pigments, and dyes is overcome which will have comparatively minimum impact on the workers engaged in the printing industry and the environment in the larger context. These inks can be stored properly in cool and dark places to use for prolonged period. Furthermore, these inks can be a suitable alternative to synthetic ink, and further studies are required for their standardization. The inks prepared in this study perhaps may be feasible for printing on paper and may be used in printers with suitable modifications. The commercial application of these synthesized inks needs to be explored. Different variations of ink shades can be produced by combining these inks with different

proportions. Future investigation needs to be attempted on using these inks in the printer, marker pens, and other such devices which are used in printing and writing thus reducing the pressure on synthetic ink production. Furthermore, other plants/natural-based products should be identified for future ink preparation so to pave the way for sustainable printing industry and a sustainable environment.

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