Websites:
http://www.sciencepub.net/nature
http://www.sciencepub.net

Emails:
naturesciencej@gmail.com
editor@sciencepub.net

Nature and Science

MARSLAND PRESS
Multidisciplinary Academic Journal Publisher

Investigation on some Physical Properties of Dry Erase Ink Formulation Produced from *Azardirachta indica* and *Carica papaya*.

[1]Mbong, E. O., [2]George, U. U., [1]Ekott, E. J., [1]Okon, H. E.

[1] Department of Environmental Biology, Heritage Polytechnic, Ikot Udoata, Eket, Akwa Ibom State
[2]Department of Fisheries & Aquaculture, Akwa Ibom State University, Obio Akpa Campus, Akwa Ibom State
Email: ubonggeorge@aksu.edu.ng

Abstract: Inks are colored fluid suspensions formulated to reproduce colorful images on sorbent surfaces. The materials used in ink making may present their undesirable unique environmental hazards and challenges. Pigments and dyes used for commercial ink brands are expensive and sometimes contain toxic substances which if inadvertently ingested by humans, especially children, may be harmful to health. Also, conventional inks are less environmentally friendly hence the need for organic and eco-friendly substitutes. This study investigates the suitability of different plant-based formulations produced from *Azardirachta indica* and *Carica papaya* as substitutes for dry erase ink. The formulations were evaluated for colour, density, pH, drying time, colour stability and viscosity. Inks formed from *A. indica* were more stable than *C. papaya*. *A. indica inks* recorded higher density (1.05-0.84 g/cm^3) than inks made from *C. papaya* (1.02-0.87 g/cm^3). Drying time ranged between 8 – 14 seconds in *C. papaya* and 6 – 12 seconds for *A. indica.* pH values were higher in *C. papaya* (6.8-7.5) but lower in *A. indica* (5.2-5.6). The viscosity was higher in *C. papaya* (8.55 -13.37$kg/m^{-1}s^{-1}$) but lower in *A. indica* (7.66 -12.87-$kg/m^{-1}s^{-1}$). Correlation and regression analysis reflected a common trend indicating that an increase in amount of plant extract led to a significant ($P<0.05$) decrease in drying time, water proof nature, density and viscosity but on the other hand it increased the pH of the formulation. Conclusively, the physical properties of these plant formulations compare favourably with standard commercial ink brands.

[Mbong, EO., George, UU., Ekott, EJ., Okon, HE. **Investigation on some Physical Properties of Dry Erase Ink Formulation Produced from *Azardirachta indica* and *Carica papaya*.** *Nat Sci* 2021;19(5):26-31].ISSN 1545-0740 (print); ISSN 2375-7167 (online). http://www.sciencepub.net/nature 3.doi:10.7537/marsnsj190521.03.

Keywords: Plant extracts, Ink, Viscosity, Alcohol, *A. indica*, *C. papaya*.

1. Introduction

Ink is a liquid or paste used to colour a surface to produce an image, text, or design. It is used for the purpose of drawing or writing with a pen, brush, or quill. Inks in paste form are used mostly in letterpress and lithographic printing. It is a complex medium composed of solvents, pigments, dyes, resins, lubricants, solubilizers, surfactants, particulate matter, fluorescents, and other materials (Nwafulugo, *et. al.* 2019). The components of inks affect its flow, thickness and appearance when dry. The components of synthetic ink which is commonly used present unique environmental hazards especially when accidentally ingested by children or toddlers (Powar, *et. al.* 2014). Ink making is a great scientific and industrial process requiring the right technology and the application of a variety of scientific disciplines and technical skills. Conventional inks are known to be linked with health and environmental problems. However, plant-based inks are seen as alternatives that are less damaging compared to the synthetic ones (Powar, *et. al*. 2014). Plant-based inks help to conserve non-renewable reserves and reducing volatile organic compounds (VOCs) emissions, while maintaining important desired ink qualities (Dagde. *et. al*. 2019). They are eco-friendly because they are obtained from plant-based derivatives. Also, in modern class and conference rooms, white boards are used. Although they may be computer boards, presenters are sometimes prompt to write on them which thereafter require cleaning. Most commercial inks do not give perfect cleaning, prompting the use of solvents, like alcohol as cleaning agents, for effective board cleaning.

Available literature reveals that ink formulations have been derived from *Citrus limonene*, *Pentas lanceolata*, beetroot and turmeric (Powar, *et. al;* 2014). Other plant species with pharmacological activities, such as *Beta vulgaris* (Beet Root) family Chenopodiaceae, *Citrus limonene* (Citrus peel) family, Rutaceae, *Amelanchier arborea* (Butterfly Flower petals), *Bauhinia purpurea* (Butterfly tree) family and Caesalpiniaceae had also been used in the preparation of herbal ink. Aside from using natural ink formulations for lithography and calligraphy, natural inks have also been adopted as confectioneries and

food additives (dyes). Such additives have been produced from saffron, Beet Root, Annatto, Elderberry, Pandan, Butterfly pea and turmeric). Plant dyes are quite useful in textile, cosmetics, leather, food and pharmaceutical industries (Kumar, *et. al.* 2012; Powar, *et. al;* 2014).
Commonly used solvents in ink formulation are xylene, toluene, mineral oil, alcohols, esters, ketones, and water. Most inks dry by heat because the solvents evaporate from the ink. Common ink solvents used for making inks are VOCs which contribute to majority of the 101,537 tons of VOCs emitted every year by ink manufacturing and printing industries (EPA, 1995). VOCs contribute to atmospheric photochemical reactions. The economic realities associated with the production, distribution and utilization of synthetic ink and its attendant environmental cost mandates the need to identify suitable and cost-effective replacements for synthetic inks using locally sourced raw materials.

2.0 Materials and Methods

2.1 Plant Collection

Plant leaves were collected from fully matured plants of *Azadirachta indica* and *Carica papaya* from the botanical garden of Heritage Polytechnic, Eket, Nigeria (Latitude $04^{0}39.30^{'}028''$ N; Longitude $07^{0}58.29'671''$ E).

2.2 Maceration and Concentration of Extract.

The fresh leaves were chopped, blended using a blending machine and oven dried at 60°C for 2 hours. They were then pulverized using a blender to obtain powder. Extraction of the plant material was then carried out using batch process according to the modified method of (Morshed, *et. al.* 2016). 100g each of the powdered plants material were placed in a 500ml conical flask; 400ml of 98% ethanol was added to enhance extraction. It was allowed to stay for 48 hours after which the solution was double filtered to remove residues. The extract solution from both plants were concentrated by taking 300ml of each solution in a 500ml beaker and placing the beakers on a water bath at 60°C until pasty concentrates of both extracts were obtained.

2.3 Formulation and Determination of Ink Properties

The inks formulations were prepared by blending the concentrated plant extracts paste with specific amounts of solvents. As seen in Table 1, three different formulations, labeled A, B and C, were made for varying amounts of each of the plant extracts.

Table 1: Formulation table

Ink Samples	Extract (ml)	Glycerol (ml)	Ethanol (ml)	Cetyl Alcohol (ml)
A	30	5.0	5.0	5.0
B	40	5.0	5.0	5.0
C	50	5.0	5.0	5.0

2.4 Density

A Pycnometer was used to conduct density test of the different samples. The empty pycnometer was weighed initially in order to ascertain its weight. Thereafter, the ink formulations were weighed accordingly to determine the weight of the product by subtracting the total weight of the product from the weight of the empty pycnometer. The true density of the different ink formulations was obtained consecutively using the formula below.

2.5 Viscosity

Physical properties of the formulated inks were accessed to determine their suitability for use. Viscosity, drying time, pH, water proof and sunlight radiation properties were determined. Viscosity was determined according to the method of Etukudo (2019). Ostwald viscometer and a stop watch were used to determine average flow time of water and average flow time of sample. The density of ink was calculated using Equation 1:

$$l_s = \frac{weight\ of\ water}{weight\ of\ ink}$$

The viscosity of the ink n_s was calculated using equation 2.

$$\frac{n_s}{n_w} = \frac{l_s\ x\ t_s}{l_w\ x\ t_w} \qquad [2]$$

Where n_s = viscosity of sample, n_w = viscosity of water, l_s = density of sample, l_w = density of water, t_s = average flow time for sample and t_w = average flow time for water.

2.6 Drying Time

After application to a writing surface, the solvent should vaporize quickly leaving the colorant and the binder so that the ink will attach to the surface but not absorbed by it. The time taken for the vaporization of the solvent is the drying time of the ink. To determine this, the inks were used for stamping on white paper and for writing on a clean white board. The drying time was measured using a stopwatch, then later erased to determine their ability to be erased. The inks were easily and sufficiently erased with a polyethene board duster. The ink was erased from the board without the application of other cleaning agents like methylated spirit or ethanol.

2.7 pH

The pH values of the ink samples were determined using a PHS-25 pH meter calibrated with acetate buffer of standard pH of 7.0.

2.8 Waterproof Test

To determine the water proof of the inks, samples of the formulated inks were written on a white paper and the paper was left out in open air for 24 hours. The paper was then held under running water for 30 seconds on each side of the page. The paper was dried by placing it between two paper towels, and then pressed with a light weight box placed on top to aid its drying. Observation and clarity of stroke and retention of original color of ink around the letters were used for judgment. The paper and paper towel that was used to dry the papers were also examined for ink transfer. The judging factors determined the score or rating of each ink, with 5 as the highest, indicating the level of ink's retention on the paper, and 1 as the lowest, indicating that the words were unreadable after test (Ryan, 2017).

2.9 Colour Retention

The formulated ink samples were exposed to sunlight radiation for several hours and the impact, in terms of colour fasting, observed over a period of six days.

2.10 Statistical Analysis

Karl Pearson Correlation Coefficient (r) between pairs of variables (extract amount X, and ink physical properties Y) was computed as follows:

$$r = \frac{n(\sum xy) - (\sum x)(\sum y)}{\sqrt{[n\sum x^2 - (\sum x)^2][n\sum y^2 - (\sum y)^2]}} \qquad \text{Equation [3]}$$

Also, all quantitative ink physical properties (Y) were predicted as a function of extract amount (Log C) using Polynomial regression analysis. These models were derived using Microsoft Excel spread sheet (Version 2016).

3.0 Results

Table 2 compares the quantitative and qualitative properties of ink formulations produced from *A. indica* and *C. papaya.* It reveals that inks formulated from *A. indica recorded* higher density (0.84 -1.05 g/cm^3) than inks made from *C. papaya* (0.87 -1.02g/cm^3). The Drying time values ranged between 14 seconds in *C. papaya* and 6 -12 seconds for *A. indica.* pH values were higher in *C. papaya* (6.8-7.5) but lower in *A. indica* (5.2-5.6). The viscosity was higher in *C. papaya* (8.55 -13.37kg/m^{-1}s^{-1}) but lower in *A. indica* (12.87-7.66 kg/m^{-1}s^{-1}). The stability of ink formulations when exposed to sunlight was evaluated as follows for 60 hours (Table 3)

Table 2. Properties of formulated ink samples.

	A. indica			***C. papaya***		
	Sample A	**Sample B**	**Sample C**	**Sample A**	**Sample B**	**Sample C**
Density (g/cm^3)	1.05	0.96	0.84	1.02	0.81	0.81
Drying time (seconds)	12	10	6	14	13	8
Water proof	5	5	4	4	4	3
pH	5.2	5.3	5.6	6.8	7.3	7.5
Erase-ability	Easily erased	easily erased	Not easily erased	easily erased	easily erased	Not easily erased
Colour	Light blue	Light blue	Blue	Light green	Lemon	green
Viscosity kg/m^{-1}s^{-1}	12.87	10.51	7.66	13.37	11.74	8.55
Readability	Less distinct and clear	Distinct and clear	Distinct and clear	Less distinct and clear	Distinct and clear	distinct and clear

Table 3. Effects of sunlight rays on formulated ink samples.

Time (hours)	*Azardirachta indica* Sample A	Sample B	Sample C	*Carica papaya* Sample A	Sample B	Sample C
0	No colour change	No colour change	No colour change	No colour change	No colour change	No colour change
6	No colour change	No colour change	No colour change	No colour change	No colour change	No colour change
12	No colour change	No colour change	No colour change	No colour change	No colour change	No colour change
18	No colour change	No colour change	No colour change	No colour change	No colour change	No colour change
24	No colour change	No colour change	No colour change	No colour change	No colour change	No colour change
30	No colour change	No colour change	No colour change	No colour change	No colour change	No colour change
36	slight colour change	slight colour change	slight colour change	slight colour change	slight colour change	slight colour change
42	slight colour change retained	slight colour change retained	slight colour change retained	slight colour change retained	slight colour change retained	slight colour change retained
48	slight colour change retained	slight colour change retained	slight colour change retained	slight colour change retained	slight colour change retained	slight colour change retained
54	slight colour change retained	slight colour change retained	slight colour change retained	slight colour change retained	Colour faded to white	Colour faded to white
60	slight colour change	slight colour change	slight colour change retained	slight colour change	Colour faded to white	Colour faded to white

Table 4 revealed high significant correlation values between extract concentration and ink physical properties. It reveals that extract concentration (*A. indica*) yielded high negative correlation coefficients with density (-0.993), drying time (-0.974), water proof (-0.845) and viscosity (-0.996). On the other hand, the correlation coefficient was positive as regarding pH (0.949). Also, Table 5 revealed high significant correlation values between extract concentration and soil physical properties. It reveals that extract concentration (*C. papaya*) yielded high negative correlation coefficients with density (-0.722), drying time (-0.918), water proof (-0.845) and viscosity (-0.975). On the other hand, the correlation coefficient was positive as regarding pH (0.980). Table 6 shows parametric regression models explaining the variations in physical parameters of ink formulations as predicted by extract concentration (Log C= log transformed amount in ml of Extract).

Table 4: Matrix reflecting inter-relationships between ink formulation properties (*A. indica*).

	Extract amount	Density	Drying Time	Water Proof	pH	Viscosity
Extract amount	1					
Density	-.993	1				
Dry Time	-.974	.994	1			
Water Proof	-.845	.904	.945	1		
pH	.949	-.980	-.996	-.971	1	
Viscosity	-.996	0.999*	.991	.892	-.974	1

Significance at P<0.05 = *

Table 5: Matrix reflecting inter-relationships between ink formulation properties (*C. papaya*).

	Extract Concentration	Density	Drying Time	Water Proof	pH	Viscosity
Extract Amount	1					
Density	-.722	1				
Drying Time	-.918	.388	1			
Water Proof	-.845	.240	.988	1		
pH	.980	-.846	-.820	-.721	1	
Viscosity	-.975	.549	.983	.943	-.910	1

Table 6: Regression models explaining inter-relationships between ink properties.

Parameters	***A. indica***	***C. papaya***
Density	Y = -0.98(log C) + 2.5	Y = -0.73(log C) + 2.1
Drying time	Y = -28(Log C) + 54	Y = -28(Log C)+ 56
Water proof	Y = -4.6(Log C) + 12	Y = -4.6(Log C) + 11
pH	Y = 1.9(Log C) + 2.4	Y = 3.3(Log C) + 1.9
Viscosity	Y = -24(Log C) + 49	Y = -22(Log C) + 47

4.0 Discussion

The results authenticate the suitability of plant derived extracts as commercial brand ink substitute. similar observation had been made by earlier researchers (powar, *et. al*; 2014, & Dagde, *et. al.* 2019). This result confirms earlier assertion by Tritrat (2015) who reported that natural colorants from plants organs possess inherent potentials to yield colors suitable for writing, printing or dyeing. The ethanol used as solvent serves to prevent growth of molds, prevents the ink from solidifying and reduces its surface tension. Glycerol is viscous and therefore improves the ink's absorbency on the board. The added acetyl alcohol acts as an emulsifier which serves as a binder to control viscosity of the ink and acts as liquid pool cover that form a surface layer to reduce evaporation (Smolinske,1992).

The prepared inks were written on white board and then exposed to direct sunlight in order to assess distinctness and stability. The evidence from this protocol revealed that these formulations were relatively stable when exposed to sunlight radiation for about 40 hours. This corroborates the reports of kumar *et. al.* (2012) with studies on the effects of UV rays and sunlight on similar plant-based formulations. They reported no visible change of colour of ink in one week of exposure.

Peculiar to this result is the fact that the intensity of colours decreased gradually beyond 40 hours. This revalidates the findings of kumar *et. al.* (2012). Colour fading varied with the plant species and extract amount. This variability is associated with the amount of resinous sap present in both extracts (Trirat, 2015). In line with this, inks formed from *Azaridacta indica* were more stable and recorded a lower drying time on white board than those of *Carica papaya*. Generally, the values of ink quality parameters recorded compares with those of commercial ink brands as reported by Dagde, *et. al.* (2019).

The correlation matrix is a testimonial of relationships which exist between the physical properties of the formulation in relation to extract amount. The negative correlation coefficient between extract amount and formulation density, drying time, waterproof property and viscosity is expected. This evidence may be interpreted to mean that the higher the amount of plant materials in the formulation, the lower the density, the faster the drying time and the more viscous the liquid. These observations may be harnessed towards improving the quality of the formulation to meet up with the properties of commercial brands. The possibility of these adjustments may be made feasible by the application of the regression models described herein.

Regression is a direct technique which seeks to obtain a relationship between pairs of variables with conceivable prediction at high accuracy levels (Dagde, *et. al.* 2019, Mbong, *et. al.* 2020). Simulated models predicting the density, drying time, water proof, viscosity and pH of formulations made from plants material were derived using experimental data. Based on the amount of extract in the formulations, the models do not only illustrate the relationships between amount of ink with its quality parameters but gives a theoretical basis on the influence of varied amount of the different plant materials on the overall quality of the ink. This results thus vindicate that the application of this technique is suitable for monitoring, predicting and simulating the characteristics of plant-based ink formulations as a substitute for commercial brands.

5.0 Conclusion

Investigations to assess the suitability of plant-based formulation as replacement for dry erase marker ink were made. The results showed that plant-based ink formulations from the two plants studied recorded values for the pH, viscosity and density and other test parameters which compared favourably with commercial marketed brands. This study brings to bear the possibility of producing a high quality environmentally friendly and cost-effective dry erase ink formulation using local plant-based materials.

6.0 References

[1]Dagde. K.K, Nwosa G. I and Ukpaka. C. P. (2019). Formulation of White Board Marker Ink Using Locally Sourced Raw Materials. EJERS, *European Journal of Engineering Research and Science*; 4(3): 107 – 119.

[2]EPA (1995). Profile of the Petroleum Refining Industry. EPA Compliance Assistance and Sector Programs Division. EPA/310-R-95-013 2911: 63.

[3]Etukudo, U. I. (2019). Formulation and production of bioplastics from local plant waste materials. Master Degree Theses. Department of Chemistry, University of Uyo.

[4]Kumar, M. R., Priya, P., Lakshmi, R., Vadivelu, A and Gopal, V. (2012). Formulation and Standardisation of Herbal Based Edible Ink. *International Journal of Advances in Pharmacy, Biology and Chemistry*; 1(2): 252 -254.

[5]Mbong, E. O., Anwana, E. D. and Ezekiel, A. G. (2020). Correlating the Abundance of woody species to environmental gradient within University of Uyo Arboretum. Journal of Earth science and Climatic change. 1(2): 1 – 9.

[6]Morshed, M. N., Deb, H., Al Azad, Shamim., Sultana, M. Z., Arun, A, and Guha, K. (2016). Aqueous and Solvent Extraction of Natural Colorants from *Tagetes Erecta L., Lawsonia Inermis, Rosa L* for Coloration of Cellulosic Substrates. *American Journal of Polymer Science and Technology,* 2(2):34-39.

[7]Nwafulugo F. U., Samuel, F, Tobias, N., Omale, S. O., Nwosibe, P. O. (2019). Marker Ink Production from Berry Seed Extract. *International Journal of Scientific & Engineering Research.* 10(5):698-704.

[8]Powar P. V., Lagad, S. B., Ambikar, R. B. and Sharma, P. H. (2014). Herbal Ink: Safe, Easy and Ecofriendly Alternative. *International Journal of Pharmacognosy and Phytochemical Research.* 6(2):146-150.

[9]Ryan, B. (2017). The best waterproof fountain pen inks. Accessed September 01, 2020. www.mountainofink.com

[10]Trirat, P. (2015). The Property of Screen Ink from Natural Mordant, Colorant, and Additive for Art. *International Journal of Business and Social Science,* 6 (11): 68 – 76.

Corresponding Author:
Dr. George Ubong
Department of Fisheries & Aquaculture,
Akwa Ibom State University,
Obio Akpa Campus, Obio Akpa
Akwa Ibom State, Nigeria
Telephone: 08032625310
E-mail: ubonggeorge@aksu.edu.ng

5/21/2021