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Engineering sustainable inks from natural biomaterials for digital printing

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Abstract

With sustainability at the fore, the textile industry is ambitiously parading toward alternative ecological materials to eliminate air and water pollution resulting from petroleum-based contenders. This study reports the potential environmental consequences of synthetic inks and addresses the issue by formulating herbal inks for blue, red, yellow, and black colors derived from plant extracts such as bio indigo, quebracho red, and from the flame of the forest plant in digital printing. These engineered herbal inks were studied for their physical and chemical properties on the viscometer, conductivity meter, and tensiometer. The findings were statistically validated by applying an Analysis of Variance (ANOVA) one-way test. The Attenuated Total Reflection - Fourier Transform Infrared Spectroscopy (ATR-FTIR) studied the stability of the developed herbal inks, indicating good dye strength following a one-month storage phase at room temperature, except for bio indigo herbal ink. The study also highlighted the significantly strong results for the quebracho red herbal ink. The outcomes of the study support implementing biodegradable materials, waterless inkjet print technology, and processing at room temperature as a driver for implementing more sustainable practices in the garment dyeing industry. For future work, experimentation with herbal inks as conductive inks and life cycle analysis is recommended.

Keywords: circular materials; cotton; herbal inks; inkjet printing; sustainable inks; wool

Introduction

The European Green Deal's one trillion projects aim to establish a climate-neutral Europe by 2050, wherein the textile industry is at the heart of a new circular economy action plan (citation). The use of natural renewable circular materials in eco-design has been suggested (European, n.d.). A growing body of literature recognizes the importance of sustainability in textile wet processing units. Concurrently, the volatile organic compounds (VOCs) from solvents, namely ethanol, methanol, toluene, and other humectants such as urea, propylene glycol, polyethylene glycols, diethylene glycol, among others (Fu, 2006), are utilized in enormous amounts in the textile print industry, with dire consequences to the environment and human health alike (Christie, et al., 2000). According to a United Nations Environment Protection Agency study, the textile printing industry releases 99 % of its total Toxic Release Inventory into the air, while the remaining 1 % of discharge is let into the water and land at a 50:50 ratio, respectively (OEcotextiles). Its aftermath is manifested as greenhouse gas effect, ozone depletion, acid rain, diseases in humans such as nausea, headache, hallucinations, psychosis moreover, diseases of the liver, kidney, and central nervous system, and even DNA mutation (Marrion, 1994; OEcotextiles, n.d.). The Ellen MacArthur Foundation (2021) reported that the textile industry produces 21 billion tons of waste, 20 % of which is highly toxic wastewater from dye houses.

The typical printing inks contain phthalates as plasticizers such as Nethyl- and N-methyl toluene sulphonamides and tris(2-Ethylhexyl) taramellite, which is notorious to the environment and human health (Ethical Rebel, 2021). The emissions and effluents from textiles industries are the second largest globally, next to big oil. Water pollution kills 1.8 million people and makes 1 billion people ill per year, more than war and violence combined. Groundwater is tough to clean and expensive, therefore often impossible. Once polluted, it can be unusable for decades (Christie, et al., 2000). In essence, the waterless technology approach will prove helpful in expanding the color range accessible with natural colors. At the same time, the amount of extract is concentrated in herbal ink formulation and hence significantly less of it is consumed for inkjet printing than in the alternative, dyeing process. Consequently, inkjet printing protects agricultural land from being jeopardized for cultivating plants for fabric dyeing. In practicality, it would save both water and plants and propel climate action.

The use of digital printing may offer a solution to reducing chemical waste and combating the climate crisis. The process of inkjet printing is an electro-mechanical process involving low emissions and effluents generation. For instance, in the inkjet print process, the carbon footprint from energy consumption is 74.26 kg CO2eq of gas emission as compared to 123.76 kg CO2eq gas emission generated with rotary printing. Currently, synthetic inks for digital printing dominate the market. However, due to the potential perils, there is significant interest in eco-friendly inks or bio-inks. Bio inks are inks produced from living sources namely vegetables, plants, insects, ores, and others; herbal inks are inks constituted from any plant's part or shrubs as raw materials. Hence, the present research on plant-based ink formulation for inkjet printing of wool and cotton fabrics for sustainable outcomes.

In conjunction with digital printing, renewable plant-based pre-treatment and ink formulations provide an opportunity to mitigate the hazards associated with synthetic contenders. In application, the use of plant-based renewable sources circumvents depleting petroleum-based fossil fuels such as glycols, alcohols, and phthalates, which are responsible for high carbon footprints. (Minney, 2016). The current plant-based color palette is unique. However, specific colors are still tricky to derive without harmful additives and modifiers, namely black and blue. The detrimental materials in fossil fuel-based ink formulations are, for example, organic compounds, namely 2- pyrrolidone and propanediol utilized as cosolvents. Also, the non-ionic surfactants, such as Surfynol®, and anionic surfactants, such as aerosol and rheology (viscosity) modifiers polyethene glycols (PEG), are damaging ingredients in use within synthetic ink formulation. The pigment inks were reported to be more deleterious than water-based inks (Fu, 2006). Therefore, this research attempts to derive practical solutions by working with the cyan, magenta, yellow, and black color (CMYK) model set-incoherence within the typical digital printer.

Background

Scientific research on natural colors from plants indicates no possible health threat or toxicity to humans and animals alike (Vankar & Shukla, 2019). The dyes are synthesized from wild fruits, flowers, stems, leaves, bark, grasses, seeds, roots, trees, and berries. The molecules of natural origin are biodegradable, whereas synthetic chemicals lack biodegradability, and remain in the environment releasing toxic fumes (Sharma, et al, 2018). The rheology (physical characteristics) and stoichiometry (chemical constitution) of inks are vital to determining their use and potential. However, there is no comprehensive literature on herbal inks for inkjet printing.

Rheology

Rheology describes the science of how materials flow and deform (Marrion, 1994). Several studies suggest that digital printing ink's physical properties, mainly density, viscosity, and surface tension, provide valuable information on the jetting quality of the fluid (Marrion, 1994; Vadillo, et al., 2011). This research incorporates a filament stretching apparatus, called Cambridge Trimaster, in combination with high-speed cinematography to evaluate the fluid's (inks) high-speed pulling, which refers to the viscoelasticity of ink to flow through the printer capillaries, and break-up behaviour, which is specific to how the ink leaves the nozzle to form a perfect ink-drop. These imply print quality. Inks with in-range viscosity and surface tension would enable avoiding ink-drop formation with heads and tails that distort the print quality (In other words dithering and satellite formation are prevented), which also leads to lower colorfastness properties. As the molecular weight of an ink increases, the droplet slows down and forms more satellites in drop-on-demand (DOD) inkjet printing (Vadillo, et al., 2011). The ink droplet could be expelled from the minute nozzle only if the kinetic energy of the ink droplet transcends the forces related to surface tension and viscosity. The viscosity of the ink influences the process by which the ink droplet diffuses, migrates, and fixes into the microfibrillar structure of a fabric, thereby affecting the print accuracy and fastness properties of the printed fabric (Gao, et al., 2019). The herbal ink formulation process implements glue, oil, vinegar, and iron kind of additives to acquire the desired consistency and color on the application for paper (Neddo, 2015). In the same vein, the botanical inks are extracted from food waste, black walnut, oak galls, goldenrod, marigold, logwood, and other materials associated with natural modifiers like lemon juice to acquire a rainbow of colors (Behan & Lightbody, 2018).

Stoichiometry

Stoichiometry is a branch of chemistry that deals with the quantitative relationship between two or more substances, primarily physical or chemical changes. During the formulation of inkjet printing inks, various chemical phenomena occur, namely dissolutions, dispersion, ionization, colloid formation, wetting, absorption, and adsorptions onto the subsequent substrate (Christie, et al., 2000; Gao et al., 2021). The inks generally have a particle size of fewer than 500 nanometers, a pH value of between 7 and 9, surface tension between 25 and 50 mN m21 (millinewton meter), and a viscosity between 1 and 25 mPa-s (millipascal-seconds). These ink specifications are corroborated with typical digital printer requirements and form blueprints for new ink formulations.

Ink stability

Each formulated ink's physical and chemical properties vary due to the different molecular structures of the components involved, leading to various intermolecular forces. The weather stability of the ink is a significant performance and logistical parameter. For example, a good ink would not precipitate or change its physical and chemical characteristics during long storage (Christie, et al., 2000; Gao, et al., 2021). Typical printing inks contain phthalates as plasticizers, such as N-ethyl- and Nmethyl toluene sulphonamides and tris(2-Ethylhexyl) taramellite, which are noxious for environmental and human health (Nerin, et al., 1993).

Inkjet printing

Inkjet printing on textile substrates with plant-based colors has had a limited evaluation, as a result, there is insufficient literature on the experimentation of inkjet printing with herbal inks. In one evaluation, Savvidis et al. (2012) tested the yellow color from annatto seeds of the achiote tree for digital printing on cotton fabric. The annatto ink was stable over a 28-day timeline. Waterbased annatto ink gave a pale-yellow color with the color corresponding absorption-scattering of light wavelength (K/S) value of 0.42. However, no in-depth analysis was provided regarding inks rheology, stoichiometry (quantities), particle size, solubility, stability, and print quality analysis. The study focused on conventional dyeing and screen printing with natural dyes. Therefore, it is of limited relevance to the present research, which is exclusively focused on waterless ecological solutions for the coloration of textiles with plantbased colors, fulfilling an existing gap of knowledge in the field.

Materials

In the present study herbal biomaterials were implemented for the formulation of herbal inks, as depicted in Table 1. The plant-extract powders utilised were obtained from Ewe and Ply, Wonky Weaver, DT Craft and Design, Wigwam Wool Work, UK, and Sodhani Biotech Pvt. Ltd. India. The herbs were selected according to the C, M, Y, and K colour theory of the DOD print system Epson SureColor PC-600.

To validate the engineered herbal inks, the digital printing with herbal inks on wool and cotton fabrics is succinctly presented. It is always challenging to secure blue and black with plant-based extracts without the input of metallic additives, namely copper, ferrous, or aluminium elements hence not sustainable. Therefore, ecologically, a systematic approach was given to acquiring black from plant extracts. The primary colour scheme was adopted to achieve the much-coveted black, as shown in (Table 2) of the supplementary materials. Initially, natural plant-based colours from woad herb, weld herb, walnut leaves, myrobalan tannin, aleppo oak tannin, and others were experimented with. However, they were eliminated due to low solubility. Mineral-based natural colours, namely lapis lazuli, ochre, sienna, azurite, mineral black colour from slate rock, and iron and aluminium-based additives and others, were eliminated as encouraging those would further keep depleting the natural mineral ores.

Table 1. Herbal biomaterials for herbal inks formulation.

Colour	**Herb**	***Botanical name***	**Part of plant**	**Colouring component**	**Colour**
C & LC	Bio indigo	*Indigofera tinctoria*	Leaves	Indigotin	Blue
M & LM	Quebracho red	*Schinopsis lorentzii*	Wood	Tannin	Red
Y	Flame of the forest/ Sacred tree	*Butea monosperma*	Flowers	Butein	Yellow
K, LK & LLK	Bio indigo + quebracho + Sacred tree = Black colour	*Indigofera tinctoria* + *Schinopsis lorentzii* + *Butea monosperma*	Leaves + Wood + Flowers	Indigotin + Tannin + Butein	Blue + Red + Yellow = Black

Abbreviations for Colours: C, Cyan; LC, Light Cyan; M, Magenta; Y, Yellow; K, Black; LK, Light black; LLK, Light, light, black.

Table 2 Stoichiometry of herbal inks to acquire C, M, Y, & K colurs.

Herbal ink	**Colour**	**Colourant**	**Water**	**Glycerol**	**Viscosity**

		(g)	(ml)	(ml)	(cP)
Bio indigo herb	C	9	76	15	L
Bio indigo herb	LC	6	74	20	M
Quebracho herb	M	6	69	25	H
Quebracho herb	LM	4	76	20	M
Flame of the forest herb	Y	6	79	15	L
Bio indigo herb + quebracho herb + the flame of the forest herb	K	11	64	25	H
Bio indigo herb + quebracho herb + the flame of the forest herb	LK	11	69	20	M

Methods

Stoichiometry of herbal inks

Eight herbal inks (see Table 2) were developed as required for the Epson SureColor PC-600 print system. However, only the results specific to CMYK are considered for this discussion.

Because the particle size of a herbal extract affects the solubility of plant extract in distilled water, it is vital to investigate the particle size and the solubility of herbal ingredients while formulating inks. The plant extracts utilized for experimentation were water soluble and herbal ink formu lations were double filtered; this process ensured that the particle size would not block print jets. The solute, a herbal extract color, forms a herbal solution when dissolved in distilled water (DW), the solvent.

The entire ink formulation process was repeated to obtain the CMYK color herbal inks. Of note, the sludge obtained after double filtration is both recyclable and biodegradable.

Results and Discussion

Assessment of Herbal Ink Properties

The physical properties of the engineered herbal inks were statistically plotted and investigated, and the consequent changes in the properties of inks were systematically documented and investigated. It is vital to note that the herbal ink for the black color was formulated from a mixture of the primary colors, namely yellow + red + blue. Two sets of inks were formulated from the plant extracts named as herbal inks 1 (HI-1) and herbal inks 2 (HI-2). The prepared inks were assessed for their rheological (Physical) properties as detailed in Table 3 and 4.

The physical properties investigated include the relative density acquired for herbal inks. The relative density differed for each color. The discrepancy could be attributed to the particle size of the plant extracts. Finally, an ANOVA one-way test results concluded the relative density value of 1.06 to be the most influential on the K/S values gained on the wool and cotton fabrics inkjet printed with herbal inks.

Rheological assessment

In this study, HI-1, see Table 3, obtained the highest viscosity value (13.73 cP) with the ink made from the quebracho herb extract for magenta color (M). The second highest viscosity value of 9.98 cP was noted with the ink made from the logwood herb for yellow color (Y), followed by the viscosity value of 9.56 cP obtained with the herbal ink formulated for the black color (K); see Table 3.

HI-2, see Table 4, demonstrated the highest viscosity value of 12.30 cP with ink constituted from the quebracho herb for magenta color (M). The second highest viscosity value of 10.70 cP was noted for the ink formulated from the quebracho herb for light magenta color (LM), followed by the viscosity value of 9.76 cP gained with the herbal ink formulated for the black color (K); see Table 4. Ultimately, an ANOVA one-way test results determined the viscosity value of 9.98 cP to be the most influential on the K/S values gained on the wool and cotton fabrics inkjet printed with herbal inks.

Table 3 Rheology of Herbal inks 1.

Herbal ink	Relative Density	Viscosity (cP)	Surface tension (mN/m)	Conductivity (mS/cm)	pH	Viscosity range
Distilled water (DW)	1	5.30	60	0.00	6.05	L
Bio indigo herb(C)	1.09	7.23	56	15.60	6.63	L
Bio indigo herb (LC)	1.06	7.70	37	16.14	6.68	L
Quebracho herb (M)	1.06	13.73	49	2.51	5.19	H
Quebracho herb (LM)	1.04	8.56	39	2.85	5.03	M
Logwood herb (Y)	1.06	9.98	55	8.63	5.31	M
Bio indigo herb+ quebracho herb + logwood herb (K)	1.12	9.56	57	18.5	5.67	M
Bio indigo herb + quebracho herb + logwood herb (LK)	1.12	7.17	53	15.3	6.01	L
Bio indigo herb + quebracho herb + logwood herb (LLK)	1.12	6.67	58	14.7	6.04	L

The rheology of formulated herbal inks was anticipated to differ with each herb procured from varied sources. Hence the analysis was of vital importance. However, graphically no drastic fluctuations were noted between the formulated HI-1 and HI-2. Overall, the physical parameters of herbal inks were in the acceptable range for both HI-1 and HI-2. Additionally, the results reinforced the reliability of the adapted stoichiometry of herbal inks. As a result, following the stoichiometry is expected to accurately yield herbal inks with appropriate physical properties.

Plant-Extract Yield

The plant extracts were filtered on dissolution in distilled water. Each plant extract gained a different yield F1c as presented in Figure 1. Further, the actual colors acquired are different from the ideal CMYK colors.

Water-based herbal inks were constituted from plant extracts and the initial volume before the filtration process was noted. The actual volume gained after purification is reported, and the difference was calculated, analyzed, and graphed in Figure 1. The quebracho herb plant extract delivered a 100 % yield on extraction in distilled water. The smallest yield was obtained with bio indigo herb herbal extract, offering only 32.89 % yield on extraction in distilled water.

Table 4 Rheology of Herbal inks 2

Herbal ink	Relative density	Viscosity (cP)	Surface tension (mN/m)	Conductivity (mS/cm)	pH	Viscosity range
Distilled water (DW)	1	5.60	73	0.02	7.21	L
Bio indigo herb (C)	1.10	7.33	61	18.90	6.48	L
Bio indigo herb (LC)	1.06	9.70	52	9.18	6.94	M
Quebracho herb (M)	1.06	12.3	60	3.18	4.90	H
Quebracho herb (LM)	1.04	10.7	53	2.51	4.90	H
Flame of the forest herb (Y)	1.06	8.80	46	2.96	5.63	M
Bio indigo herb + quebracho herb + sacred tree herb (K)	1.12	9.76	60	9.52	5.78	M
Bio indigo herb + quebracho herb + sacred tree herb (LK)	1.12	9.40	54	19.91	5.87	M
Bio indigo herb+ quebracho herb + sacred tree herb (LLK)	1.12	9.10	51	12.92	5.87	M

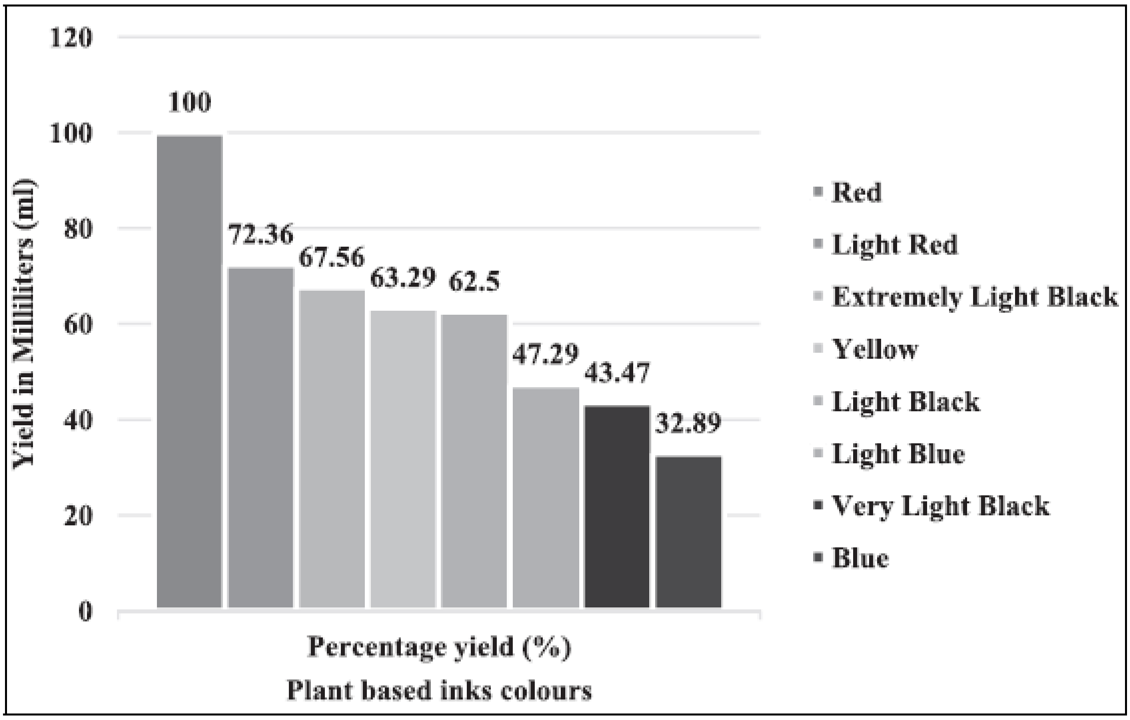

Figure 1 Color-wise percentage yield from plant extracts

There are two likely reasons for the differences between the yield acquired from the plant extracts, such as the particle size of each herb, and equally the sludge or residual obtained. Coined by Joel Hildebrand in 1916, the rule of thumb is "like dissolves like," when applied to the solubility of the solute in solvent, this is also known as a solubility parameter, thoroughly associated with the similar internal energies of the solvents-solute (Patton, 1979).

Stability of Herbal Ink to Storage Phase

The herbal inks were assayed for stability over a one-month storage phase in the shade at room temperature. Freshly prepared herbal ink is denoted as new herbal ink (NHI), while the same herbal ink following one month in storage is indicated as old herbal ink (OHI).

The CMYK-colored herbal inks were examined to observe changes in physical properties, viscosity, surface tension, conductivity, and pH parameters. The properties of NHIs were contrasted with OHIs after the one-month storage phase. The rule of thumb indicates that the smaller viscosity values, lower surface tension, and lesser conductivity values are desirable for drop-on-demand inkjet printing as this is consistent with electro-mechanical printer requirements.

In general, the herbal inks were suitable for printing after one month. For synthetic inks, it is recommended to use the ink within three months of opening while the packed ink has more than a year of shelf life. Hence, taken together, these findings have important implications for developing herbal inks with more extended stability to storage time.

Conclusions

Herbal inks can be constituted from plants, namely bio indigo, the flame of the forest, and quebracho red herb, for blue, yellow, and red colors, respectively. The black color inks can be derived from a mixture of blue, yellow, and red color herbs. In this study devised inks were successfully utilized for inkjet printing fabric and compared favourably with presently used toxic synthetic dyes.

Future Work

As a natural progression of this research, a life cycle analysis of the formulated herbal inks is suggested. The potential scope of color tweaking and engineering-conductive herbal inks with graphene/ silver/gold is recommended for future study.

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Contribution statement

Dr Danmei Sun; Supervision and editing.

Alka Madhukar Thakker; Conceptualization, Formal Analysis, Investigation, Methodology, Writing – original draft and editing.

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Disclosure statement

All authors declare that they have no conflict of interest.

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