

## Eco-ink Production from Waste Cooking Oil

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### Abstract

The production of eco-ink creates an alternative to replace conventional inks that contain toxic materials. In this study, a binder, raw materials, natural pigment, and additives are combined to create an ink formulation from waste cooking oil. The characterization of the produced eco-ink was carried out through chemical characterization using UV-Vis spectroscopy and FTIR spectroscopy. Physical characterization involved tests such as viscosity, ink flow, pH, adhesion, and water resistance to determine the suitability of the eco-ink formulation for practical applications. The results demonstrated the broad absorption band in the range of 3000–3600 cm<sup>-1</sup> corresponds to the O-H stretching mode, which indicates that the -OH group is due to distilled water, ethanol or glycerin and is present in both the charcoal and butterfly pea flower (BPF) inks during FTIR analysis, and the viscosity was found to be 4-5 mPa.s. Since the ink flow time is shorter, it indicates a lower viscosity value. The pH values were 8.5 for charcoal ink and 8.6 for BPF ink. The adhesion for charcoal ink was 44.44% and for BPF was 100%. Water resistance was 74% for charcoal ink and 54% for BPF ink. Overall, these results indicated the potential for utilizing waste cooking oil as a viable raw material for new sustainable products like eco-ink.

## 1. Introduction

The use of industry inks contains toxic materials, such as petroleum-based materials, which are non-renewable and not environmentally friendly. It contains volatile organic compounds (VOCs) such as toluene (C<sub>7</sub>H<sub>8</sub>), benzene (C<sub>6</sub>H<sub>6</sub>), and xylene (C<sub>8</sub>H<sub>10</sub>), which are organic substances with a high vapour pressure at ordinary room temperatures [1]. It will affect the environment as well as human health [2]. Besides that, according to the Malaysia Palm Oil Board (MPOB) survey, 77% of the population threw away waste cooking oil (WCO) in a less suitable way, resulting in pollution to the environment and clogging waterways. Therefore, producing ink from WCO is an alternative to utilizing waste because it has a lot of potential to be converted into useful things [3].

Previous research highlighted that palm oil is the most widely used vegetable oil in the world, exceeding soybean, rapeseed, and sunflower oil [4]. It produces 39.6% of all vegetable oils, is about four times as productive as other oil seeds and has the highest production per hectare of any edible oil. It is also the most widely used and cheapest type of vegetable oil [5]. However, ecology is affected when WCO from food preparation is disposed of improperly. These environmental problems include things like unpleasant odours and the accumulation of oil on the surface of the water, which increases biological oxygen requirements [6]. On the other hand, WCO have a great ability where recent oil chemistry research has shown that refined unsaturated vegetable oils can be used to produce high-quality printing inks for newspapers and magazines, such as lithographic, typographic, and offset

ink [7]. However, limited studies have been done to investigate WCO as a base for ink production, particularly with the use of natural pigments. This gap provides a chance to develop safer, biodegradable ink utilising readily available components while also addressing environmental pollution from wasted cooking oil.

Recent studies investigated bio-based ink formulations using other types of oils. For example, [8] effectively mixed waste soybean oil with natural pigments to create a plant-based ink formulation, illustrating the potential of food sector waste as a sustainable ink component. In another study, researchers produced offset printing inks using karanja oil as a sustainable substitute for linseed oil, which demonstrated acceptable print qualities and environmental benefits [9]. These innovations indicate the growing interest in using vegetable oils for eco-friendly ink. However, the usage of waste palm cooking oil and natural pigments for general-purpose ink remains limited in research.

This eco-friendly ink is composed of only organic material, making it safe to use, unlike conventional ink, which often includes chemicals, heavy metals and VOCs that cause negative impacts on the environment. To ensure compatibility and cost efficiency, only two types of organic pigments are used which are butterfly pea flower (BPF) and charcoal. These pigments were chosen due to their availability, low cost, and environmental advantages, making them promising options for eco-ink development. Next, gum arabic was used as the natural binder to help the ink adhere to the paper. Linseed oil was added as a fast-drying agent to improve absorption and prevent smudging. Glycerin acted as a humectant to maintain moisture and smooth flow of ink, while ethanol functioned as a preservative to prevent any microbial growth. Tween-80 was also used as an emulsifier to make sure the mixture is homogenous, and specifically, Tween-80 is chosen due to its biosafety, biocompatibility, and biodegradability. Additionally, it is proven that Tween-80 is effective in forming stable microemulsions with oils like coconut and olive oil [10].

The process of the ink consists of five phases. The first phase is the preparation of raw material, followed by preparation of binder, ink formulation, chemical and physical characteristics and lastly ink application. Furthermore, in this study, only several factors used to evaluate the ink formulation include viscosity, colour intensity, pH value, ink flow test, adhesion, water resistance, colour intensity and chemical bond. Lastly, only drawing papers are used to produce a standard substrate for the ink application.

Therefore, this research aimed to formulate eco-friendly ink using WCO. It also aims to characterise its chemical properties, such as colour intensity and chemical bonds, along with physical properties like viscosity, pH value, ink flow, adhesion, and water resistance. In addition, this study aims to evaluate the performance of eco-friendly ink that is produced by WCO on drawing paper.

## 2. Materials and Method

### 2.1 Materials

Waste Cooking Oil (WCO) was collected from households in the Batu Pahat area. Glycerin, Tween-80 emulsifier, Arabic gum, triethanolamine, linseed oil, Butterfly Pea Flower powder (BPF), charcoal powder, activated charcoal powder, cheesecloth, bleaching earth, filter paper, and an empty brush pen were purchased online. Distilled water and ethanol were provided in the UTHM Pagoh laboratory.

### 2.2 Preparation of Raw Materials

WCO was filtered using a cheesecloth to remove large impurity particles. Then, 25 g WCO was poured into a 100 ml beaker. The 25 g WCO was heated at 100 °C using a hot plate. Next, 0.5 g activated charcoal powder and 1.75 g bleaching earth were added to the 25 g WCO and mixed. The temperature was maintained at 100°C for 30 minutes. After that, the mixture was filtered through filter paper.

### 2.3 Preparation of Binder

1.5 grams of Arabic gum were weighed using an analytical balance and dissolved in a 100 ml beaker using 14 ml of distilled water. Arabic gum was stirred using a glass rod. After it had fully dissolved, 6 ml of Linseed oil, 6 ml of glycerin and 2 ml of Tween-80 were measured using a measuring cylinder and added to the dissolved gum Arabic. The mixture was mixed well.

### 2.4 Ink Formulation

The prepared binder was slowly added to the purified oil while stirring it continuously. Then the mixture was divided evenly into two 100 ml beakers, one for charcoal ink and another for BPF ink. Each beaker contained 45 mL of binder. Next, for the charcoal ink, 3.8 g of charcoal powder was gradually added to the beaker while stirring it. This was followed by the addition of 10 ml of distilled water, 4.5 ml of Tween-80 and 3 ml of ethanol, each added

one by one while continuously stirring to ensure even dispersion. For the BPF ink, 5.5 g of butterfly pea flower powder was added in the same manner, followed by 15 ml of distilled water, 5.5 ml of Tween-80 and 3 ml of ethanol. Each composition was added gradually while stirring to ensure a uniform and homogeneous mixture. Both mixtures were mixed until homogeneous, and the pH was checked. Triethanolamine was added dropwise until the range of 8.5-9.5 pH was reached.

## 2.5 Chemical Characterization

The chemical characterization involved evaluating colour intensity using UV-Vis spectroscopy by measuring the spectral reflectance of ink in the range 230-800 nm wavelength range, with the blank was ink base formulation without pigment added. Chemical bonds were identified using FTIR spectroscopy which revealed the presence of functional groups based on characteristic absorption peaks with the range wavelength 4000 and 750  $\text{cm}^{-1}$ .

## 2.6 Physical Characterization

Meanwhile, the physical characterization evaluated the usability of the ink through various tests. The viscosity of the ink was measured using the flow time method. A pipette was filled with 10 ml of ink, and the time taken for the ink to flow out completely was measured. Next, the same step was repeated for distilled water. Then, the viscosity of the ink was calculated using equation (1):

$$\eta_{\text{ink}} = \eta_{\text{distilled water}} \times \frac{t_{\text{ink}}}{t_{\text{distilled water}}} \times 100\% \quad (1)$$

where  $\eta_{\text{ink}}$  is the viscosity value of the ink,  $\eta_{\text{distilled water}}$  is the viscosity value of distilled water (1 mPa.s), time taken (t) ink is the time taken for the ink to flow out completely, and t distilled water is the time taken for the distilled water to flow out completely. Based on the viscosity test, the ink flow could also be determined. The suitable viscosity for this ink was in the range of 2–5 mPa.s. A higher viscosity of the ink indicated longer flow times, while a lower viscosity indicated shorter flow times.

The next physical characterization was the pH value. The pH value was determined using a pH meter and should fall within the range of 8.5-9.5. Then, the adhesion of the ink was measured using a tape test. The ink was applied to the paper and allowed to dry for 1 minute. After 1 minute, a grid line was drawn, and a piece of adhesive tape was pressed onto the surface of the paper. The tape was then quickly peeled off at an angle of 180 degrees. Next, the tape was examined to determine how much ink had come off with it. The adhesion was measured by using the formula of the percentage of ink remaining on the paper below:

$$\text{Percentage of ink remaining (\%)} = \frac{\text{Amount of the remaining area}}{\text{Total ink applied area}} \times 100\% \quad (2)$$

The last test used to analyse the physical characterization of the ink was the water resistance test. The ink was applied to the paper by drawing a grid line and allowed to dry for 1 minute. Two drops of distilled water were applied to the surface of the crosscut line and spread using a ruler. After that, the sample was left to dry for 30 seconds, and the distilled water was gently removed using tissue. The ink was then visually inspected for any changes, such as smudging. Equation (2) was used to determine whether the ink had good water resistance based on the percentage of ink remaining after water exposure.

## 2.7 Application of Ink

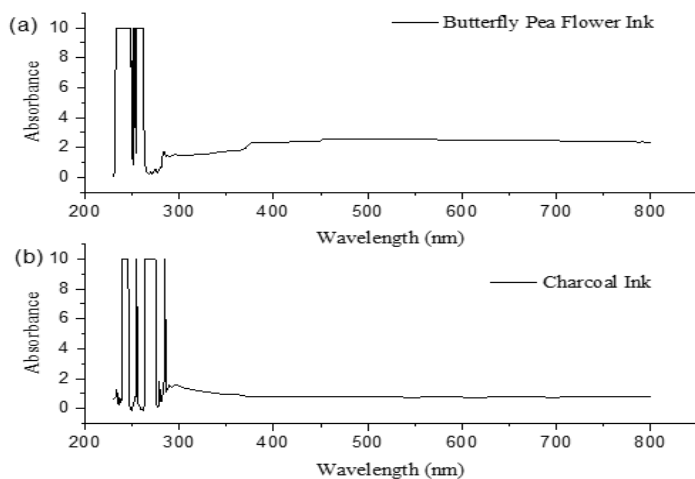
The application of ink on paper was conducted to evaluate its performance based on key properties such as spreadability and appearance. The ink was applied using a 4.5 mm brush pen on paper, which is drawing paper. Next, the ink was left to dry.

## 3. Result and Discussion

### 3.1 UV-Visible Spectroscopy

Carbon-based pigments, such as carbon black, usually show strong and continuous absorption in 180-260 nm of the UV-Vis spectroscopy region. No absorption peak ( $\lambda_{\text{max}}$ ) is observed in the visible range because they are broadband absorbers. Absorbance values are often below 2.0 A after necessary dilution to keep the instrument in its linear range [11]. In contrast, anthocyanins extracted from the Butterfly Pea flower also show a clear absorption maximum near 548 nm with absorbance values between 0.8 to 1.5 A, depending on concentration [12].

Fig. 1(a) shows that the maximum absorbance was 10.000 A observed at 269 nm, rather than where intact anthocyanins were expected at 548 nm. The source of this deviation is due to many factors, including the degradation of the pigment during formulation or storage. Absorbance values were high due to pigment concentration and lack of subsequent dilution. Fig. 1(b) shows a strong and persistent absorbance curve with not specific  $\lambda_{max}$  in the visible region, with extremely high absorbance values greater than 10.000 A, because of the lack of dilution due to high concentration, caused the spectroscopy to record values outside of its linear detection range [13].



**Fig.1** UV-Vis Spectrophotometer (a) BPF ink; (b) Charcoal ink

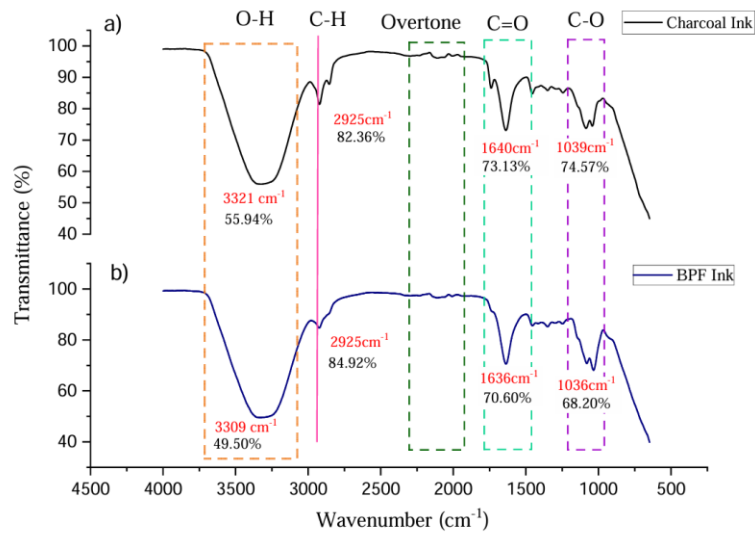
### 3.2 FTIR Spectroscopy

Fig. 2 indicates the FTIR spectroscopy for both charcoal and butterfly pea flower (BPF) ink. The functional groups for charcoal ink and BPF ink are observed by using Fourier-Transform Infrared Spectroscopy (FTIR) at a wavelength range  $4000-450\text{ cm}^{-1}$  [14]. A broad absorption band observed at  $3000-3600\text{ cm}^{-1}$  corresponds to the presence of O-H stretching, likely contributed by distilled water, ethanol or glycerin. Meanwhile, absorption peaks that observed between  $2850-2950\text{ cm}^{-1}$ , indicating C-H stretching vibrations. These are the long hydrocarbon chains, primarily present in fatty acids, which make up a major component of waste cooking oil (WCO) [15].

Additionally, the absorption band in the  $1500-1700\text{ cm}^{-1}$  region correspond to C=O stretching, which may rise due to the ester group in Tween-80 and the carboxylic group present in gum Arabic [16]. An overtone peak is observed in both inks around  $2000\text{ cm}^{-1}$ . Overtone is often related to aromatic ring structures. It could be come from carbon-based aromatic compounds in charcoal ink, or it could also come from flavonoids and anthocyanins in BPF, both of which contain aromatic rings. Next, the band observed around  $1000\text{ cm}^{-1}$ , most likely corresponds to the C-O stretching from various alcohol groups present in glycerin and ethanol, as well as the sugar backbone of gum Arabic [17].

Overall, the FTIR spectra of charcoal and BPF ink show that most peaks are the same due to the similar use of materials, such as waste cooking oil, ethanol, glycerin, Arabic gum, and Tween 80. However, slight differences were noticed, particularly in the O-H stretching area, where BPF ink showed a lower transmittance of 49.50% at  $3309\text{ cm}^{-1}$  compared to charcoal ink, 55.94% at  $3321\text{ cm}^{-1}$ . This indicates a higher number of O-H groups in BPF ink compared to charcoal ink, most likely due, more water usage during pigment preparation, and may also be due to the natural O-H groups present in anthocyanin compounds in the BPF pigment.

Furthermore, in the FTIR spectrum of BPF ink, the absorption band at about  $1036\text{ cm}^{-1}$  is attributed to C-O stretching vibrations, which are commonly associated with structural and non-structural carbohydrates, including sugars and glycosidic bonds found in anthocyanin compounds [18]. Since BPF contains a high concentration of anthocyanins, which commonly exist as glycosides, the absorbance at the C-O region is most likely due to its sugar structure. In contrast, charcoal, which is primarily carbonaceous and inorganic, lacks such structures, explaining the slight difference in transmittance found between the two inks in the C-O band.



**Fig. 2** FTIR of (a) BPF ink; (b) Charcoal ink

### 3.3 Viscosity Test

Table 1 shows the time taken for both charcoal ink and BPF ink to flow out compared by distilled water as a reference.

**Table 1** Time taken for ink flow

Ink Type	Reading 1 (s)	Reading 2 (s)	Reading 3 (s)	Average (s)
Charcoal	58.30	53.38	54.83	55.50
Butterfly Pea Flower	65.62	58.50	57.68	60.60

The time taken for the distilled water to flow out completely was 12.7 seconds. The viscosity value for both inks was determined using equation (1), with the average for each ink being the value for  $t$  ink. The viscosity for charcoal ink was 4.37 mPa.s, and for the BPF ink was 4.77 mPa.s. This shows that the charcoal ink has a lower viscosity than the BPF ink. Both ink viscosities are between the range 2-5 mPa [19], which is a suitable viscosity for a regular ink. It helps to indicate each ink flow time since a lower viscosity of the ink indicates shorter flow times.

### 3.4 Ink Flow Test

Since the viscosity test was done using the flow time method, the time taken for the ink to flow out is the result for this test. Each ink shows a decrease in seconds the more trial is run, with the Charcoal shortest time to be 53.38 seconds and the BPF 57.68 seconds, with distilled water as the blank sample (12.70 seconds). This test is required to know the time required by a fluid to flow. It shows that the inks are denser than distilled water, making it take more time to flow through.

### 3.5 pH Value

Fig. 3 (a) and (b) show the effect of adding triethanolamine (TEA) to the ink. This highlights that the presence of triethanolamine makes the ink more alkaline. As shown in Fig. 3 (a), the initial pH value for charcoal ink was 5.5, and it increases to 8.5 when TEA is added dropwise. In Fig. 3 (b), the BPF ink initial value was 5, and it increased to 8.6. The final pH value for both inks reached the targeted pH value, which is 8.5-9.5 [20]. Acidic pH may reduce pigment solubility, causing crystallization or aggregation on the paper, and improve light fastness, which is a reason why pH 8.5-9.5 is an ideal range for ink.

Additionally, after storing the ink for two months, the BPF ink started to change colour, indicating a change towards a more acidic pH. A similar finding states that natural inks, such as bio-indigo, also showed a pH drop after storage, affecting viscosity [21]. This phenomenon might be due to anthocyanins, which are natural pH indicators in butterfly pea flowers.

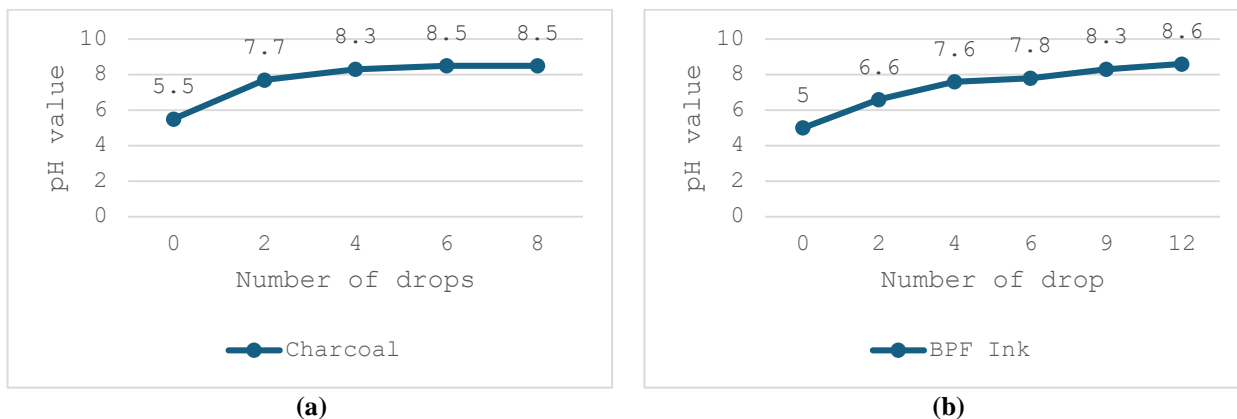


Fig. 3 Effect of adding TEA in (a) Charcoal ink, (b) BPF ink

### 3.6 Adhesion Test

Fig. 4 shows the visual grid consists of 108 small boxes drawn on the ink surface. The percentage of ink remaining on the paper was calculated using equation (2). For charcoal ink, 48 boxes contained some ink remaining after the tape test. This indicated a calculated adhesion percentage of approximately 44.44%. For butterfly pea flower ink, visual inspection suggested that nearly all boxes contain their ink after the tape test. Out of 108 boxes, nearly 108 boxes contained intact ink, representing a very strong adhesion with 100% percentage.

A lot of black residues were transferred to the tape. The charcoal ink applied in this study contains undissolved charcoal powder. Solid particles are loose at the physical level, and the tape can easily lift the charcoal particles. Consequently, the amount of residue on the tape may not represent only poor adhesion but also show the physical property of charcoal powder wanting to release [22]. For butterfly pea flower ink, there was no ink transfer, and the tape appeared clear.

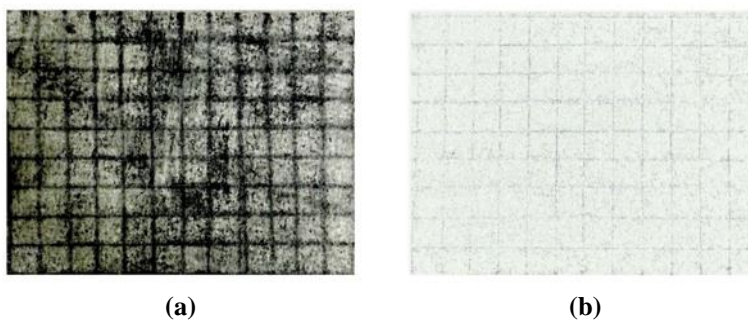


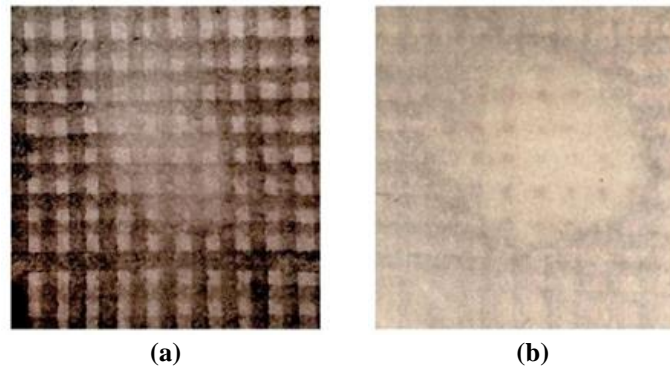
Fig. 4 Tape test after removal from the surface (a) charcoal ink, (b) BPF ink

### 3.7 Water Resistance Test

The water resistance test evaluated the durability of the ink against water exposure by measuring the percentage of ink that remained after contact with distilled water. As shown in Fig. 5, the two inks had very different responses to water exposure. The visual grid consisted of 100 small boxes drawn on the ink surface. The amount of ink remaining on the paper was evaluated using equation (2).

As demonstrated in Fig. 5 (a), the result for charcoal had 74 boxes remaining with the ink still intact after the test. This suggested a calculated water resistance percentage of approximately 74%. The grid lines remained easily visible with the application of water and removal from the paper. Only slight smudging was noted, suggesting that most of the ink remained intact. This implies that charcoal ink has reasonable water resistance, as it adhered well to the surface of the paper and did not easily dissolve or wash away.

Fig. 5 (b) demonstrated the result for Butterfly Pea Flower ink, where out of the 100 boxes, approximately only 54 boxes had ink still intact with a percentage of 54%. The ink grid lines almost completely disappeared after the same water treatment. The grid lines were hardly visible at all at the impacted area of the water treatment. This implies that water resistance of BPF ink is very poor, as a large portion of the ink got removed or smeared upon contact with water.



**Fig. 5** (a) Charcoal ink with intact grid lines, (b) BPF ink with faded grid lines after water exposure

### 3.8 Application of Ink

As shown in Fig. 6, the formulated ink applied with a 4.5 mm brush pen demonstrated good spreadability and initial appearance on paper. But after two weeks, there were sporadic and broken lines during application due to the irregular ink flow. There are probably a number of causes for this problem. First of all, clogging in the pen's feed system may result from an increase in ink viscosity brought on by solvent evaporation over time [23]. The smooth flow of ink may also be disrupted by phase separation, in which pigments sink to the bottom and the binder gets more concentrated. Additionally, eco-friendly ink formulations are vulnerable to microbial growth in the absence of suitable antimicrobial agents, which can result in the formation of biofilms and additional blockages inside the pen. It is interesting to note that the ink performed flawlessly with no indications of drying or flow disruption when applied using different application techniques, like a paintbrush and stamping.



**Fig. 6** (a) Charcoal ink application, (b) BPF ink application

## 4. Conclusion

The experimental results demonstrated key differences between Butterfly Pea Flower (BPF) ink and charcoal ink. BPF ink showed a viscosity of 4.77 mPa·s with a flow time of 60.60 seconds, while charcoal ink had a slightly lower viscosity of 4.37 mPa·s and a faster flow time of 55.50 seconds. FTIR analysis confirmed similar functional groups in both inks, including O-H, C-H, C=O, and C-O stretches. UV-Vis analysis revealed that charcoal ink had a strong absorbance peak at 269 nm with no visible  $\lambda_{max}$ , while BPF ink did not show the expected peak at 548 nm, likely due to pigment degradation. pH values for both inks were successfully raised to the target alkaline range (8.5–9.5) using triethanolamine. The adhesion test showed that BPF ink had excellent bonding with 100% ink retention, whereas charcoal ink had only 44.44% adhesion due to the presence of undissolved particles.

The development of this ink has shown promising potential in addressing both performance and environmental concerns. Laboratory testing confirmed key properties such as durability, adhesion, and colour strength, making it suitable for further development. However, it remains underdeveloped and requires more in-

depth study and testing to reach its full commercial potential. Areas for improvement include colour consistency, drying time, and spreading quality. This ink could support sustainable practices and is suitable for commercialization as a safer and more environmentally friendly alternative.

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## Conflict of Interest

Authors declare that there is no conflict of interests regarding the publication of the paper.

## Author Contribution

The authors confirm contribution to the paper as follows: **study conception and design:** Siti Nazirah Abdul Malek, Nurul Fatimah Syahronie, Nurish Alysa Sofea Roslan, Norazreen Sharip; **data collection:** Siti Nazirah Abdul Malek, Nurul Fatimah Syahronie, Nurish Alysa Sofea Roslan, Norazreen Sharip; **analysis and interpretation of results:** Siti Nazirah Abdul Malek, Nurul Fatimah Syahronie, Norazreen Sharip; **draft manuscript preparation:** Siti Nazirah Abdul Malek, Nurul Fatimah Syahronie, Norazreen Sharip. All authors reviewed the results and approved the final version of the manuscript.

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