# Synthesis and Characterization of Natural Pigmens from Butterfly Pea Flower (Clitoria Ternatea) With PVA Binder for Eco-Friendly Ink Applications

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## ABSTRACT

This research aims to synthesize and characterize natural ink from butterfly pea flower (Clitoria ternatea) pigments as a dye, using aquades was use for solvent, polyvinyl alcohol (PVA) as a binder, and alcohol as an additive for eco-friendly ink applications. The pigment was extracted through maceration using aquades and alcohol after sieving to 120 mesh. The pigment extract was mixed with varying amounts of PVA (0, 0.2, 0.4, 0.6, 0.8, 1, and 1.2 grams) to form ink with physical characteristics similar to commercial ink. Color identification was performed using a color meter, density measurement through empirical calculation, viscosity measurement with an Ostwald viscometer, and print tests using a Canon iP2770 printer. The color meter test results showed that the butterfly pea flower pigment is bluish-purple in liquid form and turns light blue when printed. The ink density with 1.2 grams of PVA was 0.95 grams/ml, equivalent to conventional printer ink. However, its viscosity was higher (27.88 cP) compared to conventional ink (1.12 cP) due to the larger particle size (0.1 mm). The addition of 1.2 grams of PVA produced a color closest to commercial ink, with an identical a* value (-0.5%), although the L* and b* values still differed. Butterfly pea flower ink demonstrated good color stability under normal storage conditions. This study indicates that the combination of butterfly pea flower pigment and PVA has potential as an eco-friendly natural dye, but formula optimization is needed to reduce particle size and improve print quality, thereby benefiting the sustainable ink industry.

**Keywords**: Natural Pigments, Butterfly Pea Flower, Ink, PVA

## INTRODUCTION

In an era where environmental sustainability is a global priority, the ink and printing industry is also experiencing a paradigm shift. One aspect that has garnered the attention of researchers and manufacturers is the use of natural dyes as a primary component of ink [1]. Natural dyes, extracted from plants, animals, and microorganisms, have been used since ancient times in various applications, including textiles [2], food [3], and cosmetics [4].

Natural dyes have attracted interest as substitutes for synthetic dyes, as evidenced by research such as that conducted by Ramadhani et al. (2024) using durian fruit peels and jengelang leaves, Bukit et al. (2022) using guava leaves, Assifa et al. (2020) using dragon fruit peels, and Wiguna et al. (2015) using leaf waste, among many others. This is due to the promising potential of natural-based inks and their lower impact on health and the environment [5].

The renewed attention to natural dyes in ink formulations is driven by several factors. First, increasing consumer awareness of the environmental impact of synthetic dyes, which are often derived from petrochemical sources and can produce toxic waste. Second, advancements in extraction and stabilization technologies for natural dyes have improved their quality and reliability. Third, the global trend toward 'green' and 'bio-based' products has created a ready market for inks based on natural dyes [6].

One promising source of natural dye for ink applications is the butterfly pea flower. Butterfly pea (Clitoria ternatea), also known as the Butterfly Pea, is a tropical plant commonly found in Southeast Asia [7]. The flower is characterized by its bluish-purple color and produces green seed pods. The color of the butterfly pea flower is due to the presence of anthocyanins, which range from red to deep purple [8]. Anthocyanins are flavonoid compounds commonly found in flowers, fruits, and leaves, capable of producing various colors such as red, purple, and blue [9]. The blue color of the butterfly pea flower has long been used as a natural dye in the food industry, including in butterfly pea tea, cake coloring, and beverages [10]. Although the butterfly pea flower has been utilized in various traditional applications, its use as a pigment in ink formulations remains underexplored.

In simple terms, ink consists of four main components: colorants (dyes or pigments), carriers, binders, and additives [11]. The colorants provide the color of the ink, the carriers help transfer the ink to the substrate, the binders help the colorants adhere to the substrate, and the additives are used to modify the properties of the ink, such as viscosity, water resistance, or drying time [12].

Polyvinyl alcohol (PVA) is a widely used chemical in various industrial applications, including as a binder in ink formulations [13]. PVA is a water-soluble synthetic polymer known for its unique physicochemical properties, such as good solubility, high adhesive strength, and the ability to form strong and flexible films. These properties make PVA highly suitable as an adhesive in ink, where the correct viscosity and consistency are crucial for optimal performance and print quality [14].

In the ink industry, binders serve to control the viscosity of the ink, ensuring that it flows well through the printing system without dripping or clumping [15]. Binders also aid in forming a uniform ink film on the surface of the print media, resulting in sharp and clear images and text. PVA has good compatibility with various types of pigments and dyes, including natural pigments, which are increasingly being used as environmentally friendly alternatives to synthetic pigments [16]. The use of natural pigments offers significant environmental benefits and reduces dependence on synthetic chemicals, which are often toxic and non-biodegradable.

The use of additives is an important aspect in enhancing ink performance. One common type of additive is alcohol. Alcohol has several advantageous properties in ink formulations, especially in water-based inks and inks used in modern printing applications such as inkjet and flexographic [17]. The addition of alcohol can help dissolve other ink components that are less soluble in water, such as certain dyes, resins, or other additives. Additionally, alcohol has a lower boiling point than water, which helps accelerate the drying of the ink after deposition on the substrate [18]. This is particularly important in high-speed printing applications or on less absorbent substrates.

This study aims to synthesize, characterize, and assess the feasibility of natural ink using pigment from butterfly pea flowers as a dye, distilled water as a solvent, PVA as a binder, and alcohol as an additive. The pigment extraction process was conducted through sieving to 120 mesh, referring to the study by Wiguna et al., (2015), followed by maceration using distilled water as a solvent. The pigment extract was then mixed with various concentrations of PVA (0 grams, 0.2 grams, 0.4 grams, 0.6 grams, 0.8 grams, 1 gram, and 1.2 grams) to determine the optimal ink formulation.

The ink characterization included color testing using a color meter, viscosity measurement using an Ostwald viscometer, density measurement using empirical calculation, and print testing using a Canon iP2770 printer to analyze the potential of ink from butterfly pea flowers.

## MATERIAL AND METHODS

The ink characterization included color testing using a color meter, viscosity measurement using an Ostwald viscometer, density measurement using empirical calculation, and print testing using a Canon iP2770 printer to analyze the potential of ink from butterfly pea flowers.

The first stage of this research involved drying butterfly pea flowers using a microwave at a moderate temperature for 30 minutes to remove the moisture content from the flowers. Subsequently, the dried flowers were ground using a mortar and pestle to reduce their size. The butterfly pea flowers, now in powder form, were then sifted using a T 120 mesh screen to produce homogeneous flower powder.

In the next step, 5 grams of butterfly pea flower powder was prepared and mixed with PVA in amounts of 0 grams, 0.2 grams, 0.4 grams, 0.6 grams, 0.8 grams, and 1.2 grams. Then, two milliliters of alcohol was dissolved in two milliliters of distilled water and stirred using a magnetic stirrer for five minutes at room temperature at 500 rpm. After that, butterfly pea flower powder and PVA were gradually added to the distilled water and alcohol solution and stirred for 20 minutes at 70°C until they became ink.

Color testing is done using a color meter, viscosity measurement using an Ostwald viscometer, density determination using empirical calculations, and print testing using a printer.

# RESULTS

This research focuses on the synthesis and characterization of natural pigments from butterfly pea flowers (Clitoria ternatea) for eco-friendly printer ink applications. The use of natural pigments is an important step towards a more sustainable ink industry, reducing dependence on synthetic pigments that can have negative environmental impacts [6].

## Extraction of Natural Pigments

The natural pigment extraction from butterfly pea flowers (Clitoria ternatea) was conducted using the maceration method with distilled water as the solvent, alcohol as an additive, and PVA as the binder, with variations. The resulting ink has a deep blue color, as shown in Figure 1. This color is attributed to the presence of anthocyanins, primarily delphinidin-3-glucoside [19]. Anthocyanins are natural pigments that can produce blue, purple, and violet colors [20].

Figure 1. Butterfly pea flower ink

The ink extracted from butterfly pea flowers was then compared with commercial ink by dropping them onto paper to test initial ink characteristics, as shown in Figure 2. The results indicated that the butterfly pea flower ink has a deep blue color (Figure 2a) compared to the brighter blue color of the commercial ink (Figure 2b). Another noticeable difference is the rate at which the ink is absorbed into the paper. Commercial ink is more easily absorbed into the paper compared to butterfly pea flower ink, influenced by factors such as viscosity, particle size, and additives used [21].

Figure 2. Comparison of inks when dropped on paper a) butterfly pea flower ink b) commercial ink

Ink that does not readily absorb into paper has several significant advantages, especially in applications requiring high print quality and durability. Clarity and color sharpness are among its main advantages, as pigments or dyes remain on the paper's surface, resulting in more vivid colors and sharper prints [22].

**Ink Characterization**

To analyze the ink produced from butterfly pea flowers, density testing, viscosity testing, color testing, and print testing were conducted.

**Density**

Density (ρ) is defined as the mass (m) of an object divided by the volume (V) occupied by the object. Mathematically, it is formulated as [23]:

$$\rho = \frac{m}{V} \qquad (1)$$

Density measurement was conducted using the pycnometer method for butterfly pea flower ink with variations of PVA binder and commercial printer ink [24].The density was measured by filling the pycnometer with the ink solution and measuring its total mass, then calculating the density based on the mass difference between the empty pycnometer and the pycnometer filled with ink, divided by the volume of ink in the pycnometer. The measurement results can be seen in the following table (Table 1).

The density of ink is one of the important parameters that determine the physical properties and performance of ink in various applications, including printing and writing. Ink density describes how dense an ink is, which directly impacts several important aspects of ink use and performance.

From Table 1. it can be seen that the density of butterfly pea flower ink increases with the increase in the added PVA mass. PVA (polyvinyl alcohol) is a water-soluble polymer commonly used as a binding or thickening agent in ink formulations [25]. The addition of PVA increases the viscosity and density of the ink, as indicated by the increase in density values from 0.6587 g/mL in ink without PVA to 0.9515 g/mL in ink with 1.2 g PVA.

Table 1 The density of ink with various PVA binders

| PVA mass (gr) | Empty pycnometer mass (g) | Ink-Filled Pycnometer mass (g) | Volume (mL) | Density (g/mL) |
|---|---|---|---|---|
| 0 | 30.50 | 37.087 | 10 | 0.6587 |
| 0.2 | 30.50 | 37.765 | 10 | 0.7265 |
| 0.4 | 30.50 | 37.903 | 10 | 0.7403 |
| 0.6 | 30.50 | 38.277 | 10 | 0.777 |
| 0.8 | 30.50 | 38.447 | 10 | 0.7947 |
| 1 | 30.50 | 38.573 | 10 | 0.8073 |
| 1.2 | 30.50 | 40.051 | 10 | 0.9515 |
| Commercial ink | 30.50 | 40.006 | 10 | 0.9506 |

The density of butterfly pea flower ink without PVA (0.6587 g/mL) is much lower compared to commercial ink. This indicates that ink without PVA is thinner and may have lower viscosity. Inks with lower density tend to penetrate more into the paper, which can result in less sharp and more prone to smudging prints [26].

With the addition of PVA, the density of butterfly pea flower ink increases gradually. With the addition of 0.2 g

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PVA, the density of the ink increases to 0.7265 g/mL, and continues to increase until reaching 0.9515 g/mL with the addition of 1.2 g PVA. This increase indicates that PVA plays a significant role in increasing the viscosity and density of the ink, approaching the characteristics of commercial ink.

The density of the commercial ink measured was 0.9506 g/mL. This value is very close to the density of butterfly pea flower ink with 1.2 g PVA (0.9515 g/mL), indicating that with the addition of PVA at a certain concentration, butterfly pea flower ink can achieve similar physical characteristics to commercial ink in terms of density.

**Viscosity**

Viscosity is one of the most important properties in the ink industry. Generally, viscosity refers to the thickness or resistance to flow of a fluid to shear force. In ink manufacturing, viscosity plays a crucial role in determining the ink flow behavior, transfer to the substrate, and the quality of the final print [27].

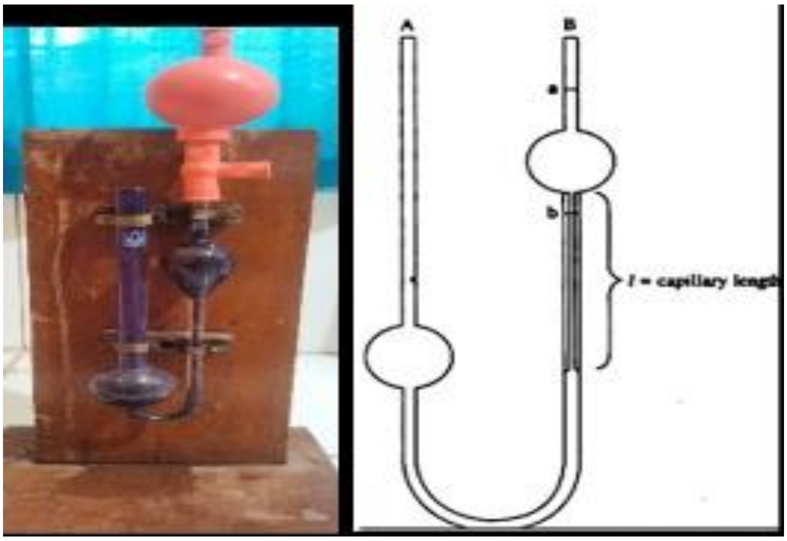


Figure 3. a) Ostwald viscometer b) Sketch of Ostwald viscometer section

The Ostwald viscometer, also known as a capillary viscometer, utilizes the principle of fluid flow through a capillary tube to determine the viscosity of a liquid [28]. This method is very practical and economical, and suitable for use in various types of inks, including water-based inks, solvents, and natural materials.

The working principle of the Ostwald viscometer is quite simple. This viscometer consists of two large bulbs connected by a capillary tube. The ink sample is introduced into the viscometer, and the time taken by the ink to flow through the capillary tube is measured accurately. This flow time is then used to calculate the viscosity of the ink relative to a standard formula used to measure viscosity using the Ostwald viscometer [29] is as follows:

$$\eta = \eta 0 \frac{(t \, . \, \rho)}{(t0 \, . \, \rho 0)} \qquad (2)$$

where $\eta$ is viscosity of the test fluid (cP), $\eta 0$ is viscosity of the reference fluid (cP), $\rho$ is density of the test fluid (gr/mL), $\rho 0$ is density of the reference fluid (gr/mL), $t$ is flow time of the test fluid, and $t0$ flow time of the reference fluid.

By using equation 2 and the previously calculated density, the viscosity of the ink can be determined. The relationship between the viscosity of butterfly pea ink and the variation in PVA mass is shown in Figure 4. It can be seen that with an increase in PVA mass, the viscosity of the ink increases significantly. The dashed red horizontal line indicates the viscosity value of commercial ink, which is at 1.12 cP. From this graph, it can be concluded that the increase in PVA mass is directly proportional to the increase in ink viscosity, indicating that the addition of PVA increases the thickness and viscosity of the ink. The viscosity of butterfly pea ink is much higher than that of commercial ink, especially at higher PVA masses.

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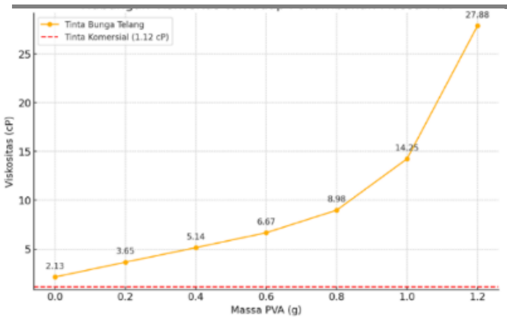


Figure 4. Graph of the relationship of ink viscosity to PVA mass gain

In addition to the increase in PVA mass, the size of the butterfly pea powder significantly affects the measured viscosity of the ink. PVA acts as a binder that forms a polymer network in the ink, increasing viscosity with the addition of its concentration. This is evident from the increase in measured viscosity, from 2.13 cP at 0 grams of PVA to 27.88 cP at 1.2 grams of PVA. Furthermore, the relatively large size of the butterfly pea powder particles (Figure 5) at around 0.1 mm also affects the measured viscosity. These large particles increase flow resistance in the ink, causing longer flow times in the viscometer and resulting in higher viscosity [30].

Figure 5 illustrates the results of butterfly pea ink strokes using a brush. In the main image, an area with uneven texture due to the ink strokes is visible. In the magnified image, the ink particles are still coarse and poorly dispersed, leading to an uneven texture of the strokes. This is attributed to the use of a T120 screen mesh with a 0.1 mm diameter, which is less effective in producing fine ink particles. A screen with a 0.1 mm hole width still allows large particles to pass through, resulting in coarse ink particles.

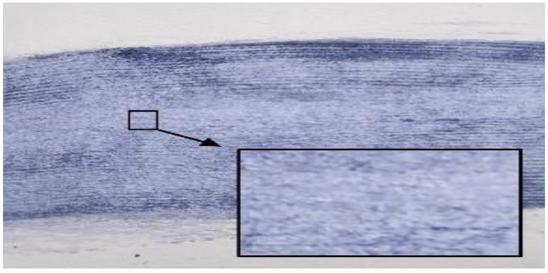

Figure 5. Depicts the application of butterfly pea ink on paper.

The national standard for printer ink viscosity requires ink to have a viscosity within a specific range to ensure optimal performance across various types of printers. According to the national standard (SNI 06-1567-1999) for printer ink, the ideal viscosity for inkjet printer ink is between 0.9 and 1.12 Cp [31]. From the measurement results, all butterfly pea ink formulations with PVA variations have significantly higher viscosities than the national standard. This is due to the relatively large size of butterfly pea particles, around 0.1 mm, which slows down ink flow and increases viscosity. Ink with high viscosity may encounter problems in inkjet printer applications as it can cause nozzle clogging and uneven ink flow [32]. Therefore, for practical applications, further optimization is needed, either by reducing the size of butterfly pea particles or by adjusting the PVA concentration to achieve viscosity that meets the national standard. This optimization will ensure that butterfly pea ink can be effectively used in inkjet printers, offering an environmentally friendly alternative that still meets the required performance criteria.

## Color Testing

Color testing on butterfly pea ink with PVA binder was conducted using a color meter to evaluate the quality and consistency of the colors produced in LAB color code. Color measurement in the LAB code provides parameters L* (brightness), a* (green-red axis), and b* (blue-yellow axis), which collectively reflect the accurate color of the ink sample. The working principle of the color meter is based on measuring the intensity of light reflected/transmitted by the sample at specific wavelengths [33]. The results of color testing using a color meter are presented in Table II.

The measurement results in Table II indicate the influence of Polyvinyl Alcohol (PVA) mass on the color parameters measured using the LAB system, namely L (brightness), A (redness), and B (yellowness). Under conditions without PVA addition (0.0 g), the brightness value (L) reaches 40.1%, with A value at -2.3% and B value at -20.4%. With the addition of 0.2 g PVA, there is a decrease in brightness to 35.6%, A value increases to -1.9%, and B value decreases to -22.0%. When the PVA mass is increased to 0.4 g, the brightness increases significantly to 44.5%, A value decreases to -1.5%, and B value decreases slightly to -22.5%. Further addition of PVA up to 0.6 g results in a slight decrease in brightness to 41.2%, A value decreases to -1.2%, and B value decreases to -23.1%. At 0.8 g of PVA, brightness drastically increases to 47.7%, but A value decreases significantly to -4.8%, and B value decreases slightly to -23.5%. At a PVA mass of 1.0 g, the brightness value remains stable at 47.7%, but A value returns to -2.3% and B value decreases sharply to -30.5%. The last addition of PVA at 1.2 g produces the highest brightness value at 48.6%, A value approaches 0 at -0.5%, and B value decreases to -35.5%.

Table II. Color measurement results with a color meter

| PVA mass (gr) | L (%) | A (%) | B (%) |
|---|---|---|---|
| 0.0 | 40.1 | -2.3 | -20.4 |
| 0.2 | 35.6 | -1.9 | -22.0 |
| 0.4 | 44.5 | -1.5 | -22.5 |
| 0.6 | 41.2 | -1.2 | -23.1 |
| 0.8 | 47.7 | -4.8 | -23.5 |
| 1.0 | 47.7 | -2.3 | -30.5 |
| 1.2 | 48.6 | -0.5 | -35.5 |
| Commercial ink | 53.3 | -0.5 | -48.5 |

For comparison, commercial ink has a brightness value (L) of 53.3%, A value of -0.5%, and B value of -48.5%. From this data, it can be seen that the addition of 1.2 g of PVA produces the same A value as commercial ink (-0.5%). Although the brightness value (L) and B value are still different from commercial ink, the addition of 1.2 g of PVA is the closest to the color of commercial ink, especially in terms of the A value that is identical to commercial ink. However, to achieve results that are truly close, further optimization is needed, especially for brightness and B value parameters.

Based on the conversion results of LAB color coordinates in Table III using Aspose Color Names, it can be seen that the colors produced by adding PVA mass vary. At PVA masses of 0.0 g and 0.2 g, the produced color is "DarkSlateBlue" with similarity levels of 0.9932 and 0.9941 respectively. With the addition of PVA up to 0.4 g, the color changes to "SteelBlue" with a similarity of 0.996, and remains at this color up to 1.2 g, with an increasingly higher similarity level reaching 0.9998. Meanwhile, commercial ink shows the color "DodgerBlue" with a similarity of 0.9988. This change indicates that the addition of PVA mass shifts the color from the darker "DarkSlateBlue" to the brighter "SteelBlue," but still within the blue spectrum, and still does not reach the bright blue of the commercial ink "DodgerBlue."

Table III. Colourmeter conversion table

| Mass PVA (gr) | Colour name | Similarity | Colors |
|---|---|---|---|
| 0.0 | DarkSlateBlue | 0.9932 | |
| 0.2 | DarkSlateBlue | 0.9941 | |

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| | | | |
|---|---|---|---|
| 0.4 | SteelBlue | 0.996 | |
| 0.6 | SteelBlue | 0.9937 | |
| 0.8 | SteelBlue | 0.9978 | |
| 1.0 | SteelBlue | 0.9982 | |
| 1.2 | SteelBlue | 0.9986 | |
| Commercial ink | DodgerBlue | 0.9988 | |

**Printing Test**

To test the performance and quality of butterfly pea ink as an environmentally friendly ink alternative, a printing test was conducted using a printer. This test aims to evaluate the compatibility of butterfly pea ink with printing technology and observe the characteristics of the resulting prints.

Based on the density measurements and analysis using a color meter that has been conducted, the addition of 1.2 grams of PVA shows that the color produced is close to commercial ink. Therefore, the addition of 1.2 grams of PVA was chosen as the formula that is closest to commercial ink, and only this formula was subjected to printing tests for further evaluation.

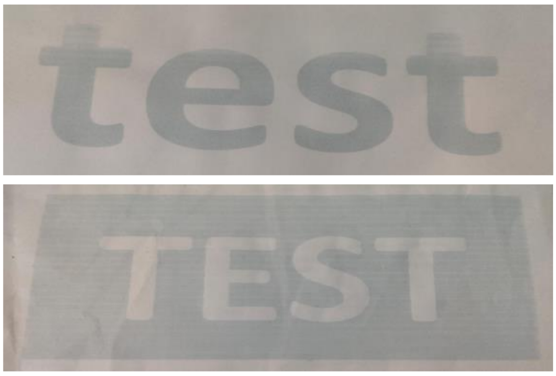


Figure 6. Printer output of butterfly pea ink

The type of printer used in this test was a Canon iP2770 inkjet printer. The printer was cleaned beforehand to avoid contamination with commercial ink. The printer cartridges were filled with prepared butterfly pea ink. The print results were then observed in terms of line sharpness, color evenness, and ink penetration into the substrate. The ink was tested using a standard inkjet printer. The print test results of butterfly pea ink using the printer can be seen in Figure 6.

Figure 6 shows the print results of butterfly pea ink using a printer, where the resulting color appears faded. This is caused by the relatively large size of the ink particles, which is 0.1 micrometers. The large particle size results in uneven ink distribution on the paper surface, leading to prints that lack sharpness and optimal color brightness. To overcome this issue, a reduction in the ink particle size is needed to achieve more even distribution, along with modifications to the ink formulation to improve viscosity and spreading properties.

Testing butterfly pea ink with the addition of 1.2 grams of PVA showed promising results despite some technical challenges that need to be addressed. The color produced is close to commercial ink, indicating that the combination of natural pigments from butterfly pea flowers and PVA has great potential as an environmentally friendly ink alternative. Color sharpness and brightness are critical factors in printing, and although the initial print results show somewhat faded colors, this is more due to the relatively large size of the ink particles than the ink composition itself.

The large ink particle size, around 0.1 mm, causes uneven ink distribution on the paper surface. However, this issue is not insurmountable. With the right technology and methods, such as ultrasonication or the use of additional dispersant materials, the ink particle size can be reduced for more even distribution. This process will improve the sharpness and brightness of the print results, making butterfly pea ink more competitive with commercial inks on the market.

Additionally, the ink formula with the addition of 1.2 grams of PVA showed good stability. This stability is important to ensure that the ink does not easily clot or settle during storage, which is a major challenge in using natural pigment-based inks. PVA not only acts as a binder that enhances ink adhesion to paper but also helps maintain the homogeneity of the ink mixture, ensuring consistent print performance.

To optimize the performance of this ink, further research focusing on several aspects is needed. First, methods for reducing the ink particle size need to be explored further. Second, testing on various types of printers and substrates needs to be conducted to ensure compatibility and consistent print quality. Third, economic and environmental analyses of the production and use of butterfly pea ink need to be carried out to ensure that the ink is not only sustainable but also economical and practical for widespread use.

With the existing potential and identified improvement steps, butterfly pea ink with the addition of 1.2 grams of PVA has a bright prospect as an environmentally friendly printer ink alternative. Investment in further research and development will ensure that this ink can compete with other commercial ink products, providing a greener solution for the printing industry in the future.

## CONCLUSION

In this study, natural pigments from butterfly pea flowers (Clitoria ternatea) have been successfully synthesized into ink formulations using distilled water as a solvent, PVA as a binder, and alcohol as an additive. The resulting ink has a deep blue color derived from the anthocyanin content, particularly delphinidin-3-glucoside. Density test results show that the addition of 1.2 grams of PVA increases the density to a value identical to commercial ink (0.95). Although the viscosity test of the 1.2-gram ink shows a very high value (27.88 cP) compared to commercial ink (1.12 cP), color test results using a color meter indicate that butterfly pea ink with 1.2 grams of PVA has LAB color code that is closest to commercial ink, with an identical A value (-0.5%) while the L (48.6%) and B (-35.5%) values are still below commercial ink (L 53.3%, B 48.5%). The still large ink particle size (0.1 mm) results in uneven ink distribution on the paper surface, leading to prints with less sharpness and suboptimal brightness. Therefore, further research is needed to address the issue of uneven ink distribution by reducing the ink particle size and implementing appropriate improvement measures. The results of this study indicate that butterfly pea ink has great potential as a sustainable environmentally friendly printer ink alternative.

### Suggestion

Ink particle size can affect print quality. In order to obtain ink particles with a small size, ultrasonication or a long ball mill method can be used to improve the dispersion and clarity of the print.

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