THE DEVELOPMENT OF GREEN PRINTING INK FOR INKJET PRINTING

Submitted in accordance with the requirement of the Universiti Teknikal Malaysia Melaka (UTeM) for the Bachelor Degree of Manufacturing Engineering (Hons.)

By

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I hereby, declared this report entitled "The development of green printing ink for inkjet printing" is the result of my own research except as cited in references.

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APPROVAL

This report is submitted to the Faculty of Industrial and Manufacturing Technology and Engineering of Technical University of Malaysia, Malacca as a partial fulfilment of the requirement for Degree of Manufacturing Engineering (Hons). The member of the supervisory committee is as follow:

………………………...

(Profesor Madya Dr. Jariah Binti Mohamad Juoi)

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ABSTRAK

Penggunaan dakwat berasaskan petroleum adalah merosakkan alam sekitar kerana pelepasan besar sebatian organik meruap (VOC). Dakwat berasaskan petroleum adalah bahan yang tidak boleh diperbaharui dan tidak boleh terurai secara semula jadi. Adalah ditemui bahawa dengan menghasilkan dakwat percetakan berdasarkan sumber yang boleh diperbaharui dan boleh terurai secara semula jadi dapat mengatasi masalah ini. Transesterifikasi adalah proses untuk menghasilkan metil ester (ME) untuk percetakan dakwat dengan tindak balas sisa minyak masak (WCO) dan metanol dengan kehadiran KOH sebagai pemangkin. Oleh itu, objektif kajian ini adalah untuk mengkaji kecekapan penukaran tindak balas dan komposisi ME, yang mempengaruhi suhu dan masa tindak balas semasa transesterifikasi metil ester pada nisbah WCO kepada metanol (1:3). Kajian ini membincangkan proses yang dilakukan untuk menilai sifat pengikatan C-H dalam WCO dan ME dan menghasilkan dakwat percetakan hijau (GPI) dengan kelikatan yang lebih rendah yang boleh digunakan dalam percetakan inkjet melalui proses transesterifikasi. Hasil kajian ini menunjukkan bahawa campuran dakwat yang dihasilkan daripada metil ester, karbon hitam, dan cetiltrimetilammonium bromida (CTAB) sebagai surfaktan dalam komposisi yang ditentukan boleh mencapai Dakwat Percetakan Hijau dengan kelikatan 19.00cP sesuai untuk percetakan inkjet dan pengikatan C-H yang lebih rendah seperti yang ditentukan oleh spektrum FTIR. Tindak balas transesterifikasi dengan suhu tindak balas pada 70°C dan masa tindak balas pada 60 minit menghasilkan kelikatan metil ester terendah yang akan digunakan sebagai pengikat. Data yang dihasilkan menunjukkan bahawa sampel menjadi pepejal semasa proses transesterifikasi dengan menggunakan suhu tindak balas dan jangka masa tindak balas yang berubah-ubah, sambil mengekalkan kepekatan pemangkin yang tetap dan nisbah molar metanol kepada WCO pada 3:1. Secara keseluruhannya, kejayaan prestasi percetakan telah dibuktikan dengan keupayaan untuk mencetak pada substrat kertas.

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ABSTRACT

Due to its large emissions of volatile organic compounds (VOCs), petroleum-based ink consumption is detrimental to the environment. Petroleum-based ink is a non-renewable and non-biodegradable material. It is discovered that by producing printing inks based on renewable and biodegradable resources can overcome this issue. Transesterification is the process to produce methyl ester for ink printing with the reaction of waste cooking oil (WCO) and methanol with the presence of KOH as the catalyst. Thus, the objective of this study is to examine the reaction's conversion efficiency and the composition of ME, which influences the reaction temperature and time during the transesterification of methyl ester at a given WCO to methanol ratio (1:3). This study discusses the process that was performed to evaluate the properties of the C-H bonding in WCO and methyl ester (ME) and produce a green printing ink (GPI) with a lower viscosity that can be used in inkjet printing through the transesterification process. As the outcome of this study, the mixture of ink produced from methyl ester, carbon black, and Cetyltrimethylammonium bromide (CTAB) as surfactant in the designated composition can achieve Green Printing Ink with a viscosity 19.00cP appropriate to inkjet printing and a lower C-H bond as determined by FTIR spectra. The transesterification reaction with reaction temperature at 70°C and reaction time at 60 minutes produces the lowest viscosity of methyl ester to become the binder. The resulting data showed that the sample produced during the transesterification process while utilizing a variable reaction temperature and reaction duration, all the while keeping the catalyst concentration constant and the molar ratio of methanol to WCO at 3:1. Overall, the success of the printing performance has been proved by the ability to print an image on paper substrate.

DEDICATION

Only

my beloved mother, Sharifah Annie Addillah binti Syed Mohd Hazam

my beloved father, Zulkarnain bin Abdul Zabil

my lovely younger siblings, Wan Putri Irdina, Meor Zulhaziq, Wan Putri Maisarah, Putri Zahra Asiyah, and Putri Auni Nadirah

my dear best friends, Siti Nurazlinda, Wan Sarafina Shafeqah, Nur Izzah Atirah, Wan Nur Hazirah, Nurul Asyikin, Nurul Syahira and my degree friends.

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UNIVERSITI TEKNIKAL MALAYSIA MELAKA

for giving me moral support, cooperation, encouragement, and understandings during my PSM journey.

I thank you with all my heart.

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TABLE OF CONTENTS

UNIVERSITI TEKNIKAL MALAYSIA MELAKA

UTeM
اونيۏرسيتي تيكنيكل مليسيا ملاك
UNIVERSITI TEKNIKAL MALAYSIA MELAKA

LIST OF FIGURES

UNIVERSITI TEKNIKAL MALAYSIA MELAKA

UTeM
اونيورسيتي تيكنيكل مليسيا ملاك
UNIVERSITI TEKNIKAL MALAYSIA MELAKA

LIST OF TABLES

UNIVERSITI TEKNIKAL MALAYSIA MELAKA

LIST OF ABREVIATIONS

CIJ	-	Continuous Inkjet
DOD	-	Drop on Demand
IOT	-	Internet of Things
FTIR	-	Fourier Transform Infrared Spectroscopy
GPI	-	Green Printing Ink
ME	-	Methyl Ester
PSA	-	Particle Size Analysis
UV	-	Ultraviolet
VO	-	Vegetable Oil
VOC	-	Volatile Organic Compound
WCO	-	Waste Cooking Oil

UNIVERSITI TEKNIKAL MALAYSIA MELAKA

LIST OF SYMBOLS

cP	-	CentiPoise
mN/m	-	Millinewtons per metre
°C	-	Degree Celcius
%	-	Percent
mPa.s	-	Millipascal second
g/cm^3	-	Grams per centimetre cube
wt%	-	Weight percent
g/mol	-	Grams per mol

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CHAPTER 1

INTRODUCTION

The study's background, problem statement, objectives, and scope are all present in the first chapter. The study's background goes into detail regarding how reaction temperature and reaction time affect the transesterification of waste cooking oil (WCO) and methanol at a fixed 3:1 molar ratio. The challenges encountered by the earlier study, which served as the basis for the research applied to the project, are outlined in the problem statement. The primary aim of this research is represented by the objectives, and the study's scope reveals its restrictions and area of concentration.

UNIVERSITI TEKNIKAL MALAYSIA MELAKA

1.1 Background of Study

The increasing demand for environmentally friendly inks because of resource limitations, regulatory requirements, and public concern about the environment is pushing ink producers to create more eco-friendly products. Most manufacturers of printing ink have shifted in recent years to produce more environmentally friendly inks. However, there is an increasing demand for both innovative and traditional printing inks as a result of the printing ink market's 3% annual growth (Robert, 2015). The substrate and printing technique both affect the makeup of printing inks. There are various types of printing inks that were created according to its specific usage such as water-based inks, oil-based inks, eco-solvent inks, digital inks etc. Additionally, the manufacturing of printing ink uses a variety of raw materials, with synthetics taking the place of natural ones. On the other hand, rising worries about the environment and health have pushed up demand for natural products. Because of problems with recycling and volatile chemicals, synthetic mineral oil is being used in decreasing quantities.

In general, inks are coloured writing or printing materials that are often liquid. It serves a variety of functions, most commonly as an instrument of instantaneous adornment and message delivery. Regardless of the texture, size, or shape of the surface, inks are incredibly adaptable and may be applied to a broad range of surfaces. Most materials are compatible with inks. Specialised liquids or mixes known as printing inks were developed to be used in a variety of printing processes in order to transfer text or images onto substrates like paper, cardboard, cloth, and other materials.

Printing inks are composed of up of the following ingredients: a binder (resin), a colourant (pigment or dyestuff), and a carrier media such as water, solvent, or oil (Agarwal et al., 2021). The type of printing and the print substrate influence the ink's composition. Ink has various uses for the vehicles, also known as binders. After binding the support material to the printing sheet of paper, they distribute the pigments through layers. The rheological and mechanical characteristics of the inks are also altered by it. Other synthetic resins, such as polyacrylates, can be used in place of the conventional phenolic resins made from tall oil rosin (colophony) or alkyd resins (Ha et al., 2012).

A dye or pigment can be used as the colourant. In printing processes, pigments, both organic and inorganic, are practically always utilised (Gürses et al., 2021). They constitute dispersed solids that the solvent (support material) can't dissolve. In comparison to pigments, dyes have a number of disadvantages, including reduced light fastness and water resistance. Chemicals are also soluble in the supporting material. Solvents dissolve the binders and modify the ink's viscosity to suit various printing techniques. They can evaporate during the printing process and have high or low vapour pressures, depending on the application. A solvent-free ink (100% system) may result from the printing process and the viscosity of the binder not requiring any additional solvent (Robert, 2015). In printing inks, the carrier media is the liquid or solvent that contains the colourants (dyes or pigments) and other ingredients, enabling the ink to be applied to a substrate while printing.

Recently, ink manufacturers are shifting towards organic oily inks because of the danger produced by these gas-emitting compounds. Volatile organic compounds, or VOCs, are hazardous organic substances that are used in ink and dyes to prevent solvent emissions (Aydemir & Özsoy, 2020). These chemicals are subject to regulations and standards today. Volatile organic compounds (VOCs) have high vapour pressures and expand and evaporate at room temperature. Since harmful organic chemicals have a greenhouse effect, their consumption is typically linked to global warming. Therefore, the major alternatives to encounter this problem is by using a bio-renewable raw materials such as vegetable oil-based products. Vegetable oils yield less oil in their inks due to their higher evaporation point, which reduces the emissions of hazardous organic compounds. Additionally, the study discovered that the ink is more resilient than its mineral-based counterpart, which offers considerably higher picture quality and dries more quickly.

UNIVERSITI TEKNIKAL MALAYSIA MELAKA

1.2 Problem Statement

The production of inks for coating, printing, and writing has increased popular, which has resulted in an increase in volatile organic compounds (VOCs) released into the environment, which in turn has caused a variety of pollutions. Numerous studies have been undertaken to develop new formulas for producing these inks with less volatile organic compounds (VOCs) in order to reduce the number of harmful compounds released. These studies have progressed beyond petroleum depending upon on the use of vegetable oils (VOs) in the ink. Waste cooking oils (WCOs) were introduced as alternatives to the vegetable oil (VO) in the alternative formula to make environmentally friendly printing ink. Thus, it is possible to reduce the instances of pollution brought on by WCOs and VOCs. Cooking oils that are left over after any type of cooking are known as waste cooking oils, or WCO.

As a result of the increased WCO content and its delayed degradation, waste management issues have arisen that pose a risk to the environment. WCO recycling into biodiesel, lubricant, and soaps is currently the subject of the majority of study.

Regarding to the previous study of using waste cooking oil (WCO) as the binders for green printing ink where Najwa (2017), Sisubalan (2018), Rawdhah (2019) and Hazmi (2020) had experimented with the method, which was modified from conventional printing to silk printing and finally to digital printing, demonstrates a significant improvement. However, various requirements apply to each type of printing method. Qualities necessary for the ink to function. Pigments, binders, solvents, and (if necessary) additives are the main ingredients of an ink. For digital printing applications, a green printing ink derived from the low viscosity ester produced should be achievable.

According to Hazmi (2020) previous study, he had studied on the effect of the temperature and duration on the transesterification process using fixed WCO to methanol ratio. In this transesterification process, WCO and methanol reaction produces methyl ester (ME). The methyl ester is used as a low viscosity binder. The methyl ester's viscosity is influenced by the molar ratio of methanol to waste cooking oil, with the lowest viscosity observed during the lowest molar ratio transesterification reaction. As a result, methyl ester formed from a molar ratio of 3:1 has a 2.67 cP viscosity and is used as an ink binder. Brookfield Viscometer was used to test the ME obtained during the transesterification. However, for this research is to reduce the reaction time and the increase the reaction temperature in while maintaining the molar ratio of methanol to WCO 3:1.

The catalyst used from the previous study was potassium hydroxide (KOH). The liquid solution of KOH is the alternative as a catalyst for the transesterification process. Alkali-catalyst like potassium hydroxide take less time to achieve the final conversion at less temperatures.

During the previous study that was run by Hazmi (2020), the GPI produced from the effect of catalyst form in the transesterification process of methyl ester was tested for printing by using the Canon Pixma 258 inkjet printer. As the result, the printing test demonstrates the printability of the formulated green printing ink with on a paper substrate. But as for this research, the characterization of the GPI deposited were tested on paper substrates for inkjet printing.

UNIVERSITI TEKNIKAL MALAYSIA MELAKA

1.3 Objectives

The objectives of this research are:

i. To analyse the effect of reaction temperature and reaction time of the transesterification of methyl ester with ratio of fixed methanol and WCO (3:1) based on the yield and the composition of the methyl ester produced.

ii. To characterize the green printing ink formulation through viscosity test and FTIR spectroscopy.

iii. To examine the printing performance of green printing ink on paper substrate using inkjet printing and the FESEM and EDX analysis of the printed image.

1.4 Scope of research

The scope of the research are as follows:

i. Research on the affect of the duration during heating and heating temperature that influence the ME produced from the transesterification process of WCO and methanol. This research was concentrated on analysing the application of the transesterification process to produce printing ink with a reduced viscosity.

ii. Waste cooking oil (WCO) is the renewable resource that is used in the composition of printing ink to replace petroleum-based ink as a green value.

iii. Applying an inkjet printer as an inkjet printing method, the final printing ink formulation was tested. Printing occurs in performed on a paper substrate using the green printing ink (GPI).

UNIVERSITI TEKNIKAL MALAYSIA MELAKA

1.5 Significance of the study

In the present research, waste cooking oil—a non-petroleum-based material—is used to formulate green printing ink for inkjet printing. This ink has a lower viscosity requirement than inks used in conventional printing processes. Since inkjet printing is the most widely used form of printing, the composition of this printing ink might be one of the ways that can decrease the difficulties of performing waste cooking oil and contribute to reducing the amount of volatile organic compounds (VOCs) in the environment. While there has been a lot of research done on the application of biodiesel made from leftover cooking oil, this study may serve as a springboard for additional research into environmentally acceptable printing ink alternatives to vegetable oil-based ink, since the worldwide printing business is expanding quickly.

1.6 Organization of the report

The theoretical overview, problem description, goals, significance, and usefulness of the analysis are covered in Chapter 1. This gives users a deeper understanding of the analysis, especially in conjunction with the article's discussion of green printing ink. In Chapter 2, the literature review is summarised. As a result, it incorporates earlier research and work on the composition of green tin, which can replace conventional tin, as well as variables influencing the needs for printing ink. While the transesterification of cooking oil, methanol, and raw materials is covered in Chapter 3. After that, Chapter 4 is going through the findings and a discussion of the transesterification experiment. The results and conclusions that can be drawn from the experiment is cover in detail in Chapter 5.

UNIVERSITI TEKNIKAL MALAYSIA MELAKA

1.7 Summary

The goal of this research is the production of green printing ink on a ceramic substrate using leftover cooking oil, which Najwa (2017) and Sisubalan (2018) have done successfully and that has a wider use than screen printing. Next, Rawdhah (2019) proceeded, giving her the assignment to concentrate on depositing ink on the printer she had successfully acquired. As for Hazmie (2020) had achieve the low viscosity of the ME formed from the transesterification process. But since the production is still insufficient, the efficiency of producing GPI from WCO still need to by study in terms of its reaction temperature and reaction time. Thus, this effort was done to improve the green printing ink made from waste cooking oil.

UNIVERSITI TEKNIKAL MALAYSIA MELAKA

CHAPTER 2

LITERATURE REVIEW

2.1 Printing

The printing press, invented by German Johannes Gutenberg in 1440, revolutionized the printing process by combining existing technologies and his own inventions. Gutenberg's hand mould enabled precise and rapid creation of metal movable type, boosting profitability. The press spread to over 200 cities in Europe, producing over 20 million volumes by 1500. It became synonymous with the printing enterprise, forming a new branch of media (McLean, n.d.). According to Cartwright (2024), Gutenberg's printing of the Latin Bible in 1456 CE demonstrated the successful combination of metal blocks, paper, and ink, resulting in a visually appealing and mechanically pressed text. This process was a significant advancement in printing.

The development of offset printing techniques in the late 19th and early 20th centuries marked a significant advancement in printing technology, introducing a more indirect method of ink transfer. Offset printing, also known as lithography, is a process that involves transferring an image to a printing plate coated with a photosensitive emulsion. The plate is exposed to light, developed, mounted on a press, and then transferred to a rubber blanket cylinder. The inked image is then transferred to the paper, producing the printed material.

The printing press significantly increased reading materials availability, ranging from informative pamphlets to romantic novels, despite being less affordable than handmade books, thereby increasing literacy rates and facilitating writing production. As the world move into the digital age, the printing industry continues to innovate, making knowledge accessible to the society.

2.1.1 Main components of printing ink

Considering the ingredients of ink is essential to creating substitutes for conventional inks. Colourant, solvent, vehicle or binder, and additives are the four components that make up all printing ink. Paints and dyes are two types of colourants; pigments are insoluble in the medium that are applied on. Pigment distribution and property modification are done through vehicles or binders. Ink viscosity can be adjusted for various printing processes and binders can be dissolved using solvents. Several additives, like biocides, driers, plasticizers, adhesion promoters, and surfactants, enhance the qualities of ink. By lowering surface tension and wetting issues, these substances aid in enhancing the ink's qualities (Robert, 2014). According to research by Shoaib Ahmad Bilal (2016), paste inks are used in letterpress and lithography, while liquid inks are utilised in printing and flexo. Paste and liquid inks are examples of mild monitor inks.

UNIVERSITI TEKNIKAL MALAYSIA MELAKA

2.1.1.1 Binders

The function of the binder has been studied mainly in the coating layers. Vehicles or binders provide a variety of objectives in the ink industry. They disperse pigments in the support material, bind it to the printing stock, and adapt the rheological and mechanical properties of the inks. Conventionally, phenolic resins derived from tall oil rosin (colophony) or alkyd resins are applied, but alternative synthetic resins, such as polyacrylates, can also be utilised (Robert, 2014).

For printing ink binders, tall oil rosin is extracted from weathered wood or pine trees. It is made up of resin acids, specifically abietic acid, which can be changed to provide different qualities. Polyesters treated with phenol formaldehyde are commonly utilised as rosin binders. Most rosin binders, however, partly renewable, nonetheless use fossil fuels as their primary energy source. New alternatives are required as a result of the unstable market and rising demand placing pressure on printing ink manufacturers. The depletion of petroleum supplies has resulted in a significant increase in the cost of petroleum-based binders, compelling road authorities to investigate alternate materials. As of right now, the widespread practice is heading in the direction of a cost-effective, environmentally friendly, and sustainable environment.

During frying, edible oil experiences physical and chemical changes that cause deterioration and quality loss. When food, air, and water combine to form steam, hydrolysis begins, and fatty acids (FFA) are produced. The heating temperature, moisture content, and storage duration are some of the variables that affect the concentration of FFA. As a result of the accelerated degradation process, the oil becomes less saturated and unfit for further frying. The acid value parameter can be used to track the quality of vegetable oil (WCO) (Elahi et al., 2021). Figure 2.1 shows the chemical composition of WCO.

UNIVERSITI TEKNIKAL MALAYSIA MELAKA

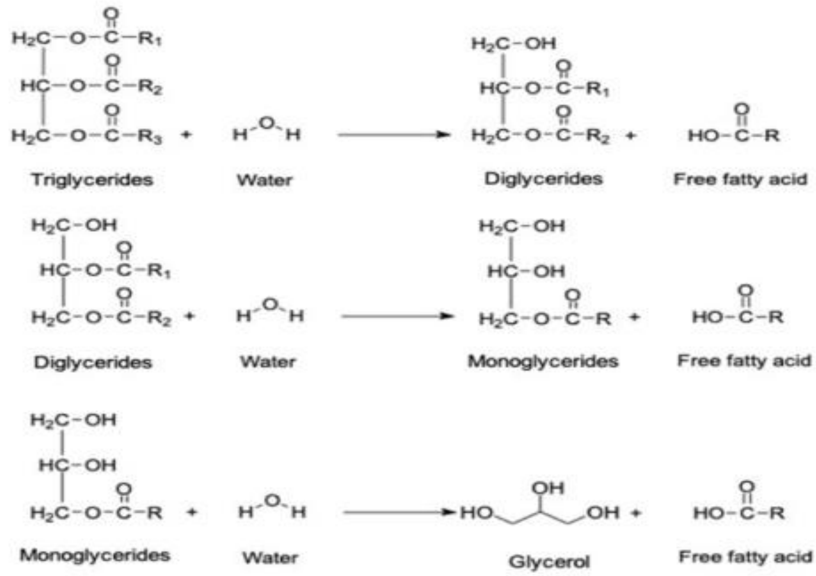

Figure 2.1: Chemical composition of WCO (Elahi et al., 2021).

In accordance with Wyatt (2008), water-based inks require two different kinds of binder. The first is the solution binder, which is made of short-chain polymers that improve the pigment particles' capacity to withstand water; the second is the emulsion binder, which is made of long-chain polymers that are unsuitable for strong mixing. They exhibit excellent drying qualities, and the emulsion binder is not absorbed by the pigments. As seen in Table 2.1, both polymers are copolymerized from distinct monomers to achieve the appropriate final characteristics.

UNIVERSITI TEKNIKAL MALAYSIA MELAKA

Table 2.1: Building Blocks of Acrylic Resins for Water-Based Inks

Contributing Monomer	**Final Property**
Methyl methacrylate	Water resistance, block resistance, hardness, gloss retention, fast dry speed
Styrene	Water, block resistance, hardness, initial high gloss, poor gloss retention, fast dry speed
Short-chain acrylates and methacrylates (R < 8)	Flexibility, stain, rub resistance, adhesion
Acrylic and methacrylic acid	Adhesion, resolubility, hardness, solvent, and grease resistance
Long-chain acrylates and methacrylates (R > 10)	Water resistance, flexibility, adhesion

2.1.1.2 Solvents

With the objective of modifying the ink's viscosity for the various printing techniques, solvents dissolve the binders. Their vapor pressure might vary, depending on the use, and they can evaporate throughout the process of printing. There may be no requirement for an additional solvent in a solvent-free ink (100% system), depending on the printing procedure and the viscosity of the binder. Triglyceride has a low level of polarity, whereas the alcohol used for producing biodiesel has a high level, hence it is preferable to utilize a substance with medium polarity like removal tools (Mohadesi et al., 2020). The quality and amount of the solvent have an impact on the rheological characteristics and viscosity of the ink that is produced, which is dependent on the printing method and substrate type. This ought to be another point to consider when choosing a solvent. Table 2.2 shows the components of solvents in safety data sheets in the printing industries.

Table 2.2: Components of solvents in safety data sheets in the printing industries (Tsai et al., 2015).

Pre-press chemicals (n = 22)				
n-Hexane (13.6%)	*n*-Heptane (13.6%)	Toluene (13.6%)	Isopropanol (13.6%)	Acetic acid (13.6%)
Ethylene glycol (9.0%)	Xylene (9.0%)	Sodium hydroxide (9.0%)	Diethylene glycol (9.0%)	Acetone (4.5%)
Methanol (4.5%)	Ethanol (4.5%)	Sorbitol (4.5%)	Ferric chloride (4.5%)	Butyl acetate (4.5%)
Nitric acid (4.5%)	Boric acid (4.5%)	Citric acid (4.5%)	Potassium carbonate (4.5%)	
Printing inks (n = 72)				
Soybean oil (84.7%)	Polybutylene terephthalate resin (84.7%)	Organic pigment (84.7%)	Mineral oil (84.7%)	Auxiliaries (84.7%)
Toluene (6.9%)	Xylene (5.6%)	Isopropanol (5.6%)	*n*-Heptane (4.2%)	Cyclohexanone (4.2%)
2-Butanone (2.8%)	Ethyl acetate (2.8%)	1-Octane (1.4%)	Phthalate esters (1.4%)	Butyl acetate (1.4%)
Fountain solutions (n = 9)				
Glycerol (66.7%)	Ethylene glycol ethers (11.1%)	Toluene (11.1%)	Gum Arabic (11.1%)	
Cleaning solvents (n = 56)				
Kerosene (41.1%)	Toluene (23.2%)	*n*-Heptane (16.1%)	Isopropanol (16.1%)	92 Gasoline (12.5%)
n-Hexane (10.7%)	1-Octane (3.6%)	Xylene (3.6%)	Cleaning naphtha (3.6%)	Methanol (1.8%)
Ethanol (1.8%)	Propanol (1.8%)	Ethylene glycol (1.8%)	Washing water (1.8%)	Butyl cellosolve (1.8%)
Sodium carbonate (1.8%)	Isoamyl acetate (1.8%)	Dichloromethane (1.8%)		
Adhesives and glues (n = 13)				
Polyvinyl acetate (100.0%)				
Dilution solvents (n = 8)				
Toluene (37.5%)	2-Butanone (37.5%)	*n*-Heptane (25.0%)	Cyclohexanone (25.0%)	Methyl isobutyl ketone (12.5%)
1-Octane (12.5%)	Xylene (12.5%)			

A good solvent selection requires that the solvent could dissolve polymers, not dissolve pigments, have an evaporation rate that is appropriate for the printing process, be well-compatible with image carriers, and be able to adjust viscosity, according to Pekarovicova A. and Husovska V. (2015). The desired amount of ink. The drying qualities and surface tension of ink can also be modified by solvents.

2.1.1.3 Colorants

Due to the wide variety of printing techniques and, by extension, the wide range of specialized printing formulations, colorants must meet expanding requirements in printing applications (Rathschlag, 2021). Acid dyes, direct dyes, reactive dyes, solvent-soluble dyes, and pigments are among the colorants taken into consideration. These dyes are either spread out or dissolved in the ink and printed using an inkjet printer onto the substrate (Lavery & Provost, 1999). The classification of colorants based on Figure 2.2 is defined. The degree of interaction between the colorant and the media is mostly determined by its chemical makeup. The ink's color will be determined by the pigment or dye contained within it. Whereas dyes are widely applied, it was subsequently found that they have poorer durability and dissolve. This is because pigments are insoluble whereas dyes are soluble in binders.

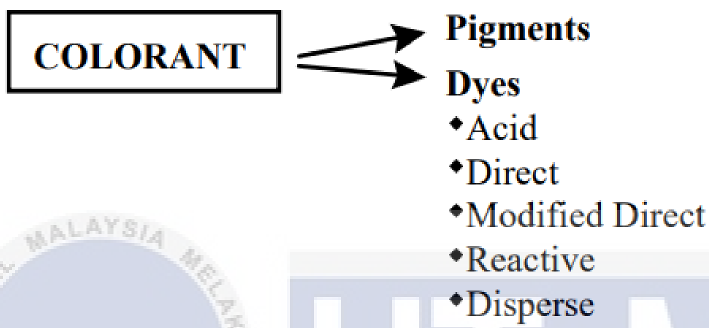

Figure 2.2: Colorants used in inkjet printing (Lavery & Provost, 1999).

Organometallic compounds are contained in the ink due to its optimized system, which includes non-toxic solvents, low viscosity, moderate surface tension, and a high color coverage capacity. When combined with ink technology and soluble heavy metal complexes, ceramic fire produces an extremely stable ink free of solid particles (Joglekar-Athavale & Shankarling, 2021).

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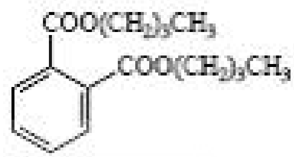
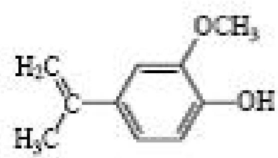
2.1.1.4 Additives

For the purpose of enhancing the ink's qualities, additives are applied. In order to reduce surface tension and the consequent wetting issue, these compounds contain surfactants. Tenderize the dried ink with plasticizers, and aid in the oxidative drying process with dryers. To point out a few, biocides stop the ink from being broken down by microorganisms and adhesion promoters or wetting agents improve the binding to the printing paper. In a more contemporary example, starch was added to easily accessible inks in higher amounts, which increased their gloss, heat resistance, water resistance, and color strength (Robert, 2015). Table 2.3 shows the common classes of printing ink additives.

Table 2.3: Common classes of printing ink additives (Wansbrough H. et al., 2007).

Type	Function	Typical example
Plasticiser	Enhances the flexibility of the printed film	 dibutyl phthalate
Wax	Promotes rub resistance	Carnauba - an exudate from the leaves of *Copernicia prunifera*. Consists of esters of hydroxylated unsaturated fatty acids with at least twelve carbon atoms in the acid chain.
Drier	Catalyses the oxidation reaction of inks which dry by oxidation	salts or soaps of cobalt, manganese or zirconium
Chelating agent	Increases the viscosity of the ink (aluminium chelate) and promotes adhesion (titanium chelate)	
Antioxidant	Delays the onset of oxidation polymerisation by reacting with free radicals formed during the autooxidation thus preventing them from reacting further	 eugenol
Surfactants[89]	Improves wetting of either the pigment or the substrate	
Alkali	Controls the viscosity / solubility of acrylic resins in water based inks	$HOCH_2CH_2NH_2$ monoethanolamine
Defoamer	Reduces the surface tension in water based inks, meaning that stable bubbles cannot exist	hydrocarbon emulsions

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2.2 Ink Properties

The vast majority of the ink's features were comprised of the following fundamental properties which are its optical qualities, its structural qualities, and its drying characteristics. Depending on the printing process and the specific requirements of the application, ink characteristics for printing varies. Chung et al. (2013) stated that one of the most essential stages in the bioprinting process is ink formulation. During and after printing, an appropriate "bio-ink" must satisfy a variety of rheological, mechanical, and biological specifications. Researchers used a variety of analytical approaches to investigate the printability, mechanical qualities, and cell survival features of alginate-based hydrogel scaffolds. The physicochemical qualities of ink are determined by the structural characteristics of the dyes, which consequently impact the quality of the inkjet print results (Li et al., 2023).

Water carries the ink particles away from the original region and is absorbed by the fibers as it travels along them. Any water that is not completely absorbed continues to flow down the fibers until it is absorbed. Therefore, the number of diffusion steps or the span of the diffusion image depends on the amount of water rather than the ink's density. The mobility of suspended ink particles is influenced by the motion of water molecules and is referred to as Brownian motion. An irreversible diffusion process occurs when two differing densities of ink merge together, causing ink particles to move from the higher-density ink to the lower-density ink. Therefore, the direction of diffusion will rely on the density of the liquid ink surrounding a given spot (J. Lee, 2001).

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2.2.1 Optical Properties

The qualities of pigments used will affect the optical characteristics of ink, including its colour and opacity. The degree of ink-created ray-refraction (transmission) is implied by transparency (Asinyo, 2019).

In printing, transparency refers to a uniform coated ink film that causes background objects to appear at the rate that corresponds to their natural color. The background is partially hidden when the ink's transparency is low; this phenomenon is also referred to as the ink's opacity because the background is partially covered. Interaction between obscurity and openness that is inversely proportionate. The thickness of the ink when all its surface is covered in ink determines the degree of transparency. It becomes less invisible and more translucent with increasing weight.

2.2.2 Viscosity of Ink

A crucial physical and chemical characteristic of inkjet printing ink is its viscosity, which is dictated by the orientation, dispersion, and induction forces among the constituent molecules (Gao et al., 2021). The temperature is the most crucial factor; nevertheless, it is inadequate, and it also affects the ink's adhesion to the substrate. The consistency of the ink during the printing process depends on higher interior temperatures. Applying the same connection to the lower temperature, viscosity would develop where it would be excessively large and result in a "split row," which is susceptible to high-speed printing.

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2.2.3 Drying characteristics

Hybrid inks combine two distinct drying characteristics into one: it reacts to UV light like UV ink but additionally dries by oxidation and substrate penetration like conventional inks. The degree of emulsification and the thickness of the ink and varnish determine the printing quality of hybrid inks that went through UV varnishing and drying (Savchenko & Velychko, 2013). Penetration has a major role in determining ink drying, and penetration curves by itself may accurately forecast the amount of time it will take for ink to dry or disappear on a plain paper surface. This is demonstrated in practice by the quality of the inkjet prints on plain sheets with different size treatments stated by Selim et al. (1997).

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2.3 Green Printing Ink

An environmentally friendly ink composition that is intended to reduce its ecological impact throughout the course of its lifecycle is referred to as green printing ink. The use of renewable and sustainable materials, lower concentrations of volatile organic compounds (VOCs), and an emphasis on environmentally friendly production techniques are among the primary features of green printing inks. The production process of printed publications involved researching into the requirement for environmentally friendly printing paper and printer ink, which are the main sources of greenhouse gas emissions from all types of printing materials. Besides environmental concerns included the operation of print shops and various machinery, the recycling process, and other associated aspects. Due to these factors, these workers in the industrial field understand the importance of environmental protection and conservation (Lee, 2012).

2.3.1 Volatile Organic Compound (VOC)

Volatile organic compounds (VOCs) emitted into the atmosphere in gas form from solids and liquids (Khan & Ghoshal, 2000). Because of its direct and secondary impacts on the environment, vapor phase organic compounds are crucial for the reduction of air pollution. As indoor and outdoor air pollutants, volatile organic compounds (VOCs) are a cause for concern. However, the emphasis of that distributing outdoors is different than indoors. The main sources of volatile organic compounds (VOCs) include oil-based heat-set web offset inks, gravure, digital and screen-printing inks, solvent compounds added for dilution, and surface coating printing inks based on organic solvents. Every stage in the ink mixing, printing, surface coating, and storage processes could result in emissions (Hettige et al., 2001). Classification of organic pollutants are illustrated in the Table 2.4.

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Table 2.4: Classification of Organic Pollutants (Okubo & Kuwahara, 2020).

Chemicals group	Boiling point (°C)	Organic chemicals
VVOCs	<0 to 50-100	Propane, methane, formaldehyde, acetaldehyde, dichloromethane, butane, methyl chloride, etc.
VOCs	50-100 to 240-260	Ethyl alcohol, acetone, benzene, toluene, xylene, isopropyl alcohol, hexanal, etc.
SVOCs	240-260 to 380-400	Chlorpyrifos, dibutyl phthalate, bis (2-ethylhexyl) phthalate, pesticides (DDT, chlordane, plasticizers), etc.

2.3.2 Green Printing Substrate

The basic building block of any printing process is ink and green printing substrates. Equally crucial to maintaining this process green are the substrates used for green printing. The broad spectrum of substrates that printing processes may utilize, including both rigid and flexible substrate. Inks frequently adhere to the substrates and attach to it, providing no exposed spaces within the printed patterns (Batet et al., 2023). Some of the type of substrate for printing inks consist of recycled paper, FSC-certified paper, bamboo paper and cotton paper.

2.3.3 Ink from renewable resources

Water-based ink, radiation-curable ink, and vegetable oil-based ink are just a few of the ink alternatives that researchers have been able to provide since environmental consciousness has become increasing in popularity.

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2.3.4 Water Based Ink

An ink is a semi-liquid substance that resembles a liquid or paste and is used for text and graphic printing, drawing, and writing. Water is the solvent used for producing a colloidal suspension of pigments or dyes in an ink, which is known as a water-based ink. In water-based inks, water serves as the primary solvent, although additional co-solvents can also be utilized. Most VOCs are these co-solvents. Water-based inks are widely used due to their functionality and have become recognized for their soft print quality. For water-based ink, pigments dissolved in water and film formers usually constitute the ingredients (McManus et al., 2017).

2.4 Waste Cooking Oil (WCO)

Vegetable oils relate to a diverse family of chemical substances called fats, or lipids, which are mostly composed of glycerol and fatty acid triesters as shown in Figure 2.3. These compounds can be processed to produce high value oleochemicals for use in a variety of industries. Both saturated and unsaturated fatty acids can be identified in vegetable oils. Vegetable oils include, but are not limited to, castor, cottonseed, linseed, jatropha, rapeseed, and soybean oils. Epoxidized vegetable oils are created through chemical modification of the unsaturation (double bond) found in vegetable oils (Yossif et al., 2017).

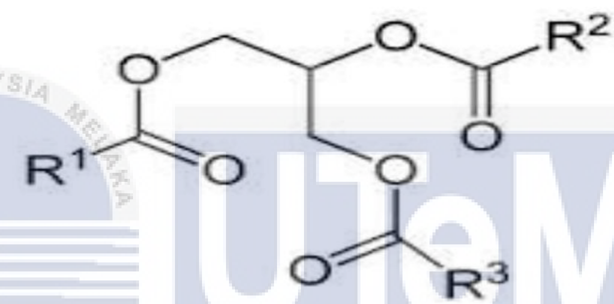

Figure 2.3: Triglyceride chain consists of a glycerol centre and three fatty acids.

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2.4.1 Characteristics and properties of waste cooking oil (WCO)

Biodiesel can be produced from a variety of materials, such as waste oil, animal fats, and edible and non-edible oils. Cooking oil is considered an essential component in the kitchen, therefore waste from the food industry is widely recognized. WCO is collected from residences, eateries, lodging establishments, and food processing facilities following frying and other meal preparation tasks. Nevertheless, the type of catalyst employed, the frying temperature, the water concentration, and the oil's provenance all have a significant impact on the physiochemical characteristics of WCO biodiesel.

Trihydric alcohol glyceride is combined with monocarboxylic acids, including saturated and unsaturated, to produce waste cooking oil. During transesterification, triglycerides are transformed into diglycerides with the aid of a catalyst. Diglycerides are next altered into monoglycerides and then to glycerol. The primary disadvantage of using WCO is the production of impurities from raw food or from chemical reactions that occur during cooking. Some of these elements that mainly affect homogeneous catalytic transesterification are water, free fatty acids (FFA), polar chemicals, and non-68 volatile compounds. The physical characteristics of biodiesel are primarily determined by the length of the hydrocarbon chain, the degree of unsaturation, and the influence of molecular packing (Abdullah et al., 2023). Table 2.5 shows the Physicochemical properties of biodiesel waste cooking oil.

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Table 2.5: Physicochemical properties of biodiesel waste cooking oil.

Type WCO	Author	Kinematic viscosity (mm²/s, 40 °C)	Density (20 °C) kg/m3)	Iodine value g (iodine/100g)	Heating value (MJ/kg)	Oxidation stability (h)	Acid Number (mg KOH/g)	Flash Point (°C)	Pour Point (°C)	Cloud Point (°C)	CFPP (°C)	Cetane Number (CN)
WCO	[58]	4.318	-	-	41.1	0.45	0.49	160	6.1	9.8	7.2	57.95
WCO	[59]	3.69	874	-	44.13	-	0.39	175	-3.4	1.6	-2.54	70.24
WCO	[60]	3.12	876	-	38.431	-	-	133	3	3	-	51.4
WCO	[61]	5.2	879	-	-	-	0.97	96	-	9	-	-
WCO	[62]	4.4	878	-	38.85	-	-	189	-	-	-	51.34
WCO	[56]	4.63	886	-	-	-	0.37	172	-10	-5	-	-
WCO	[63]	4.7	898		36.89	-	-	73	-	-	-	53
WCO	[52]	5	880	-	35	-	-	170	6	19	-	47
WCO	[19]	3.8	871	-	-	-	0.31	124	-	-	-	-
WCO	[5]	3.58	881	128.381	-	-	-	170	-8.369	-1.428	-	44.315
WCO soybean	[64]	-	-	-	-	-	0.52	92	-4	1	-	51.3
WCO	[65]	3.8	880	-	-	-	0.1	120	-2	-5	-	54
WCO sunflower	[66]	5.05	850	70.5	39.5	-	-	90	3	6	-	48
WCO	[67]	4.45	861	-	40.6	-	0.38	154	-	-	-	61.5
WCO	[68]	-	892.6	-	42.83	-	-	176	-	-	-	63.63
WCO	[69]	4	883	-	39.5	-	-	120	-	-	-	52
WCO	[70]	2.2	878	-	38.85	-	-	190	-	-	-	51.34
WCO	[71]	4.6	880	-	39	-	0.5	-	-	-	-	51
WCO	[72]	4.76	870	-	38.30	-	-	151	4	7	-	57
WCO	[73]	5.83	912	-	38.08	-	-	176	-	-	-	51.48
WCO	[74]	4.54	879	36.80	-	-	0.41	-	2	12.17	-	-
WCO	[75]	4.77	886	-	44.9	-	0.41	181	-4	2	2	-
WCO	[76]	4.71	886	-	-	-	0.53	169	-11	23	-	-
WCO	[77]	5.1	883	-	39.5	-	-	120	-	-	-	55
WCO	[78]	6	880	33	41.39	-	0.8	140	-3	-1	-	56.67
WCO	[79]	4.45	885	-	39.05	-	-	-	-	-	-	51
WCO	[80]	3.1	931	-	37.3	-	-	-	-	-	-	63.5
WCO	[81]	4.18	865	-	39.48	-	0.302	202	-4.5	1	-	-
WCO	[82]	4.75	889	-	39.48	-	-	120	-	-	-5	51.62
WCO	[83]	-	870	-	-	-	0.175	90	-3	-	-	-
WCO	[66]	5.05	850	70.5	39.50	-	-	90	3	6	-	48
WCO	[84]	4	867.3	-	-	8.2	0.32	194	-	-	-	-
WCO	[85]	4.915	884	146.44	37.114	3.45	0.48	178	-6	0	-2	49.15
WCO	[86]	4.3	871	66.52	37.4	-	0.92	133	2	6	-	46
WCO	[87]	3.23	874	-	25.81	-	-	150	-	-	-	-
WCO	[88]	4.55	851	-	40.86	-	0.55	152.5	12	-2	-	62.5
WCO	[54]	4.72	878	-	-	2.5	0.32	170	-	-	6.5	53.5

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2.5 Ink Process for digital printing

It is crucial to understand the colors and pigments that are the most crucial components of ink creation before beginning the process of making ink. More often than otherwise, pigments are employed since they are lighter than dyes, however this comes at the price of less color coverage, less color accuracy, and higher cost. Several technologies are included in digital printing, including electrophotography and inkjet. Inkjet printing uses thermal or piezoelectric printheads to drive microscopic droplets of ink which are made of water, colorants (pigments or dyes), and additives onto the substrate (Andersen, E. 2008).

2.5.1 Biodiesel

Fatty acid esters derived from vegetable or animal fat are exactly biodiesel representations. Generally, a chemical procedure called esterification reaction with an alcohol like methanol is used in the synthesis of biodiesel from vegetable oil (Komasatitaya & Taechutrakul, 2020). The formulation of inks for the additive printing of cylindrical lattices to function as heterogeneous catalysts for the synthesis of biodiesel effectively employed sodium, potassium, and mixed alkali-based geopolymers (Botti et al., 2021).

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The renewable and eco-friendly biodiesel cannot be directly employed in printing ink compositions, while being a great replacement for conventional fossil fuels. Through the process of transesterification, biodiesel is produced from renewable resources like vegetables or animal fats and is widely used as a cleaner-burning alternative to diesel fuel in industrial and transportation settings. Biodiesel is renewable and biodegradable because it is derived only from vegetable or animal fats. Additionally, sulphur, metals, and polycyclic aromatic hydrocarbons are absent from biodiesel. Diesel fuels sourced from petroleum may include up to 20% polycyclic aromatic hydrocarbons. Polycyclic aromatic hydrocarbons are up to three orders of magnitude more soluble in water than straight chain aliphatics for an equivalent number of carbon atoms. Because biodiesel does not contain polycyclic aromatic hydrocarbons, it is a safe substitute for conventional fuels for both storage and transportation (Vasudevan & Briggs, 2008).

Nevertheless, it is less typical to incorporate biodiesel or additives generated from biodiesel into printing ink formulas. Because biodiesel has a closed carbon cycle, it is renewable and does not contribute to global warming. When comparing biodiesel to diesel fuel derived from petroleum, total CO_2 emissions were found to be 78% lower (Van Gerpen, 2005).

2.6 Printing Process

The general type of image carrier used, and the image transfer method used distinguish the five main printing methods. The printed picture is either directly or indirectly transmitted to the substrate, depending on the method used. Examples of direct printing techniques include gravure, flexography, screen printing, and letterpress. In direct printing, the image is transferred straight from the image carrier to the substrate. The image is moved from the image carrier to the blanket cylinder and finally to the substrate in indirect, or offset, printing. The most common printing technology used today is lithography, which is an indirect (offset) process.

2.6.1 Digital Printing

Throughout the years, digital printing has been extensively utilized for the printing of papers and graphics. These technologies use bitmap pictures or computer-generated patterns to deposit ink onto target substrates. Recent advancements in printing technology have primarily been associated with using high-resolution, quick (high frequency jetting), low-cost processes for production printing. There has been a lot of research done to leverage digital printing for direct deposition of current advancements in practical materials. Through its additive manufacturing capabilities, direct printing technologies can have advantages over conventional photolithographic manufacturing procedures in the sense that associated manufacturing costs can be significantly decreased (Kwon et al., 2020). As shown in the Figure 2.4 is the digital printing.

Figure 2.4: A digital printer for textiles.

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2.6.2 Inkjet Printing

Using a technology called inkjet printing, ink is produced in the form of droplets that are deposited in a specific pattern onto a substrate. A picture is produced if the fluid has colorants in it. The technology of inkjet printing has developed into a single which is crucial to the graphical printing sector as well as numerous recently discovered industrial and medicinal uses. The two-way coupling from the electrical to the mechanical domain through the piezoelectric actuator, the coupling to the acoustic domain inside the ink channels to transfer the deformation into pressure waves, and the coupling to the fluid dynamic domain in the nozzle to transform acoustic energy into the kinetic and surface energy of the drop formation process are the three main processes that make up the physics of the inkjet printhead operation (Wijshoff, 2018). The main components of inkjet printer are shown in Figure 2.5.

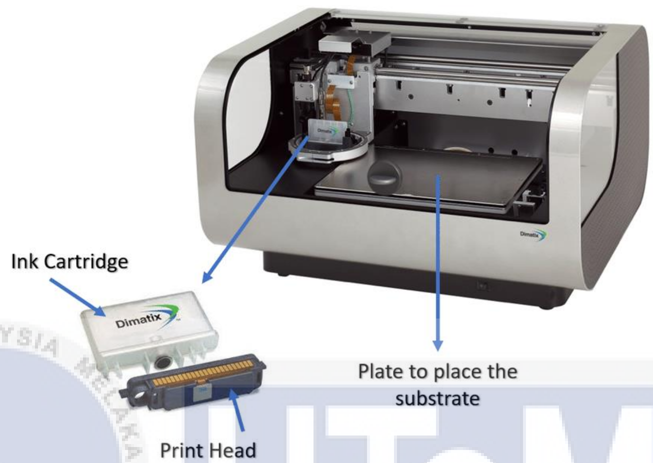

Figure 2.5: The main components in inkjet printing process consist of inkjet printer, ink, and substrate.

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2.7 Properties of digital printing

Any printing process that uses a digital file as input and does not require a fixed plate is generally referred to as digital printing. Consequently, every impression is unique. This covers a wide range of technologies, such as thermography, dry toner electrophotography, inkjet, and others. Digital printing consists of several properties which include image quality, variable data printing, wide range of substances and environmentally friendly. Although dye-based ink provides more vibrant colour than pigment-based ink, pigment-based ink dries approximately 100 times faster in inkjet printers. The most effective pigment particle size to avoid clogging issues is the smallest one. After drying, the thickness of the film layer ought to be roughly 0.5 micron.

2.8 Parameter for the GPI

For this research, there are four essential parameters that are observed during the transesterification process in order to achieve the desired ink's viscosity. Therefore, the reaction time and reaction temperature of transesterification process are the varied parameters meanwhile the concentration of catalyst and the molar ratio of waste cooking oil and methanol are the constant parameters for this research. To achieve a complete forward reaction and maximum ester generation, a higher alcohol molar ratio is required, depending on the oil, catalyst type, and process parameters (Rabu et al., 2013).

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2.8.1 Reaction Temperature

At a certain temperature, viscosity began to exhibit an increase in value, however it remained lowered as the reaction's temperature increased. Based on Cordero-Ravelo and Schallenberg-Rodríguez (2018), when it came to the reaction temperature, as long as it was over 50 °C, no discernible variations occurred in the conversion factor. Therefore, it might be said that the reaction temperature must never drop below 50 °C in order to maximize the conversion rate. At atmospheric pressure, the ordinary temperature range is between 50 and 75 C because the reaction temperature is limited by the alcohol's boiling point (Rabu et al., 2013). In this research, reaction temperature is observed as the varied parameters in transesterification process of WCO and methanol.

Triglyceride saponification accelerates by higher reaction temperatures, which results in a loss in biodiesel output when they rise above the ideal threshold. In general, in order to avoid alcohol evaporating, the transesterification reaction temperature needs to be lower than the boiling point of alcohol (Mathiyazhagan & Ganapathi, 2011.).

2.8.2 Reaction Time

As the response time increased faster, the ester's viscosity decreased. How ever, this component similarly exhibits the same pattern as the concentration of the catalysing agent. Sebayang et al. (2010) states in his research that it is readily apparent that the concentration of triglyceride dropped from the beginning point as reaction time increased. Therefore, the reaction time of transesterification process of WCO and methanol vary in this research. The yield product, such as biodiesel or mono alkyl ester, is not increased by further increasing the reaction time.

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2.8.3 Catalyst Concentration

According to Martín et al. (2016), oil transesterification requires a high concentration of catalyst. The maximum catalytic activity was demonstrated by potassium, sodium, and methoxides during the transesterification of frequently used oils. Their average concentrations ranged from 0.4 to 2 weight percent. Enhancing the amount of catalyst generally increases the rate of reaction. In addition to increasing the cost of producing biodiesel, excessive catalyst concentrations complicate product separation since excessive amounts of it may cause an emulsion to form. As a result, this variable needs to be appropriately optimized.

The concentration of catalyst has an impact on the production of biodiesel likewise. The most widely utilised catalysts to produce biodiesel are potassium hydroxide (KOH) and sodium hydroxide (NaOH). Moreover, the conversion of triglycerides into biodiesel increases in conjunction with an increase in catalyst concentration with oil samples. However, a lack of catalyst causes the conversion of triglycerides into fatty acid esters to be incomplete (Mathiyazhagan & Ganapathi, 2011.).

2.8.4 WCO to methanol molar ratio

In addition to catalysis, additional factors have been shown to influence the transesterification process. These consist of the ratio of the reactant (alcohol to oil volume or molar ratio), the temperature, time, kinetics, and mechanism of the reaction, the amount of water in the oil or fat, and the level of free fatty acids (C. Ehir, 2010).

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2.8.5 Processing ink using waste cooking oil for digital printing.

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The transesterification process is used to create the binder for ink from leftover cooking oil. Additional materials are added to the mixture to create an ink that is acceptable for digital printing.

2.8.6 Printing ink using waste cooking oil.

Soybean oil-based vehicles offer significant benefits in the manufacturing of colored inks, beyond the substitution of renewable resources for petroleum-based resins. When compared to conventional ink formulations, the vehicle's relatively pale color permits far lower pigment levels. The formulation of inks over a broader spectrum of viscosity, low rub-off characteristics, a decrease in the emission of volatile organic compounds, and a reduction in the health risks associated with inhaling "ink mist" in the workplace are additional benefits of vegetable oil-based inks (Derksen et al., 1996).

2.8.7 Formulating ink using waste cooking oil.

An environmentally responsible and sustainable method of producing ink is to formulate it with leftover cooking oil. Used cooking oils and fats, or waste cooking oil, may provide an excellent feedstock for bio-based inks. Waste cooking oil can be transformed into fatty acid methyl esters (FAME) by adapting the transesterification method, which is frequently employed in the manufacturing of biodiesel. The FAME compounds have the potential to function as a bio-based element in ink compositions. In addition to providing a way to recycle a substance that is frequently thrown away, adding used cooking oil to ink helps the ink business become less dependent on conventional petroleum-based materials.

In general, pigments, resins, solvents, and additives are used in ink compositions. Through partial substitution of waste cooking oil-derived FAME for typical petroleum-derived solvents, ink producers can produce a more environmentally friendly product. When adding waste cooking oil-based components, it is crucial to properly balance the ink's performance properties, such as adhesion, color intensity, and drying time. Furthermore, quality control procedures are necessary to guarantee that the ink exceeds industry requirements and produces trustworthy printing outcomes. Making ink from used cooking oil is an advancement towards a more socially and environmentally responsible ink manufacturing method. It also fits in with the concepts of the circular economy and resource efficiency.

2.9 Transesterification process.

Transesterification is an essential procedure before using vegetable oils in internal combustion engines. The final result of transesterification is biodiesel. An alternative fuel that is associated with efficient management, environmental preservation, sustainable development, and energy conservation is biodiesel. A number of researchers have examined the transesterification process, which produces biodiesel from various non-food oils (Ragit et al., 2011). Figure 2.6 shows the reaction equation of the transesterification process.

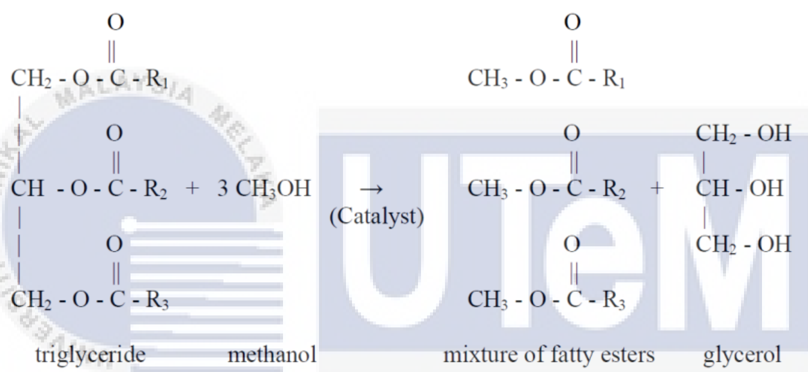

Figure 2.6: The reaction equation of the transesterification process. (Transesterification Chemical Equation, n.d.)

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2.10 Fourier Transform Infrared Spectroscopy Analysis on GPI

FTIR (Fourier transform infrared) spectroscopy has been extensively utilised to characterise organic components from many different materials, including paper and ink, which are part of historical documents (Ion & Doncea, 2014).

The vibrational bands found in the FTIR spectrum of thermochromic printing ink are typical of mineral and vegetable oils. The -CH, -CH2, and -CH3 stretching bonding vibrations of aliphatic chains contribute for the vibrational bands observed in oils, which range from 2925 to 2850. Furthermore, bands at 3010 (alkenyl CH stretching), 1463 (CH, CH2 stretching), 1166 (C-O stretching), 1101 (C-O stretching), and 721 (-CH2-rocking) cm−1 are observed in vegetable oils, along with an intense band at 1740 cm−1 that is attributed to the carbonyl group of esters. Mineral oil is indicated by the bands at 1463 cm−1 (aromatic C–H bending) and 1376 cm−1. Thus, the mineral and vegetable oils in the binder of utilised thermochromic ink cover microcapsules so completely that FTIR analysis is unable to recognise particles. Less than 5% of vegetable oils are used in the production of cold-set ink. Reagents in a binder may also be present in the ink, but the resin bands may cross over with oil vibrational bands (Rožić et al., 2020). Figure 2.7 displays the FTIR spectrum of thermochromic ink.

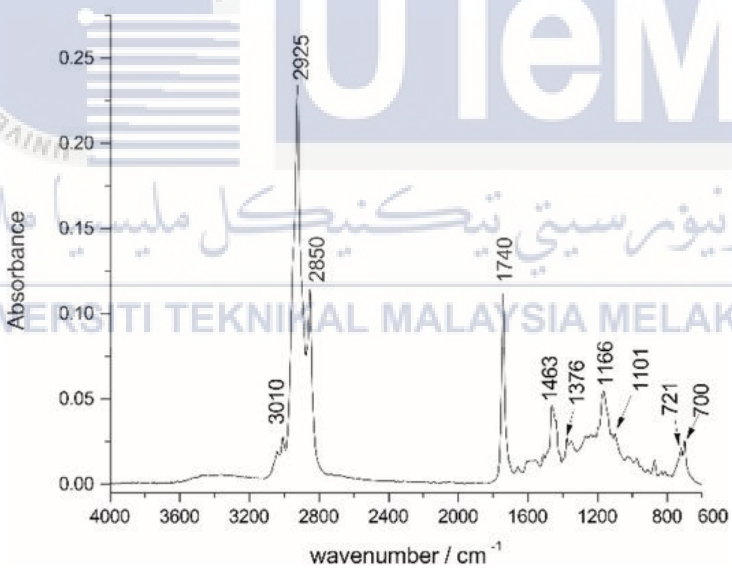

Figure 2.7: FTIR spectrum of thermochromic ink (Rožić et al., 2020).

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2.11 Scanning Electron Miscroscopy and EDX Analysis on GPI

FESEM and EDX are invaluable techniques for characterizing green printing inks used in inkjet printing. Green printing ink surface morphology can be imaged at high resolution using FESEM. Characterising the size and distribution of pigment particles is essential for maintaining consistent print quality and avoiding nozzle clogging in inkjet printers. This technique helps achieve each of these objectives. FESEM cross-sectional images demonstrated that as the number of printing cycles increased, the thickness increased (Karami et al., 2022). Conversely, EDX provides elemental analysis, which is a useful addition to FESEM. In ensuring that green inks contain only environmentally beneficial ingredients and no harmful heavy metals, EDX is a very helpful method.

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CHAPTER 3

METHODOLOGY

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3.1 Overview of methodology

This chapter outlines the steps involved in developing and producing green printing ink. The effect of the transesterification using waste cooking oil to make printing ink consist of several major steps. The processing of the raw materials, the transesterification of waste cooking oil (WCO) with methanol, and the creation of green printing ink using formula and mixture composition, followed by the tests carried out on the generated GPI. Figure 3.1 displays the flow chart of the green printing ink (GPI) process.

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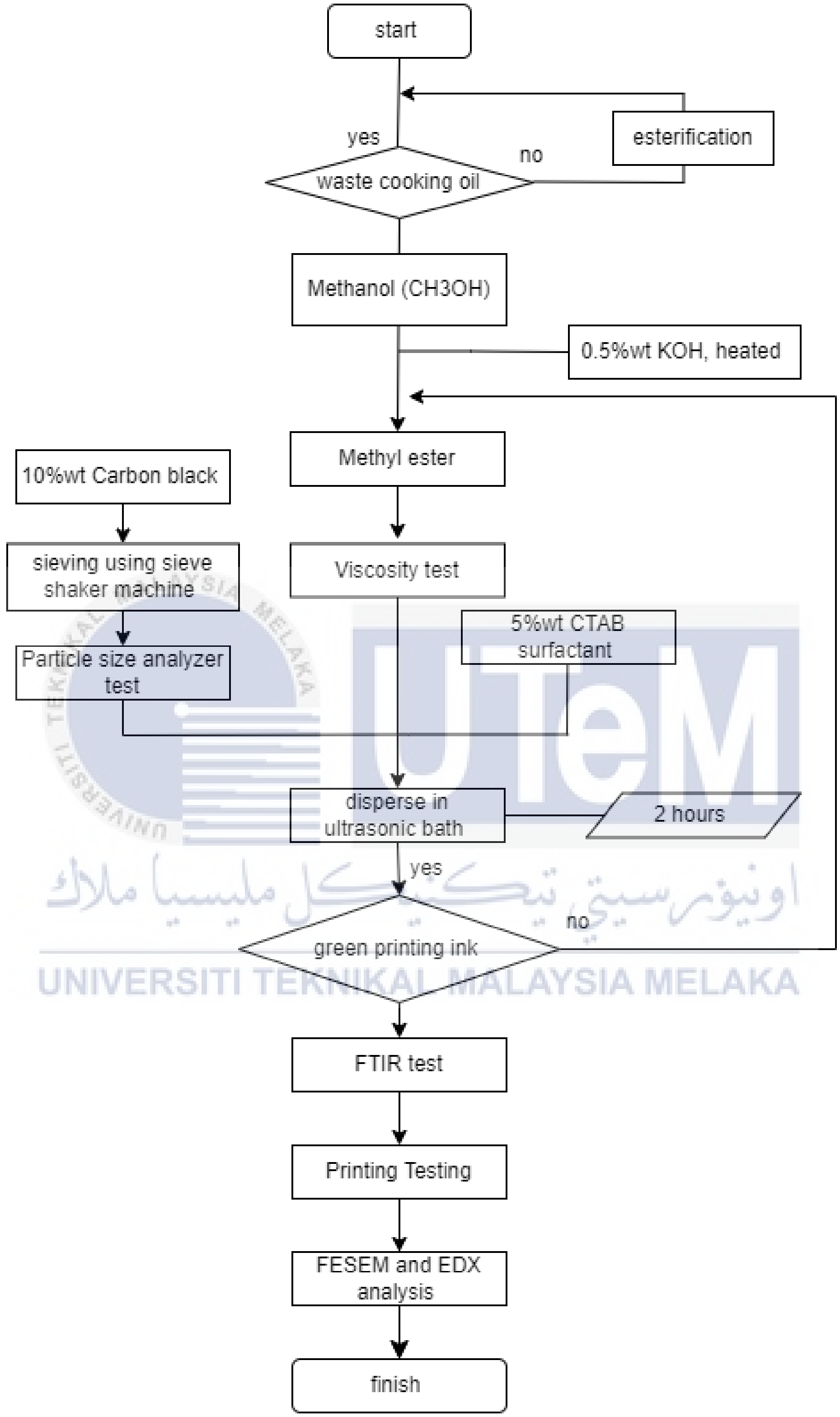

Figure 3.1: The flow chart of the GPI process.

3.2 Raw Materials

First step in chapter 3 is to describes on the raw materials used during this study, which were based on earlier research. The process of creating a green printing ink began with a list of the four primary ingredients.

The triacyl glycerides from a variety of feedstock such as nonedible oil seeds, vegetable oils, animal fats or tallow, waste cooking oil, and microbial lipids or single cell oil (from algae, oleaginous yeast, filamentous fungi, and bacteria) are converted into fatty acid methyl esters (biodiesel) in the presence of alcohol (methanol or ethanol) (Bardhan et al., 2022).

The formed methyl ester was used as the binder or vehicle of the ink. Filtered waste cooking oil was obtained and reacted with methanol (CH3OH) as the chosen solvent in the presence of 0.5%wt KOH as basic catalyst. Soluble agent of choice was methanol. because of its inexpensive cost and the lowest viscosity in the alcohol family.

Once the methyl ester was formed, 10%wt of carbon black was added into the mixture as a pigment to produce black colour to the ink. Additionally, the 5%w of hexadecyltrimethylammonium bromide (CTAB) that was act as surfactant was added into the solution.

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3.3 Free fatty acids esterification of waste cooking oil.

In order to direct the esterification reaction in the direction of the intended products, it is recommended to utilise a higher amount of alcohol than the stoichiometric ratio of 1:1 for the acid. As a result, in each example, 16 g of methanol were added for every 100 g of oil, or a 4.5:1 MeOH: oil molar ratio (Boffito et al., 2013). Sulfuric acid was utilised as catalyst and methanol was used as alcohol. The most important variables for the reaction were the temperature of the reaction, the ratio of methanol to WCO, and the amount of catalyst relative to WCO.

Furthermore, the yield of the transesterification process is decreased by higher FFA percentages in the oil.

Esterification was conducted out in a 150 mL batch reactor with three necks and a mechanical stirrer and reflux condenser at atmospheric pressure. A thermocouple was placed inside the reactor to measure the reaction temperature after the reactor was heated using a heating jacket (Ding et al., 2012). The result obtained is the WCO may seem lighter or clearer as the free fatty acids are transformed into fatty acid esters. As the reaction continues, there could occur some separation between the oil layer and a glycerol layer, depending on the particular method and WCO characteristics.

3.4 Transesterification process of WCO with methanol

The transesterification reaction between waste cooking oil and methanol has been illustrated in Figure 3.4 below. Glycerol and a fatty acid ester combination are produced when the alcohol and free fatty acids in the WCO combine. Each of the transesterification reaction experiment was carried out in a 250 mL round-bottom flask or conical flask. The mixture in the flask was heated using a hot plate that came with a magnetic stirrer arrangement (Patil et al., 2012). The waste cooking oil was heated to 60°C and then placed in a mixture of measured methanol and KOH according to the molar ratio. This process was carried out for four samples of 100g of filtered WCO with 1g of KOH powder that had been weighted and poured into a 250ml conical flask. The solution is heated at a constant temperature of 70 °C.

All three samples were left for a day following the 120-minute reaction before the phases were separated. After a day, two layers emerged, and an ink syringe was used to inject the methyl ester-containing upper layer out of the mixture. The methyl ester that was formed were preserved in a glass bottle. This process was repeated with different reaction time of 120-minute, 90-minute and 60-minute. Figure 3.2 shows the transesterification reaction of WCO and methanol.

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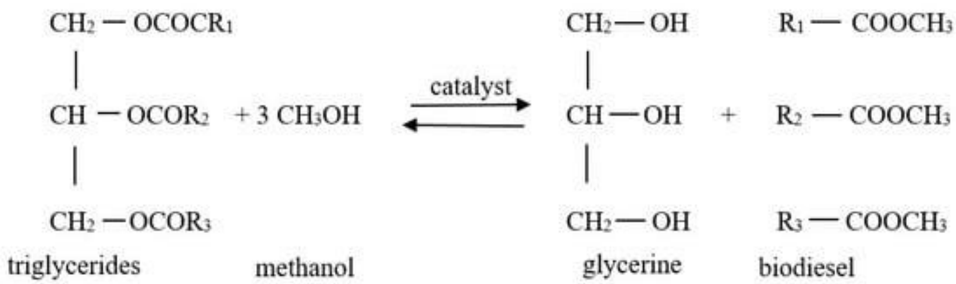

Figure 3.2: Transesterification reaction of WCO and methanol. (Ulukardeşler, 2023)

3.4.1 Waste Cooking Oil (WCO) to methanol molar ratio

According to Hazmie (2020), the molar ratio of WCO and methanol applied in the transesterification process is 19:1. He claimed that the ratio fulfils SIRIM requirements. Nonetheless, a 3:1 molar ratio of WCO to methanol is required for the purpose of this study. Since the molar ratio corresponds to one of the fixed parameters, it was maintained constant throughout the entire study.

The mass of WCO was kept constant in this study thus the masses of methanol used were varied depending on the molar ratio. Molar mass of waste cooking oil is about 882 g/mol (Encinar et al., 2005) and molar mass of methanol is 32.04 g/mol. Formula to find the molar mass of 100g of WCO is as the equation stated in Figure 3.3.

$$\text{Number of moles per } \mathbf{100g\ of\ WCO} = \frac{100\ g}{882\ g/mol} = 0.1134 \text{ mol}$$

Figure 3.3: Equation to convert mass to molar mass

3.4.2 Varied reaction temperature and reaction time

The transesterification procedure was carried out with different values of the molar ratios of WCO to methanol in Table 3.1 from the previous study by Hazmie (2020), as well as a different reaction time and reaction temperature. The objective of his experiment is to use a range of applicable parameters to obtain a viscosity of 2–40 cm^3P. As so, throughout the course of this research, the molar ratio of WCO to methanol remains constant.

Table 3.1: Varied reaction time and reaction temperature for transesterification of ME

SAMPLE	MOLAR RATIO OF WCO: METHANOL	REACTION TIME (MIN)	REACTION TEMPERATURE (°C)
A	3: 1	120	60
B	3: 1	120	70
C	3: 1	90	70
D	3: 1	60	70

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3.5 Production of green printing ink

WCO is a renewable and biodegradable raw ingredient used in the production of green printing ink. Following the production of the GPI using WCO and methanol as the binder, additional materials, such as pigments and additives, were added. Green printing ink is biodegradable and made from renewable resources. According to Aydemir et al. (2018), vegetable oil-based resources are a significant alternative for bio-renewable raw resources. These oils can effectively compete on affordability with the petrochemical-derived components typically utilised to create polymers. Specifically, within the printing ink industry, pressure has been developed in the recent several years for replacement vegetable oils and their derivatives for mineral oil in inks. There are several steps required.

Carbon black powder is selected as the pigment for this research. Carbon black is a residue of oil sludge treatment that was received from Victory Recovery Resources Sdn Bhd. Carbon black powder enhance black colour to the ink. In order to achieve the suitable size of the pigments, the particle size analyzer is used to analyzed the size of the pigments as shown in the Figure 3.4. The particle size required is 100nm. The carbon black powder is sieved by using sieve shaker machine to achieve the required size. The sieve process is conducted for 22 times/min of vibration frequency with magnitude of 9mm using a 20-micron stainless-steel sieve basket. Sieving process separates the particles of the powder (Oh et al., 2021).

The additive used for this process is Cetyltrimethylammonium bromide (CTAB) surfactant is added to the powder. There was a 15% weight increase in carbon black and a 5% weight increase in surfactant. A mixture of milled carbon black powder, CTAB surfactant, and methyl ester with the lowest viscosity was used as a binder. The mixture was evenly distributed produced the formulated GPI after two hours in an ultrasonic bath. Figure 3.5 shows the process flow chart in producing green printing ink.

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Figure 3.4: Particle Size Analyzer machine.

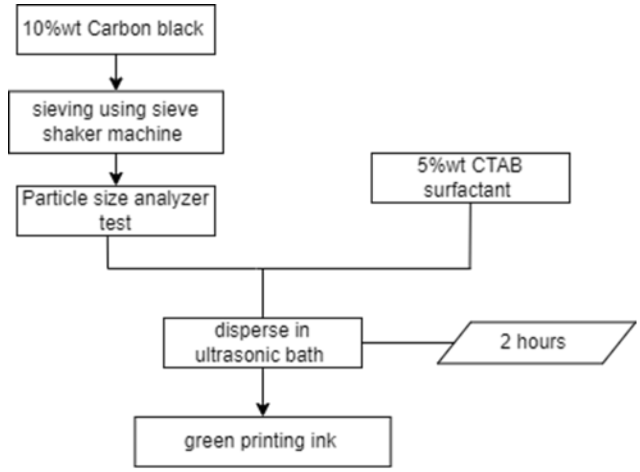

Figure 3.5: The green printing inks process flow chart.

3.5.1 Viscosity test on GPI

The initial research using the formulated GPI was a viscosity analysis to determine the final viscosity of the GPI created. The purpose of this research was to obtain GPI with an appropriate viscosity, as the inkjet printing method required viscosity ranging from 1 to 30 cP (Öhlund, 2014). This would enable GPI to be used in inkjet printing.

The machine uses to conduct the viscosity test is the Brookfield's Viscometer. The GPI is injected into a container by using a 5ml ink syringe. The viscometer spindle and rotor were placed in the container. The viscometer is starting and the result of viscosity of are displayed on the screen. The test is conduct for three times to record the data of the three sample and to calculate the mean of each methyl ester. Figure 3.6 shows the Viscotester 2 plus for viscosity testing.

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Figure 3.6: Viscotester 2 plus machine for viscosity testing.

3.5.2 Fourier Transform Infrared Spectroscopy (FTIR) test.

FTIR is implemented in the analysis of GPI characterisation. FTIR is undertaken in order to identify the chemical composition of GPI produced as well as determine the functional group of the GPI produced. Within the 400–2000 cm−1 spectrum, FTIR spectroscopy provides an accurate measurement of the ink sample and is a dependable and useful tool. The chemical elements of ink samples, such as dyes, solvents, and resins, are mostly described in this area (Sharma & Kumar, 2017). The percentage of transmittance at the C-H bond between GPI and petroleum-based ink was compared and examined, and the peaks from the GPI spectrum were recorded and calculated.

3.6 GPI performance test

The next process is carried out using an inkjet printer to determine the overall performance of the GPI. The ink of the GPI on the paper substrate is observed during the printing process.

3.6.1 Inkjet printing test

The next process is carried out using a digital printer to determine the overall performance of the GPI. The ink of the GPI on the paper substrate is observed during the printing process.

3.6.2 Physical observations

A visual inspection is conducted to determine the coverage of the printing area and the clarity of the printed image of the ink on the paper substrate. The variation between the screen image and the printed image was utilised to determine the area of the printed image. To be able to observe the final image, two printing tests are performed. Using standard printing ink for the first test, and green printing ink for the second, the tests are printed out using the inkjet printer. In order to determine out if the GPI passed the viscosity test, multiple tests are conducted.

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3.7 FESEM and EDX analysis of the GPI and conventional ink

The study focuses on the analysis of green printing ink and conventional ink using Field Emission Scanning Electron Microscopy (FESEM) and Energy Dispersive X-ray Spectroscopy (EDX). Green printing ink samples were prepared by depositing them onto suitable substrates such as paper, while conventional ink samples were applied using a consistent method. The samples were then dried under controlled conditions to remove residual solvents (Momenzadeh et al., 2017). Sputtering process was conducted as a function to reduce the electric charging of SEM samples and attain the highest quality of imaging possible (Ramesh et al., 2023).

FESEM was used to examine the surface morphology of the ink samples, with the samples mounted onto aluminium stubs and coated with gold or platinum to enhance conductivity (Ramesh et al., 2023). EDX was used to analyze the elemental composition of the ink samples, with the EDX detector collecting X-ray signals emitted from the sample (Xu et al., 2016). The analysis helped compare the elemental profiles of green and conventional inks, providing insights into their formulations and potential environmental impacts. The FESEM images were analyzed to assess particle size distribution, surface roughness, and morphological characteristics, while EDX spectra determined the relative abundance of elements in each ink sample.

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CHAPTER 4

RESULTS AND DISCUSSIONS

4.1 Overview

The purpose of this research was to apply the transesterification process to create an environmentally friendly printing ink for digital printing applications that has a lower viscosity from leftover cooking oil. Every outcome was examined in accordance with the objectives and methods of the research.

The experimental data were examined and compared with the findings of previous studies as well as petroleum-based ink. The results were arranged in an order that was consistent with the objectives of the research. Using methanol and excess oil from cooking in a fixed molar ratio, four transesterification samples were made, and their viscosities were measured and compared over time. Following that, the methyl ester with the lowest viscosity was chosen and blended with the rest of the ingredients.

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The results of the Fourier Transform Infrared Spectroscopy (FTIR) analysis of the chemical bonds present in the formulated green printing ink were compared with the results for the petroleum-based ink. The formulated green printing ink was also tested for printing performance using a Canon Pixma 258 inkjet printer on a paper substrate. The surface morphology and microstructure of the printed images were analysed using a scanning electron microscope (SEM) equipment on the printed images from the green printing image and the printed image from the petroleum-based ink.

4.2 Transesterification analysis

Differentiating methanol to oil molar ratio in transesterification process to produce a lower viscosity of methyl ester was the first objective in this study. The transesterification of methanol and waste cooking oil produced a layered solution in which the upper layer in Figure 4.1(a) was the methyl ester and the bottom in Figure 4.1 (b) layer consist of glycerol, excess methanol, remaining base catalyst and soaps formed during the transesterification reaction as stated by Encinar J.M. et al. (2005). The methyl ester produced functioned as binder in the ink. Figure 4.1 shows the layers of the solution and Figure 4.2 shows the results of transesterification of different molar ratios.

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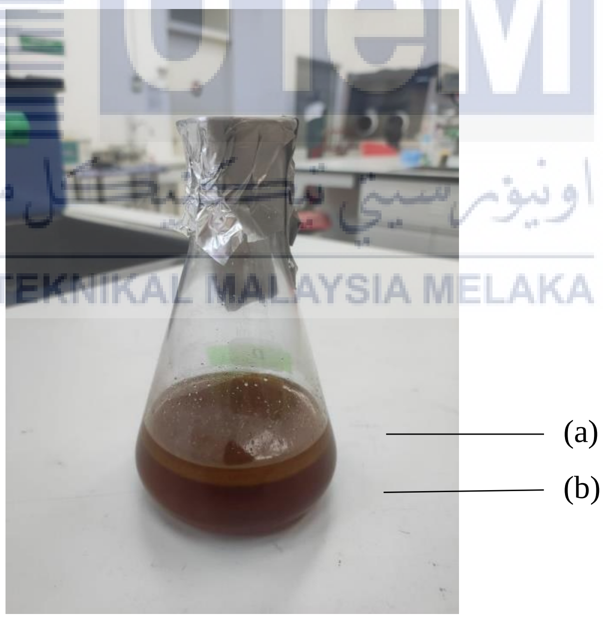

Figure 4.1: Product of transesterification of methanol and waste cooking oil (a) glycerol, excess methanol, remaining catalyst and soaps formed (b) methyl ester.

Figure 4.2: Samples of methyl ester from transesterification of different reaction temperature and reaction time.

4.2.1 Viscosity testing of methyl ester

In this experiment, viscosity test was measured for methyl ester, which is the result of four samples that were produced by the transesterification process of methanol and waste cooking oil, each with a different reaction temperature and reaction time determined in Chapter 3. The methyl ester which was obtained throughout the transesterification process was tested using a viscometer. After three repetitions of the viscosity test, the mean viscosity was obtained. The methyl esters' viscosity results are displayed in Table 4.1.

The results of the viscosity showed that the viscosity of methyl ester was the lowest with reaction temperature at 70°C and reaction time at 60 minutes compared to reaction temperature at 70°C and reaction time at 90 minutes, reaction temperature at 70°C and reaction time at 120 minutes and reaction temperature at 60°C and reaction time at 120 minutes with mean viscosity of 0.483cP, 0.843cP, 0.987cP and 0.443cP respectively. The trend of viscosity test of methyl ester with varied reaction temperature was shown in Figure 4.3.

Table 4.1: Viscosity results of methyl ester from different reaction time and reaction temperature.

SAMPLE	REACTION TIME (min)	REACTION TEMP(°C)	VISCOSITY OF METHYL ESTER (cP)				REMARKS
			1st	2nd	3rd	Mean	
A	120	60	0.48	0.49	0.48	0.483	High viscosity
B	120	70	0.83	0.86	0.84	0.843	High viscosity
C	90	70	1.03	0.97	0.96	0.987	Highest viscosity
D	60	70	0.43	0.44	0.46	0.443	Lowest viscosity

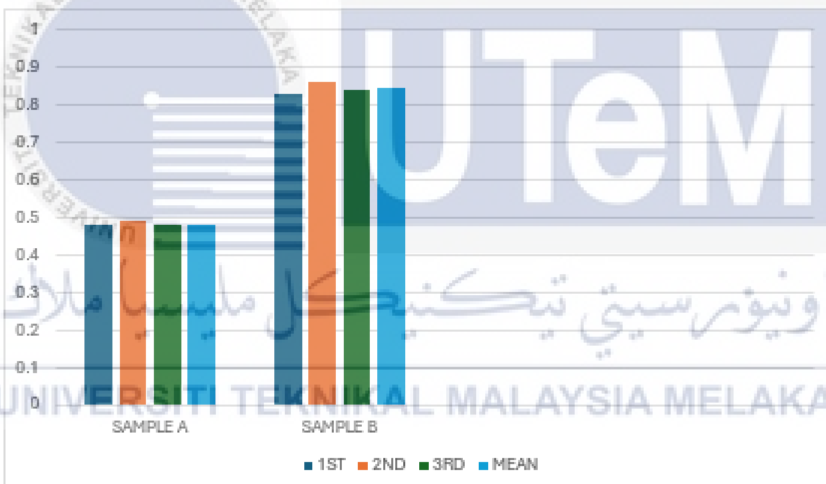

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Figure 4.3: Graph of viscosity of methyl ester at 60 minutes with different reaction temperature.

The effect of reaction temperature (°C) on the viscosity of methyl ester was presented in Figure 4.3. At 70 °C, faster reaction kinetics may occur due to poorly mixed reaction mixture or optimal catalyst concentration, while at 60 °C, slower reactions may proceed more evenly. Increasing the reaction temperature has significant effects on the transesterification reaction (Alhassan et al., 2014). Altering the operating pressure enables the maintenance of higher temperatures (Arachchige et al., 2021). Parts of the glycerol may remain in the ester layer at higher temperatures due to the excess methanol causing it to be difficult to separate the glycerol layer by gravity. Furthermore, during separation, glycerol may draw towards the ester layer; this would contribute to the graph's trend indicating the formation of methyl ester viscosity.

The effect of reaction time on the viscosity of methyl ester is shown in Figure 4.4. The 60-minute reaction time is efficient for transesterification with minimal side reactions, while extending the time may not significantly increase conversion and may introduce unwanted by-products. Reaction equilibrium develops more faster at higher temperatures, and the ME content begins decreasing as reaction time increases (Wan & Hameed, 2011). One of the most crucial reaction conditions in the biodiesel production process is reaction time (Alhassan et al., 2014). The quantity of heterogeneous catalyst generally has a significant positive affect on the number of active sites available for the reaction during the transesterification of vegetable oil to methyl ester (Wan & Hameed, 2011).

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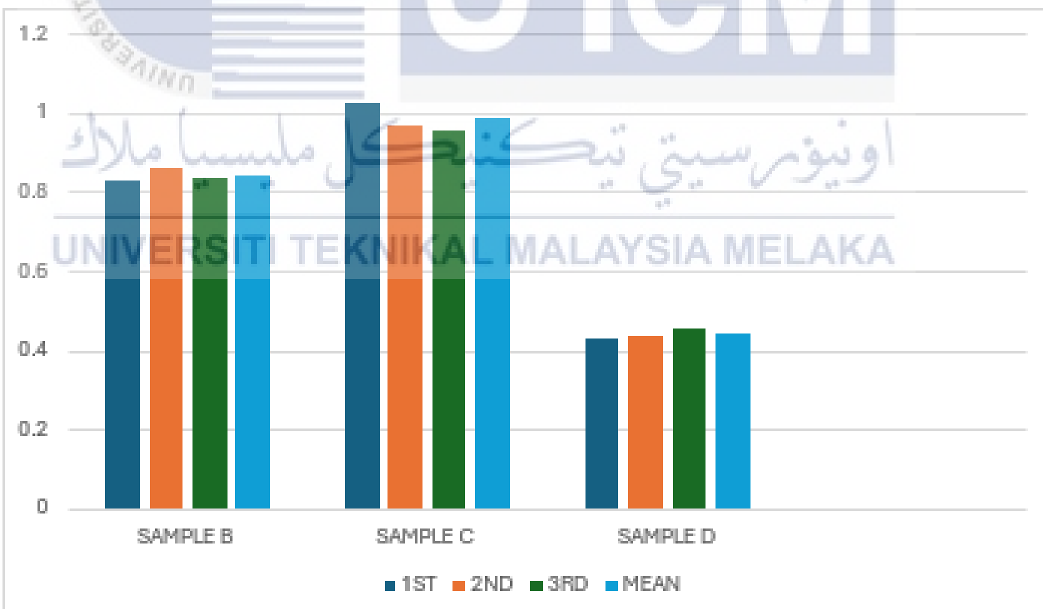

Figure 4.4: Graph of viscosity of methyl ester at 70 °C with different reaction time.

4.3 Formulated ink analysis

The second objective of this study was to characterize the green printing ink formulation through viscosity test, Fourier Transform Infrared spectroscopy (FTIR), Field emission scanning electron microscopy (FESEM) and Energy dispersive X-ray analysis (EDX). The GPI is composed of binder, colorants and surfactant as additive which were mixed with a suitable mixture composition percentage. The formulated ink was tested for viscosity, presence of chemical bond for comparison, to obtain high-resolution imaging of the ink's surface morphology and structure and to perform elemental analysis of the ink in order to achieve the second objective.

At a reaction temperature of 70 °C and a reaction time frame of 60 minutes, the sample of methyl ester produced during transesterification had the lowest viscosity. As a result, methyl ester of 0.443 cP, which operated as a binder, has been combined in a specified weight percentage with other ink compositions. As shown in Figure 4.5, the carbon black powder size decreased after being sieve for 22 minutes using the sieve shaker machine. The powder's mean size decreased from 5.545 µm to 2.701 µm. The Particle Size Analyzer (PSA) was used to measure the powder's size. Referring to the graph on Figure 4.5, the standard deviation of 0.138 indicates moderate variability.

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Green printing ink (GPI) with a black colour was produced by combining 80 % methyl ester, 15 % carbon black powder, and 5 % CTAB surfactant. The components were then distributed in an ultrasonic bath, as seen in Figure 4.6.

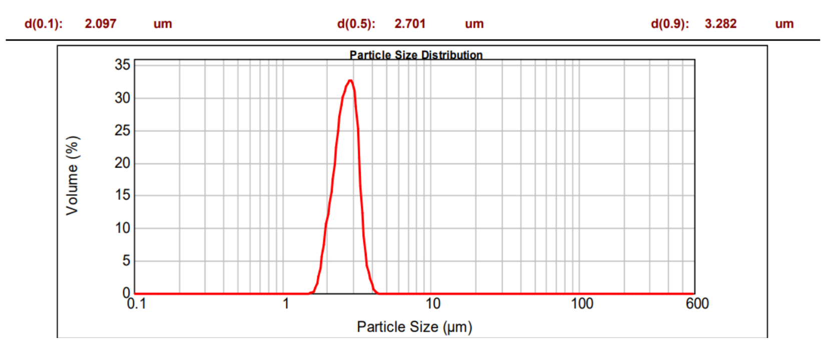

Figure 4.5: PSA results of sieved Carbon Black powder.

Figure 4.6: Formulated Green Printing Ink

4.1.1 GPI viscosity test

The GPI was tested for viscosity using Brookfield Viscometer and the test was repeated three times. The results were tabulated, and the mean viscosity of GPI was calculated as in Table 4.2. The final viscosity of GPI was 19.0 cP which was within the range of viscosity suitable for inkjet printing application. Thus, the mixture compositions of the formulated GPI succeeded in producing inkjet printing ink within the range of desired viscosity for inkjet printing application.

Table 4.2: Viscosity of formulated GPI

GPI/Viscosity	Viscosity of GPI (cP)			
	1st	2nd	3rd	Mean
GPI	18.7	18.6	19.9	19.0

4.3.1 Chemical bond analysis

Fourier Transform Infrared Spectroscopy (FTIR) was used to conduct chemical bond analysis in order to determine the functional groups and chemical bond of GPI. FTIR spectra of the prepared GPI are represented in Figure 4.7. The result of the spectroscopy test was shown in Figure 4.7 and chemical bonds Table 4.3.

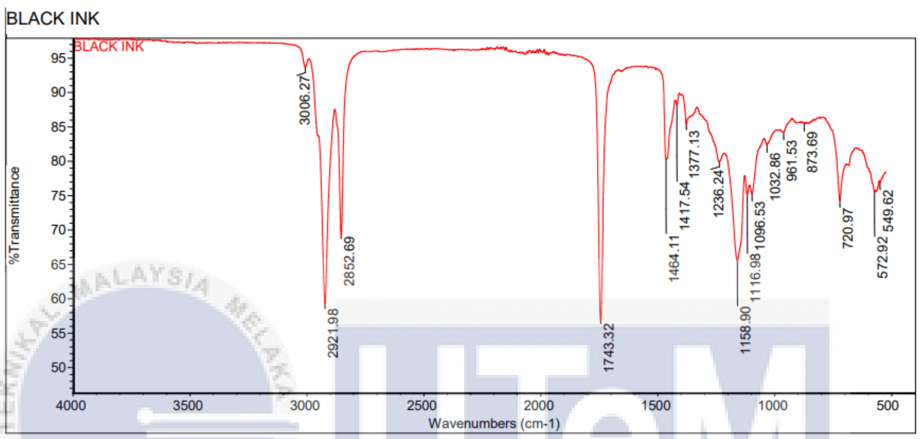

Figure 4.7: FTIR spectra of formulated GPI.

Table 4.3: Chemical bonds in formulated GPI.

	Wavenumber (cm⁻¹)	Bond
1	**3001.27**	C-H
2	**2921.69**	C-H
3	**2852.60**	C-H
4	**1732.92**	C=O
5	**1464.14**	C-H
6	**1377.13**	C-H
7	**1262.31**	C-O
8	**1116.95**	C-O
9	**1036.85**	C-O
10	**874.59**	C-H
11	**720.67**	(CH_2) n where n > 4

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Figure 4.7 shows the FTIR spectra of the produced GPI. The methyl ester was created by transesterifying waste cooking oil and methanol, and it was mixed with carbon black powder as a colorant and hexadecyltrimethylammonium bromide (CTAB) as a surfactant. As demonstrated in Figure 4.7, the black color obtains highest peak at the wavenumbers of 3001.27 cm^{-1}. It is typically associated with C-H stretching in alkanes or aromatic compounds. The black ink formulation includes a combination of hydrocarbon chains (both aliphatic and aromatic), esters, and possibly fatty acids or glycerides. This mixture would contribute to the ink's physical properties such as viscosity, drying behaviour, and adhesion characteristics. Proper formulation and quality control are essential to ensure consistent performance in printing applications.

The conventional ink formulation includes a combination of hydrocarbons chain consisting of both aliphatic and aromatic, carbonyl groups consisting of esters, ketones and aldehydes, amines or amides, ethers and methylene deformation. The slower rate of microcapsule disintegrates and less visible color shift in the print were caused by the thicker layer of ink binder (including classic pigment) on the surface of the microcapsules on recycled paper. The slowest rate of print biodegradation is caused by the thickest layer of conventional pigment and ink binder on synthetic paper surfaces (Vukoje et al., 2018). Saturated hydrocarbons enable volatile organic compounds (VOCs) to develop during printing, which can lead to air pollution and leave printing facility employees' health at risk (Hu et al., 2021). Saturated hydrocarbons have adverse effects on the environment and human health, which emphasises the need for more sustainable printing ink alternatives. FTIR spectra of the UV thermochromic ink at 25 °C and 50 °C, measured in transmission mode were presented in Figure 4.8 and Table 4.3 shows the chemical bonds in commercial ink.

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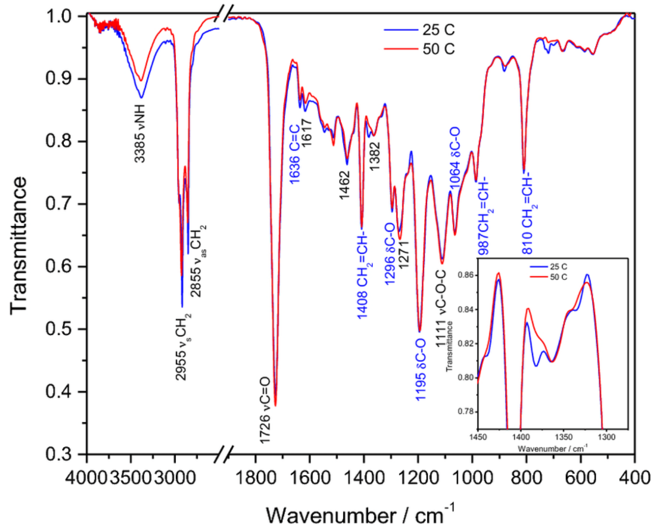

Figure 4.8: FTIR spectra of the UV thermochromic ink at 25 °C and 50 °C, measured in transmission mode (Vukoje et al., 2018).

Table 4.3: Chemical bonds in commercial ink

	Wavenumber (cm⁻¹)	**Bond**
1	**3385**	N-H
2	**2955**	C-H
3	**2855**	C-H
4	**1726**	C=O
5	**1636**	C=C
6	**1408**	C-H
7	**1296**	C-O
8	**1064**	C-O
9	**1111**	C-O-C
10	**987**	C-H
11	**810**	C-H

A lower peak of spectrum of a bond depicts a lower presence of the bonds thus from the comparison of transmittance percentage of C-H bonds between GPI and petroleum-based ink, GPI had a lower peak than petroleum-based ink showing a greener or better properties due to lower VOC emissions to air.

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4.4 FESEM and EDX analysis

FESEM and EDX analysis was conducted in order to visualize the surface morphology and microstructure of the sample at very high resolutions of the ink particles and provides quantitative information on the elemental composition of the sample as shown in the Figure 4.9 and Figure 4.10.

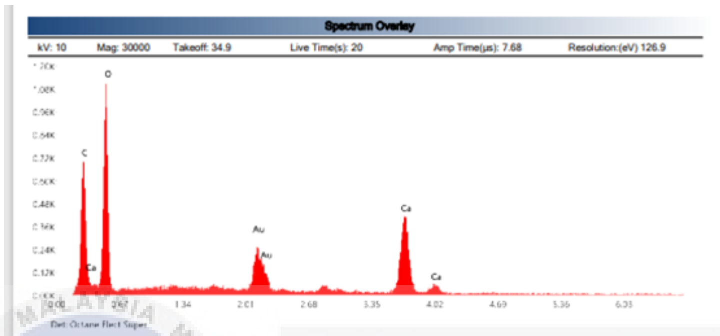

Figure 4.9: The EDX spectra of green printing ink image on paper substrate

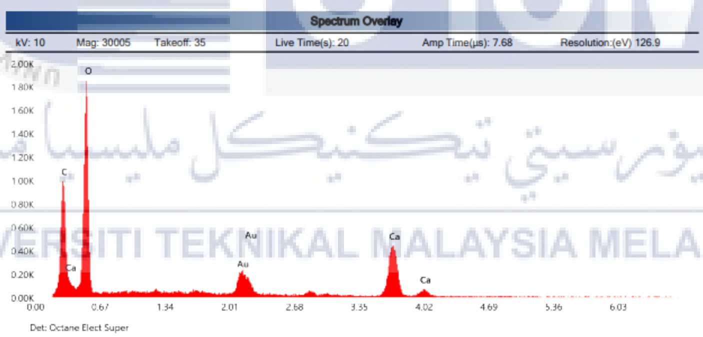

Figure 4.10 : The EDX spectra of conventional ink image on paper substrate.

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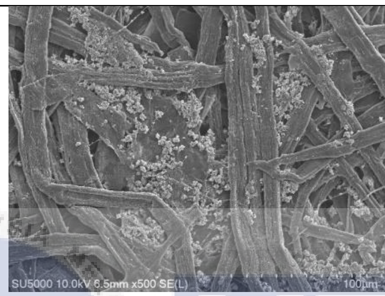
FESEM is used to visualize the surface morphology and microstructure of the sample at very high resolutions. The SEM images for GPI printing image on paper and conventional ink printing image on paper are both shown in the Table 4.4. The comparison of SEM images between conventional ink printing and green printing ink printing indicates several of significant modifications. Paper fibres are visible in both inks, indicating similar paper substrates. But compared to the small, asymmetrically formed particles in standard ink, the green printing ink has more of the particles and distributes particles more evenly. An altered formulation or finer milling of the pigment particles might have contributed to the more consistent and even surface texture of the green printing ink. FESEM cross-sectional scans demonstrated that as the number of printing cycles increased, the thickness increased as well (Karami et al., 2022).

Table 4.4: The comparison FESEM images of GPI and conventional ink

SEM SAMPLE	GREEN PRINTING INK	NORMAL INK PRINTING
SEM IMAGES		
FIBROUS STRUCTURE	Like the normal ink printing image, the paper fibers are visible, indicating a similar paper substrate.	The cellulose fibers of the paper are clearly visible, with a mesh-like structure.
INK PARTICLES	The particles seem more abundant and possibly finer compared to the normal ink. They also appear to be more uniformly distributed over the fibers.	There are numerous small, irregularly shaped particles scattered across the fibers, typical of pigment particles in normal printing ink.
SURFACE TEXTURE	The ink particles in this image have more even and consistent distribution, potentially indicating a different formulation or finer milling of the pigment particles	The ink particles have a granular appearance and are well-distributed over the fibers.
MAGNIFICATION AND SCALE	The magnification and scale are the same as in the first image (500x100 micrometers).	The magnification is 500x, and the scale bar indicates 100 micrometers.

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The distribution of the ink pigment was determined by through energy dispersive X-ray analysis (EDX) (Heard et al., 2004). The EDX analysis provides quantitative information on the elemental composition of the sample. Carbon is the most common element in the GPI, accounting for around 43.1% of the total elements, according to the elements analysis performed using energy-dispersive X-ray (EDX) equipment as shown in Figure 4.9. At different concentrations, certain amounts of additional atomic elements become visible (Striani et al., 2021). Other elements such as O, Ca and Au are also present in the ink surface. The components characterization showed that the carbon black ink from the green printing ink contained methyl ester, which is remarkably comparable to the ink from conventional ink that is based on petroleum.

4.5 Printing performance analysis

The final objective of this study was regarding the printing performance of the formulated GPI using an inkjet printer printed on a paper substrate. The sole way to propose recommendations for improvement for future research is by analyse the ink's performance after printing on a substrate. For a future study on green printing ink made from waste cooking oil, the performance test results could serve as a benchmark for improvement.

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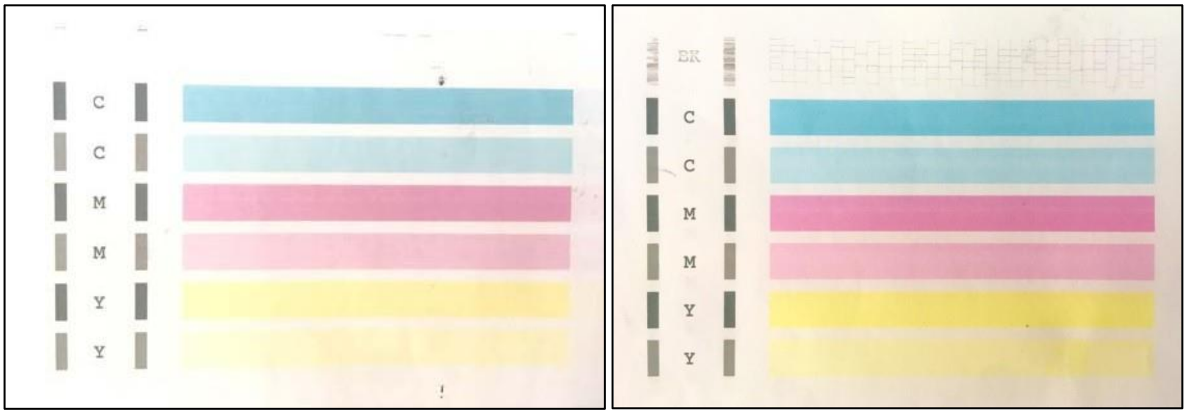
The printing test started with proving that the results of printed images were from the injected GPI into black cartridge of Canon Pixma258 Inkjet printer. The printer was set to print the nozzle check pattern as this setting or pattern could determine whether the ink was ejected properly out of the print head nozzle. Comparison of the pattern before and after injection of GPI into the printer was as shown in Figure 4.11.



(a) (b)

Figure 4.11: Nozzle check pattern; (a) without ink in black cartridge (b) with GPI in black cartridge

The UTeM logo was test printed on paper using an inkjet printer to examine the ink's ability to print images. This test was carried out in order to analyze the printing performance test, which involved physical observation, after the nozzle check pattern test confirmed the presence of the formulated green printing ink in the cartridge.

Two different renditions of the UTeM logo were printed on the paper: the first print used the colour version of the logo, while the second print used grayscale. The first print displayed the UTeM logo in its optimal colour and the black colour logo print was perfectly defined according to the shape of the logo. Both printing images can be observed in Figures 4.11 and 4.12.

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Figure 4.12: Printed UTeM logo using conventional ink.

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Figure 4.13: Printed UTeM logo using GPI ink.

Since the printed logo was produced on a paper substrate, a quick physical analysis of the printed image was conducted. There were no ink smudges due to the image being printed clearly and the ink was not dispersed when contacted with the finger, demonstrating the ink's strong rub-resistance. There were lines of unprinted sections merged with the equally cloudy seemingly insignificant black colour. An approximate calculation indicated that 86 % of the image was printed on the paper.

According to Figure 4.13, the printing performance tests result from nozzle pattern depicted that the formulated GPI was able to pass through the cartridge nozzle and printed on the paper substrate. Initial testing in printing applications demonstrated the developed inks' good performance (Robert, 2015). In the article, Soleimani-Gorgani, A. (2015) stated that despite the advantages of inkjet printing technology, nozzle blockage has invariably been the main issue since inkjet ink contains insoluble micro- or nanoparticles that can aggregate and precipitate. One of the challenging factors for researchers with respect to inkjet printer ink processing was nozzle blockage.

Based on the physical observation of the printing image of the GPI in Figure 4.13, there are variations in the ink density, with certain areas appearing lighter than others, indicating inconsistency in the ink application. The uneven distribution of colours and unclear or faint prints might result from the inconsistent dispersion of pigments in waste cooking oil. Besides, the methyl ester-based ink's viscosity and chemical composition might vary compared to conventional petroleum-based inks, which could have an impact on the ink's adhesion and flow characteristics. The stable dispersion of the particles obtained in suspension by chemical or electrostatic treatment to be important criteria other than the size of the particles (Soleimani-Gorgani, A., 2015).

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CHAPTER 5

CONCLUSIONS AND RECOMMENDATIONS

5.1 Conclusions

The first objective of this research is to analyse the effect of reaction temperature and reaction time of the transesterification of methyl ester with ratio of fixed methanol and WCO (3:1) based on the yield and the composition of the methyl ester produced. The data collected are analysed and significant conclusions for this objective as follows.

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- Transesterification process between methanol and waste cooking oil produce methyl ester that functioned as binder for the ink with a low viscosity.

- As the reaction temperature and reaction of time of the methyl ester is varied, each of the produced methyl esters has different viscosity from this research. The lowest viscosity with 0.443 cP of methyl ester produced was chosen which methyl ester with reaction temperature at 70 °C and reaction time at 60 minutes.

- Thus, methyl ester produced from the reaction temperature at 70 °C and reaction time at 60 minutes has a viscosity of 0.443 cP and is selected as the binder of the ink.

The second objective of this research is to characterize the green printing ink formulation through viscosity test, FTIR spectroscopy, FESEM and EDX analysis. Printing ink is composed of four main components and is mixed to produce a green printing ink. The significant conclusion of the second objective is as follows.

- The formulated GPI mixture composed of 80 wt% methyl esters as binder, 15 wt% carbon black powder with size of 2.706 µm and 5 wt% CTAB achieve the suitable application of inkjet printing.

- The FTIR spectra of GPI compared to petroleum-based ink obtain a lower percentage of transmittance of C-H bonds confirmed a lower amount of C-H bonds presence in GPI. Therefore, GPI has better properties due to lower VOC emissions to air, making it a greener alternative.

The final objective of this research is to examine the printing performance of green printing ink on paper substrates using inkjet printing. As the results are analyzed, the significant conclusion based on the third objective of this research is as follows;

- The process for producing a green printing ink from waste cooking oil has been successful since the printed test shows that the ink can be printed on paper and can pass through the print head nozzle to print the UTeM logo.

- Based on the FESEM images, the green printing ink exhibits a homogeneous distribution of particles, indicating a well-milled formulation, and the printing image's surface texture is consistent, demonstrating strong adherence to the substrate and a smooth finish.

- By the EDX analysis, the ink primarily contains environmentally friendly elements derived from waste cooking oil, confirming the sustainable nature of the ink. Essential pigments are present in adequate amounts, contributing to the ink's colour properties.

5.2 Recommendations

Research is a subject where there is always space for improvement, and this study is no exception. Both the printing procedure and the formulation of a green printing ink created from waste cooking oil require significant development and improvement. The following are the important recommendations that should be made for future research, out of the few that ought to be put forward

i. Using various coloured colourants, which could be based on minerals and plants, widened the colour spectrum of the ink generated. These organic-based colourants are potentially able to assist in managing the excessive use of inorganic colourants in coatings, paints, and inks.

ii. Apply more ecologically friendly ingredients to substitute the mixture of components in the produced green printing ink. For instance, bio-surfactants can be utilised as a surfactant alternative in the formulation of green printing ink. As natural resources are utilised for producing ink, a less hazardous composition mixture lowers the risk to human health from the inorganic solvents employed in the process and lowers harmful emissions to the environment.

iii. Applying additives that might improve the printing qualities and homogeneity of the ink mixture will help to improve the quality of the final output, with the focus of this research was the quality of the printed image.

iv. Implement other techniques or utilize ball milling to achieve nanosized

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pigments as required for inkjet printing and most of other digital printing applications. One of the techniques to improve is by wet milling in which solvents or surfactants are added during the milling process.

v. Enhance the resistance and colour accuracy of green pigments to ensure print results remain vibrant and consistent. As part of this, pigment particle size and dispersion are optimised for smooth printing that does not clog printheads.

5.3 Sustainability Element

This research's focus is on the utilizing waste cooking oil in producing ink. The topic of this research, formulation of green printing ink from waste cooking oil for digital printing itself clearly manifests a development of a sustainable product.

There are 5 stages in a product's life cycle which begin with pre-production, production, distribution, usage and disposal. Waste cooking oil is a product at its disposal stage as the use stage of the cooking oil has already happened. The waste cooking oil should not be used back in food industry without treatment due to health risk and thus, the used cooking oil would be lavishly disposed and increase the waste in our country.

Thus, to sustain the life of cooking oil, this research which also involves waste management is a very practical approach. Besides, the waste cooking oil comes from plants and thus it does not disturb the resources of petroleum.

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