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Investigation of Surface Free Energy of Palm Oil-Based Offset Printing Ink on Coated Paper

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Abstract. The widely use of petroleum-derived mineral oils for the production of offset printing inks has resulted in several environmental impacts. The suitable approach to develop alternative resources is palm oil fatty acid methyl ester (FAME) as a substitute for petroleum-based solvent. This study aimed to investigate the surface properties of palm oil-based offset ink on gloss-coated paper surfaces in terms of printability. We developed different Cyan color inks with linseed oil (FAME-LO) and soybean oil (FAME-SO) as drying oil. Printing tests were carried out using the IGT-A2 printability tester. Contact angles of water, methanol, and hexane on the surface of printed ink were measured using Contact Angle Analyzer. The surface free energy (SFE) and its components were calculated using Girifalco-Good-Fowkes-Young, Owens-Wendt, and van Oss-Chaudhury-Good methods. The results showed that as the liquid contact angle increased, the surface energy decreased. The surface energy of FAME-SO ink printed samples 36.36 mJ/m2 was higher than the unprinted paper 35.84 mJ/m2. Since high surface energy will affect the bond strength of the ink to the substrate, this will be an advantage for multicolor prints requiring good ink adhesion, which will enlarge the acceptance of other colors to be printed on the previous color.

1. Introduction

Petroleum-based solvents are used as raw materials for the production of varnish, which is one of the main components of offset printing inks. One of the approaches to developing alternative renewable resources is palm oil fatty acid methyl ester (FAME) as a substitute for petroleum-based solvent. There have been numerous studies on the use of renewable resources as raw materials for the printing industry [1–5]. As vegetable oils, palm oil are biodegradable, non-toxic, non-pollutant since the vegetable oils contain a higher percentage of desirable fatty acid [5]. The use of palm oil in printing ink development can increase the diversification of palm oil end products as a substitute for petroleum-based products, as palm oil has an abundant source in Indonesia. Moreover, it has the potential to reduce the dependency on imported printing ink raw materials.

In the present work, palm oil fatty acid methyl ester (FAME) is applied with two types of drying oil to develop eco-friendly varnish and ink. In terms of the printability of inks, this study aimed to investigate the surface properties of palm oil-based offset ink on gloss-coated paper surfaces in comparison with conventional ink. Printability evaluations on papers with specified surface properties were carried out according to the criteria of contact angle and surface free energy.

2. Methods

2.1 Materials

Rosin-modified phenolic resin (Arakawa Chemical Industries Ltd., Japan) was used as the resin. Soybean oil and Linseed oil were applied as drying oil. Palm oil ester - FAME (fatty acid methyl ester) prepared by the base-catalyzed transesterification of palm olein triglycerides. For the ink preparation, phthalocyanine beta blue (CI Pigment Blue 15:3, Solafast Blue BG-WS, India) was used as a pigment. The additives used include Triethanolamine (TEA), Butylated hydroxytoluene (BHT), Polyethylene (PE) wax compound (Hubergroup India Pvt. Ltd. India), Polytetrafluoroethylene (PTFE) wax paste (Hubergroup India Pvt. Ltd. India), and Special Drier (Hubergroup India Pvt. Ltd. India). As conventional offset ink, we used Inkredible Surprize TK Cyan from Hubergroup India.

2.2 Preparation of varnishes and inks

The varnish and printing ink preparation were slightly modified from Leach and Roy [3,6] and formulated as the composition given in Table 1 and Table 2. Drying oil was heated to 150°C in a five-necked one-liter glass kettle fitted with a mechanical stirrer, thermometer, condenser, and nitrogen inlet. The resin was added, heated to 180°C, and cooked for 30 minutes. At 160°C, FAME was added, followed by TEA. BHT was then added and stirred for 5 minutes to allow the varnish to properly mix.

Table 1. Varnish formulation.

Raw Materials	Varnish (%w/w)	
	FAME-LO	FAME-SO
Linseed Oil	23	-
Soybean Oil	-	23
Resin	43	43
FAME	33	33
Additives[a]	1	1

[a] TEA and BHT

The inks were prepared by mixing the composition listed in and then milled with the help of a Laboratory 3-roll mill (Zili Chemical Machinery Co., Ltd., China) to obtain fine size particles.

Table 2. Formulation of the printing ink by using different varnishes.

Raw Materials	Ink (%w/w)	
	FAME-LO	FAME-SO
Varnish FAME-LO	72	-
Varnish FAME-SO	-	72
Pigment[a]	18	18
FAME	4.5	4.5
Additives[b]	5.5	4.5

[a] Blue 15:3
[b] PTFE Wax, PE Wax, BHT, and Drier

2.3 Tests and measurements

Printing tests of inks were carried out using the IGT-A2 printability tester (IGT Testing System, Netherland) on 120 g/m^2 gloss-coated paper at a pressure of 400 N with an average ink transfer amount on paper 1.08 g/m^2. The contact angle and surface energy values of the printed and unprinted papers were measured and analyzed by the sessile drop method, using Phoenix 300 Contact Angle Analyzer

(Surface Electro-Optics, Korea). Tables 3 and 4 show the test liquids used in this study. Water is represented as the polar liquid, methanol as the semi-polar liquid, and hexane as the nonpolar liquid [7]. Contact angles were determined by capturing droplet images with a camera-based contact angle analysis system.

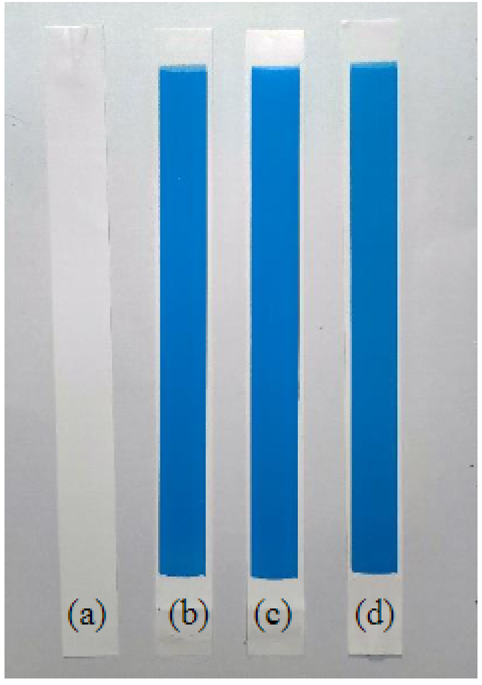

Figure 1. The samples of (a) unprinted paper (gloss-coated paper), (b) printed FAME-LO ink, (c) printed FAME-SO ink, and (d) printed conventional ink.

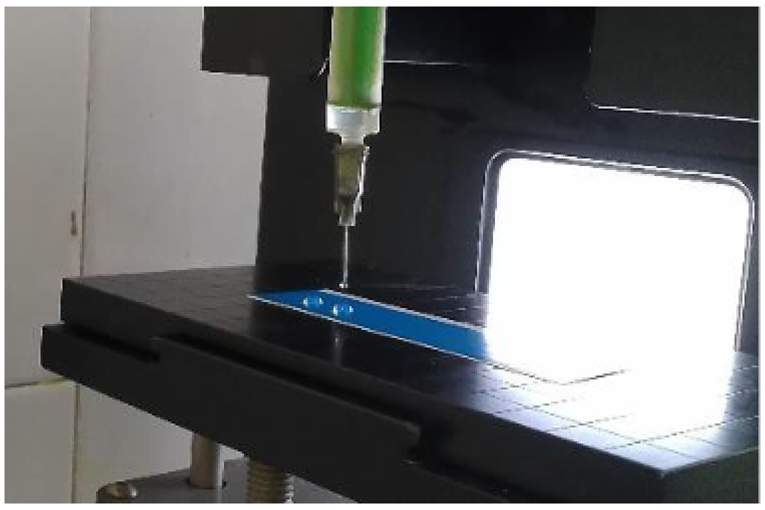

Figure 2. Sessile drop method of liquid on the sample surface

The surface free energy (SFE) and its components were calculated using Surfaceware 8 software with different methods, Girifalco-Good-Fowkes-Young (GGFY), Owens-Wendt (OW), and van Oss-Chaudhury-Good (vOCG). Water with a known total liquid SFE value of 72.8 mJ/m^2 was used for GGFY method. The probe liquid parameters used for the determination of the printed and unprinted surface energy are listed in tables 2 and 3, for Owen-Wendt and vOCG, respectively.

Table 3. The probe liquids used for Surface Energy calculation according to the Owen-Wendt method [7,8]

Liquid	γ_l	γ_l^p	γ_l^d
Water	72.8	21.8	51
Hexane	18.4	18.4	0

Table 4. The probe liquids used for Surface Energy calculation according to the vOCG method [7,8]

Liquid	γ_l	γ_l^{LW}	γ_l^{AB}	γ_l^{+}	γ_l^{-}
Water	72.8	21.8	51	25.5	25.5
Methanol	22.5	18.2	4.3	0.06	77
Hexane	18.4	18.4	0	0	0

The relationship between a static contact angle and the surface energy forces is due to the Young equation [9]. It is given as:

$$\gamma_l \cos\theta = \gamma_s - \gamma_{sl} \tag{1}$$

where γ_l and γ_s denotes SFE of liquid and solid, respectively. γ_{sl} means the interfacial free energy between the solid and θ is the contact angle.

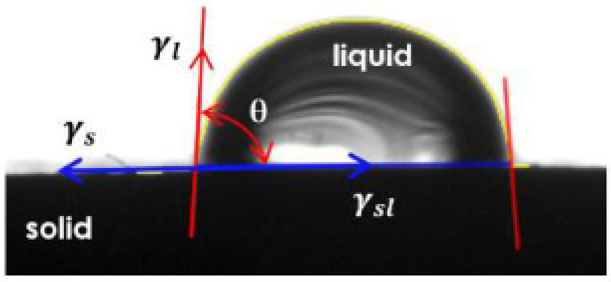

Figure 3. Contact angle of a liquid on a solid surface

According to Girifalco-Good-Fowkes-Young method, the surface energy was calculated by presented equation [10,11]:

$$\cos\theta = -1 + {2\left(\gamma_s^d \gamma_l^d\right)^{1/2}} \Big/ {\gamma_l} \tag{2}$$

where $\gamma_l = \gamma_l^d + \gamma_l^p$ represents the combined contributions of the dispersion and polar components of the aqueous phase and γ_s^d is the surface energy of the solid having only a dispersion component. Then Owens and Wendt suggested the following form [12]:

$$(1 + \cos\theta)\gamma_l = 2(\gamma_s^d \gamma_l^d)^{1/2} + 2(\gamma_s^p \gamma_l^p)^{1/2} \tag{3}$$

where γ_s^d and γ_s^p are the dispersion and polar force components of γ_s, respectively.

The latest version of SFE calculation is the Lifshitz-van der Waals/acid-base approach, initiated by van Oss, Chaudhuri, and Good [13]. This vOCG method split the polar component (γ^{AB}) to the acid component (electron-acceptor: γ^+) and the base component (electron-donor: γ^-), in such a way that $\gamma^{AB} = 2(\gamma^+\gamma^-)^{1/2}$. This method requires contact angle from three liquids, then the following correlation expressed as:

$$(1 + \cos\theta)\gamma_l = 2\left(\sqrt{\gamma_s^{LW}\gamma_l^{LW}} + \sqrt{\gamma_s^+\gamma_l^-} + \sqrt{\gamma_s^-\gamma_l^+}\right) \tag{4}$$

3. Results and Discussion

Contact angles of the test liquids (water, methanol, and hexane) on the surface of the unprinted paper, printed FAME-LO, FAME-SO, and conventional inks as control are given in Figure 4. The value of each contact angle was the average of three droplets. Contact angle measurement shows the wetting performance of liquids applied to solid surfaces and describes the hydrophobic or hydrophilic behavior of a material [14,15]. As it can be seen in Figure 4, the contact angle of the water drops on the paper surfaces increased in the printed FAME-LO ink. It could be indicated that printed FAME-LO ink on paper surfaces has greater water resistance than unprinted paper surfaces and other printed ones. Meanwhile, the values of water contact angles indicate that the lowest wettability was determinate for printed FAME-LO ink. Larger contact angles demonstrate low wettability, whereas smaller contact angles demonstrate high wettability [16]. Printed FAME-SO and conventional inks have great wettability on coated paper. Furthermore, either printed or unprinted paper was nonpolar and hydrophobic materials, due to the droplets of nonpolar liquid (hexane) on its surface being wet quickly, reacting to the sample's surface.

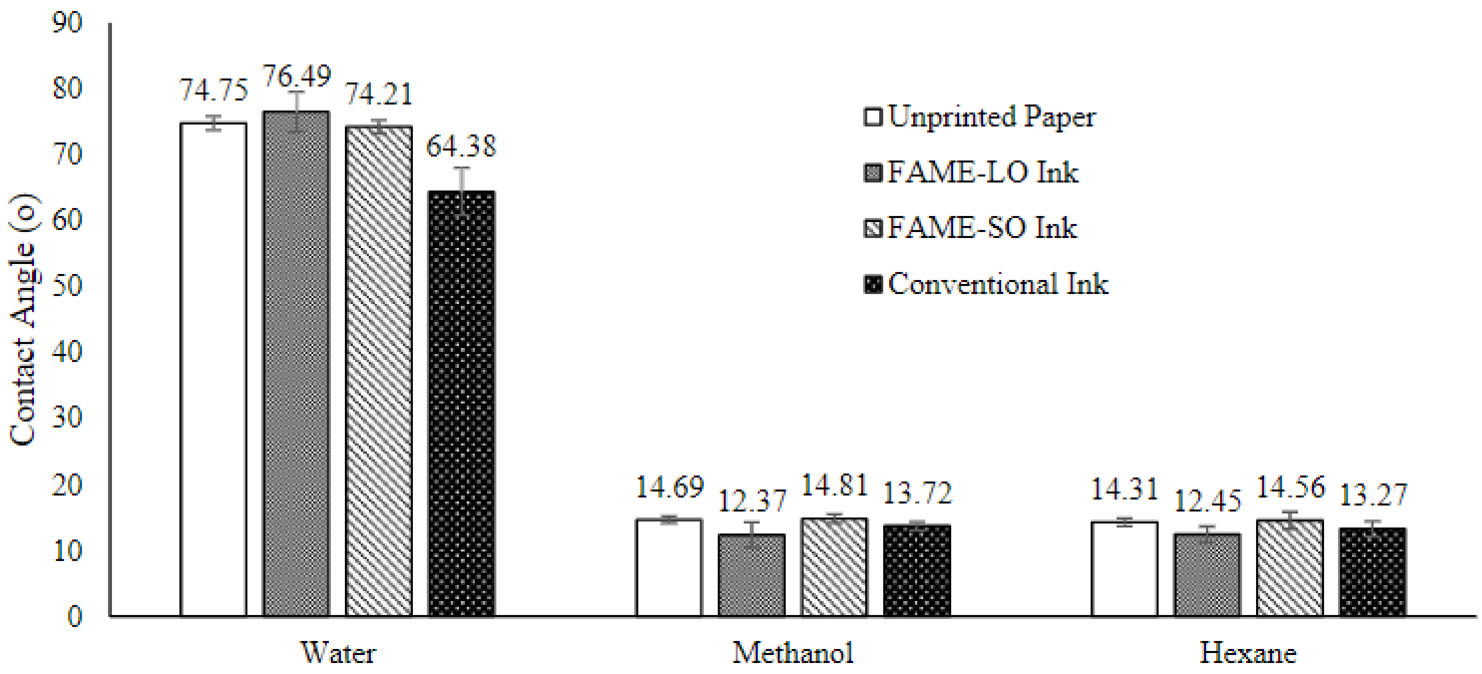

Figure 4. Contact angles of the test liquids on the printed and unprinted paper

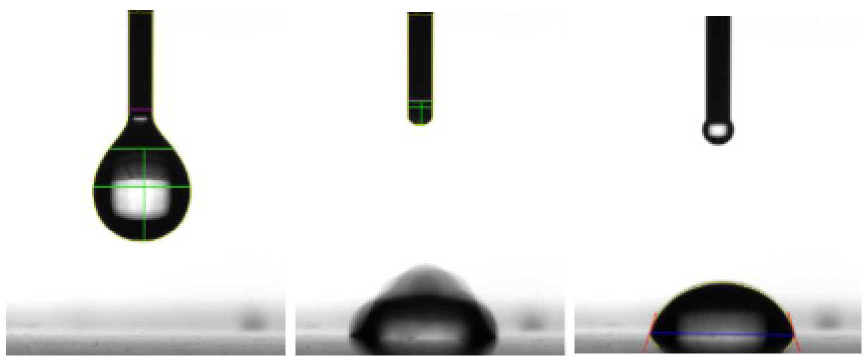

Figure 5. Representative figures of contact angle measurements of printed FAME-SO inks 73.47°

According to the contact angle values, the surface free energy (SFE) of samples and their components were calculated using three previously known methods, Girifalco-Good-Fowkes-Young (GGFY), Owen-Wendt (OW), and van Oss-Chaudhury-Good (vOCG). The force available on a solid surface to attract liquid molecules is referred to as SFE. Unlike surface tension, SFE cannot be measured directly; it must be calculated using contact angle measurements [15]. The values of the SFE and its components are presented in Table 5.

Table 5. SFE and its components (in mJ/m^2) of ink film surface calculated with various methods

Samples	GGFY	Owen-Wendt			vOCG				
	γ_s	γ_s	γ_s^d	γ_s^p	γ_s	γ_s^{LW}	γ_s^{AB}	γ_s^+	γ_s^-
Unprinted Paper	35.84	31.35	17.83	13.52	21.08	17.83	3.24	0.11	23.68
FAME-LO ink	34.19	30.33	17.97	12.36	21.16	17.97	3.19	0.12	21.42
FAME-SO ink	36.36	31.68	17.81	13.87	21.09	17.81	3.27	0.11	24.36
Conventional ink	46.10	38.47	17.91	20.56	21.57	17.91	3.66	0.09	37.37

The values of total SFE (γ_s) of printed inks was higher than control unprinted paper surface, except for printed FAME-LO ink calculated by GGFY and OW methods. It was revealed that the surface energy of printed gloss-coated papers decreased by FAME-LO ink than that of other inks. Since SFE will adversely affect the bond strength of the ink to the paper, higher SFE will be advantageous, especially for multicolor and dense prints requiring good ink adhesion. This will increase the acceptance of other colors to be printed on the previous color in multicolor prints [14]. Furthermore, it was realized that there is a relation between contact angle and surface energy. When the contact angle is high, surface energy decreases and the absorbency of the ink-film lowers. These results are in line with several previous studies [14,15,17,18].

The dispersive components (γ_s^d) obtained by the OW are similar with vOCG (γ_s^{LW}) method and higher than its polar components. The values of water contact angle strongly affect the values of polar components of surface free energy. In general, the printing process increases the values of polar and dispersive components and also the total SFE increase [19]. It can be observed as well from vOCG method calculation values that Lewis base (γ_s^-) SFE component is higher than Lewis acid (γ_s^+) for all samples. Owing to the insignificant γ_s^{AB} value it may be presumed that these surfaces will show properties of nonpolar materials. Surfaces with high SFE have a greater tendency to adsorb atmospheric materials (e.g., dust particles or moisture), which reduces wettability [16]. It can also be observed that there is great similarity between the polar dan dispersive component of inks and paper surface, which indicates the better the wetting and adhesion on the surface of printing substrate [20].

4. Conclusion

Palm oil-based cyan offset printing inks have been successfully synthesized and its surface properties on gloss-coated paper surfaces in terms of printability have been investigated. This study shows that as the liquid contact angle increased, the surface energy decreased. On the gloss-coated paper, the printed FAME-LO ink showed the lowest surface free energy value, while the surface energy of FAME-SO ink printed samples 36.36 mJ/m^2 was higher than the unprinted paper 35.84 mJ/m^2, although lower than conventional ink. Since high surface energy will affect the bond strength of the ink to the substrate, this will be an advantage for FAME-SO ink on multicolor prints requiring good ink adhesion, which will enlarge the acceptance of other colors to be printed on the previous color.

5. Funding

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