Synthesis of mono diacyl glycerol from palm fatty acid distillate and glycerol as antistatic agents on plastics

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Abstract. Palm fatty acid distillate (PFAD) production as a CPO refining by-product reaches 1 million tons per year. PFAD can increase its added value by making it as mono- diacylglycerol (MDAG). MDAG is a nonionic surfactant. It has biodegradable properties and not irritant compared to cationic and anionic surfactants. This study aimed to determine the best temperature and reaction time for the synthesis of MDAG, to characterize the synthesized MDAG, and to apply it as antistatic agent on plastic. MDAG was produced through chemical synthesis by reacting PFAD and glycerol through an esterification process with PFAD to glycerol molar ratio 1:6. The catalyst that used in this study was para-toluenesulfonic acid (pTSA). Synthesis was carried out under various conditions of temperature (100 °C, 120 °C, and 150 °C) and reaction time (90 min and 120 min). The best conditions of synthesis reaction were at temperature 150 °C, molar ratio 1:6, and 90 min reaction time. The yield of MDAG was 14.33 %, pH value of 5, hydroxyl value 344.01 mg KOH/g that contain 93.29 % MAG, and 6.71 % DAG. The MDAG obtained from synthesis was white, dry texture, and odorless. The synthesized MDAG has met the standard because the minimum standard MAG content at commercial MDAG was 91 %. Tests on plastics showed that synthesized MDAG and commercial MDAG showed the same surface resistivity value of 1.0x1012 ohm/sq.

Key words : monodiacyl glycerol, palm fatty acid distillate, glycerol, esterification, antistatic

1. Introduction
MDAG belongs to the partial glycerides group because MDAG is an ester of glycerol with free fatty acids that have no reacting hydroxyl groups. MDAG consists of two acylglycerol fractions, namely monoacylglycerol and diacylglycerol. Monoacylglycerol has two free hydroxyl groups, while diacylglycerol has one free hydroxyl group. This free hydroxyl group which allows MDAG is also used as an antistatic agent, both in bio-composite material and in plastics.

Antistatic agent is one of the most important additives in plastics that serves to avoid the phenomenon of static electricity between two different materials. Static electricity occurs due to excess or lack of electrons found on the surface. This imbalance can be created by the friction of two materials or through the process of induction with ionized air [1]. Static electricity is very detrimental to plastic production because it often causes the plastic molding machine to disrupt production. In addition, static electricity also damages the aesthetics of plastic because it causes dust and dirt to empt
on the plastic surface. MDAG has a free hydroxyl group which has hydrophilic properties so that it forms a thin layer and reduces surface conductivity thereby avoiding the accumulation of static electricity on the surface.

The amount of monoacylglycerol and diacylglycerol fractions in MDAG is determined by the success of the MDAG synthesis process, one of which is influenced by temperature and time. The aim of this study was to determine the best temperature and time for MDAG synthesis, the characteristics of MDAG synthesized, and to make a plastic PE prototype with antistatic MDAG synthesized. This research is expected to support the government's plan to prioritize industries based on PFAD, as well as add value to PFAD, and also make the synthesized MDAG an additive for bio-composite materials, given its biodegradable nature.

2. Materials and Methods

2.1. Materials
The equipment used were a stirred tank reactor with a capacity of 2 kg/batch, a pilot scale reactor with a capacity of 20 kg/batch, a purification reactor with a capacity of 5 L/batch, injection molding, Work Surface Tester ST-4 (Simco Japan, Inc.), a funnel Buchner glass cup and stirrer hotplate. The materials used in this study were PFAD, glycerol with a purity of 99.7%, para-toluene sulfonic acid (pTSA), zeolite, NaHCO₃, n-hexane, ethanol 96%, Whatman filter paper No.42, and polypropylene plastic seeds.

2.2. Characterization of Raw Materials
Characterization of raw materials was carried out on PFAD which included free fatty acid, melting points and pH value.

2.3. Synthesis of Crude MDAG
The process was performed by reacting PFAD and glycerol in the esterification reactor with PFAD and glycerol molar ratio of 1:6, 1.5% (w/w) pTSA catalyst and 5% (w/w) zeolite adsorbent. The process was carried out under vacuum conditions with temperatures of 100 °C, 120 °C, 150 °C and reaction time of 90 and 120 min. The characterization of crude MDAG included free fatty acid, melting points, and pH values.

2.4. Purification of Crude MDAG with Solvent Extraction
The purification process began with the addition of a hexane-ethanol solvent mixture with a 1:1 volume ratio into a crude MDAG. The weight and volume ratio of crude MDAG and a solvent was 1:5. Neutralization process was performed by adding NaHCO₃ 0.3% (w/w) to neutralize the fatty acid and residual catalyst.

The excess NaHCO₃ then precipitated and removed from the solvent fraction. The solvent fraction was crystallized at 60°C for 72 hours. From the crystallization process, purified MDAG flakes will be precipitated and can be separated from the solvent through a vacuum filtration. Purified MDAG then washed with ethanol to remove the remaining glycerol. The yield was calculated by dividing the purified MDAG by crude MDAG. Characterization of MDAG included color, aroma, texture, free fatty acid, melting point, pH value and hydroxyl number.

2.5. MDAG Synthesis in a Pilot Scale
MDAG synthesis in a pilot scale was conducted using a reactor with a capacity of 20 kg/batch. The process conditions used in the pilot scale were the best reaction conditions at the laboratory scale. The mono and diacylglycerol content of MDAG was determined by Thin Layer Chromatographic analysis based on Sherma and Fried (2005). The chromatographic patterns were then calculated using ImageJ software.
2.6. MDAG Application on Plastic Plates
Application of MDAG as an antistatic agent was carried out at PT DIC Astra Chemicals, Pulogadung. 300 g of polypropylene was mixed with 0.3 g purified MDAG then put into an injection molding machine. The molding process was performed at a temperature of 235 °C. The surface resistance was measured by Work Surface Tester ST-4 (Simco Japan, Inc.), then was converted to surface resistivity value.

2.7. Data analysis
The experimental design in this study used a factorial completely randomized design (CRD) with 2 factors and 2 replications. This randomized design was used to determine the effect of time and temperature to the characteristics of the crude MDAG. The data analysis was performed using SPSS 25 software with ANOVA test at 0.05 followed by Duncan test.

3. Results and Discussion

3.1. Characterization of Raw Materials
PFAD is a by-product from the Crude Palm Oil (CPO) refining process. It was produced from the palm oil deodorization process. The characteristics of PFAD used in this study are presented in Table 1.

| Parameter               | Result | SNI 01-0015-1987 Crude PFAD |
|-------------------------|--------|-----------------------------|
| Fatty Acid Content (%)  | 81.67  | min 80                      |
| Melting Point (°C)      | 47     | -                           |
| Water Content (%)       | 0.89   | max 1.0                     |
| pH                      | 5      | -                           |

The free fatty acid content of PFAD was 81.67 %. The level of free fatty acids will determine the characteristics of crude MDAG. The higher free fatty acid presence, the more fatty acid will be converted into monoacylglycerol and diacylglycerol. Melting point according to Mardaweni et al. [2] will be determined by the fatty acids content of oil or fat. PFAD is dominated by saturated fatty acids (palmitate), making them solid at room temperature. Water content of PFAD was below the maximum value of Indonesian Standard (SNI). Based on the free fatty acids and water content, it can be concluded that PFAD used in this study has a good quality. The results of pH value showed that the PFAD was acidic.

3.2. Synthesis of Crude MDAG
MDAG can be produced through the hydrolysis process using enzymes [3], transesterification [4] and esterification [2]. MDAG in this study was synthesized through the esterification process. This process was faster reaction times and more economical than enzymatic hydrolysis or transesterification.

Esterification is classified as a reversible endothermic reaction so 100 % convection is not possible. The solution to increasing conversion of esterification is by making excess reactants, so that the equilibrium of the reaction will be shifted to the right which causes higher conversion values and produces more products [5]. The mechanism of MDAG synthesis by esterification is shown in Figure 1. The crude MDAG resulted from the esterification process then characterized by free fatty acid content, melting points and pH values.
3.3. Free Fatty Acid Content of Crude MDAG
Free fatty acid content (FFA) of crude MDAG was in the range of 4.26-72.72 % (Figure 2). High FFA content indicated that the esterification process was not optimal. This can be caused by the lack of substitution of hydrogen groups by free fatty acids [6]. Meanwhile, the lower levels of FFA indicated that the conversion of FFA into mono-, di- and triacylglycerol was increased [7]. Based on ANOVA test results with a significance level of 5 % (P < 0.05), temperature and time had a significant effect on FFA content. Duncan’s tests showed that each treatment gave different results of FFA.

3.4. Melting Point of Crude MDAG
The melting point of MDAG will figure the fatty acid constituents. The MDAG has a higher melting point compared to PFAD. According to McClements and Decker [8], changing of free fatty acids into MAG (monoacylglycerol), DAG (diacylglycerol), and TAG (triacylglycerol) will change the melting point and other physical properties. Melting point of MDAG at the treatment of 100 °C for 90 min, 100 °C for 120 min, 120 °C for 90 min, 120 °C for 120 min, 150 °C for 90 min, and 150 °C for 120 min was 51 °C, 50 °C, 50 °C, 51 °C, 50 °C, 52 °C, 50 °C, 50 °C, and 50.50 °C, respectively. ANOVA test results with a significance level of 5 % (P < 0.05) showed that the temperature and reaction time did not affect the melting point of MDAG.

3.5. pH value of Crude MDAG
Measurement of pH values aimed to determine the degree of acidity of the crude MDAG. The crude MDAG in each treatments had a pH value of 5. The pH value is only relevant for knowing the acidity of a material or ingredient. To determine the content of organic acids, free fatty acids content are more relevant for measuring both dissociated and undisassociated fatty acid. While pH measurement with a strip indicator or pH meter measuring only dissociated fatty acid [9]. The results of ANOVA test with a significance level of 5 % (P < 0.05) showed that the temperature and reaction time did not give a significance effect on the pH value of crude MDAG.
3.6. Purification of MDAG

The MDAG purification process aimed to remove residuals from the reaction, such as unreacted free fatty acids, impurities, as well as catalyst residues and triacylglycerol (TAG) as a reaction product that cannot be avoided. Thus, the purity of MDAG can be increased up to above 90 %. Free fatty acids must be removed from the end product because according to Ketaren [10], oil products must have the lowest levels of free fat. A free fat will causes rancidity due to its easily to oxidized. Meanwhile, TAG is not desired on the product because it causes an oily texture. Triacylglycerol also does not have a free hydroxyl group thereby reducing the antistatic and emulsifier properties of MDAG.

Purification of MDAG can be performed by several methods include molecular distillation, column chromatography and solvent extraction. Purification method of crude MDAG adopted in this study was based on the Setyaningsih et al. [11] with modifications. It was started with solvent extraction, followed by alkaline saponification and crystallization. The solvents used were hexane and alcohol. Hexane was used to dissolve TAG, while alcohol was used to dissolve saponified soap, residual glycerol and improve the physical characteristics of MDAG. Saponification aimed to neutralized free fatty acids, which was applied by the addition of NaHCO3 as much as 0.3 \% (w/w) of the crude MDAG. The alkaline concentration which used in this study was lower than previous studies because based on Mardaweni et al. [2], when the higher the alkaline added, the lower yield of pure MDAG obtained. The purification process ended with crystallization at 6 °C. At that temperature, MDAG will become solid while TAG and free fatty acids remained in a solvent fraction [11].

3.7. Yield of Purified MDAG

Crude MDAG that can be refined only 3 samples out of a total of 6 samples. MDAG with the treatment of 100 °C with reaction time of 90 and 120 min, and at 150 °C with reaction time 120 min did not form MDAG flakes after crystallisation, so that the yield of pure MDAG could not be calculated. This was caused by two different factors. At the treatment of 100 °C with the reaction time of 90 and 120 min, the absence of yield caused by the low conversion of free fatty acids into MDAG. This can be seen from high FFA content on crude MDAG, which was 71.49 \% for the treatment of 100 °C 90 min and 72.72 \% for the treatment 100 °C 120 min. Meanwhile, at the treatment of 150 °C 120 min, the absence of yield was caused by the esterification process leads to TAG formation, so there was no MDAG flakes formed during the crystallization process. This result is consistent with a previous study [12] which stated the higher temperature and reaction time used produces more oil or triacylglycerol (TAG). Yield of purified MDAG is shown in Figure 3.

![Figure 3. Yield of purified MDAG.](image-url)
3.8. Visual and Physical Characteristics of MDAG

The physical characteristics of MDAG are expected to be the same as the commercial product which has a white colour, no specific odour and dry texture. The visual characteristics of purified MDAG are shown in Table 2. MDAG which has same physical characteristics as a commercial one was MDAG obtained at 150 °C with a reaction time of 90 min.

Table 2. Visual and physical characteristics of MDAG.

| MDAG          | Odour         | Colour      | Texture  | Visual Appearance |
|---------------|---------------|-------------|----------|-------------------|
| Commercial    | No odour      | White       | Dry powder |                   |
| 120 °C 90 min | No odour      | Yellowish-white | Oily      |                   |
| 120 °C 120 min| No odour      | Yellowish-white | Oily      |                   |
| 150 °C 90 min | No odour      | White       | Dry powder |                   |

3.9. Free Fatty Acid Content of Purified MDAG

Purified MDAG must have low free fatty acid (FFA) content because these FFA determine product stability during storage. Products which have high levels FFA will be easily oxidized, causing a rancid odour on the product [10]. MDAG synthesis was performed at a temperature of 150 °C for 90 min to produce MDAG with the lowest free fatty acid content (0.51 %), while FFA in commercial MDAG were not detected. The results of FFA content from each sample are shown in Figure 4.

![Figure 4. Free fatty acid content of pure MDAG.](image)
The results of the ANOVA test at the significance level of 5 % (P < 0.05) showed differences in the FFA content of the purified MDAG. Duncan's tests showed that MDAG which was synthesized at 120 °C with 90 and 120 min reaction time had the same FFA, but different from commercial MDAG and MDAG which was synthesized at 150 °C for 90 min.

3.10. Melting Point of Purified MDAG
The melting point of MDAG after purification tends to increased compared to crude MDAG. This can be caused by the changing of fatty acid distribution at the crude and purified MDAG [8]. The higher melting point indicated that MDAG was composed of saturated fatty acids, while the lower melting point indicated that MDAG was composed of unsaturated fatty acids. The melting point of MDAG which was synthesized at 120 °C with a reaction time of 90 min, 120 °C-120 min, and 150 °C-90 min was 56.50 °C, 57 °C and 57 °C, respectively. While the melting point of commercial MDAG was 66 °C. The melting point of MDAG from this study corresponds to the MDAG melting point range in general, which is 54 - 64 °C according to Kishore [13].

The results of the ANOVA test at the significance level of 5 % (P < 0.05) showed a difference in the melting point of the MDAG. Duncan's tests showed the MDAG synthesized under various conditions were not significantly different from each other, but significantly different from the commercial MDAG. This can be caused by commercial MDAG was synthesized from hydrogenated free fatty acids.

3.11. pH value of Purified MDAG
The pH value of MDAG did not change before and after purification. All pH values of the MDAG was 5, as well as the commercial MDAG. The commercial MDAG used in this study had a different pH value from a previous study [14] which uses MDAG with a pH value of 6. This difference will not affect the ability of MDAG as a surface-active agent, especially for non-ionic surfactants group [15]. This is because the ability of MDAG will be influenced by the composition of MAG and DAG, in which the higher the MAG, the MDAG will have better surfactant ability [13]. The results of the ANOVA test at significance level of 5 % (P < 0.05) showed no difference between pH value of MDAG sample.

3.12. Hydroxyl Number of Purified MDAG

![Figure 5. MDAG hydroxyl numbers.](image)
The hydroxyl number test shows the number of free hydroxyl groups on surfactants. The hydroxyl number also determines the quality of MDAG, i.e., a higher hydroxyl number provides better antistatic properties. The hydroxyl number is related to the MAG and DAG fractions. A higher hydroxyl number indicates a higher MAG fraction. The best hydroxyl number was obtained from MDAG synthesized at 150 °C-90 min with a hydroxyl number of 349.03 mg KOH/g (Figure 5).

The results of the ANOVA test at the level of 5% (P < 0.05) showed that differences in the hydroxyl number in each MDAG sample. Duncan's test showed that MDAG synthesized under various conditions produced hydroxyl numbers that different from each other.

3.13. Duplicating MDAG Synthesis in the Pilot Scale

Synthesis of MDAG in the pilot scale was performed by the esterification condition that produces the best characteristics of MDAG at the laboratory scale, i.e., 150 °C for 90 min. Modifications were conducted to purification process in the pilot scale, by changing the crystallization time and the washing process. Purification of MDAG in a pilot scale uses the addition of hexane before washing with technical ethanol. So, there were two cycles of solvent extraction with hexane. This step was applied because of the high content of TAG on the crude MDAG. Thus, additional extraction with hexane was required.

The duration of crystallization for purification at pilot scale was reduced from 72 hours to 24 hours to reduce the amount of TAG trapped in the MDAG fraction. A comparison between purification process at the laboratory and pilot scale can be seen in Table 3.

| Process                        | Laboratory scale | Pilot scale |
|--------------------------------|------------------|-------------|
| Crystallization                | 72 h             | 24 h        |
| Second hexane extraction       | -                | 24 h        |

The pilot-scale MDAG has a white-dry powder and no specific odour. Free fatty acids were not detected on the pilot scale MDAG. This indicated that the MDAG will be stable during storage, and also not easily oxidized due to low free fatty acids [10]. The melting point of the pilot scale MDAG was lower than the commercial product, but it still in the MDAG melting point range in general, which is 54 - 64 °C [13]. This is due to the composition of its constituent fatty acids. Commercial MDAG is produced from hydrogenated fatty acids so that they are composed by saturated fatty acids, thus it has a higher melting points [16].

Pilot scale and commercial MDAG had pH value of 5. Pilot scale MDAG had a lower hydroxyl number than commercial MDAG. This can be caused by monoacyl glycerol (MAG) content on the pilot scale MDAG was lower than the commercial one. Pilot scale MDAG contained 93.29 % MAG, while commercial MDAG contains 100 % MAG. A comparison of commercial MDAG and pilot scale characteristics is shown in Table 4.

The quality standard of MDAG according to US Food and Drugs Administration (FDA) [17] is MDAG has at least 91 % of MAG content with FFA content no more than 0.5 %. therefore, MDAG which synthesized on this study suitable with FDA standards with undetectable FFA and MAG content up to 93.29 %. High hydroxyl numbers also indicated that MDAG from this study have good antistatic properties.
Table 4. Comparison of commercial MDAG and pilot scale characteristics.

| Parameter            | Pilot scale MDAG | Commercial MDAG |
|----------------------|------------------|-----------------|
| Texture              | Dry powder       | Dry powder      |
| Colour               | White            | White           |
| Odour                | No specific odour| No specific odour|
| Visual appearance    |                  |                 |
| Yield (%)            | 14.33            | -               |
| FFA (%)              | -                | -               |
| Melting point (°C)   | 57               | 66              |
| pH                   | 5                | 5               |
| Hydroxyl number (mg KOH/g) | 344.01        | 456.53          |
| MAG (%)              | 93.29            | 100             |

3.14. MDAG Application as Antistatic on Plastic

MDAG as an antistatic agent diffuses to the surface and attracts water molecules to the surface. Free hydroxyl groups on MDAG that have hydrophilic properties will cause MDAG to form a thin layer of water on the surface which will reduce the surface resistivity, thereby preventing the accumulation of static electricity. Antistatic agents reduce surface resistivity in the range of $10^{10}$ to $10^{12}$ ohms/sq. Antistatic agents are commonly used in films, PET bottles and cosmetics [13].

The amount of antistatic agent addition commonly used in plastics according to Hahladakis et al. [18] is 0.1-1.0 % (w/w) plastic pellets. MDAG application as an antistatic agent was performed by mixing polypropylene plastic seeds with MDAG as much as 1 % (w/w), then molding at a temperature of 235 °C to form a plastic plate (Figure 6). From the measurement of surface resistivity, plastic plate which added with MDAG and commercial product produced a same surface resistivity, which was $10^{12}$. Meanwhile, control plastic plate without MDAG addition has a surface resistivity values of $10^{13}$ ohms/sq (Figure 7).

![Figure 6. Plastic plates with MDAG addition as antistatic.](image-url)
4. Conclusion
The research results showed that the best conditions for the synthesis of MDAG through esterification were obtained at 150 °C with a reaction time of 90 min. Purified MDAG had a white-dry powdery texture and no specific odour with a yield of 17.60%. The MDAG produced had FFA content of 0.51%, melting point of 57 °C, pH value of 5 and hydroxyl number of 349.3 mg KOH/g. The reaction conditions used in the laboratory scale were tested in the pilot scale and succeeded in producing MDAG that has the same visual characteristics as MDAG in the laboratory scale, with undetectable FFA, melting point of 57 °C, pH value of 5, and hydroxyl number of 344.01 mg KOH/g. Testing of antistatic properties on plastic plates showed that MDAG addition can change the surface properties of plastic. The synthesized MDAG reduced the surface resistivity of polypropylene plastic plates to $10^{12}$ ohms/sq with the addition of 0.1 % (w/w) MDAG.

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