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Hydrogenated Palm Fatty Acid Distillate as Raw Materials for Magnesium Stearate Alternatives

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Highlights:

- PFAD was used as raw material to produce a solid lubricant, or anti-adherent, for confectionery or pharmaceutical products.
- Solid lubricant Mg-PFAD was produced through saponification of PFAD with MgO, while solid lubricant Mg-HPFAD was produced through hydrogenation of PFAD followed by saponification with MgO.
- The hydrogenated PFAD produced by catalytic transfer hydrogenation (CTH) had a lower iodine value than that produced by gaseous hydrogenation (GH).
- Paracetamol tablets with Mg-PFAD or Mg-HPFAD lubricant showed higher dissolution of active compounds with similar friability, frictiability, and hardness compared to paracetamol tablets with Mg-stearate.

Abstract. Palm fatty acid distillate (PFAD) was used as raw material to produce solid lubricant, or anti-adherent, for confectionery or pharmaceutical products. To improve the degree of saturation, the PFAD was hydrogenated by using two methods: gaseous hydrogenation (GH) and catalytic transfer hydrogenation (CTH) using ammonium formate to produce hydrogenated PFAD (HPFAD). The HPFAD was saponified with MgO to produce magnesium salts of hydrogenated PFAD (Mg-HPFAD). The objective of this research was to investigate the effect of hydrogen concentration and reaction temperature on the iodine value of HPFAD, and to investigate the characteristics of paracetamol tablets when using Mg-HPFAD as lubricant compared to commercial Mg-stearate. The HPFAD produced by CTH had a lower iodine value than the HPFAD produced by GH. The lowest iodine value was obtained after CTH using 3.6 M ammonium formate at 90°C. Paracetamol tablets with Mg-PFAD or Mg-HPFAD lubricant showed higher dissolution of active compounds with similar friability, frictiability, and hardness compared to paracetamol tablets with Mg-stearate.

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1 Introduction

Palm fatty acid distillate (PFAD) is a by-product of crude palm oil (CPO) deodorization during physical refining. PFAD has a high fatty acid content of 82-95%-wt with a yield of approximately 4-6%-wt of CPO [1,2], or approximately 1.86 million tons per year in Indonesia [3]. However, PFAD utilization in Indonesia is still limited; it is mostly exported in crude form. Meanwhile, the production of metallic salts of fatty acids such as magnesium stearate has not yet been developed in Indonesia. Magnesium stearate, or magnesium salt of stearic acid, is used as anti-caking agent, binder compound, excipient, and solid lubricant in the industrial manufacture of medicinal tablets and hard candies [4-8]. PFAD has high potential as raw material for magnesium stearate, but it naturally has a low stearic acid content. According to the commercial specification of stearic acid it has to contain a minimum of 40%-wt stearic acid and 90%-wt of total stearic and palmitic acid [9]. Therefore, PFAD is hydrogenated to increase its stearic content by converting the unsaturated C18 fatty acids into stearic acid.

Lipid hydrogenation can be conducted using two alternative methods: gaseous hydrogenation (GH), which uses high-pressure gaseous hydrogen as a hydrogen donor [10-12], and catalytic transfer hydrogenation (CTH), which uses a hydrogen-donor compound [13-15]. In the GH method, increasing the temperature will increase the hydrogenation reaction rate on the surface of the catalyst, where the highest degree of saturation is obtained at 14 atm pressure and 180°C temperature [10,11]. Smidovnik, *et al.* [13-15] used the CTH method with soybean oil as substrate and ammonium formate as hydrogen donor compound at a temperature of 90°C and catalyst loading of 2% (w/w). It was shown that CTH-hydrogenation of soybean oil by using 10%-wt Pd/C as catalyst resulted in hydrogenated oil with a lower iodine value compared to hydrogenation using 5%-wt Ru/C or 5%-wt Pd/Al_2O_3 [13-16]. Tike, *et al.* [17] investigated the effect of hydrogen donor compounds, donor concentration, substrate concentration, and temperature on CTH using soybean oil as substrate and showed that hydrogenation using 4.2 M ammonium formate as hydrogen-donor resulted in the lowest iodine value of the product.

Production of magnesium stearate is carried out by reacting vegetable or animal oils or fatty acids with alkali or metal oxide. Common methods to produce magnesium stearate are double decomposition, fusion (wet and dry), modified fusion, and modified saponification. In modified fusion, a small amount of water is added to the fatty acid and metal oxide/hydroxide mixture as catalyst to increase the reaction rate at lower temperature. The amount of water added is 0.3-

5.0 moles per mole of MgO. The advantage of the modified fusion method is that the reaction takes only about 3-5 minutes at lower temperature (65-121°C) [18,19].

In general, the raw material for magnesium stearate is obtained from animal or vegetable fats. However, animal or vegetable fats have limited availability and the process requires high energy. Due to its high availability and high free fatty acid content, PFAD is a potential raw material for magnesium stearate production. The objective of this research was to investigate the effect of operating conditions (hydrogen concentration or temperature) on the iodine value of hydrogenated PFAD (HPFAD) and to investigate the characteristics of paracetamol tablets with magnesium salt of HPFAD (Mg-HPFAD) as anti-adherent.

2 Methods

PFAD (iodine value 52.2 g I_2/100 g; slip melting point 45-50°C) was obtained from PT. Tunas Baru Lampung Tbk., Lampung, Indonesia. Ammonium formate, 10% Pd/C catalyst and other chemicals for acid and iodine value analysis were purchased from Sigma Aldrich. Ni/Al_2O_3 catalyst for gaseous hydrogenation of PFAD was provided by the Center for Catalysis and Reaction Engineering (CaRe), Department of Chemical Engineering, Institut Teknologi Bandung, Indonesia. The research was conducted in three stages: 1) production of hydrogenated PFAD (HPFAD); 2) production of magnesium salts of PFAD and HPFAD; and 3) tablet characterization using magnesium salts of PFAD and HPFAD as lubricants.

The fatty acid composition of the PFAD was analyzed by using gas chromatography-mass spectroscopy (GC-MS) at the Department of Chemistry, Universitas Pendidikan Indonesia (UPI) (Table 1).

Table 1 Fatty acid analysis of PFAD using GC-MS.

Components		Mr	GC-MS	
			Composition (%-wt)	X_{FFA} Mr
C14	Miristic acid	228.37	2.27	5.18
C16	Palmitic acid	256.42	46.78	119.95
C18:0	Stearic acid	284.48	3.90	11.09
C18:1	Oleic acid	282.46	35.26	99.59
C18:2	Linoleic acid	280.44	10.87	30.48
C18:3	Linolenic acid	278.43	ND	ND
			Mr PFAD	266.31

2.1 Production of hydrogenated PFAD

Production of hydrogenated PFAD was conducted using two methods: gaseous hydrogenation (GH) and catalytic transfer hydrogenation (CTH). GH used gaseous hydrogen as hydrogen donor at T = 180°C and P = 14 bar by using 19% Ni/Al_2O_3 catalyst [11]. CTH used 4.2 M ammonium formate solution and 10% Pd/C catalyst at temperature 90°C and 600 rpm agitation following the schematic reactor from Tike, *et al*. [17]. The PFAD and HPFAD were analyzed for their iodine and acid value [20].

2.2 Saponification of PFAD or Hydrogenated PFAD with Magnesium Oxide

Saponification of the PFAD was carried out based on the modified fusion method [18], which was further adjusted following the method from Listianingrum, *et al.* [19]. The PFAD and the HPFAD were melted briefly at 60 ºC, followed by MgO addition with an MgO to PFAD molar ratio of 1.1 mol MgO/mol PFAD. Next, a small amount of water was added (0.3-5 moles of water per mole of MgO). After the reaction was completed, the product was removed from the reactor, dried, and milled into fine particles. The product was stated as magnesium salt of PFAD (Mg-PFAD) or magnesium salt of hydrogenated PFAD (Mg-HPFAD), which were used as lubricants in paracetamol tablets for further characterization. The PFAD, HPFAD, Mg-HPFAD, and Mg-stearate were characterized using a Perkin Elmer FT-IR spectrometer using a potassium bromide (KBr) plate in the wavelength range of 4500 cm^{-1} to 400 cm^{-1}.

2.3 Characterization of Mg-PFAD and Mg-HPFAD as Lubricants in Paracetamol Tablets

The magnesium salt of PFAD (Mg-PFAD) and the magnesium salt of hydrogenated PFAD (Mg-PFAD) were tested as lubricants in paracetamol tablet formulation. Paracetamol tablet manufacturing was carried out at the Laboratory of Pharmaceutical Technology for Solid Preparation of the School of Pharmacy School, Institut Teknologi Bandung. The paracetamol tablets were made using wet granulation with the formulation consisting of 92%-wt inner phase, consisting of 500 mg paracetamol, 75 mg dried amylum, 22.5 mg polyvinylpyrrolidone K-25, and 92.5 mg lactose, and 8%-wt of outer phase, consisting of 1%-wt lubricant, 2%-wt talc, and 5%-wt Amprotab. Next, the Mg-PFAD and Mg-HPFAD were used as tablet lubricants and compared to commercial Mg-stearate lubricant. The tablet evaluation parameters were friability, frictiability, and hardness. Friability (%-wt loss) is the tendency of an uncoated tablet to crumble due to knocking, which is calculated from the difference in mass of tablets before and after the test using an Erweka friability tester. Frictiability (%-wt loss) is the resistance of tablets when rubbed against

each other, which is calculated from the difference of mass of tablets before and after the test by using an Erweka frictiability tester. Tablet hardness (kp) is a measure of the tablet's resistance to deformation due to pressure as measured by using a Hardness Tester PharmaTest PTB 111.

3 Result

3.1 Production of Hydrogenated PFAD

The fatty acid composition of the PFAD was analyzed by using gas chromatography-mass spectroscopy (GC-MS) (Table 1). The fatty acid composition was used to estimate the molecular weight of the PFAD. Next, hydrogenation of the PFAD was conducted using two methods: gaseous hydrogenation (GH) and catalytic transfer hydrogenation (CTH). The hydrogenated PFAD (HPFAD) was analyzed for its iodine value, acid value, and color, as presented in Table 2.

Table 2 Process parameters of PFAD and Hydrogenated PFAD (HPFAD) after Gaseous Hydrogenation (GH) and Catalytic Transfer Hydrogenation (CTH).

Process Parameter **Operating conditions**	**PFAD**	**HPFAD-GH,0** P = 14 bar; T = 180 °C	**HPFAD-CTH,0** [NH_4COOH] = 4,2 M; T = 90 °C
Iodine value (g I_2/ 100 g sample)	52.2 ± 1.7	29.2 ± 2.7	14.0 ± 0.2
Reaction conversion (%)	–	44.0 ± 5	73.0 ± 0.5
Acid value (mg KOH/ g sample)	188.4 ± 2.2	187.4 ± 3.5	188.5 ± 1.5
FFA (%)	90.3 ± 1.0	89.9 ± 1.7	90.4 ± 0.7
Color	Yellow	Brownish yellow	Yellowish white

Acid value analysis determined the FFA content, which may indicate the occurrence of secondary reactions such as lipid hydrolysis or fatty acid oxidation. The hydrolysis of fatty acids increases the FFA content, while oxidation reduces the FFA content due to its conversion into other derivative products, such as fatty alcohol. Based on the acid value of the HPFAD in Table 2, the hydrogenation process did not result in a significant change in acid value of the HPFAD, which indicates that no secondary reaction occurred during the hydrogenation of the PFAD. The hydrogenated PFAD produced by CTH had a lower iodine value (higher saturated fatty acid content) than the HPFAD produced by GH (Table 2). This indicates that the HPFAD produced by CTH had a higher saturated fatty acid content due to higher conversion of unsaturated oleic (18:1), linoleic (18:2), and linolenic acid (18:3) to saturated stearic acid (18:0), following the overall hydrogenation reaction according to Naglic [16] in Eq. 1:

$$Linolenic\ acid \xrightarrow{k_1} Linoleic\ acid \xrightarrow{k_2} Oleic\ Acid \xrightarrow{k_3} Stearic\ Acid \quad (1)$$

The overall CTH reaction mechanism with catalyst Pd/C and hydrogen donor of ammonium formate followed Eq. 2 proposed by Tike, *et al.* [17]:

$$HCOONH_4 + R_1\text{-}CH{=}CH\text{-}R_2 \rightarrow R_1\text{-}CH_2\text{-}CH_2\text{-}R_2 + CO_2\uparrow + NH_3\uparrow \quad (2)$$

Rai [21] proposed a calculation method to estimate the composition of saturated and unsaturated fatty acids of hydrogenated oil product by using the iodine value (Eqs. 3-5), with the two following assumptions:

1. An iodine value of HPFAD ($0 < IV_{HPFAD} < IV_{oleic\ acid}$) indicates that the linolenic/linoleic acid has been converted completely into oleic/stearic acid.
2. Triglyceride and unsaponifiable compounds do not affect the iodine value.

$$\%Oleic\ acid = \frac{IV\ product}{IV\ oleic\ acid}\ x\ 100 \quad (3)$$

$$\%Other\ compound = \ 100\% - \%FFA \quad (4)$$

$$\%Saturated\ FA = \ 100\% - \%other\ comp. - \%Oleic\ acid \quad (5)$$

The effect of temperature and concentration of ammonium formate on the conversion of the hydrogenation reaction using CTH was further investigated; the result is presented in Table 3. The estimate saturated and unsaturated fatty acid composition of the HPFAD is shown in Figure 1, which shows that hydrogenation significantly reduced the iodine value. However, the HPFAD had a slightly lower saturated fatty acid content than commercial Mg-stearate.

Table 3 Iodine value of HPFAD produced by CTH at 10% Pd/C; Catalyst loading 2%-wt PFAD; 600 rpm; reaction time 90 minutes.

Hydrogenated PFAD	Process Parameter		Iodine value (mg I_2/g sample)	C=C Conversion (%)
	Temperature (°C)	NH_4COOH Concentration (M)		
PFAD (feed)			52.2 ± 1.7	0
HPFAD CTH-A70	70	4.2	19.1 ± 1.2	63
HPFAD CTH-A80	80	4.2	16.1 ± 1.4	69
HPFAD CTH-A90	90	4.2	14.0 ± 0.2	73
HPFAD CTH-B	90	2.3	7.1 ± 0.9	86
HPFAD CTH-C	90	3.6	5.1 ± 0.8	90

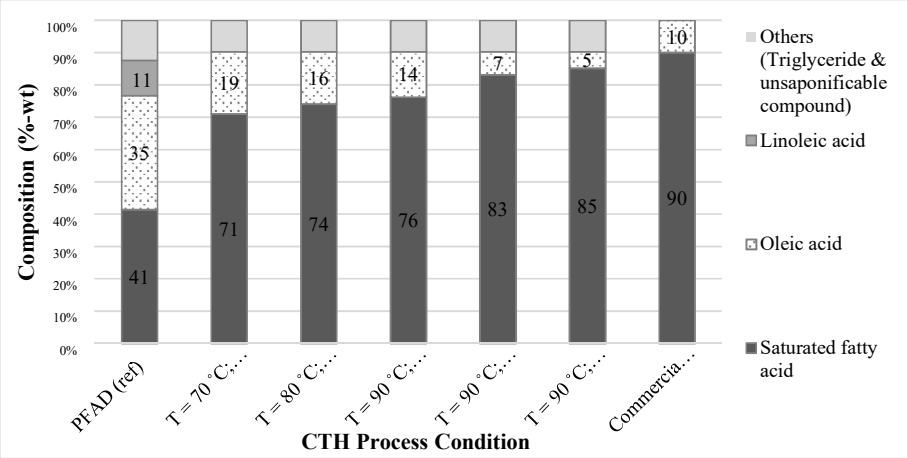

Figure 1 Estimated composition of hydrogenated PFAD produced by CTH at various temperatures and donor concentrations.

Table 3 and Figure 1 show that a higher temperature promoted higher conversion, which resulted in a higher saturated fatty acid content in the HPFAD. The HPFAD CTH-C that was produced through CTH with 3.6 M ammonium formate at 90 °C had the lowest iodine value and the highest saturated fatty acid content; it was used as lubricant in paracetamol tablets for further characterization.

3.2 Characterization of Mg-PFAD and Mg-HPFAD as Magnesium Stearate Alternatives

The PFAD and HPFAD CTH-C were saponified using magnesium oxide (MgO) to produce magnesium salts of PFAD (Mg-PFAD) or magnesium salts of hydrogenated PFAD (Mg-HPFAD). The PFAD, HPFAD, Mg-PFAD, Mg-HPFAD were analyzed using FTIR and compared to Mg-stearate (Figure 2).

Jun [22] states that the peak at 1704 cm^{-1} may be attributed to the stretching of the C=O group, while the peak near 3000 cm^{-1} may be attributed to stretching of the -OH group. This result corresponded well with the absorption bands at around 1700 cm^{-1} in PFAD and HPFAD, which may be contributed by C=O bond stretching of the fatty acid and did not occur in the spectra of the Mg-HPFAD and the Mg-stearate due to the saponification reaction. The absorption band at 1321 cm^{-1}, which occurred in the Mg-HPFAD spectra but was absent in those of the Mg-stearate, may be attributed to the C=C bond. The peaks at 547, 686, 1100, and 1577 cm^{-1} occurred in the spectra of the hydrogenated PFAD, but not in the spectra of the PFAD. This may be correlated to the changes during the hydrogenation reaction.

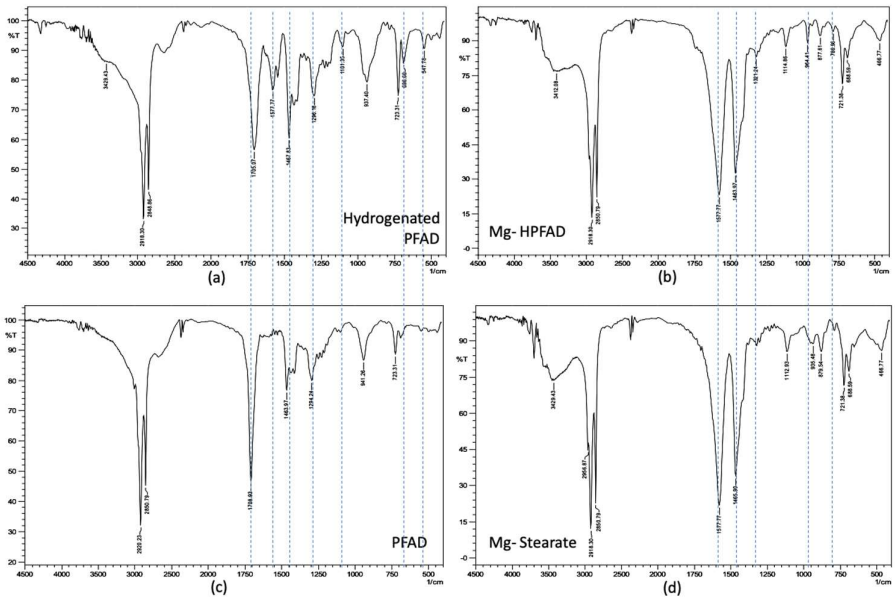

Figure 2 FTIR spectra of (a) hydrogenated PFAD, (b) Mg-PFAD, (c) PFAD, and (d) Mg-stearate.

3.3 Evaluation of Paracetamol Tablets Using Various Lubricants

Magnesium salt of PFAD and magnesium salt of HPFAD were used as lubricants in paracetamol tablet formulation and compared with commercial magnesium stearate. The tablets weighed approximately 756 mg and had a thickness of 4.8 mm and a diameter of about 13.05 mm and were evaluated for hardness, friability, and frictiability (Table 4).

Table 4 Evaluation of 500-mg paracetamol tablets with Mg-PFAD or Mg-HPFAD as lubricants compared to commercial Mg-stearate and standard.

Lubricants	Hardness ($kp_)$	Friability (%-wt loss)	Frictiability (%-wt loss)	Disintegration time (s)		Dissolution after 30 minutes (%)
				Range	Average	
Mg-PFAD	9.41 ± 2.36	0.53	0.40	91-160	125 ± 49	96.33 ± 4.93
Mg-HPFAD	9.96 ± 1.74	0.53	0.53	108-157	132 ± 35	96.56 ± 3.85
Mg Stearate	10.11 ± 1.66	0.40	0.40	58-124	91 ± 47	90.02 ± 9.57
Standard	7-14	< 1.0	< 1.0			

Friability is the tendency for tablets to crumble due to impact. This parameter is used to see if tablets will not crush easily in a bottle but can still be disintegrated

well in the digestive system. The permissible friability value is less than 1%-wt loss [4,6-8]. Based on the result in Table 4, tablets with Mg-PFAD or Mg-HPFAD lubricant had slightly higher friability (0.53%) compared to tablets with commercial magnesium stearate (0.4%). This indicates that the tablets with Mg-PFAD or Mg-HPFAD lubricant had slightly lower abrasion resistance than the commercial magnesium stearate tablets, which may be due to the higher content of unsaturated fatty acids in the PFAD and HPFAD. The friability value for all tablet variations, including tablets with Mg-PFAD or Mg-HPFAD lubricant, were below 1% and met the pharmacopoeia standards. Frictiability is the resistance of tablets when rubbed against each other. The frictiability value of the tablets with Mg-HPFAD lubricant (0.53%) was higher than that of the tablets with Mg-PFAD and Mg-stearate lubricants (0.4%), which indicates that the tablets with Mg-HPFAD lubricant had slightly higher resistance to friction than those with Mg-PFAD or Mg-stearate lubricant. Tablet hardness is a measure of the resistance to deformation due to stress, preferably in a range between 7 and 14 kp, and may vary for different tablets [9,23]. The tablets with Mg-PFAD or Mg-HPFAD lubricant had slightly lower hardness compared to the tablets with commercial Mg-stearate (Table 4).

The presence of unsaturated fatty acids in the Mg-PFAD and Mg-HPFAD increased the salt particle size and improve the ability to reduce friction. This corresponds well to the result from Gnanasekaran and Chavidi [24], who state that a high content of monounsaturated fatty acids facilitates good lubrication ability. In addition, the Mg-HPFAD lubricant produced the tablets with the longest disintegration time (132 s). After 30 min of digestion, the amount of released active compounds from the tablets with the Mg-PFAD and Mg-HPFAD lubricants (96%) was higher than that of the tablets with the Mg-stearate lubricant (90%). These results show that paracetamol tablets with Mg-PFAD or Mg-HPFAD as lubricant showed similar hardness, friability, and frictiability compared to paracetamol tablets that used commercial Mg-stearate lubricant.

4 Conclusion

Based on the result of this study, we conclude that the catalytic transfer hydrogenation (CTH) method using [NH_4COOH] of 4.2 M; T= 90°C produced hydrogenated PFAD with a lower iodine value than when using the gaseous hydrogenation (GH) method at P = 14 bar and T = 180°C. The increased temperature of CTH improved conversion of double-bond saturation and reduced the iodine value of the hydrogenated PFAD, with the lowest iodine value produced with 3.6 M ammonium formate solution at 90°C. The dissolution of active compounds from the tablets with Mg-PFAD or Mg-HPFAD lubricant was higher than from the tablets with Mg-stearate. The paracetamol tablets with Mg-PFAD or Mg-HPFAD as lubricant showed similar friability, frictiability, and

hardness compared to the paracetamol tablets with commercial magnesium stearate lubricant. These results indicate that Mg-PFAD or Mg-HPFAD salts are potential plant-based alternatives to Mg-stearate as solid lubricants.

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