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Conversion of industrial waste coconut fatty acids to methyl ester sulfonated-based detergents

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Abstract: This research focuses on the production of methyl ester sulfonate (MES) from waste coconut fatty acids (WCFAs) via sequential esterification of lauric acid as model compound for WCFAs and the sulfonation of methyl ester (ME). Firstly, lauric acid was converted to ME by esterification with methanol. Then, ME was converted to methyl ester sulfonic acid (MESA) by sulfonation with either methanesulfonic acid (MSA) or sulfuric and followed by bleaching. Next, MESA was neutralized with 20%w/v sodium hydroxide to convert into MES. Finally, MES was synthesized from WCFAs with the optimum condition of esterification and sulfonation that was considered earlier. It was found that esterification of lauric acid with methanol, using molar ratio of lauric acid and methanol at 5:1 provided the highest lauric acid conversion. Sulfonation of ME from lauric acid with 30%v/v MSA at 150 °C for 3 hours provided higher ME conversion than sulfonation with 30%v/v sulfuric acid at 140 °C for 3 hours. Next, esterification of WCFAs by using molar ratio of WCFAs and methanol at 10:1 provided WCFAs conversion around 98.6%. Sulfonation of ME from WCFAs with MSA provided higher ME conversion compared with sulfonation with sulfuric acid at 87.1% and 79.4%. The MES selectivity from sulfonation of ME from WCFAs with MSA and sulfuric was 83.2% and 73.8%. The MES from WCFAs with MSA and sulfuric acid had critical micelle concentration (CMC) at 4.8 and 2.7 mM, surface tension at CMC was 25.9 and 21.7 mN/m, and Krafft point was 10 and 8 °C.

Keywords: Coconut fatty acid, esterification, sulfonation, methanesulfonic acid, methyl ester sulfonate.

1. Introduction

Coconut fatty acid is a chemical in coconut oil, consisting of saturated fatty acid and unsaturated fatty acid. The unsaturated fatty acid may convert into trans unsaturated double bonds via hydrogenation process. The consumption of oils with trans unsaturated double bonds increased the risk of cardiovascular disease, infertility, endometriosis, gallstones, Alzheimer's disease, and some cancers. Moreover, the unsaturated fatty acid is a cause of rancid oil from the oxidation or hydrolysis process [1]. Therefore, the manufacturing of coconut oil is required to eliminate the coconut fatty acid, especially unsaturated fatty acid from the coconut oil to prevent the problem as mentioned earlier. The production of coconut oil tends to increase every year. Thus, the coconut fatty acid which is the waste from the manufacturing of coconut oil is also increased [2]. However, the coconut fatty acid can be added value by using it as raw material to produce detergent, soap, biodiesel, lubricant, etc. [3].

Linear alkylbenzene sulfonate (LAS), which is derived from petroleum, is the most used detergent in household and industrial [4]. However, the consumption of petroleum tends to be increased, while the natural reserve of petroleum is decreased [5]. Although LAS is a biodegradable detergent, it can cause health issue from the contamination in the food industry. World health organization has been reported that human daily intake of LAS from food is estimates vary from 4.5 to 14.5 mg/day [6]. Vomiting and diarrhea of dark blood may occur when LAS was ingested. Moreover, LAS may cause severe and permanent damage to the digestive tract, gastrointestinal tract burns [7]. Thus, LAS might be inappropriate for cleaning the equipment in the food industry. Therefore, the alternative detergent with a lower effect on the health issue should be studied.

There are various types of biodetergent such as methyl ester sulfonate (MES), alcohol ether sulfate (AES), etc. [8]. MES become an interesting detergent because it is an anionic detergent derived from vegetable oils or fatty acid such as coconut oil, palm oil, etc., which are environmentally friendly. Besides, Ishak et al. have been stated that MES is low toxicity compared with LAS [9]. Moreover, petroleum price which has increased in recent years leading these renewable resource detergents have become more attractive. The MES market is further expected to grow at a compound annual growth rate (CAGR) of 14% between 2021 and 2026 to reach a volume of almost 1.5 million metric tons by 2026 [10].

The commercial production of MES is sulfonation of methyl ester (ME) with excess gaseous sulfur trioxide (SO_3) to produce methyl ester sulfonic acid (MESA) followed by purification of MESA with methanol (CH_3OH) to remove excess sulfur trioxide. Then, MESA was bleached with hydrogen peroxide (H_2O_2). Finally, MESA was converted to MES by neutralization [11]. In addition, MES can produce from animal fats or vegetable oils such as soybean, sunflower oil, palm oil, etc. The fats or oils were converted into ME before starting the sulfonation. Potassium hydroxide (KOH), sodium hydroxide (NaOH), sodium methoxide (CH_3ONa) can be use as catalyst in transesterification [12]. Furthermore, MES also produce from fatty acid. The fatty acids were converted into ME before starting the sulfonation. Sulfuric acid (H_2SO_4) can be use as catalyst in esterification [3]. However, using sulfur trioxide, which has high corrosion and toxicity, in sulfonation is quite dangerous. So, this research focus on MES production from lauric acid and waste coconut fatty acids (WCFAs) by esterification and sulfonation with other acids i.e., MSA, sulfuric acid. Lauric acid was selected as model compound coconut fatty acid because the main composition in waste coconut fatty acids (WCFAs) around 53% was lauric acid.

The objective of the research is to study the conversion of model compound coconut fatty acid, which is lauric acid, to MES by esterification follow by sulfonating and to investigate the optimum conditions aiming to maximize the yield of MES. Then, WCFAs are used to synthesized MES by using the optimum condition of each reaction. The MES properties are also included.

2. Materials and Method

2.1 Chemical and raw materials

A raw material, lauric acid Hi-LR™, was purchased from HiMedia and waste coconut fatty acids (WCFAs) was collected from Ampol Food Processing Company Limited, Thailand. Analytical grade chemicals were used in the experiments. They include methanol (99.9%), tetrahydrofuran (99.5%) was purchased from Daejung Chemicals, sulfuric acid (98%), methanesulfonic acid (99%), and sodium hydroxide (98%), was purchased from Sigma-Aldrich, hydrogen peroxide (30%) was purchased from Chem-Supply.

2.2 Experiment

The experiment was mainly divided into 4 parts which include (1) Esterification of fatty acid, (2) Sulfonation of ME, (3) Bleaching of MESA, (4) Neutralization of MESA.

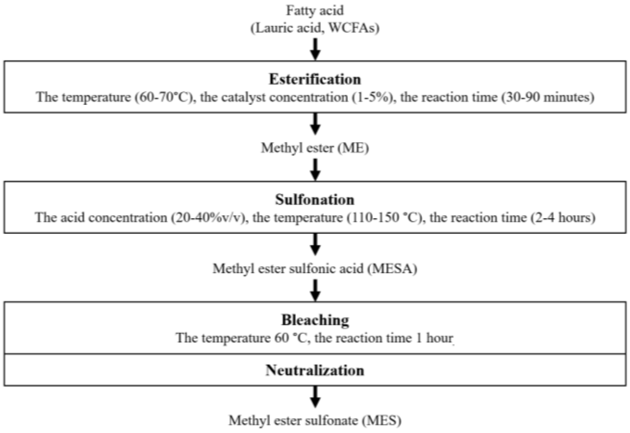


Figure 1. Flowchart of the whole experiment.

Figure 1 shows the flowchart of the whole experiment. In esterification step, fatty acid was converted to ME. Then, sulfonation step, ME was converted to MESA. Next, MESA was bleached. Finally, MESA was converted to MES by neutralization.

2.2.1 Esterification of lauric acid

Firstly, experimental reactions were conducted by using a test tube and then Parr reactor (stirred reactor) was used to scale up. For test tube as an apparatus set, nitrogen gas was flow at 1cc/min with stirring speed at 360 rpm. The setting of test tube apparatus was shown in figure 2. For Parr reactor as an apparatus set, the nitrogen gas was purged until the pressure was 10 barg with stirring speed at 200 rpm.

The esterification reaction shows in figure 3. There were three variable factors included in this experiment: the reaction temperature (60-70 °C), the catalyst concentration (1-5 %v/v), and the reaction time (30-90 minutes). Lauric acid was converted to ME via esterification with methanol by using MSA as a catalyst. First, the molar ratio of methanol to lauric acid was 5:1, and 3% volume of MSA per volume of lauric acid (%v/v) was used. The reaction temperature was maintained at 65 °C for 30 minutes. The product was washed with distilled water to remove impurities such as methanol and MSA. Finally, the remaining water was removed from the product by using a separating funnel. For studying the effect of the catalyst and the effect of the reaction time, the temperature was fixed at 65 °C according to most research [13]. Moreover, to prevent the probably of evaporation of methanol out of the reaction.

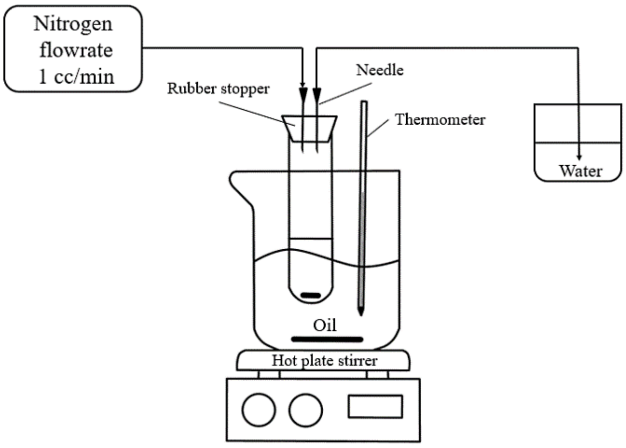
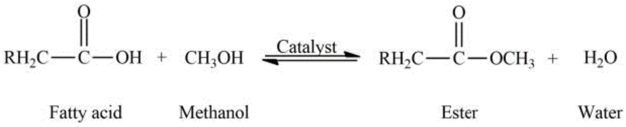


Figure 2. Test tube apparatus setting.

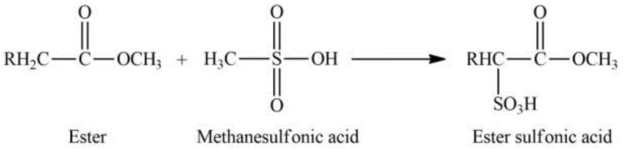


$$RH_2C-\overset{O}{\overset{\|}{C}}-OH + CH_3OH \xrightleftharpoons{\text{Catalyst}} RH_2C-\overset{O}{\overset{\|}{C}}-OCH_3 + H_2O$$

Figure 3. Esterification reaction of fatty acid with methanol.

2.2.2 Sulfonation of ME from lauric acid

ME obtained from the previous step, which was methyl laurate, was converted to MESA via sulfonation of ME with acid. The sulfonation reaction shows in figure 4.



$$RH_2C-\overset{O}{\overset{\|}{C}}-OCH_3 + H_3C-\underset{\underset{O}{\|}}{\overset{O}{\overset{\|}{S}}}-OH \longrightarrow \underset{\underset{SO_3H}{|}}{RHC}-\overset{O}{\overset{\|}{C}}-OCH_3$$

Figure 4. Sulfonation reaction of ME with MSA.

The experimental reactions were conducted by using a test tube with stirring speed at 360 rpm. The acid used, which was MSA, and sulfuric acid was varied in each experiment to compare the results.

First, 20 %volume of MSA per volume of ME (%v/v MSA) was added dropwise into ME. The reaction temperature was maintained at 110 °C for 3 hours. Then, there were three variable factors included the acid concentration (20-40 %v/v), the temperature of reaction (110-150 °C), and the reaction time (2-4 hours) was varied. Then, all of experiments were repeated with 20 %volume of sulfuric acid per volume of ME (%v/v sulfuric acid).

2.2.3 Esterification of WCFAs

WCFAs was converted to ME via esterification with methanol by using MSA as a catalyst. The molar ratio of methanol to WCFAs was 10:1. The temperature of the reaction, the catalyst concentration, and the reaction time which was the optimum condition of esterification of lauric acid was used.

2.2.4 Sulfonation of ME from WCFAs

ME obtained from the previous step was converted to MESA via sulfonation of ME with MSA and sulfuric acid. The acid concentration, the temperature of reaction, and the reaction time which was the optimum condition of sulfonation of ME from lauric acid is used.

2.2.5 Bleaching and neutralization of MESA

MESA obtained from the sulfonation step was bleached by adding 30% H_2O_2 into MESA with stirring speed at 360 rpm and stirred at 60 °C for 1 hour. Then, 20 %w/v NaOH was used to neutralized until the pH of the product is 8-9. The neutralization reaction shows in figure 5.

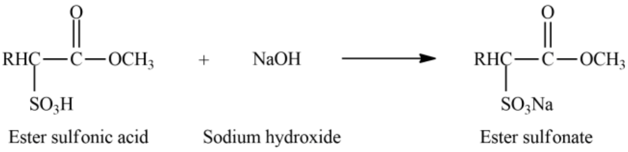


Figure 5. Neutralization reaction of MESA.

2.3 Analytical method

ME from esterification of lauric acid and MESA from sulfonation of ME from lauric acid was analyzed by using gas chromatography equipped with flame ionization detector (GC-2010 Shimadzu) to calculate the conversion according to equation 1 and 2, respectively.

$$\%\text{Lauric acid conversion} = \frac{\text{Lauric acid fed} - \text{Lauric acid unreacted}}{\text{Lauric acid fed}} \times 100 \quad (1)$$

$$\%\text{ME Conversion} = \frac{\text{ME fed} - \text{ME unreacted}}{\text{ME fed}} \times 100 \quad (2)$$

The column equipped was a capillary column DB-5 (30 m × 0.25 mm × 0.25 μm). ME and MESA was prepared by dissolve in tetrahydrofuran (THF). First, the temperature of GC was initially set at 150 °C, hold for 3 minutes. Then, the temperature was raised up to 300 °C with a rate 10 °C per minute and hold for 20 minutes. The injector and detector temperature were set at 250 °C and 300 °C, respectively.

WCFA was analyzed the selectivity of fatty acids by using gas chromatography–mass spectrometry (GC–MS 2010 Plus Shimadzu). The column equipped was the same as column specification in GC. The WCFA prepared by dissolve in THF. The sample was injected into the GC–MS. The temperature of GC–MS was initially set at 150 °C, hold for 5 minutes. Then, the temperature will be raised up to 240 °C with a rate 5 °C per minute and hold for 10 minutes. Lastly, the temperature was raised up to 300 °C with a rate 10 °C per minute and hold for 2 minutes.

ME from esterification of WCFAs and MESA from sulfonation of ME from WCFAs was also analyzed by using gas chromatography equipped with flame ionization detector (GC-2010 Shimadzu) to calculate the conversion. The conversion of WCFAs and ME was mainly calculated basis on lauric acid and methyl laurate (12-carbon atom chain), respectively.

The synthesized MES has analyzed the selectivity of MES by using gas chromatography equipped with flame ionization detector. The column equipped was a capillary column DB-5MS.

The properties of MES were analyzed. Critical micelle concentration (CMC) of MES was determined from the relationship between the electrical conductivity and the concentration of MES. Similarly, Krafft point was determined from the relationship between the electric conductivity and the temperature of MES. The electrical conductivity of MES was carried out using a digital conductivity meter. Surface tension was determined by using tensiometer.

3. Results and Discussion

3.1 Synthesis of ME from lauric acid

3.1.1 Effect of reaction temperature

Figure 6 shows the lauric acid conversion at different reaction temperatures in °C. The lauric acid conversion linearly increases from 83.5% to 95.7% with rising of the reaction temperature from 60 °C to 70 °C.

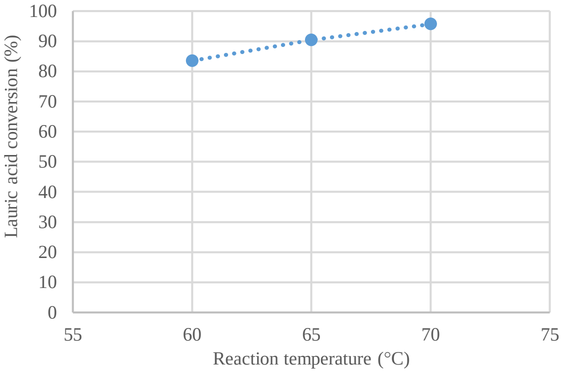


Figure 6. Laurie acid conversion versus reaction temperature.
(Condition: lauric acid to methanol ratio = 1:5;
reaction time = 30 minutes; catalyst concentration = 5%v/v)

In general, an increase in reaction temperature causes an increase in reaction rate due to the average kinetic energy of the reactant being raised. However, this experiment has a limitation on the boiling point of methanol (65 °C), so the reaction temperature should not be too high to prevent the evaporation of methanol, which is a main reactant, from the reaction.

3.1.2 Effect of catalyst concentration

In general, higher catalyst concentration leads to an increase in reaction rate and the equilibrium shift towards the product, which is ME. However, adding too high concentration of catalyst may cause the shifting of equilibrium back towards the reactant, which is lauric acid. As shown in figure 7, the lauric acid conversion increases from 80.4% at 1%v/v to 91.3% at 3%v/v, respectively. However, the lauric acid conversion decreases to 90.4% with increasing of catalyst concentration from 3%v/v to 5%v/v, respectively.

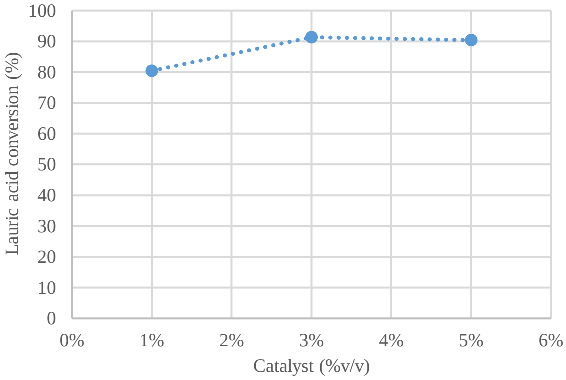


Figure 7. Lauric acid conversion versus catalyst concentration.
(Condition: lauric acid to methanol ratio = 1:5;
reaction time = 30 minutes; reaction temperature = 65 °C)

3.1.3 Effect of reaction time

Figure 8 shows the lauric acid conversion at different reaction time in minute. The lauric acid conversion slightly increases from 91.3% to 92.8% with increasing of the reaction time from 30 minutes to 60 minutes, while the lauric acid conversion at 90 minutes decreases to 80.4%.

Basically, the product of each reaction increases with the reaction time. However, the product of esterification reaction may decrease with longer reaction time because when the product increases too much, the equilibrium shifts back towards the reactant.

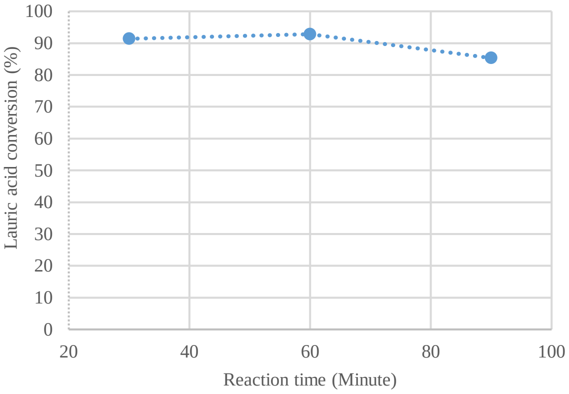


Figure 8. Lauric acid conversion versus reaction time.
(Condition: lauric acid to methanol ratio = 1:5;
reaction temperature = 65 °C; catalyst concentration = 3%v/v)

3.1.4 Effect of apparatus set

The comparison of lauric acid conversion from differently apparatus set which is test tube and Parr reactor shows in figure 9. The results shows that the lauric acid conversion obtained from parr reactor is 96.6%, while the lauric acid conversion obtained from test tube is 92.8% and using Parr reactor provides higher lauric acid conversion due to methanol cannot loss from the system, while using test tube methanol may loss from the system because the boiling point of methanol is lower than reaction temperature. The loss of methanol from the system might cause the shifting of equilibrium back towards the reactant. Moreover, the pressure in Parr reactor, which is a close system, is higher than the test tube. The higher temperature also promotes the reaction.

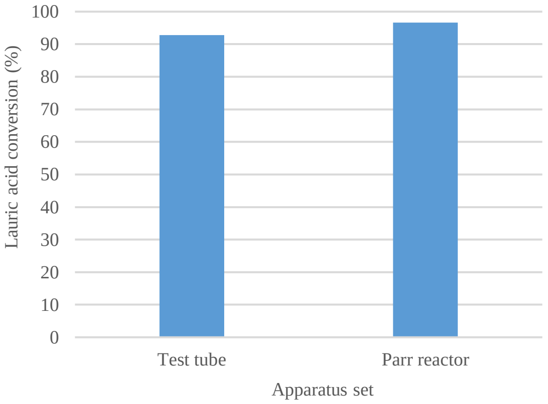


Figure 9. Comparison of lauric acid conversion from differently apparatus set.
(Condition: lauric acid to methanol ratio = 1:5; reaction temperature = 70 °C; catalyst concentration = 3%v/v; reaction time = 60 minutes)

3.2 Sulfonation of ME from lauric acid

3.2.1 Effect of acid and ME ratio

Figure 10 shows the ME conversion of ME sulfonation with MSA and sulfuric acid at different acid and ME ratio in percentage of acid volumetric in ME volumetric (%v/v). For the sulfonation with MSA, the ME conversion increases from 61.6% at 20 %v/v to 72.7% at 30 %v/v. Then, the ME conversion slightly increases to 72.9% with increasing of acid and ME ratio to 40%v/v. For the sulfonation with sulfuric acid, the ME conversion increases from 70.2% to 76.4% with increasing of acid and ME ratio from 20%v/v to 30%v/v. Although increasing of acid and ME ratio to 40%v/v, the ME conversion slightly decrease to 75.6%. An increase in acid and ME ratio causes an increase in reaction rate due to acid which is a reactant is increased. In detail, MSA and sulfuric acid show slightly different ME conversion. Sulfonation with sulfuric acid achieves higher ME conversion.

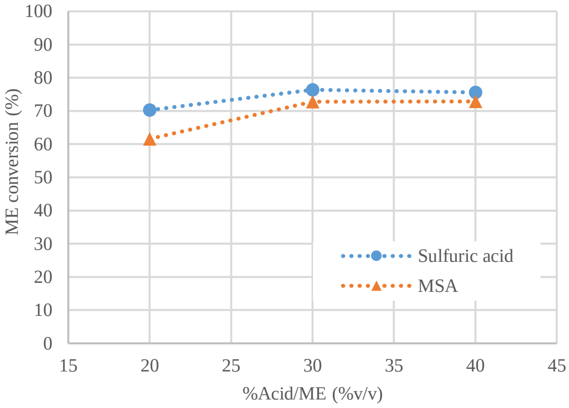


Figure 10. ME conversion versus acid and ME ratio.
(Condition: reaction time = 3 hours;
reaction temperature = 120 °C)

3.2.2 Effect of reaction temperature

The ME conversion of sulfonation with MSA increases from 56.4% to 97.9% while the ME conversion of sulfonation with sulfuric acid increases from 65.3% to 89.1% with rising of the reaction temperature from 110 °C to 140 °C. Then, the ME conversion slightly decreases to 88.0% with increasing of reaction temperature to 150 °C. Increasing reaction temperature has more affecting on ME conversion from sulfonation of ME with MSA than sulfonation of ME and sulfuric acid as shown in figure 11.

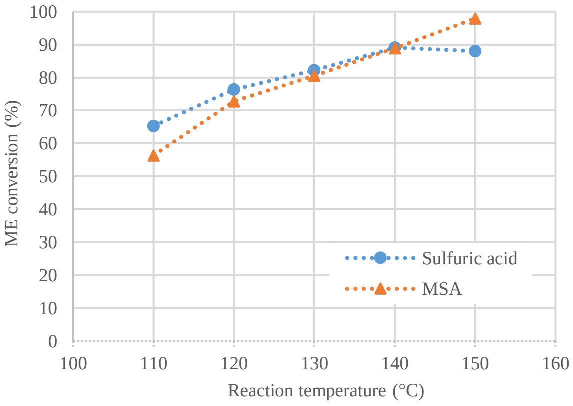


Figure 11. ME conversion versus reaction temperature
(Condition: Acid and ME ratio = 30%v/v;
reaction time = 3 hours)

3.2.3 Effect of reaction time

Figure 12 shows the ME conversion at different reaction time in hour. The ME conversion from sulfonation of ME with MSA linearly increases from 61.4% to 72.7% with increasing of the reaction time from 2 hours to 3 hours. Then, the ME conversion slightly decreases to 72.5% with increasing of reaction time to 4 hours. The ME conversion from sulfonation of ME with sulfuric acid also increases from 64.7% at 2 hours to 76.4% at 3 hours and slightly increases to 76.5% at 4 hours.

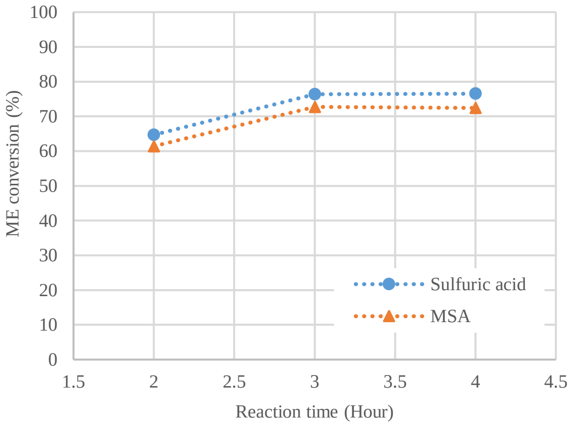


Figure 12. ME conversion versus reaction time.
(Condition: Acid and ME ratio = 30%v/v;
reaction temperature = 120 °C)

3.3 Fatty acid distribution of WCFAs

The composition of WCFAs was mainly lauric acid. The fatty acid distribution of WCFAs shows in table 1. However, WCFAs also had the presence of ketone group, ester group, alcohol group, and other chemical compounds.

Table 1. Fatty acid distribution of WCFAs

Fatty acid	Distribution (%)
Caprylic acid ($C_{8:0}$)	8.5
Capric acid ($C_{10:0}$)	7.0
Lauric acid ($C_{12:0}$)	53.7
Myristic acid ($C_{14:0}$)	16.9
Palmitic acid ($C_{16:0}$)	7.2
Stearic acid ($C_{18:0}$)	1.7
Oleic acid ($C_{18:1}$)	5.0

3.4 Synthesis of MES from WCFAs

The conversion of ME from esterification of WCFAs with methanol, using the molar ratio of methanol to WCFAs at 10:1 with 3% MSA as catalyst at 70 °C for 1 hours, was 98.6%. Esterification of WCFAs has required a higher molar ratio of methanol to fatty acid than esterification of lauric acid due to the complicated composition of WCFAs had fatty acids with a higher number of carbon atoms compared with lauric acid as demonstrated in the article by Saravanan et al., which the main composition of reactant was palmitic acid [14] and oleic acid in Brinks et al. [15] that used higher molar ratio of methanol to fatty acid.

Sulfonation of ME from WCFAs with 30%v/v MSA at 150 °C for 3 hours also provide higher ME conversion compare with sulfonation with 30%v/v sulfuric acid at 140 °C for 3 hours at 87.2% and 79.4%, respectively. Even though the sulfonation condition of ME from WCFAs was the same as the sulfonation condition of ME from the lauric acid, the conversion of ME from WCFAs is lower than the conversion of ME from lauric acid due to WCFAs had more impurities that might interfere the reaction.

3.5 MES selectivity

The selectivity of MES from sulfonation of ME from lauric acid with 30%v/v MSA at 150 °C for 3 hours was 86.4%, while the selectivity of MES from sulfonation of ME from lauric acid with 30%v/v sulfuric acid at 150 °C for 3 hours was 78.2%. For WCFAs, the selectivity of MES from sulfonation of ME with 30%v/v MSA at 150 °C for 3 hours was 83.2% and the selectivity of MES from sulfonation of ME with 30%v/v sulfuric acid at 140 °C for 3 hours was 73.8%.

3.6 MES properties

The properties of MES, which was synthesized from lauric acid with MSA, and sulfuric acid had the critical micelle concentration (CMC) at 5.3 and 3.6 mM. Surface tension at CMC was 26.1 and 22.9 mN/m. Krafft point was 12 and 8 °C.

The properties of MES, which was synthesized from WCFAs with MSA, and sulfuric acid has CMC at 4.8 and 2.7 mM. Surface tension at CMC was 25.9 and 21.7 mN/m. Krafft point was 10 and 8 °C.

4. Conclusions

The optimum condition of lauric acid esterification with methanol was using molar ratio of lauric acid and methanol at 5:1 with 3% MSA as catalyst at 70 °C for 1 hour in Parr reactor. The lauric acid conversion was 97.5%. The optimum condition of sulfonation with MSA was using 30%v/v MSA at 150 °C for 3 hours. The optimum condition of sulfonation with sulfuric acid was using 30%v/v MSA at 140 °C for 3 hours. The yield of MES from sulfonation with MSA and sulfuric acid was 84.6% and 69.7%, respectively. For WCFAs, MES was synthesized through an esterification and sulfonation process. The lauric acid conversion was 98.6%. The ME conversion from sulfonation of WCFAs with MSA and sulfuric acid was 87.1% and 79.4%. The yield of MES from sulfonation with MSA and sulfuric acid was 72.5% and 58.6%. Sulfonation with MSA provide higher conversion of ME and selectivity of MES compared with sulfonation with sulfuric acid. Moreover, using MSA is more appropriate for manufacturing than sulfuric acid because sulfuric acid still has a higher corrosive problem to the machine compared with MSA.

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