FATTY ACID METHYL ESTER PRODUCTION FROM *Calophyllum inophylllum* OIL BY TWO-STEP PROCESS USING SULFONATED TIO_2-SIO_2 CATALYST

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Abstract

The worldwide search for sustainable alternative energy sources has been sparked by the depletion of fossil fuel supplies and the environmental issues related to their production and burning. Fatty acid methyl esters (FAME), often known as biodiesel, is one of the sustainable fuels recently promoted. This fuel which usually uses edible oil or non-edible oil as feedstock has emerged as a promising substitute for conventional fossil fuels due to its renewable nature, lower emissions profile, and potential for carbon neutrality. Usually, homogeneous catalysts are often used in this production of biodiesel; however, heterogeneous catalysts become preferable due to ease of separation and cost effectiveness. Heterogenous solid acid catalyst on the other hand, has been proven to increase the reaction process, separate easily and be recycleable while supported by the catalyst from biomass waste. Rice husk ash (RHA) which produces silica from biomass waste, can be considered affordable, widely available and able to eliminate waste problem. The potential of sulfonated SiO_2-TiO_2 as a binary catalyst that could address both the challenges of high FFA content and improve biodiesel yields through enhanced catalytic activity is underexplored. Therefore, in this study, sulfonated TiO_2-SiO_2 catalyst was optimized for the esterification and transesterification of *Calophyllum inophyllum* oil into methyl esters. A two-step reaction process was performed by using sulfonated TiO_2-SiO_2 as solid acid catalyst in esterification process and followed by base-catalysed transesterification using potassium hydroxide, KOH. The analysis on the optimization of the reaction parameters revealed that the higher conversion of biodiesel production of 65.50% was achieved using 9:1 methanol to oil ratio, 3 wt% of catalyst loading at 65°C for 1 hour. Gas chromatography mass spectrometry (GC-MS) showed seven of the methyl ester peaks that were obtained at optimized reaction parameters. Therefore, solid catalyst, sulfonated TiO_2-SiO_2, has the ability to be used as an alternative catalyst for FAME production from *C. inophyllum* oil.

Keyword: *Calophyllum inophyllum* oil, fatty acid methyl ester, heterogeneous catalyst

Introduction

Biodiesel is a renewable, energy-efficient, and biodegradable resource that has the potential to provide all the world's energy needs. It is a feasible alternative to fossil fuel, which is responsible for the depletion of the ozone layer and environmental damage. It is a renewable fuel that burns cleanly and can be used in existing petroleum diesel engines without requiring any modifications. It is also known as eco-diesel. Typically, biodiesel is identified by its primary feedstock, which include vegetable oils and animal fats (Zurqarnain et al., 2021).

Biodiesel is more environmentally benign than petroleum-based fuels, more affordable, and capable of creating greener energy, which has favourably impacted the economy (Alsaiari et al., 2023).

Nevertheless, one of the main obstacles to the synthesis of FAME is the need for a sufficient feedstock/oil and catalyst. There have been concerns over the use of edible oils such as palm, sunflower, coconut, soybean, and cottonseed oils as the basis for FAME synthesis as this process competes with the food availability and this frequently results in higher prices for edible vegetable oil, which adds to the relatively expensive FAME production (Adenuga et al., 2020; Chimezie et al., 2022). Hence, to identify and evaluate locally accessible non-edible seed oils as acceptable feedstocks for the synthesis of FAMEs, concerted research efforts are focused on jatropha oil, linseed oil, rubber seed oil, avocado peel oil and *C. inophyllum* oil as the second-generation feedstocks (Chimezie et al*.,* 2022; Phydimalla et al., 2023). The significant characteristics of this second-generation biodiesel that attract researchers to them are that it is environmentally friendly, lower in production costs, eliminates food inequity and requires less area for cultivation (Singh et al., 2020).

C. inophyllum is seen as a promising biodiesel source because it has a lot of oil in its seeds containing up to 73% oil content. It also produces many fruits quickly, is easy to grow, and can thrive in various climates (Adenuga et al., 2021). However, the oil from *C. inophyllum* contains a high level of free fatty acid (FFA) and viscosity. Therefore, before the transesterification process, a pretreatment step involving acid-catalyzed esterification is necessary for crude oil with a high FFA level. The higher the FFAs, the more difficult it is in conversion to the biodiesel production process and the lower the yield. To address this issue, researchers devised a two-step procedure that included pre-treatment processes before transesterification using an acid catalyst. The esterification process transforms most FFAs into the corresponding FAME during the pre-treatment stage. However, triglycerides remained in the pre-treated oil, necessitating a transesterification process to complete the biodiesel generation (Sathyaselvabala et al., 2011). Marso et al., (2017) studied the production of biodiesel from metal modified graphene oxide composite catalyst via pre-esterification of *C. inophyllum* oil. The optimum conditions were obtained at 3 h reaction time in the presence of 8% of catalytic dose at 65°C. In concordance with the result of other studies, it can be said that 65°C of reaction temperature worked the best in converting higher percent yield. However, higher temperatures also tend to speed up processes but can also cause unintended side effects and product deterioration.

The research on biodiesel production using sulfonated TiO_2-SiO_2 as a catalyst, specifically with *C. inophyllum* oil is limited, despite its potential advantages. Few research has explored combined effect of single-component catalyst such as sulfonated silica or titania, as most research focused on them separately. Thus, there is a lack of comprehensive studies that explore the potential optimum condition, catalyst's reusability, and effectiveness in high FFA oils. Limitation of non-sulfonated TiO_2-SiO_2 is insufficient catalytic activity in biodiesel production, especially when involving high FFA content oils. The introduction of sulfonic acid (-SO_3H) groups significantly enhances the acidity of the catalyst, improving its ability to esterifying FFAs and facilitating transesterification reactions. Combining TiO_2 and SiO_2 as catalyst provides benefits such as SiO_2 offering a high surface area that facilitates TiO_2 dispersion, while TiO_2 contributing its strong acidity to enhance the catalytic activity by increasing the number of accessible active sites. This combination leads to higher conversion rates, better catalytic activity, and a more stable reaction environment (Bagheri et al., 2014). Previous research by Brindhadevi et al. (2024) studied the comparison of photocatalytic versus conventional transesterification for biodiesel preparation from fish waste oil using green TiO_2/SiO_2 catalyst. The result showed higher catalytic activity in both methods at reaction conditions methanol to oil ratio 5:1 and mass of catalyst 30 mg.

In this study, a simple method has been introduced to produce SiO_2 from rice husk. Some solid catalyst support such as synthetic zeolite has high cost and involve multiple steps in synthesizing it. As such, it is important to create inexpensive, sustainable heterogeneous catalysts that are simple to recycle. Rice husk is one of the agricultural wastes that is abundant in developing countries. The expense of collecting and disposing of rice husks increases as the amount of rice husks produced increases yearly. In Malaysia, it is estimated that about 1.2 million tons of agricultural waste are disposed every year. Rice husk waste is dumped or burned, causing significant environmental issues. Thus, applying Rice Husk Ash (RHA) can reduce waste product problems while improving biodiesel quality. RHA typically contains 85% to 95% silica (SiO_2) by weight, depending on the burned conditions (Della et al., 2002). Rice husk which is burned at controlled temperatures between 500°C and 700°C usually has the highest purity of silica content. In this study, sulfonated TiO_2-SiO_2 catalyst was optimized for the esterification and transesterification of *C. inophyllum* oil into methyl esters.

Materials and Methods

C. inophyllum fruits were collected from Bandar Pusat Jengka, Pahang. The *C. inophyllum*'s seed oil was obtained from fruit kernel and used as the feedstock for production of FAME. The rice husk was obtained from Selangor. The analytical reagent (AR) grade of the methanol was purchased from HmbG Chemical while AR grade of n-hexane was from Astral Scientific. AR grade of toluene, isopropyl alcohol and hydrochloric acid, HCl (fuming 37%) were purchased from R&M Chemical, and for potassium hydroxide (KOH) and sulphuric acid, H_2SO_4 (98%) were purchased from Friedemann Schmidt Chemical and Loba Chemie, respectively.

Seed preparation and drying process

The seed of *C. inophyllum* was cracked open manually by using a hammer and the waste husk was removed. The outer layers were detached to expose the kernels. The separated seed kernels were cleaned and stored at room temperature. The seed kernels were dried at 65°C to 70°C for 3-4 days in the oven (Bhuiya et al., 2015).

Extraction oil preparation

The dried seed kernels were grounded by using blender to get a fine consistency to maximize surface area. The ground seed kernels were placed into conical flasks. After that, n-hexane was added at ratio of 2:1 in which the weight and volume of the kernel and hexane used was 300 g and 600 ml of hexane, respectively. The mixture was stirred initially to certify the kernel were wetted with hexane. The conical flask was covered with aluminum foil. The samples were left to run for up to 3 days. The hexane/oil mixtures were collected, filtered, and transferred into aluminum foil containers for solvent evaporation. The mixture was placed under the fume hood for 20-24 hours (Bhuiya et al., 2015).

SiO_2 preparation process

50 g of rice husks (RH) was weighted with electronic balance. By using conical flask and filter paper, RH was washed with distilled water to remove dirt. The cleaned RH was mixed with 0.4 M of hydrochloric acid (HCl) in the ratio of 50 g of rice husks towards 500 ml of HCl. Then, the mixture was stirred for 20 minutes and heated at 110°C for 3 hours. The RH was rinsed with distilled water to remove HCl acid and dried using oven for 3 hours at 110°C. After being dried, the RH was placed in alumina crucibles and was burned for 6 hours at 550°C to obtain

white Rice Husk Ash (RHA) (Mostafa et al., 2015).

TiO_2-SiO_2 catalyst preparation process

The weight ratio of TiO_2:SiO_2 used was 3:1. 30 g of TiO_2 was added slowly to 2.0 M H_2SO_4 solution until dissolved completely. Then, 10 g of SiO_2 was added into the solution. The mixture was stirred for 6 hours at 600 rpm. After that, a solid precipitate was obtained, and the precipitate was filtered and calcined for 4 hours at 500°C with heating rate of 10°C per min (Embong et al., 2016).

Characterization of the catalyst

The catalyst was characterized by using Fourier Transform Infrared Spectrometer (FTIR). The functional group present in the sulfonated TiO_2-SiO_2 was analysed by Perkin Elmer infrared spectrometer using attenuated total reflection (ATR) accessory between the peak from 400 to 4000 cm^{-1}.

Esterification acid catalysed of *C. inophyllum* oil

A three-neck round-bottom flask was used to conduct the esterification reaction with a thermometer, condenser and magnetic stirrer placed in water bath. Parameter of catalyst loading, and reaction time were studied to find the higher percent yield of methyl ester. 15 g of CIO was added into the methanol and catalyst mixture. The mixture was refluxed while being constantly stirred magnetically at 400–600 rpm. The conversion of free fatty acid was performed at 65°C by using 9:1 methanol to oil ratio and varied at catalyst loading (1 wt%, 2 wt%, 3 wt%, 4 wt%, 5 wt% and 6 wt%) and reaction time (1 hours to 6 hours) (Ramli et al., 2015; Koria et al., 2010). After that, the mixture was transferred into centrifuges tubes to separate the product and the by-product which is glycerol and solid acid catalyst and was centrifuged at 4000 rpm in 15 minutes. Then, the mixture was separated and the layer that contained methyl ester was removed by using dropper and stored in beaker to heat. The heating process of the methyl ester was done at temperature above 80°C in 2 to 5 minutes to remove excess methanol. The esterification process was repeated 2 times until the acid value of the mixture was below 2 mg KOH/g.

Base-catalyzed transesterification

After esterification process, the esterified CIO was mixed with 1 wt% of alkali catalyst (KOH) and 6:1 methanol to oil ratio. The mixture was pre-heated to the temperature of 60°C at 300 rpm and stopped after 1 hour reaction (Sathyaselvabala et al., 2011). After the reaction stopped, the mixture was centrifuged at 4000 rpm in 15 minutes to separate the layer and the reaction mixture was transferred into a separating funnel for separation. Glycerol and contaminants were found in the bottom layer, which was removed. 500 ml of hot distilled water was used to wash the biodiesel on the upper layer. Following the last washing, the top layer was transformed into the biodiesel product, with the bottom layers being disposed of. The final product was weighted, and the biodiesel product was placed in a hot air oven set at 110°C for one hour to eliminate water (Thiruvengadaravi et al., 2012). The catalyst was washed with the n-hexane for 4-5 times and were centrifuged at 4000 rpm in 5 minutes for reusability. After that, the reused catalyst was dried in the oven at 110°C in 1 hour.

Characterization of methyl ester

The produced methyl esters produced were measured by gas chromatographic mass spectrometry (GC-MS). GC-MS was conducted using gas chromatography (GC) (QP2010 Ultra, Shimizu, Shizuoka, Japan) equipped with RTX-5MS column. The samples were injected at a column temperature of 80°C. The column temperature was maintained at 200°C for 26 min and increased to 270°C at a rate of 5°C/min, then maintained for 40 min.

Results and Discussion

Fourier Transform Infrared Spectrometer (FTIR) Analysis

As shown in **Figure 1**, the presence of the different functional groups on the surface of synthesized catalyst was investigated by using Fourier Transform Infrared Spectrometer (FTIR) of the TiO_2, SiO_2 and TiO_2-SiO_2 catalyst. The peak at wavenumber of 804.03 cm^{-1} indicates the symmetrical stretching of Si-O bonds (Shao et al., 2013; Nizar et al., 2017). In addition, the intense broad peaks at wavenumber of 1056.06 cm^{-1} correspond asymmetrical stretching of the Si-O bonds (Sathyaselvabala et al., 2011). As for TiO_2, the main absorption band observed indicates O-Ti-O bonds at 722.88 cm^{-1} in TiO_2, confirming the presence of the TiO_2 structure. Band at 1250 cm^{-1} is related to the vibration of Si-O-Ti bonds. Generally, the FTIR spectra of the TiO_2-SiO_2 series are comparable to one another and appear to be combination spectra of SiO_2 and TiO_2 (Nizar et al., 2017). In the process of synthesis of TiO_2-SiO_2, tetrahedral framework formation is shown by the Si-O-Ti bonding which is generated as the result of chemical reaction between SiO_2 and TiO_2. The mechanism for the formation of Ti_xSi_y oxides as described by Teamsinsungvon et al. (2022).

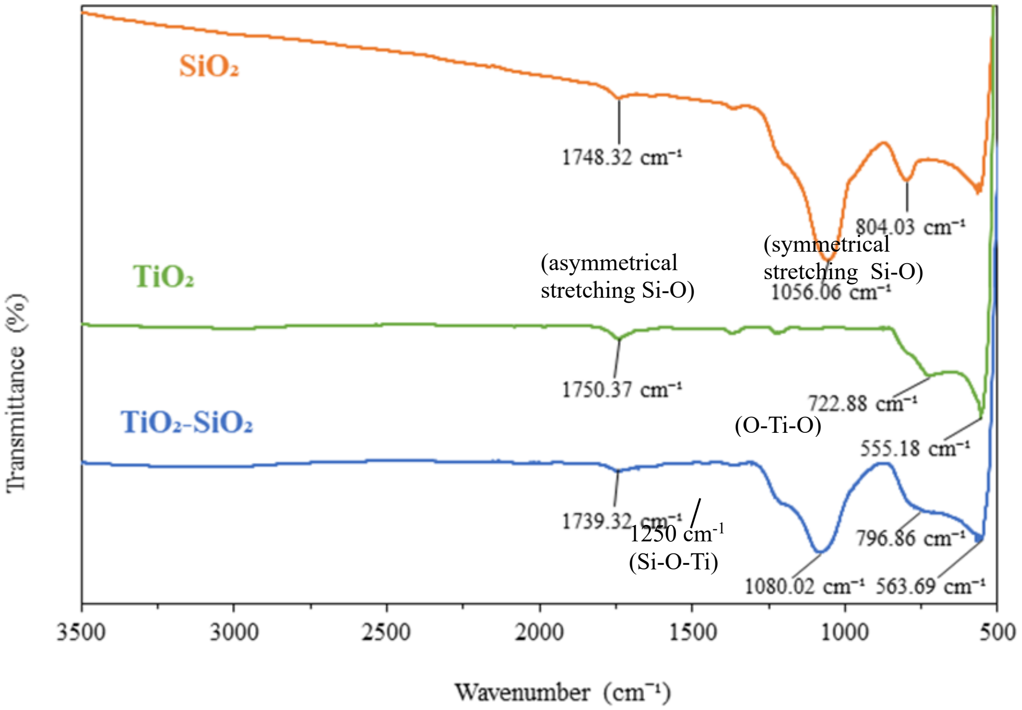

Figure 1 FTIR spectra of SiO_2, TiO_2 and TiO_2-SiO_2

Effect of catalyst loading in two-step reaction

The introduction of pre-treatment process reduced the acid value of CIO to the lower level which is less than 2 mg KOH/g immediately and makes it fit to proceed with base catalysed transesterification. In this study, the acid value of crude oil of *C. inophyllum* is 34.96 mg KOH/g was reduced to 1.18 mg KOH/g after esterification process. Esterification of *C. inophyllum* oil was performed in 1 hour at 65°C by using a 9:1 methanol to oil ratio to determine the effect of catalyst loading to obtain methyl ester yield. The biodiesel yield obtained via esterification process with varied catalyst loading of 1 wt%, 2 wt%, 3 wt%, 4 wt%, 5 wt% and 6 wt% are shown in **Figure 2**. The findings suggest that an optimal catalyst concentration of approximately 3 wt% leads to the highest biodiesel yield. The immiscibility of triglyceride and ethanol implies that increasing the catalyst concentration facilitates the transesterification process, resulting in a rapid enhancement of the yield (Kamran et al., 2020). However, at very low or very high catalyst concentrations, soap formation occurs, impacting the separation of glycerol from biodiesel and potentially reducing the overall yield. Inadequate catalyst usage can lead to incomplete reactions and diminished product yield (Mukhtar et al., 2021). Therefore, the maximum yield is achieved when an adequate catalyst concentration is employed. Besides, increasing the amount of catalyst used can lead to undesirable side effects and greater costs rather than increasing the yield (Hundie et al., 2022). In this study, the optimal parameter for catalyst loading based on this study was 3 wt%, methanol to oil ratio of 9:1, reaction temperature of 65°C in 1 hour.

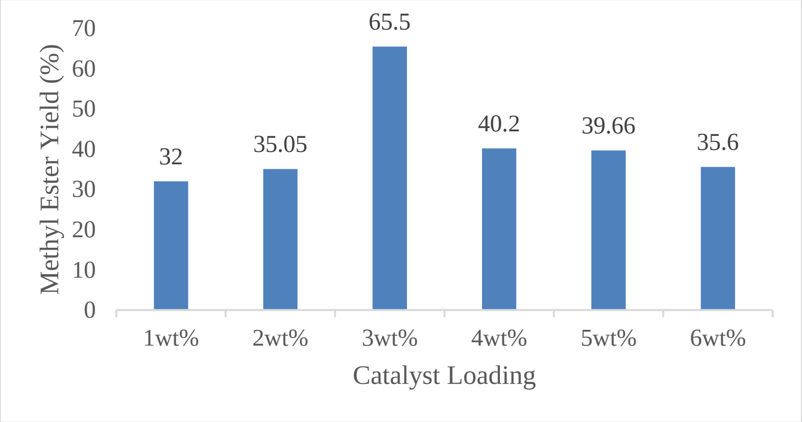

Figure 2 Effect of catalyst loading in two step reaction at 1 hour of reaction time, 9:1 of methanol to oil ratio in 65°C of reaction temperature

Effect on reaction time in two step reaction

Reaction time plays an important role which can affect the percent yield of the biodiesel. Thus, a series of experiments were performed at different reaction times from 1 h to 6 h to investigate the effect of reaction time to obtain methyl ester yield. Based on the result in **Figure 3**, the yield of methyl ester decreases after 1 hour of reaction. This could be because of hydrolysis and deterioration of the fatty acid methyl ester when increasing reaction times. The fatty acid methyl esters in the oil could undergo hydrolysis over time, particularly if water is present in the reaction mixture. After achieving equilibrium, a reversible reaction might happen which could reduce the yield of the reaction and raise the probability of saponification (Kedir et al., 2023). In this study, the optimal reaction condition for reaction time is 1 h which has a percent yield of 65.50%.

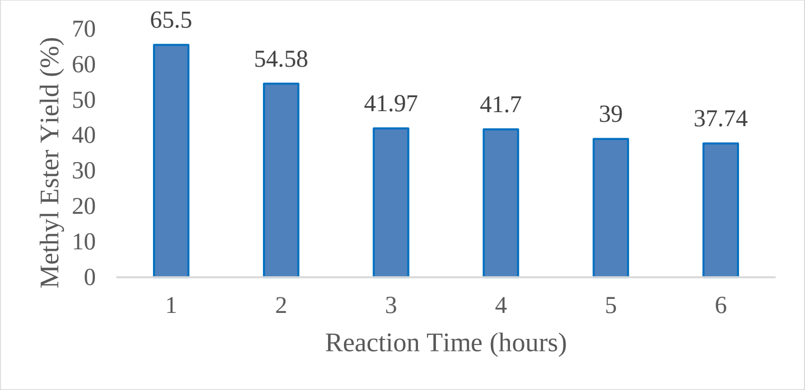

Figure 3 Effect of reaction time in two step reaction at 3 wt% of catalyst loading, 9:1 of methanol to oil ratio in 65°C of reaction temperature

Qualitative analysis of methyl ester

In this study, the techniques for quantitative analysis were analysed by using gas chromatography mass spectroscopy. The fatty acid composition in FAME obtained were analysed which include lauric acid methyl ester, myristic acid methyl ester, palmitic acid methyl ester, stearic acid methyl ester, oleic acid methyl ester, linoleic acid methyl ester and eicosanoic acid methyl ester as shown in **Table 1** and **Figure 4**.

Table 1 Fatty acid composition in fatty acid methyl ester (FAME)

Retention Time (min)	Methyl Ester Compound	Common Name	Chemical Formula
15.213	Dodecanoic acid methyl ester	Lauric acid methyl ester	$C_{13}H_{26}O_2$
19.880	Methyl tetradecanoate	Myristic acid methyl ester	$C_{15}H_{30}O_2$
24.365	Hexadecenoic acid methyl ester	Palmitic acid methyl ester	$C_{17}H_{34}O_2$
27.502	Methyl stearate	Stearic acid methyl ester	$C_{19}H_{38}O_2$
27.985	9-Octadecanoic acid methyl ester	Oleic acid methyl ester	$C_{19}H_{36}O_2$
28.866	9,12-Octadecanoic acid (Z,Z) – methyl ester	Linoleic acid methyl ester	$C_{19}H_{34}O_2$
30.109	Methyl 18-methylnonadecanoate	Eicosanoic acid methyl ester	$C_{21}H_{42}O_2$

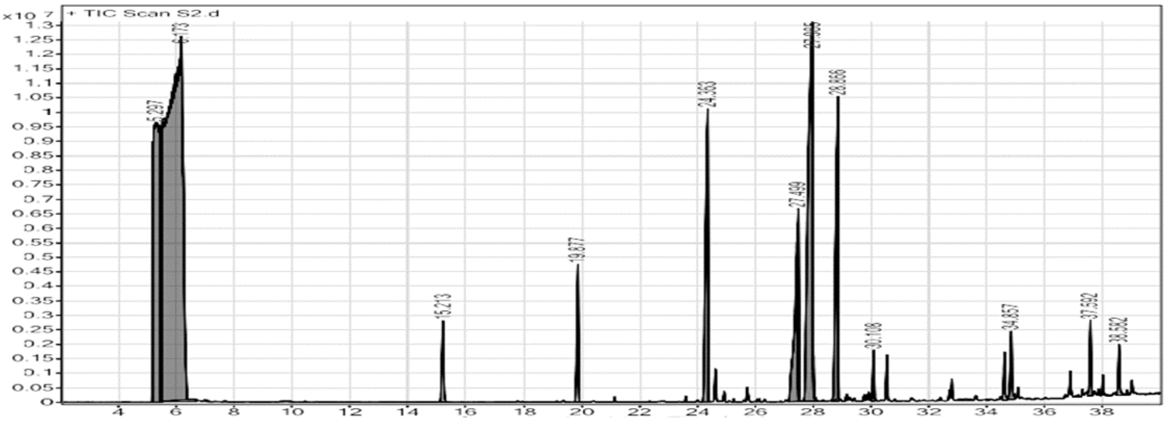

Figure 4 GC chromatogram for FAME obtained at 3 wt% of catalyst loading, 9:1 of methanol to oil ratio in 65°C of reaction temperature and 1h reaction time

Reusability of catalyst

Table 2 shows the result for reusability of the optimum parameters to produce fatty acid methyl ester. The percentage yield of the biodiesel declined from 65.50% to 53.01% after 4 cycles. The decreasing value of percent yield of the fatty acid methyl ester might be caused from the methyl ester residue formed from the previous reactions clogging the catalyst pores (Burmana et al., 2023).

Table 2 Effect of catalyst reusability on under optimum parameters

Cycle	Percentage yield (%)
1	65.50%
2	54.58%
3	53.76%
4	53.01%

Conclusion

In conclusion, heterogeneous catalyst sulfonated TiO_2-SiO_2 from rice husk waste biomass, is capable of producing methyl ester. The catalyst was able to produce 65.50% of methyl ester in 1 hour with methanol to oil ratio of 9:1 and catalyst loading of 3 wt%. This type of catalyst has a good potential to produce FAME from *C. inophyllum* oil.

Ethic Statement

The research does not require research ethics approval.

Authors Contribution

Writing - draft preparation, Kamarullah, S.H., Mustafa, N.A.; Writing - Review and editing, Kamarullah, S.H., Mustafa, N.A.

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Conflict of interests

Author declares no conflict of interest.

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