Continuous Biodiesel Production from Waste Soybean Oil Using a Nano-Fe$_3$O$_4$ Microwave Catalysis

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Abstract: In this study, we conducted an efficient microwave-assisted transesterification process combining homogeneous and heterogeneous catalytic phases to produce biodiesel from waste soybean oil. A cylindrical quartz reactor packed with nanoparticles of Fe$_3$O$_4$ as a co-catalyst was applied to improve the reaction. The process was carried out with a methanol-to-oil molar ratio of 6:1, power of 560 W, and residence time of 30 s. The specifications of the biodiesel produced in this study were compared with two standards, i.e., ASTM D6751 and EN 14214. We found that the continuous conversion of waste soybean oil to methyl ester was approximately 95%. The biodiesel showed a higher flash point and a higher carbon residue content than that of both standards, and the viscosity (5.356 mm$^2$/s) and density (898.1 kg/m$^3$) were both at a high level. Compared to a conventional heating plate, the energy consumption was significantly reduced by nearly 93%. It is expected that these findings will provide useful information for green and sustainable processes for the regeneration and reuse of oil.

Keywords: microwave catalysis; nano-Fe$_3$O$_4$; waste soybean oil; transesterification; biodiesel

1. Introduction

Due to the shortage of petroleum products, biodiesel has become more and more attractive as an alternative fuel resource. The advantages of biodiesel include renewability, biodegradability, environmental friendliness, low emissions, and a lack of toxicity; furthermore, biodiesel can be used in normal diesel engines [1–3].

Transesterification is a well-established chemical reaction whereby vegetable oils and animal fats react with an alcohol to form fatty acid alkyl esters and glycerol [4–6]. The biodiesel yield via a transesterification reaction depends on many parameters such as the concentration of catalyst, the presence of water and soap formation, the presence of free fatty acids (FFAs), and separation of glycerol [7]. Transesterification is also known as alcoholysis, and methanol and ethanol are the most common alcohols used in the reaction. A stoichiometrically alcohol-to-oil molar ratio of 3:1 is required to complete the reaction. Since the transesterification reaction is a reversible reaction, excess alcohol is used in practice to shift the equilibrium to the product side, and, therefore, increase the biodiesel yield. A lower alcohol-to-oil molar ratio affects the conversion of the triglycerides to the methyl esters, while a higher molar ratio reduces the yield [8–10].

The amount of FFAs present also affects the fuel properties and biodiesel yield [11–13]. Several methods for producing biodiesel have been developed, and the transesterification process can be carried out in either the presence or the absence of a catalyst. A catalyst
is used to accelerate the reaction and to produce biodiesel of a higher quality [14]. Both alkaline and acid catalysts have been used depending on the characteristics of a feedstock. Alkaline catalysts are preferred because they require a lower reaction temperature and pressure, have a higher reaction speed, and give a higher conversion efficiency as compared with acid catalysts [15–17]. When a feedstock with an FFA content greater than 1% is used, acid catalysts are better than alkaline catalysts because they are insensitive to the presence of FFAs [8], and product recovery becomes complicated in an alkali-catalyzed transesterification reaction because of the saponification that occurs [18,19].

In general, transesterification is carried out by conventional heating using either a carbon-based fuel or electrical power for the heating process. Thus, to produce a high yield of biodiesel the large amount of energy consumed, and the long reaction time required makes this method inefficient. Microwave heating is unique and offers a number of advantages over conventional heating including non-contact heating, efficient energy transfers rather than heat transfer, rapid heating, material selective heating, heating from interior of the material body, a higher level of safety, and the ability to automate the process. Therefore, heat transfer using microwaves is more effective than conventional heating and in such circumstances the reaction can be completed within a shorter time period [20–22].

The microwave-assisted transesterification of various feedstocks such as soybean oil [23], cotton seed oil [24], Pongamia pinnata seed oil [25], algal oil [26], and waste frying palm oil [27] has been reported by several studies [28]. These studies have shown that the chemical reactions can be accelerated by microwave radiation and high product yields can be achieved within a short time. In the transesterification process, the mixture of feedstock oil, methanol, and metal hydroxide contains both polar and ionic components, therefore, rapid heating is observed under microwave irradiation and because the energy interacts with the sample at the molecular level, very efficient heating is obtained [29]. Various technologies that use a microwave-assisted technique have been developed to reduce the production costs for biodiesel. These studies have focused on transesterification by different catalysts and using different reaction conditions with the microwave heating system. In a 2016 study, researchers used NaOH, KOH, CH₃ONa, and CH₃OK as the homogeneous alkali catalysts for biodiesel production from soybean oil. At a methanol-to-oil molar ratio of 6:1 and catalyst concentration of 1% at 60 °C, they found that alkali metal alkoxides were more active transesterification catalysts than hydroxides. In addition, they found that sodium methoxide was the most efficient catalyst, and about 98.93% conversion of soybean oil was obtained [30]. Standish and Worner reported that particle size was also an important factor during microwave heating but did not necessarily have to be consistent [31]. They observed that the heating rate of fine particles was faster than that of coarse particles of Al₂O₃, but that coarse particles of Fe₃O₄ were heated faster than fine particles. Their results also showed that microwave heat treatment enhanced the reduction rates and they estimated that the cost of microwave heating was 15–50% lower than conventional methods. In our previous study, we reported that Fe₃O₄, as a microwave co-catalyst, performed effectively for reducing energy consumption during the heating process [32]. The nano surface of particles can be used to produce catalysts with extremely high activity, selectivity, and high stability to obtain a high yield of the reaction product, by changing the surface functionality, elemental composition, or number of atoms in the particles [19]. Lipase immobilized on Fe₃O₄ magnetic nanoparticles has the integrated advantages of traditional immobilized lipase and free lipase for its rather fast reaction rate. A biodiesel yield of 81–89% has been achieved by lipase immobilized on Fe₃O₄ compounds’ magnetic nanoparticles [33].

In this study, we used a microwave catalytic system as a fixed bed to produce biodiesel from waste soybean oil using an alkaline microwave-assisted transesterification process. The nano surface of Fe₃O₄ particles was used as the microwave co-catalyst and the surface morphology of Fe₃O₄ was analyzed by a scanning electron microscopy (SEM). Using an ethanol-to-oil molar ratio of 6:1, the reaction time, microwave power, properties of
biodiesel, and energy consumption were compared to results obtained using a conventional heating plate.

2. Materials and Methods

2.1. Microwave Catalytic Reactor Design

A quartz column (50 mm I.D. by 53 mm O.D. by 27.5 cm L) was used as the microwave co-catalyst packed column (approximately 120 mL) and filled with granules of Fe$_3$O$_4$ (50 g, approximately 40 mL in volume). A domestic microwave oven (RE-079R, Sampo, Taoyuan, Taiwan), with a frequency of 2450 MHz, was modified to contain the co-catalyst-filled quartz column and was used as the microwave catalytic reactor.

2.2. Manufacturing of the Fe$_3$O$_4$ Microwave Co-Catalyst

FeCl$_3$ (1.4 g, 97%, Sigma-Aldrich, St. Louis, MO, USA) and FeCl$_2$·H$_2$O (0.86 g, 97%, Sigma-Aldrich, St. Louis, MO, USA) were added to a solution of oleic acid and acetone and stirred for 3 min at 56 °C. Then, ammonia (NH$_4$OH) (5 mL, 5 N, Hayashi Pure Chemical, Osaka, Japan) and oleic acid (1 mL) were added and stirred for 5 min to produce Fe$_3$O$_4$ precipitate. After washing with a solution (10 mL) of acetone (99%, Kyotowako Pure Chemical, Kyoto, Japan) and methanol (99.5%, Hayashi Pure Chemical, Osaka, Japan) (volume ratio 1:2) three times, the Fe$_3$O$_4$ was dried at 60 °C for 2 h. Finally, the Fe$_3$O$_4$ precipitate was sintered at 800 °C for 10 min, and then sintered at 1050 °C for 4 h with a temperature increase of 1 °C/min in order to form the solid catalyst. In addition, the metal oxide of Fe$_3$O$_4$ microwave co-catalyst was analyzed by scanning electron microscopy (S-3000H, Hitachi High-Tech, Tokyo, Japan) in order to observe its surface morphology.

2.3. Performance Testing of the Microwave Catalytic Reaction System

2.3.1. Processes in Batch Mode

A schematic of the microwave catalytic reaction system is shown in Figure 1. The waste soybean oil used in this study was expired salad oil (unopened) obtained from the Taiwan Sugar Corporation, Taiwan. A catalytic and non-catalytic temperature analysis was carried out in batch mode at different levels of microwave power (266, 532, and 800 W) in order to compare the temperatures in the microwave reactor with and without Fe$_3$O$_4$ co-catalyst using the same volume of 80 mL.

![Figure 1. Schematic diagram of the microwave catalytic system. (1) mixing flask; (2) peristaltic pump; (3) microwave reactor; (4) thermometer; (5) separating funnel; (6) washing flask; (7) separating funnel; (8) oven.](image-url)
2.3.2. Processes in Continuous Mode

A microwave catalytic continuous test was carried out at a power of 560 W with a methanol-to-oil molar ratio of 6:1 using a 1.0% NaOH-methanol solution. The flow rate was set at 170 mL/min, with an effective volume of the quartz reaction tube to fill the catalyst of 80 mL, and with a hydraulic retention time of 30 s. A peristaltic pump (MasterFlex Replacement motor model 7553-80, Cole Parmer, Vernon Hills, IL, USA) was used to quantitatively pump the soybean oil (700 g) and NaOH-methanol solution (7 g NaOH dissolved in 200 g methanol) in order to control the hydraulic retention time. A thermometer was set up at the outflow port of the reactor to ensure that the reaction temperature reached 60 °C. After the reaction, the upper layer of the sample was separated and collected using a separating funnel, and then washed with distilled water (at a volume ratio of 1:1) for several times to remove residual glycerol. Finally, the washed sample was put into an oven (60 °C) for further refining.

2.4. Analysis of the Physical and Chemical Properties of the Biodiesel

An analysis of the physical and chemical properties of biodiesel was carried out based on the standard test methods supplied by ASTM International [34], which included density (at 15 °C, ASTM D4052), flash point (ASTM D93), viscosity (at 40 °C, ASTM D445), sulfur ash (ASTM D5453), copper strip corrosion (ASTM D130), cetane number (ASTM D613), water and sediment content (ASTM D2709), distillation temperature (90% and 95%, ASTM D86), and carbon residue (ASTM D4530). The parameters of the American Standard Specification for Biodiesel (ASTM D6751) and the European Standard for Biodiesel (EN 14214) were used as the standards compared with the biodiesel made by this study [3,11,13].

3. Results and Discussion

3.1. Surface Morphology of the Fe₃O₄ Co-Catalyst

The Fe₃O₄ metal oxide co-catalyst used in this study consisted of circular granules with a diameter of approximately 0.3 cm (Figure 2) and was adopted as the microwave heating co-catalyst. The surface morphology of the Fe₃O₄ co-catalyst was analyzed by SEM, since particle size is an important factor in microwave heating [31,33,35]. Before being loaded into the SEM chamber, the Fe₃O₄ co-catalyst was coated with a film of Au for 120 s to avoid electron accumulation and discharge due to its low electrical conductivity. The surface morphology of the Fe₃O₄ co-catalyst is shown in Figure 2. In the SEM microphotograph (Figure 2B), it can be observed that the Fe₃O₄ nanoparticles are mostly uniform with a size of approximately 100 nm.

Figure 2. (A) exterior image of the Fe₃O₄ catalyst; (B) SEM microphotograph (10,000×) of the Fe₃O₄ catalyst taken in secondary electron imaging (SEI) mode with a working distance (WD) of 10.1 mm.
3.2. Temperature Comparisons of the Microwave Process with and without Fe$_3$O$_4$ Co-Catalyst

A batch test of microwave heating was carried out with and without Fe$_3$O$_4$ co-catalyst at a methanol-to-oil molar ratio of 6:1 using a 1.0% NaOH–methanol solution [25,30,36]. Since the reaction was carried out in a closed system, the volatilization of methanol could be ignored. As shown in Figure 3, the temperature increased with time over a period of 20 to 80 s at microwave powers of 266, 532, and 800 W. The increase in the temperature of the feedstock added with Fe$_3$O$_4$ co-catalyst was higher than that without Fe$_3$O$_4$ co-catalyst added in the batch microwave heating test. When tested without Fe$_3$O$_4$ co-catalyst and heating for 80 s at a microwave power of 266 W, the temperature increased from 25 to 47 °C, whereas the temperature increased from 25 to 58 °C when the Fe$_3$O$_4$ co-catalyst was present under the same conditions. At the microwave power levels of 532 and 800 W without Fe$_3$O$_4$ co-catalyst, the temperature increased from 25 to 67 °C and from 25 to 75 °C, respectively. At microwave powers of 532 and 800 W with the Fe$_3$O$_4$ co-catalyst applied, greater temperature increases from 25 to 78 °C and from 25 to 98 °C were observed, respectively. These results indicate that microwave heating could be further improved with the application of Fe$_3$O$_4$ co-catalyst. The highly effective increase in temperature in the microwave heating system could be attributed to the direct adsorption of the microwave radiation by the metal oxide [37].

![Figure 3. Comparisons of the co-catalytic and non-co-catalytic temperatures at microwave powers of (A) 266 W, (B) 532 W, and (C) 800 W for various batch tests (presence or absence of 50 g of Fe$_3$O$_4$ catalyst with a feedstock oil volume of 80 mL).](image)

3.3. The Performance of Fe$_3$O$_4$ Microwave Catalysis during Continuous Biodiesel Production

The continuous microwave catalytic system was operated with a reaction time of 0.5 min at a microwave power of 560 W, and the results are shown in Figure 4. Without the addition of Fe$_3$O$_4$ co-catalyst, the temperature increased up to 45 °C after 4 min and remained almost constant thereafter. Obviously, the temperature at 45 °C was not sufficient to complete the transesterification reaction. On the other hand, when the Fe$_3$O$_4$ nanoparticles were used, the temperature rapidly increased to above 60 °C in only 2 min at the same microwave power of 560 W, and then remained at approximately 61 °C after 4 min.
These results indicate that a microwave catalytic heating process with the addition of Fe$_3$O$_4$ nanoparticles shortens the reaction time and provides better efficiency in transmethylation.

![Figure 4](image_url). The temperature curve changes during continuous testing at a microwave power of 560 W with or without Fe$_3$O$_4$ co-catalyst. The reaction was carried out in triplicate, and the average is shown in the figure.

In this efficient system, which combined the heterogeneous Fe$_3$O$_4$ co-catalyst with the homogeneous catalyst CH$_3$ONa, the feedstock conversion rate for glyceride transesterification was estimated to be approximately 95%. The heat transfer with high efficiency could be attributed to the absorbance of microwaves by Fe$_3$O$_4$ nanoparticles [37]. Some experimental factors applied in microwave-assisted transesterification processes were summarized in Table 1. The system used in the study applied a relatively short reaction time (30 s) and a moderate microwave power (560 W) for the process. Since the continuous biodiesel production was only carried out in a short-time test and 1.0 L-scale, further persistent tests with a longer period of time and scale-up tests should be carried out to confirm for industrial application.

| Feedstock                        | Alcohol  | Molar Ratio (Alcohol/Oil) | Catalyst                        | Reaction Time | Microwave Power (W) | Temperature (°C) | References |
|----------------------------------|----------|---------------------------|--------------------------------|---------------|---------------------|-----------------|------------|
| Sunflower oil                    | Methanol | 6:1                       | NaOCH$_3$ (1 wt%)               | 5 min         | -                   | 60              | [20]       |
| Mixture of waste cooking oil and | Methanol | 5.96:1                    | KOH (0.774 wt%)                 | 7.15 min      | 850                 | 100             | [22]       |
| *Calophyllum inophyllum* oil     | Methanol | 6:1                       | CH$_3$ONa (0.75 wt%), KOH (1.5 wt%) | 3 min         | 750                 | -               | [23]       |
| Soybean oil                      | Methanol | -                         | NaOH (0.5 wt%), KOH (1 wt%)     | 3–10 min      | 125-300             | 60              | [24]       |
| Cottonseed oil                   | Methanol | -                         | NaOH (0.5 wt%), KOH (1 wt%), NaOCH$_3$ (1 wt%), BaO (1.5 wt%), SrO (20 wt%); KOH (1 wt%); NaOH (0.5 wt%) | 60 s (KOH, NaOH), 4 min (BaO, SrO) | 800 | - | [25] |
| *Camelina Sativa* oil            | Methanol | 9:1                       | NaOH (3 wt%), NaOH (0.5 wt%), BaO (1.5 wt%), SrO (20 wt%) | 30 s | 800 | 78 | [26] |
| Palm oil (fried)                 | Ethanol  | 12:1                      | KOH (1 wt%), NaOH (3 wt%)       | 10 min        | 500                 | 60              | [27]       |
| Yellow horn oil                  | Methanol | 12:1                      | Ce$_{2}$H$_2$PW$_{12}$O$_{40}$ (1 wt%) | 10 min        | 500                 | 60              | [38]       |
| Waste soybean oil                | Methanol | 6:1                       | CH$_3$ONa (1 wt%)               | 30 s          | 560                 | 60–61           | This study * |

*: Continuous microwave catalytic system with addition of Fe$_3$O$_4$ nanoparticles.
3.4. Analysis of the Physical and Chemical Properties of the Biodiesel

The parameters of the ASTM D6751 and EN 14214 were used to evaluate whether the biodiesel produced in this study meets the specifications of diesel fuel, and the results are shown in Table 2 [39–41]. When biodiesel was produced using Fe₃O₄ microwave co-catalytic converter technology, the cetane index was 51.1, the flash point was 182 °C, the sulfur content was 0.18 ppm, the distillation temperature was 345–346 °C, the density was 0.8981 g/mL, the water and sediment content was 0.05 vol%, the kinematic viscosity was 5.356 mm²/s, the copper strip corrosion was a mild color, the pour point was 0 °C, and the amount of carbon residue was 0.60 wt%. The water and sediment content and carbon residue of the biodiesel produced in this study show higher levels than those of both the ASTM D6751 and EN 14214 standards. The cetane number of the biodiesel is slightly over the minimum level of the EN 14214 standard, i.e., by 0.1. Although the density fits the range of the EN 14214 standard, it shows a rather high level. The viscosity of the biodiesel fits the range of the ASTM D6751 standard, however, it is over the maximum level of the EN 14214 standard by 0.356. Moreover, the lower limit of ester content (or methanol fatty acid methyl esters, %FAME) according to the EN 14214 standard is 96.5%, the biodiesel in this study had a lower level of 95% (estimated by glyceride transesterification). Above all, the biodiesel produced in this study cannot be regarded as an ideal biodiesel ready for use, and further modifications (such as elongation of reaction time in the process) is necessary. Nevertheless, the Fe₃O₄ microwave catalytic heating system proposed in this study was convenient for processing and has the potential to become a good practice that could reduce the cost of producing biodiesel.

Table 2. Biodiesel specifications *1 and the properties of the biodiesel made in this study.

| Items                                | ASTM D6751 | EN 14214 | Biodiesel (This Study) |
|--------------------------------------|------------|----------|------------------------|
| Density at 15 °C (kg/m³)             | 889        | 860–900  | 898.1                  |
| Flash point (°C)                     | >130       | >101     | 182                    |
| Viscosity at 40 °C (mm²/s)           | 1.9–6.0    | 3.5–5.0  | 5.356                  |
| Sulfur (ppm)                         | <15        | <10      | 0.18                   |
| Copper strip corrosion *2            | Class 3 max.| Class 1 rating | Class 1                |
| Cetane number                        | >47        | >51      | 51.1                   |
| Water content and sediment           | <0.005 (vol%) | <500 (mg/kg) | 0.05 (vol%)            |
| Distillation temperature (°C)        | 360        | °3       | 345–346                |
| Carbon residue (wt%)                 | <0.05      | <0.3     | 0.6                    |

*1, references of [39–41]. *2, Class 1 = mild discoloration; Class 1 rating = light orange; Class 3 max = almost just polished copper. °3, no data.

3.5. Comparisons in Terms of Energy Consumption

An energy consumption comparison between the traditional hot plate approach and the microwave catalytic heating method approach was carried out at the same conversion rate of 95%. The measurements and calculations were based on electricity consumption at a laboratory scale. An amount of 1.0 L soybean oil at a methanol-to-oil molar ratio of 6:1 using a 1.0% NaOH-methanol solution was applied as a benchmark. We found that the conventional heating plate method at a power of 420 W took 120 min to complete the reaction, whereas the continuous test of the new Fe₃O₄ microwave catalytic heating system at a power of 560 W, took only 6 min to reach the same conversion rate, that is, to reach the same conversion rate, the conventional heating plate method consumed 0.84 kWh while the new Fe₃O₄ microwave catalytic heating system consumed 0.056 kWh. In other words, the power consumption ratio between the Fe₃O₄ microwave catalytic heating system in this study and a conventional heating plate method was about 1 to 15. At a scale of a 1.0 L sample, the production cost of the nano-Fe₃O₄ microwave catalytic heating system was significantly reduced in terms of energy input by nearly 93% as compared with a traditional heating plate method. Chang et al. reported about 56% reduction in energy consumption by using an Fe₃O₄ co-catalyst with a microwave heating process [31]. Patil et al. also discovered that around 95% of energy was saved using a microwave heating method as
compared with a conventional method [42]. The results based on the microwave heating method showed high energy efficiency as compared with conventionally heated reactors, which was similar to the results described by Moseley and Woodman at the meso scale (from 1 L to 3 L) for organic reactions [43].

4. Conclusions

In summary, this study demonstrated that biodiesel can be produced from waste soybean oil using a microwave-assisted transesterification system. The system was modified and improved by the addition of Fe₃O₄ nanoparticles, and the heat transfer with better efficiency was confirmed by the tracing of heating temperature. In the batch system, after heating for 80 s with the Fe₃O₄ co-catalyst applied, the temperature increases were enhanced from 47 to 58, 67 to 78, and 75 to 98 °C at microwave powers of 266, 532, and 800 W, respectively. In the continuous system, it took only 2 min to reach the steady temperature of 60 to 61 °C at the power of 560 W, and the feedstock conversion rate for glyceride transesterification was estimated to be approximately 95%. As for the specifications of biodiesel, the biodiesel produced in this study showed a higher level of viscosity than that of the EN 14214 standard and fit the range of the ASTM D6751 standard. Since the water content and sediment and carbon residue content were higher than that of both standards, the biodiesel produced in this study cannot be regarded as an ideal fuel ready for use. To improve the qualities of the biodiesel, further modifications of the process are necessary. Nevertheless, this study proposed an efficient biodiesel production process that reduced energy input by nearly 93% as compared with that for a conventional heating plate method. These findings could promote the development of green and sustainable processes for the effective utilization of reusable resources.

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