

BIODIESEL PRODUCTION FROM WASTE COOKING OIL BY USING ULTRASONIC TUBULAR REACTOR

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Abstract

The aim of this research is to find an optimum of synthesis biodiesel from waste cooking oil (WCO) using ultrasonic tubular reactor. The experimental studies explored the variations in reaction time, molar ratio WCO to methanol (MeOH), amount of catalyst, frequency of ultrasonic and output power ultrasonic on the ester contents. Comparisons of type ultrasonic and also mechanical stirring method based on time reaction were investigated. The optimum results of biodiesel process is the reaction time of 5 minute, NaOH catalyst 1%wt of WCO, molar ratio WCO to MeOH of 1:6, frequency ultrasonic of 20 KHz and output power ultrasonic of 650 W. The reaction time reduced 12-24 times compared to both of method and the yield of ester contents was obtained at 96.54%wt.

Keywords: Ultrasonic tubular reactor; biodiesel; waste cooking oil; Sonochemistry; cavitation; transesterification

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1. Introduction

Biodiesel is an alternative diesel fuel from renewable feedstock such as vegetable oils or animal oil [1]. Many advantages of biodiesel compared to diesel fuel base on petroleum oil such as biodegradable, renewable, nontoxic and cleaner engine emissions [2]. Biodiesel feedstock's in Malaysia is mainly refined palm oil [3], due to Malaysia being an exporter of palm oil and about 17.7 million tons of palm oil in 2008 and of that 40% of the palm oil are made into cooking oil, margarine, specialty fats and oleochemicals [4]. However, biodiesel from palm oil is not competitive cost wise because the feedstock is expensive. Therefore, WCO is one alternative feedstock for biodiesel process due to the price of WCO is 2.5–3.5 times cheaper than palm oil and non-edible oil, thus this can significantly reduce the total manufacturing cost of biodiesel [5] and it was estimated 0.5 million tones/year of WCO are disposed off yearly in Malaysia without treatment as wastes [6].

Biodiesel is produced by transesterification of triglyceride (TG) with an alcohol, such as methanol (MeOH) or ethanol, in the presence of a catalyst, mainly alkaline (NaOH, KOH and CH₃ONa), to biodiesel or FAME (fatty acid methyl ester) and glycerol as side product. The alternative technology

for biodiesel process such as ultrasonic technique [7-15], co-solvent [16], microwave [17], catalytic conversion [18], enzymatic production [19], and supercritical MeOH [20] have been studied intensively with the objective of improving the conversion, reaction time and material consumption with the higher yield [9].

Ultrasonic technology has been recognized as an effective method to enhance mass transfer rate between immiscible liquid-liquid phases within a heterogeneous system [10]. When a liquid is irradiated by a strong ultrasonic wave, large quantity of tiny gas bubbles appear and collapse violently, which is a phenomenon known as acoustic cavitation [21]. The tiny bubbles repeatedly expand and contract according to the pressure oscillation of an ultrasonic wave [22]. The speed of the bubble collapse sometimes increases up to the sound velocity of the liquid, and the temperature and the pressure inside a bubble increase dramatically because such a strong collapse is nearly adiabatic [24,25]. As a result, ultrasonic in transesterification has proven to be an efficient mixing tool and provides sufficient activation energy to initiate the reaction [11]. Ultrasonic-assisted transesterification does not only shorten reaction time, but also minimize the molar ratio of alcohol to oil and reduce

energy consumption compared to conventional mechanical stirring method [25]. There are many researchers on transesterification of vegetable oils at laboratory scale have reported excellent biodiesel yield by ultrasonic technique. Many type of conventional ultrasonic such as ultrasonic cleaner [7,12], ultrasonic horn [13,14], and ultrasonic processor [14-15,26] for biodiesel process has been investigated. However, conventional ultrasonic systems are based on the relatively-fixed resonant frequency of the transducers used, small capacity and inefficient sonochemistry reactors. Therefore, an ultrasonic tubular reactor is one of the alternative tool for reaction of biodiesel. It is tool have piezoelectric transducers type at tube with the longitudinal vibration. The tube is driven by the transducers and can effectively transform the longitudinal vibration into the radial vibration; moreover, the frequency and the output power ultrasonic can be adjustable.

The objective of this research was to find optimum synthesis of biodiesel from waste cooking oil using ultrasonic tubular reactor as well as to find the effect of the reaction time, molar ratio WCO to MeOH, amount of catalyst, frequency of ultrasonic and power output of ultrasonic on the ester contents using ultrasonic tubular reactor, and to compare process time completion, product quality and time of reaction with conventional ultrasonic cleaner and mechanical stirring method.

2. Experimental and Procedures

2.1 Materials

WCO was collected from a local Malaysian potato crackers factory in Parit Raja, Batu Pahat, Johor. It was then filtered through normal sieve to remove any suspended food particle and then heated to evaporate water residue in it. As the free fatty acid contents in the WCO obtained was observed to be very little (less than 3%) [14], the WCO was used directly. The chemical and physical property of WCO is reported in Table 1. Methanol pure analytic grades (p.a), sodium hydroxide (NaOH) (p.a), potassium hydroxide (p.a), phenolphthalein indicator (p.a), ethanol (p.a) were purchased from HmbG Chemical in Malaysia.

Another chemical such as heptanes (p.a), Pyridine (p.a), MSTFA (N-methyl-N-trimethylsilyl trifluoroacetamide) used as solvent for gas chromatography (GC) and purchased from Fluka, Malaysia. Glycerin, 1-Mono [cis-9-octadecenoyl]-rac-glycerol (monoolein), 1,3-Di [cis-octadecenoyl] glycerol (diolein), 1,2,3-Tri [cis-octadecenoyl] glycerol (triolein), (S)-(-)-1,2,4-Butanetriol (Internal

Standard 1), 1,2,3-Tridecanolyglycerol (tricaprin) (Internal Standard 2), and methyl heptadecanoate were supplied from Sigma-Aldrich Malaysia.

Table 1. Properties and composition of WCO

Properties and composition	WCO
Appearances	Dark brown
Density at 40°C (g/ml)	0.8936
Kinematic Viscosity at 40°C (cm ² /s)	37.3771
Acid value (mg KOH/g oil)	1.5630
Free fatty acid as oleic (% wt)	0.7850
Myristic acid (% wt)	3.27
Palmitic acid (% wt)	21.06
Stearic acid (% wt)	7.22
Oleic acid (% wt)	35.31
Linoleic acid (% wt)	17.20
Linolenic acid (% wt)	15.50

2.2 Experiment

2.2.1 Equipment

The ultrasonic tubular reactor type MSG.1200.IX-LF and conventional ultrasonic cleaner laborette 17 are shown in Fig. 1a, 1b, respectively. It used to perform the transesterification reaction from WCO. The specification of ultrasonic tubular reactor have diameter 60 mm x 580 mm length with quick flanges and covers (material SS316L), maximal output ultrasonic power 1300 W and adjustable frequency range 17.5 kHz to 28.5 kHz. Meanwhile ultrasonic cleaner laborette 17 have specification as follows: diameter 24.5 cm, 13 cm deep, 5.6 liters capacity and fixed frequency at 35 kHz.

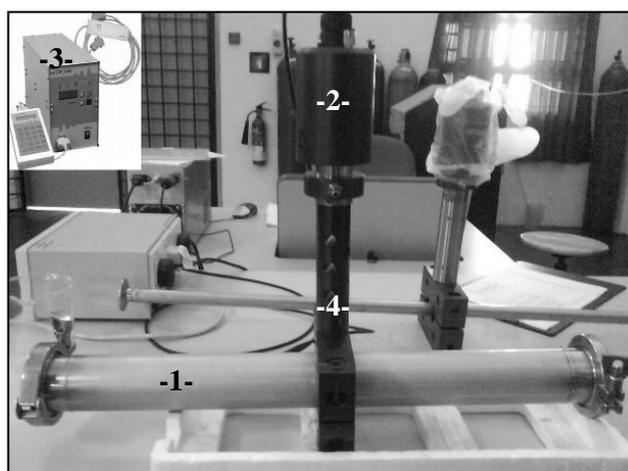


Fig. 1a. Experimental set-up of ultrasonic tubular reactor for biodiesel process. (1) Tube reactor, (2) Transducer, (3) Ultrasonic Generator, (4) Acoustic wave guide

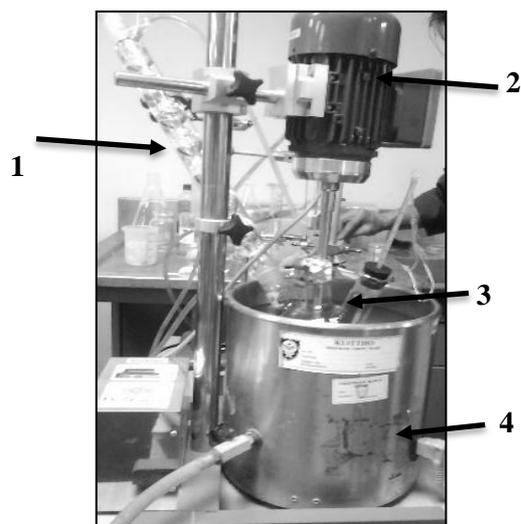


Fig. 1b. Experimental set-up of conventional ultrasonic cleaner for biodiesel process. (1) reflux condenser, (2) motor stirrer, (3) Three-neck flask, (4) Ultrasonic bath

2.2.2 Experimental procedure

The experiment was carried out using an ultrasonic tubular reactor which it is included mastersonic software for control the ultrasonic parameter. Ultrasonic tubular reactor with frequency of 20 kHz was arranged on display ultrasonic generator. 100 g of WCO was first added to the reactor. The powder of NaOH first dissolved completely in methanol (MeOH) and then the mixture of MeOH – NaOH was added to the tube reactor. As soon as the mixture of MeOH – NaOH was added to the WCO, the time was noted and the reaction was carried for about 5 minutes. After completion, the mixture was transferred into a separator tunnel for phase separation. The biodiesel mixture formed at the upper layer and glycerol form in the lower layer. The traces of catalyst and MeOH in biodiesel layer were washed out with warm water until pH 6-7. Finally, the ester layer was dried and analyzed.

Different operating parameters were used in this research, by optimizing the ester contents of WCO based on reaction time at 3, 5, 10, and 15 minute, including WCO to MeOH molar ratio at 1:6, 1:9, and 1:12, the amount of catalyst (0.5%wt, 0.75%wt, 1 %wt and 1.25%wt based on WCO), the ultrasonic frequency were 18 kHz, 20 kHz and 22 kHz and the output power ultrasonic were 325 Watt, 650 Watt and 975 Watt.

Experiments were also performed using conventional ultrasonic cleaner and mechanical stirring. The optimized parameter from ultrasonic tubular reactor was applied to conventional ultrasonic cleaner and mechanical stirring.

Experimental procedures for conventional ultrasonic and mechanical stirring were similarly adopted for ultrasonic tubular reactor method.

2.2.3 Chemical and Physical Properties

The chemical and physical properties of WCO and biodiesel were evaluated in numerous tests carried out to determine if the biodiesel produced met the European Standard EN 14214 and ASTM D 6751 (ASTM = American Society for Testing and Materials) such as :

1. The density measurements were carried out at temperature 40°C, using a precision balance Mettler AE 200 and a Gay Lussac type pycnometer.
2. The kinematic viscosity of samples was determined using Hydromotion Viscolite 700. This test method involves measuring the temperature conducted with water bath for control the temperature (40°C).
3. The flash point was measured by the Pensky-Martens closed cup method according to ASTM D 93-08.
4. Methyl ester contents were analyzed using a gas chromatography (Perkin Elmer GC Model Clarus 500) equipped with an Elite-Famewax, 30 m x 320 μm x 0.25 μm film. The analytic conditions for ester contents were as follow: the column temperature was used 210°C, temperature flame ionization detector (FID) of 250°C, pressure of 80 kPa, flow carrier gas of 1 ml/min, temperature injector of 250°C, split flow rate of 50 ml/min, time for analysis 20 minute and volume injected of 1 μl , referred to European Standard EN 14103.
5. Analysis of total glycerol and free glycerol referred to European Standard EN 14105. The composition of triglyceride, diglyceride, monoglyceride and glycerol was analyzed using a Perkin Elmer Gas Chromatography (GC) Model Clarus 500, equipped with a DB-5 HT capillary column (0.53 mm x 5 m) J&W Scientific. The following condition of GC are: the column temperature was started at 50°C held for 1 min, programmed 1 at 15°C/min up to 180°C, programmed 2 at 7 °C/min up to 230°C, programmed 3 at 10°C/min up to 370°C, final temperature held for 5 min, detector temperature at 380°C, carrier gas pressure (hydrogen) at 80 kPa, volume injected used 1 μl .
6. Free fatty acid (FFA) and acid value content were determined by a standard titrimetry method referred to AOCS Ca 5a-40.

3. Results and Discussion

3.1 Effect of WCO to MeOH Molar Ratio

At first, the effect of WCO to MeOH molar ratio on the ester contents is investigated to understand the optimum condition using ultrasonic technique. Fig. 2 is showed the changes in the ester contents at various molar ratios of WCO to MeOH with NaOH catalyst of 1%wt under ultrasonic tubular reactor. The optimum ester contents were observed of 96.54%wt, 85.24%wt, 92.55%wt at 5 minutes for molar ratio (WCO to MeOH) of 1:6, 1:9 and 1:12, respectively.

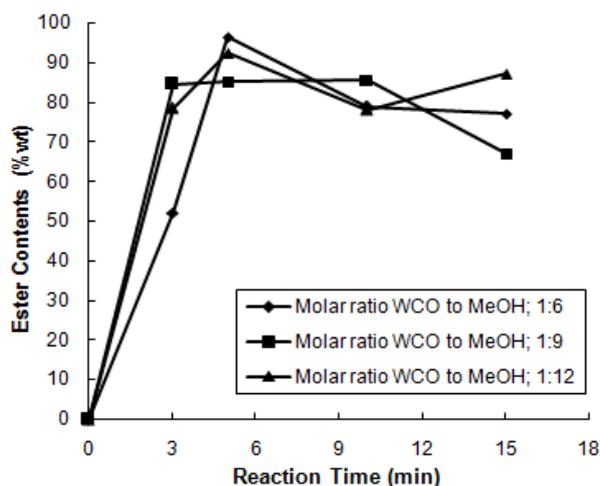


Fig. 2. Effect of molar ratio of WCO to MeOH on ester contents (%wt) at NaOH concentration of 1 %wt, output power ultrasonic of 650 W and frequency ultrasonic of 20 kHz

The reactant of MeOH and WCO are immiscible liquids, the transesterification reaction form on the layer between the WCO and MeOH and it will occurred the FAME and glycerol as by product. Due to glycerol and MeOH are polar compounds, they can be dissolve each other at any ratio. Then, the presence of glycerol will be absorbs significant amounts of MeOH, requiring large amounts of MeOH for the transesterification reaction. However, the use of large amounts of excess MeOH has adverse effects on the phase separation of glycerol and FAME, and increases the energy and time consumption for the recovery of excess MeOH [27]. It was indicated that the ultrasonic frequency form a fine emulsion at 5 minute, because the interface area between MeOH to FAME was increased, it caused the rate of the transesterification can be enhanced, and reduce the amount of excess MeOH required. On the other hand, the ester contents at molar ratio 1:6, 1:9, 1:12

were decreased at 10 to 15 minutes. It could be deduced from the study that to increase the yield of biodiesel [3], the excess amount of alcohol needed was 1:6 to increase the ester yields to maximum and beyond this limit (maximum yield), the excess alcohol slightly decreased the yields and this could be due to the dilution of the FAME with the alcohol [28]. The reaction of transesterification is reversible. It was indicated that the reaction reverse toward to TG due to the equilibrium reaction occurred in 5 minutes.

3.2 Effect of Catalyst Amount

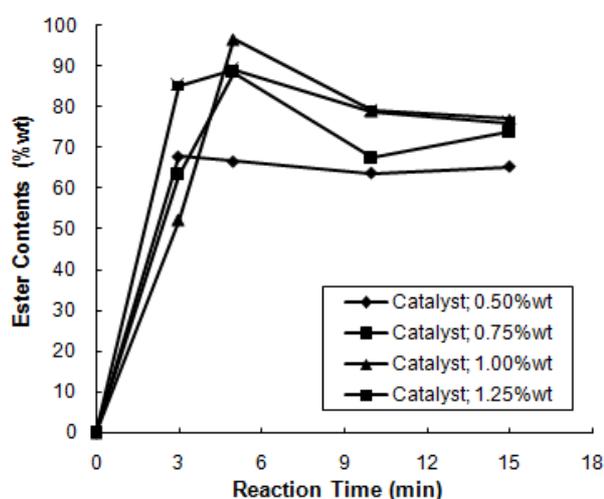


Fig. 3. Effect of amount of NaOH catalysts on ester contents (%wt) at molar ratio WCO to MeOH of 1:6, output power ultrasonic of 650 W and frequency ultrasonic of 20 kHz

Fig. 3 showed the effect of ester contents and reaction time with various amount of catalyst under ultrasonic tubular reactor. The maximum ester contents were observed at 1% wt of NaOH catalyst in molar ratio WCO to MeOH of 1:6 at 5 minute reaction time. In this case, the ester contents on various amount of catalyst were the same when increased at 3 to 5 minutes of reaction time and decreased at 5 to 15 minutes of reaction time. Excess catalyst reacted with the WCO, leading to the formation of soap, thus as the catalyst concentration increased, the separation of esters become more difficult. A minimal amount of 0.5%wt to 0.75%wt NaOH catalyst was adequate in propelling the transesterification reaction using ultrasonic tubular reactor, however an amount of 1% wt gave the best yields. As shown in the figure, and similar results with Encinar et al [29], with high concentration of NaOH catalyst (1.25%wt) form soaps in the presence of large residues of fatty acids resulting in

emulsion formation between soaps and water molecules, thus leading to low yields of ester contents.

3.3 Effect of Ultrasonic Frequency

This step was to establish the influence of ultrasonic frequency on the ester contents. From the obtained results in Fig. 4, the reactions performed under ultrasonic at 20 kHz produced higher ester contents than 18 and 22 kHz in 5 minutes reaction. Usually there are no important differences in the formation of the cavitation bubbles at 18 and 22 kHz, but in this case, at 20 kHz the reaction time was shorter to raise the equilibrium reaction. After less than 3 minutes of sonication, the mixture form emulsification. This is due to collapse of the cavitation bubbles and ultrasonic jets that impinge WCO to MeOH, disrupting the phase boundary and causing emulsification. On the other hand, at 5 minutes when the frequency ultrasonic increased from 20 to 22 kHz, the emulsion was formed in a higher amount and some ester is trapped into emulsion, causing the ester contents to decrease. At higher frequencies, the collapses of cavitation bubbles are not strong enough to impinge one liquid to the other [12]. The MeOH and WCO is immiscible and also its very poor hence emulsification does not occur, then transesterification process takes place mainly at the boundary between the two layers.

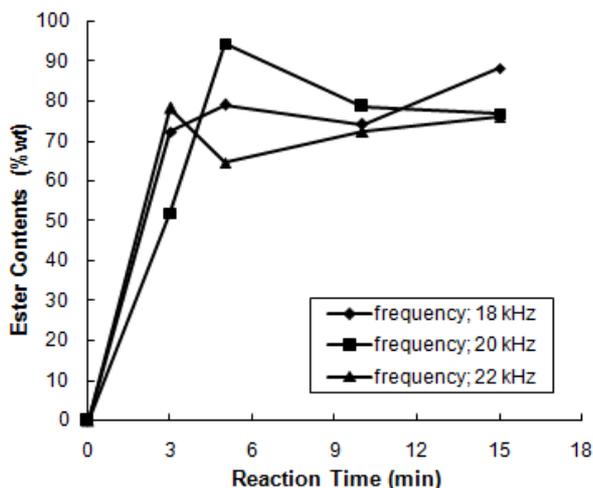


Fig. 4. Effect of frequency ultrasonic on ester contents (%wt) at molar ratio WCO to MeOH of 1:6, amount of NaOH catalyst of 1% and output power ultrasonic of 650 W

3.4 Effect of Ultrasonic Power

The optimal parameters such as WCO to MeOH molar ratio (1:6), NaOH catalyst (1% wt), ultrasonic

frequency (20 kHz) were kept constant and the output ultrasonic power was varied from 325 W to 975 W for the same operating volume of the reaction mixture. The results have presented in Fig. 5. At output power of 350 W, the obtained ester contents were around 75.19%wt at 5 minutes of reaction but when the output power was increased to 650 W, the ester contents also increased to about 96.54%wt. This can be explained, the ultrasonic output power (intensities) provoke larger amplitude at the vibrating surface in contact with the liquid [30]. On the other hand, increase in the output power ultrasonic from 650 W to 975 W at 5 minutes, the ester contents was decreased. This can be attributed to the fact that at higher output power ultrasonic usually creates more and larger bubbles, which may coalesce and lead to less transient events. But according to Hingu et al., higher power level ultrasonic will decreased transfer of energy into the system and hence lower cavitation activities [14].

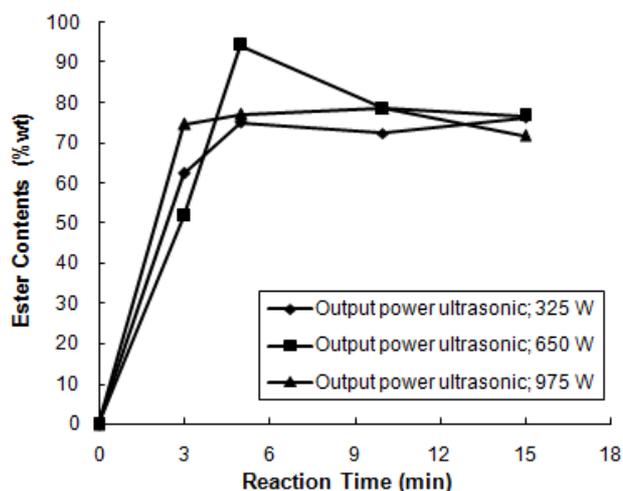


Fig. 5. Effect of output power ultrasonic on ester contents (%wt) at molar ratio WCO to MeOH of 1:6, amount of NaOH catalyst of 1% and frequency ultrasonic of 20 kHz

3.5 Comparison of Type Ultrasonic and Mechanical Stirring Method

The results of optimization using ultrasonic tubular reactors were compared with the conventional ultrasonic cleaner and also mechanical stirring method. The obtained results were shown in Fig. 6. Conventional ultrasonic cleaner and mechanical stirring experiments were performed using the same amounts of reactants and molar ratio WCO to MeOH used in the ultrasonic tubular reactor. The conversion of FFA in the WCO into FAME (fatty acid methyl ester) was calculated from the mean of acid value (Av) of the oil layer by the following equation (1) [31].

$$\text{Conversion (\%)} = \left[1 - \frac{Av_{oil}}{Av_{wco}} \right] \times 100 \quad (1)$$

where Oil and WCO refers to FAME layer and waste cooking oil, respectively.

It can be seen from the figure that ultrasonic tubular reactor was obtained 98.40 %wt of conversion from WCO to FAME whereas the conventional ultrasonic cleaner method results is much lower to the extent of conversion (30.07%wt) over similar time of operation at 5 minutes. This indicated that the reaction of transesterification using ultrasonic tubular reactor is quicker to form biodiesel compared to conventional ultrasonic approach. because the agitation intensity appears to be of particular importance for alcoholysis process [32]. The phenomenon reaction was occurred due to ultrasonic tubular reactors which have a close system. It generates a wide range of cavitation bubble sizes and a greater density of cavitation bubbles. This provides faster and better cleaning, faster sonochemical reactions, faster physical reactions, and faster liquid degassing [33]. But ultrasonic (tubular reactor and cleaner) gave high ester contents compared to mechanical stirring.

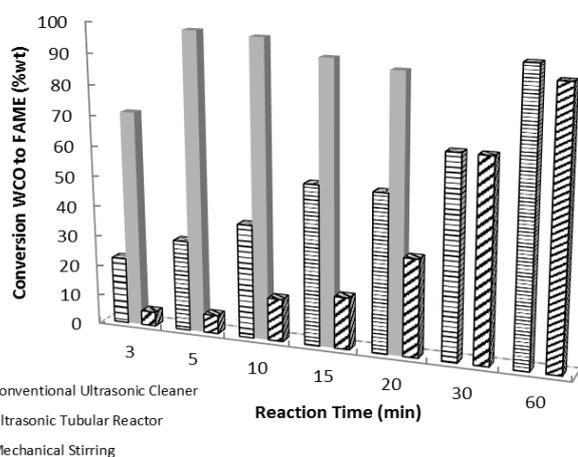


Fig. 6. Comparison of type ultrasonic and mechanical stirring method on conversion WCO to FAME. Reaction conditions: WCO to MeOH ratio of 1:6 and NaOH amount 1 %wt; Ultrasonic tubular reactor: frequency 20 kHz, output power ultrasonic 650 W, close system. Conventional ultrasonic cleaner: frequency 35 kHz and input power ultrasonic 140 W, open system. Mechanical Stirring: stirrer speed 350 rpm, open system

Furthermore, scale-up of ultrasonic tubular reactor to meet industrial-scale operations is more efficient than the conventional ultrasonic cleaner by reason of its less sensitivity to the geometric dimension of the reactor and much more efficient.

The results of chemical and physical analysis are shown in Table 2. All produced samples fulfilled the European standard limit in terms of acid value, kinematic viscosity, density, flash point, free glycerin, triglyceride, diglyceride, monoglyceride and methyl ester content. These results suggested that ultrasonic tubular reactors are the most effective method for the production of quality biodiesel from waste cooking oil.

Table 2. Chemical and physical properties of biodiesel from WCO using ultrasonic tubular reactor.

Parameters	Biodiesel	Biodiesel Standard (EN 14214 and ASTM D6751)
Appearance	Pale dark yellow	
Density at 40°C (kg/m ³)	853.1	-
Kinematic Viscosity at 40°C (cm ² /s)	3.90	3.5 – 5.0
Acid value (mg KOH/g oil)	0.067	<0.5
Flash point (°C)	150	>120
Methyl ester contents (% wt)	96.54	>96.5
Free glycerol (% wt)	-	<0.020
Triglycerides (% wt)	-	<0.2
Diglycerides (% wt)	0.0101	<0.2
Monoglycerides (% wt)	0.1016	<0.8

4. Conclusion

This research has explored the application of ultrasonic tubular reactor to synthesis biodiesel from waste cooking oil in a close system reactor. In the optimal process it showed that molar ratio WCO to MeOH of 1:6, NaOH catalyst of 1%wt, frequency ultrasonic of 20 kHz, output power ultrasonic of 625 W and 5 minute of reaction, produced ester contents of 96.54%wt. Experimental results indicated that high ester contents yields with shorter reaction time, smaller amount of MeOH and NaOH catalyst. Comparison between ultrasonic tubular reactors with conventional ultrasonic cleaner had similar enhancement effect on the transesterification reaction. However, both methods gave shorter reaction time (12-24 times) than the mechanical stirring method. Ultrasonic tubular with close system reactor were proven to be an efficient method for production of biodiesel from WCO. In conclusion, ultrasonic tubular reactor has proved to be highly efficient method to produce biodiesel at bench scale and definitely have a high potential in industry scale.

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