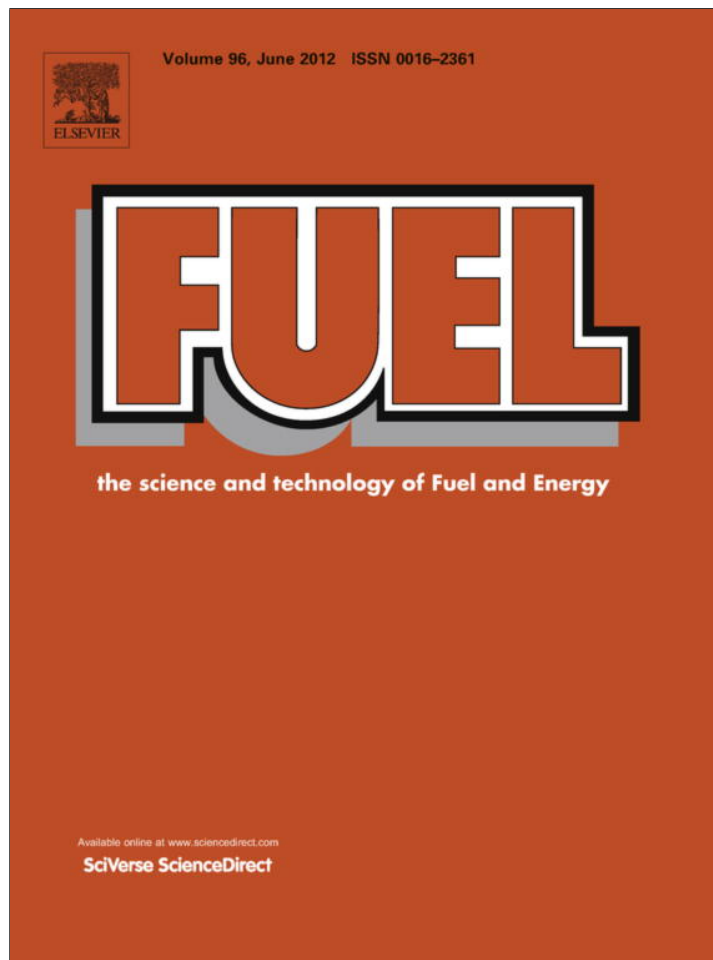


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## Short communication

## A continuous-flow bubble column reactor for biodiesel production by non-catalytic transesterification

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## ABSTRACT

A continuous-flow bubble column reactor for biodiesel production by non-catalytic transesterification of triglycerides has been developed. The principle of a bubble column reactor for transesterification is similar to reactive distillation, where the reaction products in the gas phase are continuously removed from the reactive zone, while oil, as the reactant, is retained in the reactive zone (liquid phase). A dedicated laboratory-scale continuous-flow bubble column reactor, with a predetermined 200 mL liquid volume of palm oil in the reactor, has been used for this study. In this study, the effects of the methanol feed flow rates (1.5, 3.0, and 6.0 mL/min) and reaction temperatures (250, 270, and 290 °C) on the biodiesel productivity and methyl esters content in the biodiesel product were evaluated. The biodiesel productivity increased with MeOH feed flow rate and reaction temperature correspondingly. On the other hand, the purity of the methyl ester in the biodiesel product decreased.

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## 1. Introduction

Bubble column reactors (BCRs) have been widely used for conducting gas–liquid reactions in numerous industrial applications. However, utilization of BCR for biodiesel production has only been reported by Yamazaki et al. [1] and Joelianingsih et al. [2,3]. Those reports showed that the BCR can be used for catalyst-free biodiesel production at atmospheric pressure. The reaction was carried out in semi-batch mode, where bubbles of superheated methanol (MeOH) vapor were continuously blown into the oil-containing reactor. The advantage of this method is that it requires no catalyst, and thereby transesterification of oils with high free fatty acid content can be accomplished without prior treatments. Joelianingsih et al. [2] studied the effect of reaction temperatures (250, 270 and 290 °C) on the rate constant, conversion of reaction, yield of biodiesel and methyl esters (MEs) content in the biodiesel product on the kinetics of the non-catalytic transesterification of palm oil at atmospheric pressure. For larger capacity commercial production, a continuous transesterification processes are preferred over batch processes due to the requirement of consistent product quality and low capital and operation costs per unit of product.

A continuous catalytic transesterification of vegetable oils to mono-alkyl esters has been proposed as early as 1940s [4,5] and

studied until recently [6–10]. In this study, the BCR was used for the continuous mode of non-catalytic biodiesel production. The effect of MeOH feed flow rates (1.5, 3.0, and 6.0 mL/min) and reaction temperatures (250, 270, and 290 °C) on the biodiesel productivity and ME content in the biodiesel product were evaluated.

## 2. Materials and methods

## 2.1. Materials

Refined palm oil was obtained from Spectrum Chemical Mfg. Corp., Gardena, New Brunswick, with the following characteristics: iodine value 50–55, free fatty acid (as oleic acid) 0.1% w/w, myristic acid, 0.5–5.9% w/w; palmitic acid, 32–47% w/w; stearic acid, 2–8% w/w; oleic acid, 34–44% w/w; linoleic acid, 7–12% w/w. Methanol p.a. (Pro analysis) of minimum 99.8% purity was utilized from Merck, Germany. Benzene and hexane with high grade, used for TLC/FID (thin layer chromatography/flame ionization detector) analysis, were purchased from Wako Pure Chemical Industries, Ltd., Japan. Benzene was used as the developing solution, and hexane as the solvent. Squalane (C<sub>30</sub>H<sub>62</sub>), the internal standard used in the TLC/FID analysis, was purchased from Sigma–Aldrich Japan. Triolein, diolein, mono-olein, oleic acid and methyl oleate, used as standard solutions in the TLC/FID analysis, were purchased from Sigma Chemical, St. Louis, MO.

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## 2.2. The continuous-flow BCR system

The experiment was conducted using a laboratory-scale continuous-flow BCR system with a constant liquid volume (200 mL) in the reactor. Palm oil and MeOH were pumped continuously into the reactor and the gas phase product was removed continuously from the reactor. The schematic flow diagram of the laboratory-scale continuous-flow BCR system used in the experiment is shown in Fig. 1.

The reactor is a BCR, equipped with an inner tube inside. The inner tube is covered with two perforated plates both at the bottom and the top. Each of the perforated plates has 30 holes, with each a diameter of 4 mm. The inner tube has a diameter of 43 mm and a height of 90 mm. The inside and outside diameter of the column is 55 mm and 71 mm, and the height of the column is 210 mm. The liquid hold-up capacity in the column is 200 mL. The material of the column and inner tube is stainless-steel (316SS). The vaporizer, super-heater and reactor are equipped with five electrical heaters and five temperature controllers. The level controller is equipped with an inner pipe. The glass container for the sample collector is equipped with one pipe each on the bottom and on the top.

## 2.3. Procedure and conditions

The reactor was initially filled with 200 mL of refined palm oil via the level controller and valve-5. Prior to charging, nitrogen gas was entrained to fill up the MeOH pipe and the reactor via valve-1 and to fill up the oil pipe via valve-2. During charging of the oil, nitrogen gas flow to the oil pipe was stopped, but nitrogen gas flow to the MeOH pipe was maintained to prevent oil backflow to MeOH area. The oil in the reactor was heated to the desired temperatures (250, 270 or 290 °C) and atmospheric pressure (0.1 MPa). MeOH was pumped out from the storage glass at various flow rates (1.5, 3.0, and 6.0 mL/min) to the vaporizer for vaporization (flow of nitrogen gas was stopped). Initially, the effect of the MeOH feed flow rates on the biodiesel productivity was studied at the same reaction temperature (290 °C) to obtain the optimum flow rate condition. Then, the effect of the reaction temperatures (250, 270 and 290 °C) on the biodiesel productivity was studied at the optimum MeOH feed flow rate and atmospheric pressure (0.1 MPa). The MeOH vapor was entrained through a super-heater and continuously purged out into the reactor. Palm oil was continuously pumped into the reactor via valve-3 to maintain the level of oil

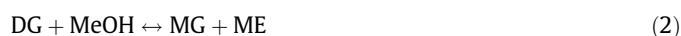
in the reactor. The excess of oil was removed via the inner pipe in the level controller. The oil consumption rate was calculated from the difference between the feed flow rate and excess flow rate via inner pipe O3. Products in the gas phase were condensed and collected in the glass container for sampling collection. The reaction products were taken from the glass container via a pipe at the bottom of the glass container every 30 min and then weighed (sample A). During the reaction time (300 min), 10 samples were collected for analysis.

## 2.4. Analysis

The methanol content in the A samples was evaporated using a rotary evaporator and then the biodiesel products were obtained and weighed for productivity calculations (sample B). The contents of TG (triglycerides), DG (diglycerides), MG (monoglycerides), free fatty acids (FFAs), and ME in the B samples (devoid of MeOH) were analyzed with TLC/FID [11]. Analysis was performed with an Iatroscan MK-5 Analyzer (Iatron Laboratories, Inc., Japan). The FID utilized hydrogen and air with flow rates of 160 mL/min and 2000 mL/min, respectively. Type SIII Chromarods were used as the thin layer. Before being spotted, rods were scanned as blank on the instrument to obtain the proper degree of hydration. The B samples (20–30 mg) were diluted with 1 mL solvent (250 mg squalane in 50 mL hexane) and 1 µL of the solution was spotted on each rod. Five replicates were used for each solution. The rods were developed for 30 min (they were stored in a glass chamber in which the atmosphere was saturated with benzene vapor), oven dried at 110–130 °C for 5 min and then analyzed on the Iatroscan. The mass fractions of TG, DG, MG, FFA and ME in the B samples were calculated based on the concentration of the internal standard.

## 3. Results and discussion

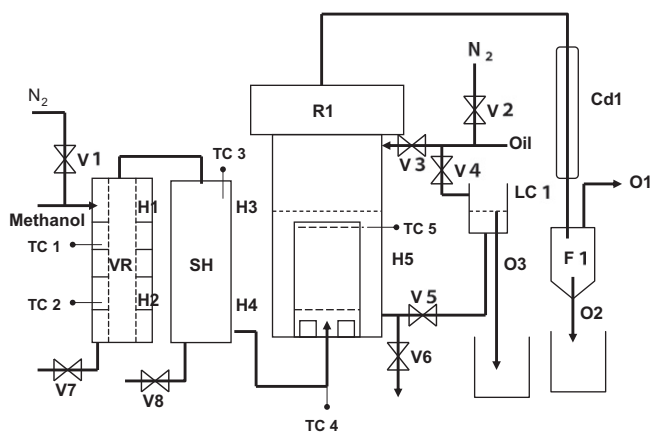
Biodiesel which consists of fatty acid methyl esters (FAME, sometimes called ME only) can be produced by transesterification of TG or esterification of fatty acids with short-chain alcohol, mainly MeOH. Studies of the mechanism and kinetics of transesterification have shown that this reaction consists of a number of consecutive and reversible reactions as presented in the following equations [12,13]:



According to the reaction above, biodiesel consists not only of ME but also of other impurities, such as TG, DG, and MG.

In a BCR, the role of MeOH is not only as reactant but also as gas bubbles and a carrier gas for the reaction product (ME and GL) in the gas phase. The role of MeOH as a carrier gas is similar to the steam in the steam distillation process. Methanol vapor will extract and bring ME and GL into the vapor phase so that both these compounds will be separated from TG, MG, and DG, as reactants in the liquid phase. This role implies a shift of the reaction direction to the right and an increase in the conversion rate of the reaction. Therefore, the completion of the reaction depends on multiple parameters, including the MeOH feed flow rate, temperature, reaction time, and properties of the feedstock and all of the components in the reaction mixture.

In general, the operation of a BCR reactor for biodiesel production by non-catalytic transesterification of TG is complicated because it consists of several processes such as diffusion of MeOH gas into liquid (oil), transesterification reaction of TG in the liquid



**Fig. 1.** The schematic flow diagram of the continuous-flow BCR system. VR: vaporizer, SH: superheater, R1: bubble column reactor, LC1: level controller, Cd1: condenser, F1: glass container for sample collector, V1–V8: valves, H1–H5: electrical heaters, TC1–TC5: temperature controllers, O1: pipe on the top of F1, O2: pipe on the bottom of F1, O3: inner pipe in the LC1

phase and distillation. Accordingly, optimum operating conditions have to be determined from the result of systematic investigations of all operating parameters. However, the complexity of the BCR system can be minimized when applied to the biodiesel production for two reasons. First, the difference between the boiling temperatures of the products (GL and ME) and TG was large enough to provide easier separation of the product from the product mixture. Second, the transesterification reaction occurred in the liquid phase only. The super-heated MeOH vapor will dissolve in the liquid bulk of oil soon after it passes through the liquid–gas interface, and react with the non-volatile component (TG, DG, and MG) [14].

### 3.1. Effect of the MeOH feed flow rate on biodiesel productivity

Productivity of biodiesel primarily refers to the amount of biodiesel (kg) produced per unit of liquid volume in the reactor (L) per unit time (h). Fig. 2 shows the mass of the biodiesel product every 30 min reaction time under the various MeOH feed flow rates at 290 °C reaction temperature.

As presented in Fig. 2, productivity of the biodiesel increased with the reaction time and became almost constant after about 300 min of reaction time. Higher MeOH feed flow rate resulted to higher productivity of biodiesel. At 300 min of reaction time, the productivity obtained was 0.028 kg/L/h of biodiesel for a 1.5 mL/min flow rate of MeOH. For a 3.0 mL/min flow rate of MeOH, productivity reached 0.128 kg/L/h of biodiesel. On the contrary, for a 6.0 mL/min flow rate of MeOH, productivity attained 0.180 kg/L/h.

This result indicates that the higher the MeOH feed flow rate, the higher the mass of MeOH that will react with palm oil.

### 3.2. Effect of reaction temperature on biodiesel productivity

In order to analyze the effect of reaction temperature on the productivity of biodiesel, various temperatures were used (250, 270, and 290 °C) with a 3.0 mL/min flow rate of MeOH. The 3 mL/min flow rate was used utilized because it resulted in a complete reaction previously, as indicated by the two layer formation (Eqs. (1)–(3)).

Changes in mass of the biodiesel product with reaction time under the various reaction temperatures are shown in Fig. 3. The productivity of biodiesel was calculated based on the data shown in Fig. 3. The respective productivity at reaction temperatures of 250, 270 and 290 °C were 0.010, 0.052 and 0.128 kg/L/h. The productivity of the biodiesel was calculated at a 300 min reaction time.

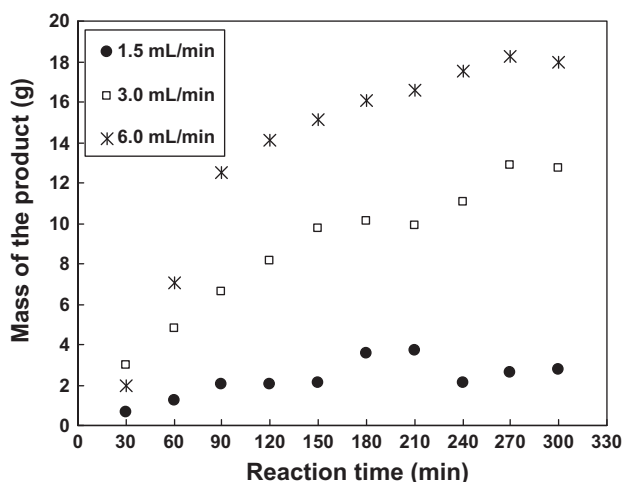


Fig. 2. Mass of the biodiesel product under the various MeOH feed flow rates at 290 °C.

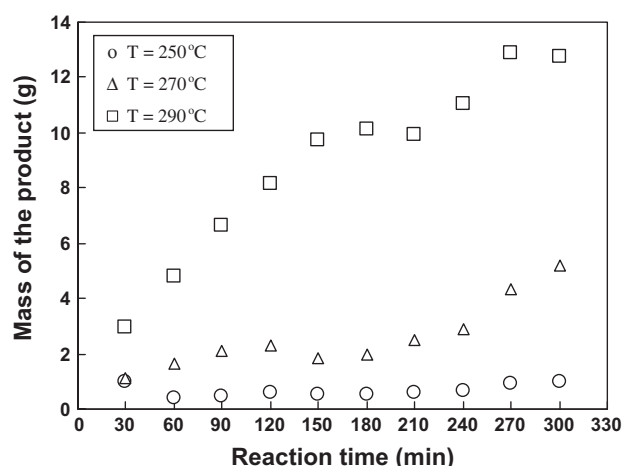


Fig. 3. Mass of the biodiesel product under the various reaction temperatures.

The temperature effect can be clearly seen, i.e. that the productivity of biodiesel increases with reaction temperature. This occurrence is in accordance to the reaction kinetics principles.

### 3.3. Effect of the MeOH feed flow rate on ME content

The effect of the MeOH feed flow rate on the ME content in the biodiesel product at a given reaction temperature is presented in Tables 1–3.

After 300 min of reaction time, the ME content in the biodiesel product decreased with the increase of MeOH feed flow rate. This result indicates that at a higher MeOH feed flow rates, more DG and MG content is carried out to the biodiesel product.

### 3.4. Effect of reaction temperature on ME content

The effect of the reaction temperature on ME content in the biodiesel product at a given MeOH feed flow rate (3.0 mL/min) is presented in Fig. 4.

Fig. 4 shows that ME content in the biodiesel product was superior at the higher reaction temperature during the initial stage of the process, but after 210 min of reaction time the lower temperature showed higher ME content. To attain 94% w/w of ME in biodiesel, a higher reaction temperature requires shorter reaction time. Reaction temperatures of 250, 270 and 290 °C required 240, 180 and 90 min of reaction time, respectively. According to reaction kinetics principles, reaction rates increase with reaction temperature. Hence, the time needed to achieve equilibrium reaction is shorter. After 300 min of reaction time, the ME content in the

Table 1

The composition of the biodiesel product at 290 °C with a 1.5 mL/min of MeOH flow rate.

Reaction time (min)	Composition of the product (% w/w)				
	ME	TG	FFA	DG	MG
30	67.4 ± 0.2	3.9 ± 0.4	0.0 ± 0.0	15.7 ± 2.9	13.0 ± 2.3
60	78.5 ± 1.7	2.8 ± 0.2	0.0 ± 0.0	8.7 ± 0.6	10.0 ± 1.4
90	88.1 ± 0.8	0.0 ± 0.0	0.0 ± 0.0	6.1 ± 0.7	5.8 ± 0.9
120	94.6 ± 1.3	0.0 ± 0.0	0.0 ± 0.0	3.4 ± 0.3	2.0 ± 0.6
150	96.6 ± 0.6	0.0 ± 0.0	0.0 ± 0.0	2.0 ± 0.2	1.4 ± 0.3
180	97.1 ± 0.2	0.0 ± 0.0	0.0 ± 0.0	1.8 ± 0.1	1.1 ± 0.1
210	97.7 ± 2.9	0.0 ± 0.0	0.0 ± 0.0	1.3 ± 0.5	1.0 ± 0.5
240	97.6 ± 1.9	0.0 ± 0.0	0.0 ± 0.0	0.9 ± 0.5	1.5 ± 0.2
270	96.1 ± 0.9	0.0 ± 0.0	0.0 ± 0.0	1.5 ± 0.4	2.4 ± 1.0
300	97.1 ± 0.6	0.0 ± 0.0	0.0 ± 0.0	0.9 ± 0.4	2.0 ± 0.4

**Table 2**

The composition of the biodiesel product at 290 °C with a 3.0 mL/min of MeOH flow rate.

Reaction time (min)	Composition of the product (% w/w)				
	ME	TG	FFA	DG	MG
30	81.4 ± 1.9	4.1 ± 0.2	0.8 ± 0.2	10.3 ± 0.9	3.4 ± 0.6
60	92.0 ± 1.2	0.0 ± 0.0	0.0 ± 0.0	5.1 ± 0.3	2.9 ± 0.2
90	95.3 ± 2.1	0.0 ± 0.0	0.0 ± 0.0	2.4 ± 1.1	2.3 ± 0.7
120	95.3 ± 1.8	0.0 ± 0.0	0.0 ± 0.0	2.2 ± 0.8	2.5 ± 1.2
150	95.2 ± 1.1	0.0 ± 0.0	0.0 ± 0.0	2.2 ± 0.1	2.6 ± 0.4
180	94.2 ± 1.5	0.0 ± 0.0	0.0 ± 0.0	2.0 ± 0.1	3.8 ± 0.1
210	95.4 ± 2.8	0.0 ± 0.0	0.0 ± 0.0	1.8 ± 0.5	2.8 ± 0.4
240	94.8 ± 2.7	0.0 ± 0.0	0.0 ± 0.0	1.7 ± 0.1	3.5 ± 0.4
270	95.3 ± 1.7	0.0 ± 0.0	0.0 ± 0.0	1.4 ± 0.4	3.3 ± 0.4
300	94.9 ± 1.8	0.0 ± 0.0	0.0 ± 0.0	1.6 ± 0.2	3.5 ± 0.5

**Table 3**

The composition of the biodiesel product at 290 °C with a 6.0 mL/min of MeOH flow rate.

Reaction time (min)	Composition of the product (% w/w)				
	ME	TG	FFA	DG	MG
30	83.0 ± 0.4	1.2 ± 0.3	1.2 ± 0.8	8.4 ± 0.7	6.2 ± 0.3
60	86.3 ± 1.4	0.2 ± 0.1	0.0 ± 0.0	7.8 ± 1.1	5.7 ± 0.6
90	88.8 ± 1.9	0.0 ± 0.0	0.0 ± 0.0	6.6 ± 1.8	4.6 ± 0.7
120	90.3 ± 1.6	0.0 ± 0.0	0.0 ± 0.0	6.4 ± 0.3	3.3 ± 0.3
150	90.7 ± 1.5	0.0 ± 0.0	0.0 ± 0.0	6.2 ± 0.2	3.1 ± 0.1
180	91.5 ± 1.2	0.0 ± 0.0	0.0 ± 0.0	5.3 ± 0.6	3.2 ± 0.4
210	91.9 ± 1.0	0.0 ± 0.0	0.0 ± 0.0	5.0 ± 0.7	3.1 ± 0.4
240	93.5 ± 1.8	0.0 ± 0.0	0.0 ± 0.0	3.5 ± 0.8	3.0 ± 0.6
270	94.2 ± 1.5	0.0 ± 0.0	0.0 ± 0.0	3.6 ± 0.4	2.2 ± 0.5
300	92.7 ± 2.2	0.0 ± 0.0	0.0 ± 0.0	4.0 ± 0.2	3.3 ± 0.3

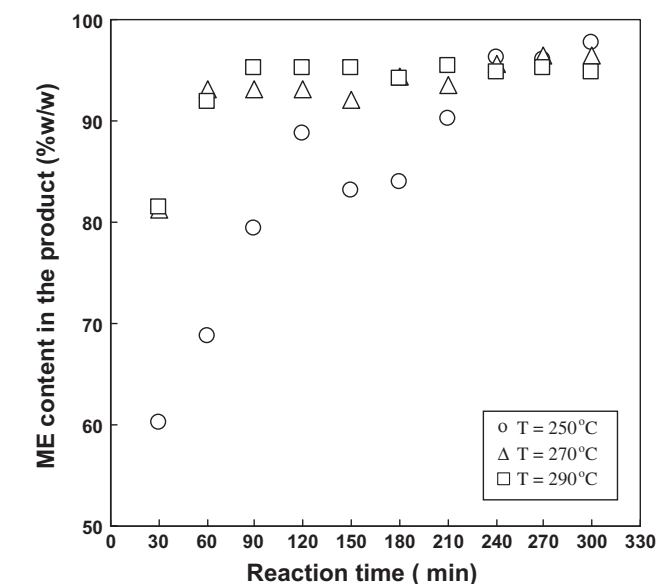


Fig. 4. Effect of reaction temperature on the ME content in the biodiesel product.

product at 250, 270 and 290 °C was 97.7, 96.5 and 94.9% w/w correspondingly. The principles of reaction thermodynamics (chemical reaction equilibrium) on exothermal characteristics of transesterification [15] explain the reason why this phenomenon occurs. The characteristic of the exothermal reaction is that with a decrease in reaction temperature the equilibrium shifts to the product, so that the ME content increases with the decrease in reaction temperature at the end of the reaction time. In addition, the principle of TG transesterification by BCR is similar to reactive distillation (RD), where the reaction products in the gas phase (GL

**Table 4**

The composition of the biodiesel product at 250 °C with a 3.0 mL/min of MeOH flow rate.

Reaction time (min)	Composition of the product (% w/w)				
	ME	TG	FFA	DG	MG
30	60.2 ± 0.4	9.5 ± 0.8	0.8 ± 0.2	23.9 ± 1.7	5.6 ± 1.2
60	68.7 ± 1.1	8.5 ± 0.6	0.1 ± 0.2	16.5 ± 1.8	6.2 ± 0.9
90	79.3 ± 1.6	3.1 ± 0.4	0.0 ± 0.0	9.2 ± 2.3	8.4 ± 1.2
120	88.7 ± 0.9	2.3 ± 0.2	0.0 ± 0.0	5.0 ± 0.7	4.0 ± 0.3
150	83.2 ± 2.3	0.4 ± 0.4	0.0 ± 0.0	8.1 ± 0.4	8.3 ± 1.0
180	84.1 ± 2.4	0.4 ± 0.3	0.0 ± 0.0	7.9 ± 0.7	7.6 ± 0.7
210	90.2 ± 2.2	0.0 ± 0.0	0.0 ± 0.0	3.8 ± 1.2	6.0 ± 0.8
240	96.3 ± 1.5	0.0 ± 0.0	0.0 ± 0.0	1.1 ± 0.1	2.6 ± 0.2
270	96.0 ± 0.9	0.0 ± 0.0	0.0 ± 0.0	2.0 ± 0.6	2.0 ± 0.3
300	97.7 ± 1.9	0.0 ± 0.0	0.0 ± 0.0	1.0 ± 0.5	1.3 ± 0.2

**Table 5**

The composition of the biodiesel product at 270 °C with a 3.0 mL/min of MeOH flow rate.

Reaction time (min)	Composition of the product (% w/w)				
	ME	TG	FFA	DG	MG
30	81.2 ± 1.9	0.9 ± 0.2	0.1 ± 0.0	10.6 ± 0.8	7.2 ± 0.8
60	92.9 ± 1.6	0.2 ± 0.1	0.0 ± 0.0	4.1 ± 0.3	2.8 ± 0.6
90	93.2 ± 2.1	0.0 ± 0.0	0.0 ± 0.0	3.7 ± 0.4	3.1 ± 0.9
120	93.2 ± 2.6	0.0 ± 0.0	0.0 ± 0.0	3.4 ± 0.4	3.4 ± 0.2
150	92.1 ± 0.7	0.0 ± 0.0	0.0 ± 0.0	4.0 ± 0.1	3.9 ± 0.4
180	94.3 ± 1.7	0.0 ± 0.0	0.0 ± 0.0	3.1 ± 0.2	2.6 ± 0.2
210	93.4 ± 2.1	0.0 ± 0.0	0.0 ± 0.0	3.2 ± 0.1	3.4 ± 0.2
240	95.6 ± 0.9	0.0 ± 0.0	0.0 ± 0.0	1.9 ± 0.1	2.5 ± 0.2
270	96.5 ± 0.9	0.0 ± 0.0	0.0 ± 0.0	1.6 ± 0.1	1.9 ± 0.3
300	96.5 ± 2.0	0.0 ± 0.0	0.0 ± 0.0	1.3 ± 0.4	2.2 ± 0.3

and ME) are continuously removed from the reactive zone, while the reactants (TG) remain in the reactive zone (liquid phase). But, the content of DG and MG in the product is higher at higher temperatures as showed in Tables 2, 4 and 5. Therefore, at the end of the reaction, the ME content is higher at lower temperatures because the content of DG and MG in the product is lower.

According to distillation principle, the DG and MG content in the product increase with reaction temperature, because DG and MG are more volatile than TG. BCR is a process unit that enables reaction and distillation in a single unit, and could be an excellent option for biodiesel production since it can achieve 100% reaction conversion of TG under ideal conditions.

#### 4. Conclusions

In a continuous-flow BCR for ME production by non-catalytic transesterification, the productivity of biodiesel increased with MeOH feed flow rate and reaction temperature. On the other hand, the purity of ME in the biodiesel product decreased. The result of this study is similar to previous research in the semi-batch process.

For further research, experimental work need to be done experimental using a solid catalyst in order to obtain higher productivity. Through the addition of a catalyst, reaction temperature can be lowered so that the generated products contain ME that meet quality standards, while still maintaining the high productivity.

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