Transesterification of palm oil with methanol in milli-channel reactor: Effects of tube size and shape (straight and bent)

Nurul Asyikin MOHD AZAM, Yoshimitsu UEMURA*, Mohamad Azmi BUSTAM (Faculty of Engineering, University of Technology PETRONAS), Katsuki KUSAKABE (Faculty of Engineering, Sojo University)

SUMMARY
Biodiesel can be produced through transesterification of oil with methanol in the presence of catalyst. Transesterification process could be done using either a batch-wise reactor or continuous reactor. A few studies have reported a common trend that milli-channel reactor with a smaller internal diameter shows a larger oil conversion than that with a larger diameter. However, there are few studies on the effects of tube size and shape on the oil conversion. In this study, the effects of tube internal diameter and tube shape (straight and bent) on the oil conversion were investigated. Also, the results from the milli-channel reactors were compared with that from a batch-wise reactor.

[1] INTRODUCTION
Biodiesel made up from vegetable oils and animal fats is becoming significant to be used due to depleting petroleum reserves [1] and the global warming issues [2]. Biodiesel can be produced by transesterification process; the process whereby oil reacts with alcohol in the presence of catalyst to produce fatty acid methyl ester and glycerol as by-product [1]. By using milli-channel reactor, the reactants are mixed and react in the flow reactor throughout the process [3]. In this work, an experimental study was conducted by using refined palm oil to investigate the effects of tube internal diameter and tube shapes (straight and bent). In order to compare with milli-channel reactor, transesterification using a batch reactor was also conducted.

[2] EXPERIMENTAL
Fig. 1 shows a schematic diagram of the milli-channel reactor system. Transparent PTFE tubes with three different internal diameters (0.58, 1.0, and 1.6 mm; length=1000 mm) were used. Two syringe pumps were used to feed the two liquids (palm oil and methanol containing 0.57 wt% KOH). Both liquids were mixed at a T-shape joint prior to introduction into the milli-channel reactor which was placed on a hot plate.

Fig. 1 Milli-channel reactor system.

Transesterification was carried out at 21:1 methanol to oil molar ratio at 60 °C and the residence time of 60, 120 and 180 s. The product was analyzed using a HPLC with silica-gel column (Shimpack CLC-SIL) and a RI detector.
[3] RESULTS AND DISCUSSION

Fig. 2 shows the plot of $-\ln(1-x)$ versus residence time. The variable $x$ denotes the conversion of oil. The reaction rate constant $k$ can be estimated directly from the slope of each plot. But it should be noted that this $k$ is an apparent reaction constant which may contain mass transfer effect, since the two reactants are existing in two different phases: oil and methanol. Anyway, the reaction rate constant, $k$, increases when the tube internal diameter decreases. The result from 0.58 mm tube internal diameter shows the highest $k$, which is $1.93\times10^{-4}$ s$^{-1}$ followed by 1.0 mm ($1.17\times10^{-5}$ s$^{-1}$) and 1.6 mm ($6.51\times10^{-4}$ s$^{-1}$) of tube internal diameter. The straight milli-channel reactor exhibits somewhere in between, $1.25\times10^{-3}$ s$^{-1}$. Batch reactor shows the lowest $k$ of $5.04\times10^{-4}$ s$^{-1}$.

![Fig. 2 Plot of $-\ln(1-x)$ versus residence time.](image)

If mass transfer step mentioned above influences the overall reaction, the reaction rate constant, $k$ may increase with increase in the interfacial surface area, $s$ per unit volume of methanol, $v$. In order to discuss the trends in Fig. 2 from this point of view, the reaction rate constant, $k$ is plotted against $s/v$ in Fig. 3. In this figure, the results from the bent milli-channels with three sizes (closed circle key) can be approximately correlated by a straight line. This trend means that there is a considerably strong mass transfer effect on the overall reaction rate. The result of the straight milli-channel with 1.6 mm in diameter is also represented by the straight line. In other words, there is no significant difference between bent and straight tubes.

The last important point for discussion is comparison of milli-channel reactor with batch reactor. It is obvious that the result from batch-wise reactor is quite far from the common trend of milli-channel reactor, as can be seen from Fig. 3. Even the batch reactor shows $47,000$ m$^{-1}$ of $s/v$ (around five to ten times of the milli-channel reactor), the apparent reaction rate constant is merely $5.09\times10^{-4}$ s$^{-1}$, which is similar to the result of 1.6 mm milli-channel reactor, which has only $4,300$ m$^{-1}$ of $s/v$. This big discrepancy between the milli-channel reactor and batch reactor may be attributed to the existence of internal circulation flow inside of methanol droplets in milli-channel reactor [4], which may enhance the mass transfer of oil inside of the methanol droplets.

![Fig. 3 Apparent reaction rate constant vs. $s/v$.](image)

[4] CONCLUSIONS

It was found that apparent reaction rate constant, $k$ of two-phase transesterification in milli-channel reactor is approximately proportional to the interfacial surface area per unit volume of methanol ($s/v$). This means there is considerably strong mass transfer limitation for the reaction system under the conditions in this study. But higher mass transfer limitation was observed for batch-wise reaction system. This discrepancy may be attributed to the existence of circulation flow in methanol droplets for milli-channel reactor, which may not exist in methanol droplet for impeller-mixing type batch reactor.


[Acknowledgement] This research was supported by the Mitsubishi Corporation Education Trust Fund.

[Contact] yoshimitsu_temura@petronas.com.my