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Performance Analyses of Fatty Acids in Reactive Distillation Process for Biodiesel Production

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Abstract - Different materials are used for biodiesel production via esterification or transesterification processes. In esterification reaction, an organic acid is reacted with an alcohol to produce an ester (biodiesel) and water (by-product). It then implies that the fatty acid used determines the type and the purity of the biodiesel obtained. As such, the performances of the fatty acids available and used for biodiesel production are not, of course, likely to be the same. So, it is good to have some knowledge about the performances of the fatty acids available for biodiesel production so as to know the type of a fatty acid to choose for the production of a particular biodiesel product at any time. Therefore, this work has been carried out to investigate the performances of some fatty acids used for the production of biodiesel using reactive distillation column. The fatty acids considered were oleic acid, which was discovered, according to Kusmiyati and Sugiharto (2010), to give biodiesel that had the quality required to be a diesel substitute, and some other ones (stearic acid, linoleic acid and palmitic acid) found to be present in jatropha oil. Methanol was used as the alcohol of the reaction. The study was carried out with the aid of Aspen HYSYS that was used to develop the model of the reactive distillation column in which both the reaction for biodiesel production and the separation of the unreacted materials and by-products were simultaneously carried out. The results obtained from the study revealed that, among the fatty acids investigated, the one with the highest performance was palmitic acid because it was able to give the biodiesel with the highest mole fraction of 0.8358.

Keywords: Biodiesel, Reactive Distillation, Aspen HYSYS, Oleic acid, Stearic Acid, Linoleic acid, Palmitic Acid

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1 INTRODUCTION

It has become necessary for governments, research communities, and private organizations around the world to look for alternative and renewable sources of energy due to the depletion of petroleum reserves, increase in energy demands, unpredictability of fossil oil production, and increased concerns of rising greenhouse gas emissions. To date, many alternatives have been researched into and demonstrated but only a few have proven to be practically feasible in terms of availability, economics, public and environmental safety, and simplicity of use. One such alternative, which has been discovered to have all the good characteristics necessary to be a good fuel, is biodiesel from vegetable oils, used at 100% or blended with diesel fuel for compression-ignition type engines. Several studies have shown that biodiesel is a better fuel than fossil-based diesel in terms of engine performance, emission reduction, lubricity, and environmental benefits (Peterson *et al.*, 1997; Canakci and VanGerpen, 2000, Singhetal, 2004).

Biodiesel, as an alternative fuel, has been currently receiving much attention due to the limited availability of conventional petroleum diesel and environmental concerns. It can be directly used to replace petroleum diesel without modifying diesel engines since their properties, e.g., specific gravity, cetane number, viscosity, cloud point, and flash point, are similar (Simasatitkul *et al.*, 2011). It is a promising alternative or extender to conventional petroleum based diesel fuel. It has a number of

advantages – it can be derived from a renewable domestic resource (vegetable oil), reduces carbon dioxide emissions by about 78%, when compared to diesel fuel on lifecycle basis, and is nontoxic and biodegradable. All these its benefits have made it a very good environmentally benign fuel (Wang *et al.*, 2004, Jaya and Ethirajulu, 2011).

Biodiesel is produced mostly from vegetable oils, which are edible oils, such as palm oil, sunflower, and soybean via transesetrification process using sodium hydroxide as a catalyst. However, the commercialization of biodiesel production from those vegetable oils still have drawback due to high cost of vegetable oil and the purification of the formed biodiesel product. Therefore, it is necessary to develop a process to produce biodiesel more efficiently and economically. One approach for this has been carried out by using Jatropha oil instead of edible oils as feedstocks. Jatropha oil contains 20% saturated fatty acids and 80% unsaturated ones. In the unsaturated fatty acid, oleic acid, is the most abundant (44.8%) followed by linoleic acid (34%), palmitic acid (12.8%) and stearic acid (7.3%) (Shah et al., 2004, Kusmiyati and Sugiharto, 2010). According to Kusmiyati and Sugiharto (2010), one kind of fatty acid derived from Jatropha oil, oleic acid, could be used as a raw material to produce biodiesel using esterification reaction involving alcohol such as methanol, etc. At present, the esterification of fatty acid and alcohol into fatty acid ester is usually conducted in a batch reactor (Omota et al., 2003). However, the production of biodiesel from esterification reaction in the conventional batch reactor has many problems because of its low conversion, heavy capital investments and high energy costs. So, this process has been found not to be economical (Gao et al., 2007). To resolve the problems, an advanced technology for biodiesel production has been developed. This method is known as a reactive distillation (RD) (Kusmiyati and Sugiharto, 2010).

Reactive distillation is a fascinating way for biodiesel production due to the fact that the process is able to reduce investments and energy costs (Lai *et al.*, 2006, Kusmiyati and Sugiharto, 2010). Generally, reactive distillation is a process that combines both separation and chemical reaction in a single unit. It is especially very attractive whenever conversion is limited by reaction equilibrium (Balasubramhanya and Doyle III, 2000; Giwa, 2012, Giwa, 2014). This is because it (reactive distillation) combines the benefits of equilibrium reaction with a traditional unit operation (in this case, distillation) to achieve a substantial progress in not only promoting the reaction conversion through constant recycling of unconverted materials and removal of products but also reducing capital and operating costs in one way by reducing the number of equipment units (Giwa and Karacan, 2012a, Giwa and Giwa, 2013b). Other advantages of reactive distillation include suppression of side reactions, and utilization of heat of reaction for mass transfer operation, especially if the reaction involved is an exothermic type (Giwa, 2013). It has also been reported that reactive distillation has scopes for difficult separations and avoidance of azeotropes (Jana and Adari, 2009; Giwa and Karacan, 2012b, Giwa and Giwa, 2013a, Giwa *et al.*, 2013).

The information obtained from the literature has revealed that Santander *et al.* (2010) employed response surface methodology and Aspen Plus to simulate the production of biodiesel using castor oil in a reactive distillation column in order to find the best conditions for producing large amount of fatty acid esters. Also, Simasatitkul *et al.* (2011) used Aspen HYSYS to investigate the production of biodiesel from the transesterification reaction between soybean oil and methanol in a reactive distillation column and analysed the effects of some operating and design parameters on biodiesel production. Furthermore, Samakpong *et al.* (2012) used Aspen PLUS to simulate a design of esterification of rubber seed oil and methanol catalyzed by sulphuric acid using reactive distillation column, and they were able to obtain the optimum values of the process operating parameters such as molar distillate rate and molar reflux ratio. Besides that, Orifici *et al.* (2013) used Aspen-HYSYS to simulate and optimize the process of biodiesel production from the transesterification reaction of crude palm oil with methanol. As can be observed from the different researches carried out so far on the production of biodiesel, no work has tried to compare the amount or the purity of the biodiesel obtained from the different fatty acids present in the raw materials used for the production.

Therefore, this work has been carried out to validate one of the findings of Kusmiyati and Sugiharto (2010) that oleic acid, one of the fatty acids present in jatropha oil, could be used as a raw material for

biodiesel production, and further apply some other fatty acids present in jatropha oil to biodiesel production in a reactive distillation column with the aid of Aspen HYSYS. The performances of the different fatty acids considered were, as well, compared in this work.

2 METHODOLOGY

In this research work, Aspen HYSYS (Aspen, 2012) was employed to develop a model (shown in Figure 1) for the reactive distillation column used to carry out the esterification process that was used to produce biodiesel from oleic acid, as well as some other fatty acids and alcohol. In this work, methanol was the alcohol used, and the other fatty acids, apart from oleic acid, used were stearic acid, linoleic acid, and palmitic acid. The fatty acids considered were, actually, chosen based on the contents of jatropha oil given in the work of Shah *et al.* (2004) because it was stated in the work that the unsaturated fatty acid of jatropha oil contained those constituents.

The chemical equations for the production of biodiesel using the different fatty acids are as given in Equations (1) to (4) below.

For the reaction between oleic acid and methanol:

$$C_{18}H_{34}O_2 + CH_3OH \leftrightarrow C_{19}H_{36}O_2 + H_2O$$
(1)

For the reaction between stearic acid and methanol:

$$C_{18}H_{36}O_2 + CH_3OH \leftrightarrow C_{19}H_{38}O_2 + H_2O$$
⁽²⁾

For the reaction between linoleic acid and methanol:

$$C_{18}H_{32}O_2 + CH_3OH \leftrightarrow C_{19}H_{34}O_2 + H_2O$$
 (3)

For the reaction between palmitic acid and methanol:

$$C_{16}H_{32}O_2 + CH_3OH \leftrightarrow C_{17}H_{34}O_2 + H_2O \tag{4}$$



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Figure 1. Reactive distillation process column for biodiesel production

In developing the Aspen HYSYS model of the reactive distillation column, the fatty acids were fed into the column in liquid form at a temperature of 350 °C and a pressure of 5 atm while the alcohol (in this case, methanol) was fed in a vapour form at a temperature and a pressure of 150 °C and 1 atm, respectively. The feeds were fed that way so as to achieve the penetration of methanol vapour into each of the liquid fatty acids in order to ensure high reaction rate for the biodiesel production. The column used had, excluding the condenser and the reboiler, 30 trays, counted from the top, and each of the fatty acid feeds was passed into the column at the eighth (8th) tray whereas the tray for the methanol feed was the nineteenth (19th) one. The section covered by the 8th and the 19th tray was made the reaction section while the section on top of the reaction section was the rectification section and the one down it was the stripping section. The column also had a condenser and a reboiler. The condenser used was a total type and its pressure was set to 1 atm. Also, the pressure of the reboiler was set to the same atmospheric pressure (1 atm). Owing to the reactive distillation process involved, Sparse Continuation Solver was used as the algorithm for the simulation of the column. Furthermore, in each of the simulations carried out with each of the fatty acids, a reflux ratio of 3 and a reboiler duty of 250000 kJ/s were used.

After the simulations of the Aspen HYSYS biodiesel production model, which was carried using Wilson model as the property package, the performances of the fatty acids used were discussed and compared as given and discussed thus.

3 RESULTS AND DISCUSSIONS

As mentioned earlier, this work has been carried out to investigate the performances of some fatty acids present in jatropha oil for biodiesel production. The fatty acids considered were oleic acid, stearic acid, linoleic acid, and palmitic acid. Each of the fatty acids was tested for biodiesel production under the same conditions, for example, using the same molar flow rates of the feeds, and the results obtained from the biodiesel production when oleic, stearic, linoleic and palmitic acids were used are as shown in Figures 2-5, respectively, below.

As can be seen from Figure 2, which is showing the component mole fraction profiles obtained when oleic acid and methanol were reacted to produce methyl oleate (a biodiesel), in the reaction section of the column, oleic acid, one of the reactants, was found to have the highest mole fraction while the second reactant, methanol had the lowest mole fraction there. This is showing that, if the reaction were to proceed to completion, methanol was likely to get used up before oleic acid. In other words, methanol has been discovered to be the limiting reactant of the process, in this work. As such, it was discovered that, in order to obtain very high conversion, it is necessary to feed the process with excess methanol so that more oleic acid can be converted to produce more biodiesel. Considering what happened in the rectification section of the column, it was discovered that water, one of the products of the process, had the highest mole fraction there, while the mole fraction of methanol was very small there, those of methyl oleate and oleic acid were very negligible in that particular section of the column. However, in the condenser section of the column, methanol had the highest mole fraction followed by water.

As can be observed, methyl oleate and oleic acid, being heavy components, were found to be present in negligible amount in the top (rectification) section of the column while the light components (methanol and water) were present in relatively higher mole fractions there. This was actually expected like this for a normal distillation column working in a proper manner, and this was an indication that the developed model of the rectification section of the reactive distillation column is working, properly, in line with the normal principles of distillation. Although methanol is lighter than water, its mole fraction in the rectification section was found to be lower than that of water because it (methanol) is one of the reactants of the process and, therefore, much of it had been consumed in the reaction section before the remaining (unreacted) parts moved to the rectification section for separation. Furthermore, of course, in the condenser section of the column, the lightest component (methanol) of the process was the one that had the highest mole fraction. Also worth considering are the variations of the mole fraction profiles of the components at the stripping section of the developed reactive distillation column. According to the results obtained and shown in Figure 2, it was discovered that the mole fractions of all the components involved in this process were almost constant at the (stripping) section. Also discovered concerning this section was that oleic acid was the one with the highest mole fraction there, followed by the main product of the process, methyl oleate, and water was the component with the least mole fraction in this section of the column. The component mole fraction profiles had this same trend in the reboiler section of the column.



Figure 2. Component mole fraction profiles obtained from biodiesel production using oleic acid as the fatty acid

Shown in Figure 3 are the mole fraction profiles of the components involved in the production of biodiesel when stearic acid was reacted with methanol. According to the figure, at the rectification section, just like it was obtained in the case of the reaction between oleic acid and methanol, water had the highest mole fraction, but in this case, the mole fractions of all the other (three) components involved were found to be negligible there. In the reaction section, the component with the highest mole fraction was stearic acid (one of the reactants) while, in this case, the other reactant (methanol) almost completely disappeared. At the upper part of the reaction section, methyl stearate, the main product of the process, was actually negligible, but it started forming from the middle of the reaction section towards the stripping section of the column. At the end of the process, the component with the highest mole fraction at the stripping section, and, of course, at the reboiler section of the column was found to be the main product, methyl stearate.

Also considered was the performance of another fatty acid, linoleic acid, when it was reacted with methanol, in the production of biodiesel using a reactive distillation column, and the results obtained from this investigation are as given in Figure 4. The observations obtained in this case of the linoleic acid were found to be very similar to those of the reaction between stearic acid and methanol discussed above (Figure 3) except that the values of the mole fractions obtained were found to be different, but, as can be observed by comparing Figures 3 and 4, the trends of the profiles were



similar. For instance, in this case, also, water and methyl linoleate had the highest mole fractions at the rectification section and the stripping section, respectively.



Figure 3. Component mole fraction profiles obtained from biodiesel production using stearic acid as the fatty acid



Figure 4. Component mole fraction profiles obtained from biodiesel production using linoleic acid as the fatty acid



Figure 5. Component mole fraction profiles obtained from biodiesel production using palmitic acid as the fatty acid

Figure 5 shows the component mole fraction profiles obtained from the reaction between palmitic acid and methanol for the production of methyl palmeate (also, a biodiesel). As can be seen from the figure, the main product of the process (methyl palmeate) was found to have a very high mole fraction in the bottom section of the column. Also discovered from the mole fraction profiles obtained from the production of methyl palmeate was that, at the bottom section of the column, the mole fractions of the other three components, apart from that of methyl palmeate, were low and close to each other, and each of them was less than 0.1.

At the top (rectification) section of the column, methanol, having being discovered to be the limiting reactant of the process, was found to be negligible there. Other components found negligible at the top section of the column were methyl palmeate and palmitic acid. Water, being the lighter of the two products formed, was discovered to be present at the top section of the column in very high purity.

Now, in order to know how the conversions of the reactants occurred, the profiles of the fatty acids (oleic acid, stearic acid, linoleic acid and palmitic acid) and those of the alcohols (methanol) used for the production of biodiesel inside a reactive distillation column are compared and shown in Figures 6 and 7, respectively, for the fatty acids and the alcohol.

It was discovered from Figure 6 that the mole fractions of all the fatty acids used were negligible at the top section of the column. Their highest mole fractions were found to occur in the reaction section, as expected. Thereafter, it decreased down the reaction section and towards the stripping section of the column. At the stripping section, their mole fractions were approximately constant and very little changes occurred to this constant values at the reboiler section of the column. According to the results obtained and shown in Figure 6, linoleic acid was found to possess the lowest mole fraction in the bottom section of the column, followed by palmitic acid, and oleic acid had the highest mole fraction among the components coming out of the bottom section of the column. It can, therefore, be said that the least converted among the fatty acids considered was oleic acid while the one with the highest conversion was linoleic acid.



Figure 6. Mole fraction profiles of the fatty acids involved in the reactions



Figure 7. Mole fraction profiles of the alcohols used for the reactions

Given in Figure 7 are the mole fraction profiles of the methanol (alcohol) used with the different fatty acids to produce biodiesel. As can be seen from the figure, it was discovered that the methanol remaining from the production of methyl oleate had the highest mole fraction at the top section of the column while those (methanol) remaining from the production of the other ones (methyl stearate, methyl linoleate and methyl palmeate) were found to be negligible at the top section of the column.

Another thing discovered from the studies carried out was that the methanol remaining from the process and that was obtained at the bottom section of the column for each biodiesel produced was higher than that obtainable at the top section. Furthermore, the methanol remaining from the production of methyl stearate was found to have the highest mole fraction at the bottom section of the column. The mole fractions of the methanol from each of the other ones were very close to one another.

Also compared in this work was the mole fractions of the products (methyl oleate, methyl stearate, methyl linoleate, methyl palmeate and water) obtained from the different reactions occurring between fatty acids and methanol that were used. The results (profiles) obtained are as given in Figures 8 and 9.

Given in Figure 8 are the mole fraction profiles of the main products (methyl oleate, methyl stearate, methyl linoleate and methyl palmeate) obtained from the reactions between each of oleic acid, stearic acid, linoleic acid and palmitic acid and methanol. According to the results, the mole fractions of all the main products were found to be negligible at the top section of the column while, at the bottom section, methyl palmeate had the highest mole fraction. Following that (the mole fraction) of methyl palmeate was the mole fraction of methyl linoleate. The main product with the lowest mole fraction at the bottom section of the column was found to be methyl oleate. At this point, it has been discovered that the highest mole fraction of biodiesel was obtained from the reaction between palmitic acid and methanol. In other words, among the fatty acids investigated, palmitic acid was found to be the one that gave the highest mole fraction of biodiesel product.



Figure 8. Mole fraction profiles of biodiesel produced from the different fatty acids used

Shown in Figure 9 are the profiles of water products obtained from the processes (methyl oleate reactive distillation process, methyl stearate reactive distillation process, methyl linoleate reactive distillation process, and methyl palmeate reactive distillation process,) that were used for the production of biodiesel in a reactive distillation column. From the figure (Figure 9), it was discovered that the mole fraction of water given in the course of producing of methyl stearate, methyl linoleate and methyl palmeate were approximately the same and equal to 1 at the top section of the column, whereas that obtained from the production of methyl oleate was less than that (1). At the bottom

section, the water given by each of the processes was found to be different from one another, but the highest of them was less than 0.2 while the lowest, which is the water obtained from the production of methyl oleate, was discovered to be negligible. This has shown that, even though the behaviours of the mole fraction profiles of water obtained from each of the productions of methyl stearate, methyl linoleate and methyl palmeate were the same with one another at the top section of the column, they were not the same at the bottom section.



Figure 9. Mole fraction profiles of water given by the reaction for biodiesel production



Figure 10. Temperature profiles for the production of the different biodiesels investigated

IJSER © 2014 http://www.ijser.org Having considered all the profiles of the mole fractions of the various components involved in the processes for the production of biodiesel from the different fatty acids and methanol, and having discovered that there were differences in the behaviours of the different fatty acids with methanol, it is very necessary to consider the factor that might be responsible for these differences. As such, since it is known from the literature that distillation is greatly influenced by temperature, the temperature profiles of the processes used for the production of biodiesel from the different fatty acids were then obtained and plotted as shown in Figure 10.

The results given in Figure 10 has shown that the trends of the temperature profiles for the four fatty acids considered at the reboiler and the stripping sections of the column were the same, although there were differences in their values. At the reaction section, the trends of the temperature profiles of the production processes for methyl stearate and methyl linoleate were found to be alike while those of the other two were discovered to be different from them and each other. Also found out, at the top section of the column, was that the trends of the temperature profiles were the same except that in which oleic acid was used. According to the results obtained from the comparisons of the mole fractions of the similar components of the processes (Figures 6, 7, 8, and 9), it was discovered that the trends of the temperature profiles were similar at the bottom section of the column, a little bit different at the reaction and not the same at the top section of the column, just as it has been obtained in this case of the temperature profiles and the component mole fraction profiles of the processes. This is confirming the information available in the literature and, thereby, giving another evidence that the developed model of the reactive distillation column is a valid one.

4 CONCLUSIONS

The results obtained from this research study have revealed that one of the findings of Kusmiyati and Sugiharto (2010) has been validated because biodiesel has been produced from oleic acid, with the aid of Aspen HYSYS. In addition, the other fatty acids - stearic acid, linoleic acid and palmitic acid - present in jatropha oil have also been used for biodiesel production. It was found that the mole fractions of biodiesel obtained from oleic acid, stearic acid, linoleic acid and palmitic acid were 0.3572, 0.671, 0.8026 and 0.8358, respectively. Therefore, among the fatty acids considered, the one with the highest performance has been discovered to be palmitic acid because it was the one that produced the biodiesel that had the highest mole fraction among all.

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