Utilization of Waste Cooking Oil for Biodiesel Production Using Alumina Supported Base Catalyst

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***Abstract*---** Utilization of vegetable oil as feedstock of biodiesel may compete with food industry and increase of the production cost. Meanwhile, using homogeneous catalyst offer a huge amount of waste water to separate and clean the catalyst from the biodiesel. Therefore, for the environmental concern and to reduce the biodiesel production cost, in this work, biodiesel was prepared from waste cooking oil (WCO) which was transesterified using heterogeneous base catalyst. WCO was collected from fast food restaurant of California Fried Chicken (CFC). The experiments were conducted by three step process that consist of treatment of WCO, synthesis of catalyst, and transesterification process. The effect of treatments on FFA content of WCO was investigated in the treatment step. Meanwhile, the effect of reaction temperatures on yield of biodiesel was observed in the transesterification process. The results reveal that WCO very promising to be transesterified into biodiesel using heterogeneous alumina base catalyst.

***Keywords*---** Alumina Supported Based Catalyst, Biodiesel Production, Trasesterification, Waste Cooking Oil

I. INTRODUCTION

MORE than two centuries the world energy demand had depended on crude fossil oil [1] [2]. Due to its non-renewable nature, the use of fossil fuel continuously over many years causes the limitation of fossil oil reserve. Moreover, it has resulted to the rise in global temperature levels which also known as global warming, as well as increased others environmental problem [3]. Increased awareness to the rapid depletion of energy resources and environmental issues, has led researchers to develop an alternative energy. Biodiesel is one of the alternative energies has got tremendous attention over the last few years, due to its ability of reducing pollutants associated with the environment if compared to the conventional diesel derived from fossil oil [4] [5]. Generally, biodiesel is produced through transesterification reaction of vegetable oils or animal fats with short chain of alcohol. Methanol is more preferable, due to technical and economic consideration [2].

Nowadays, the use of vegetable oils as fuel feedstock needs to be considered seriously because they compete with food production [5] [6].Therefore, in the future it is impossible to rely on their existence for biodiesel production. Moreover, these oils would be more expensive to use as fuel, and it will be effect on biodiesel production cost [5]. It has been reported that production cost of biodiesel are 80-90% depend on cost of feedstock [6]-[8]

Currently, biodiesel was produced by conventional process, which is the reaction conducted with presence of homogeneous base catalyst usually hydroxide of sodium or potassium. However, this process has several limitations. The catalysts cannot be regenerated and reused. The recovery of catalyst is difficult. It needs complex and expensive step to separate the catalyst from the product, where the product needs to be neutralized with mineral acid such as Sulfuric acid, hydrochloric acid or phosphoric acid, and also should be washed with a lot of water. This will produce huge amount of waste water which leads to environmental problem as well as addition in the production cost [3] [9]. These problems can be overcome by using cheap raw materials, and an effective catalyst. Hence, the contribution of waste cooking oil will be promising to use as feedstock of biodiesel production due to availability and its low cost [6]. Meanwhile, the emergence of heterogeneous catalyst seems to be an effective solution to overcome the weaknesses of homogeneous catalyst [3] [6] [9].

Heterogeneous catalyst offers several advantages such as: it requires mild conditions, easy to be separated, it doesn't need washing and neutralization, it can be reused and regenerated [3] [10]. These advantages can reduce the production cost of biodiesel, so the price of biodiesel can compete with diesel oil. Various heterogeneous catalysts have been applied in transesterification process by previous researcher, such as zeolites [11] [12], hydrotalcites [13] and metal oxides [3] [6] [14]-[16]. Calcium oxide has been reported as a potential solid base catalyst in trasesterification process [3] [15] [16]. [c]Asri et al. [4] reported that a very good result of 95% yield of biodiesel at reaction temperature of 65ºC, catalyst amount of 6%, reaction time of 5 h, and molar ratio oil-methanol of 1:42 on transesterification of palm oil with refluxed methanol using double promoted catalyst $CaO/KI/\gamma\text{-}Al_2O_3$. That catalyst, have been also introduced on transesterification process of palm oil at sub and supercritical condition. The same result (95% yield of biodiesel) was obtained at reaction temperature of 290ºC, catalyst amount of 3%, and molar ratio of oil to methanol of 1:24 over treatment only 1 h [3]. However, both of work was conducted with refined palm oil as feed stock, resulting cost of biodiesel production is higher than that of diesel oil. Instead of that,

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waste cooking oil can be used effectively for biodiesel synthesis, due to its low cost and sustained of availability.

Therefore, in this work we are focusing on introducing double promoted catalyst CaO/KI/γ-Al_2O_3 on transesterification of WCO with refluxed methanol in batch type reactor. Several factors were investigated, such as the effect kind of treatment on FFA content of WCO and effect of reaction temperature on yield of biodiesel.

II. Literature Review

A. Biodiesel

Biodiesel is an alternative diesel fuel derived from vegetable oils or animal fats [2] [8]. Nowadays, biodiesel production has been drastically increased in each country over the world. This was triggered by the depletion of fossil fuel reserves and the increasing awareness of environmental issues because the frequent usage of fossil fuels [3].

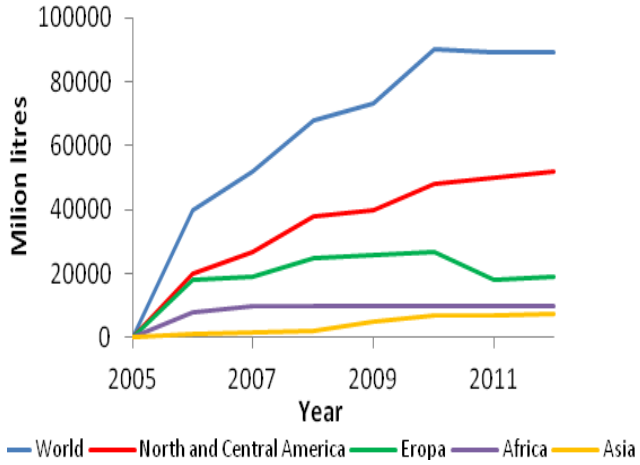

Fig. 1 Biodiesel production over the world in recent years.

Fig. 1 depicts biodiesel production over the world in recent years. It shown that world biodiesel production is linearly increases from 2005 to 2010. There is no significant increase on biodiesel production in 2011 to 2012. However, it has been predicted that in 2020, biodiesel production will be contributed by Brazil, China, India and South East Asia countries such as Indonesia and Malaysia as much as 20% to total world biodiesel production [8]. Meanwhile, the production of biodiesel from different feed stock is shown in Fig. 2. It is clear that the soybean oil is in the first position, while palm oil is in second position. However, in Indonesia, palm oil is the main feedstock for biodiesel production, because its availability is abundant. Nowadays, palm oil production of Indonesia is number one in the world.

Direct used of vegetable oils and animal fats as a fuel in compression engine was very limited, due to their high kinematic viscosity and low volatility nature [8] [17]. To reduce the viscosity can be done by converting vegetable oil into methyl esters by transesterification reaction.

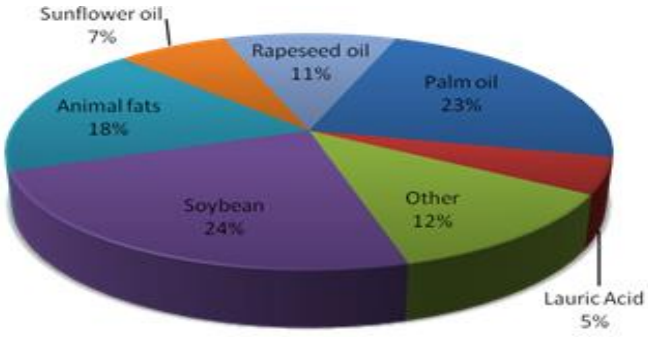

Fig. 2 Biodiesel production from different feedstock [17]

B. Waste Cooking Oil (WCO)

Currently, most biodiesel plant over the world especially in Indonesia are using refined vegetable oils as their main feedstock's, therefore cost of refined vegetable oil contributed as much as 80-90% on total cost of biodiesel production [6]. Compared to diesel oil, the high cost of diesel oil is the main obstacle to its commercialization. It cost approximately one and half time higher than that diesel oil [18]. In order to reduce the production cost of biodiesel, the manufacturer of biodiesel now focusing on using low cost feedstock. The most promising one is waste cooking oil (WCO). The WCO price is two to three times cheaper than vegetable oils [18]. The use of WCO as biodiesel feedstock will reduce the cost of waste product removal and treatment, as well as it can significantly decrease the amount of farmland which is necessary for producing of biodiesel crops [19].

The WCO has been categorized by the FFA content to two groups are the yellow grease (FFA content <15%) and the brown grease (FFA content >15%) [19]. The prices of these two types of WCO are ($ 0.04 to $ 0.09) and ($ 0.004 to $ 0.014) for yellow grease and brown grease, respectively [19] [20]. Most of vegetable oil is used for deep frying processes, because it improves the taste of the food. In the fraying process, oil is heated under atmospheric condition at temperature of 160-190ºC in relative long period of time [8]. In developing country, frying oil is used for several times due to economic reason. Some physical properties of oil will change after it continuously used for frying. The two of most important are an increase in viscosity and a change in colour [8]. Furthermore, during frying process, the presence of heat and water accelerate the hydrolysis of triglyceride, result the increase of FFA content in the oil. The FFA and water content of WCO must be taken into consideration, because they will give a negative effect to the transesterification process. Those negative effects include: separation of fatty acid methyl esters (FAME) from by-product (glycerol and excess of methanol), the oil viscosity significantly increases due to the formation of dimers and polymers, as well as the molecular mass and the iodine value decrease [18]. Meanwhile, partly of soap formed consumes the catalyst and reduces the yield of biodiesel. Until now, there is no a systematic method for managing waste cooking oils from household. It was simply discharged through the drains, sewers or direct dumped to the lands, which leads to environmental pollution. Moreover, more than 80% of WCO is produced in households. Therefore, it needs high cost

and huge amount of investment to manage its disposal [21] [22].

III. Materials And Methods

A. Materials

Waste cooking oil was collected from California Fried Chicken (CFC) in Surabaya, East Java. The properties of CFC's WCO have been analyzed, it consists of: 53.01% of palmitic acid and 46.99% of oleic acid. Meanwhile, FFA, density and water content of CFC's WFO were 2.82%, 0.92 g/cm^3 and 0.69%, respectively. Technical grade of Activated Carbon and bleaching earth for treating WFO were supplied by local supplier Bratacho-chem. Coconut coir, another cheap adsorbent was obtained from local coconut processing mill in Lamongan city, East Java Province. Analytical grade of γ--alumina was used as catalyst support, while KI and CaO as promotor were purchased from Merck, Germany. Meanwhile, NaOH, methanol, and acetic acid were provided by local supplier Bratacho-chem.

B. Pretreatment of Waste Cooking Oil

Waste frying oil that collected from fast food restaurant CFC (California Fried Chicken) was treated by adsorption, esterification, and washing process. Adsorption was done by several type of adsorbent such as technical grade activated carbon (AC), bleaching earth (BE), coconut coir powdered (CC) and mixture of activated carbon and bleaching earth, as well as a mixture of activated carbon and coconut coir powdered.

C. Preparation of Catalyst

Catalyst used in this work was γ-alumina supported catalyst CaO/KI/γ-Al_2O_3. Catalyst was synthesized by precipitation and impregnation method. First of all γ-alumina was precipitated on CaO with 30% of CaO loading (wt. % to alumina). As well as 35% of KI solution was impregnated to the mixture of CaO/γ-alumina with 30% of KI loading (wt. % to alumina). The formed slurry was drying overnight in an oven at 105-110ºC. Yellowish white crystal was milled into powder and then calcined in the muffle furnace at 650 ºC with flowed by air for 5 h. The prepared catalyst was characterized by means of three methods include X-ray diffraction (X-RD), Brenauwer-Emmet-Teller (BET) and Scanning Electron Microscopy (SEM). The procedure of preparation and characterization of CaO/KI/γ-Al_2O_3 have been described in detail at the previous work [3].

D. Transesterification Process

Trasesterification was carried out in 250 mL three neck glass batch type reactor, which equipped with reflux condenser, water batch, temperature controller and magnetic hot plate stirrer. Treated WCO and methanol with molar ratio oil to methanol of 1:15 was pouring into the flask. Subsequently, 6% of CaO/KI/γ-Al_2O_3 catalyst (wt. % to the WCO) was added to the mixture. The flask was heated in various temperature reaction of 35-65ºC in 10º of range and stirred at constant speed for 5 h. After the reaction, the mixture was centrifuged at 6000 rpm for 10 minutes to separate the product biodiesel from the excess methanol, by-product glycerol and solid catalyst. Three layers will be appeared which consist of the excess of methanol, the biodiesel and the solid catalyst CaO/KI/γ-Al_2O_3 and glycerol are in the upper, in the middle and the bottom layer, respectively. Eventually the liquid was put into a separating funnel let it overnight until the liquid separated into two phases. The excess of methanol is in the upper layer and biodiesel in the bottom. Biodiesel produced was analyzed by gas chromatograph GC-14B, Shimadzu. Yield of biodiesel determined using the result of GC analysis as according to the Equation 1.

$$Yield\ of\ biodiesel\ (\%) = \frac{w_b}{w_o}\ xC_{bs} \quad (1)$$

Where, w_b is the actual weight of biodiesel produced (mg), w_o is weight of oil used in the experiment (mg) and C_{bs} is concentration of biodiesel in the sample (wt. %).

IV. Results And Discussion

A. The Effect of Treatment Methods on FFA Content of CFC's WCO

FFA and moisture content are the two key parameters that determine the success of WCO to be transesterified into biodiesel using solid catalyst. Several Studies reported that solid acid catalyst is ideal for transesterification of WCO for FFA content more than 1% [18]. The advantages of using solid acid catalyst are insensitivity to the FFA content of the oil, esterification and transesterification could be occur simultaneously and lower catalyst requirement. However, this process was carried out at high temperature and pressure. Solid base catalysts are also very promising to convert of WCO into biodiesel, although its insensitivity was slightly lesser than that of acid catalyst process (FFA content <1%). However, the process is carried out at much lower condition compared to acid process.

In order to reduced the FFA content of CFC's WCO, several methods have been applied on pretreatment process include: Washing with hot water (WHW), adsorption process using (AC) of 2% + WHW, BE of 10%+ AC of 10%, CC of 5%+ WV), esterification (Est.) and a combination between AC of 10% with Est. Fig. 3 showed the effect of treatment methods on FFA content of CFC's WCO. It seen that the initial the FFA content of WCO (WTR= waste cooking oil without treatment) was 2.82%. Treatment with washing process using hot water (WHW) can reduced the FFA content into 2.38%. The FFA content reduced to be 1.52% using AC of 2%+WHW. There was no a significant effect on treatment of BE of 10%+AC of 10%. Meanwhile, with esterification process (Est.) and a combination of AC of 10% + esterification (Est.), the FFA content reduced into 1.32 and 1.08%, respectively. However, with 5% of coconut coir (CC) in vacuum condition, the FFA content drastically reduced into 0.81%. This is possible because the porosities of coconut coir are relatively higher than that of the activated carbon or bleaching earth, and it will lead the decreasing of FFA level of WCO. It was reported that coconut coir to be composed of cellulose, pentosan, furfural and lignin [23].

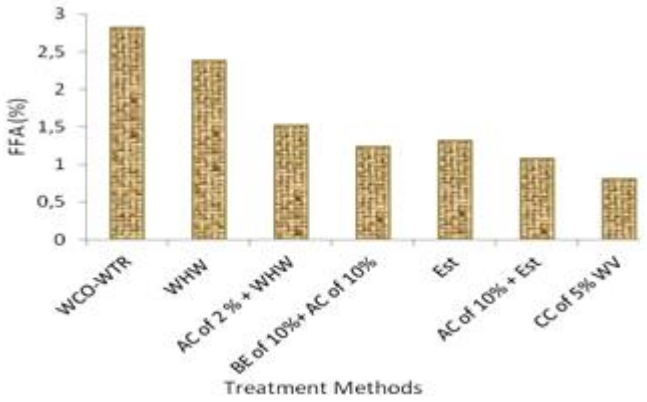

Fig. 3 Effect of treatment methods on FFA content of CFC's WCO: WTR = without treatment; WHW= washing with hot water; AC of 2%+WHW= treatment using 2% of activated carbon (wt. % to WCO) + washing with hot water; BE of 10% + AC of 10% = treatment using a mixture of bleaching earth of 10% (wt.% to WCO) and Activated carbon of 10% (wt.% to WCO); Est. = treatment using esterification process; AC of 10% + Est.= treatment using 10% of activated carbon + esterification process; CC of 5% wv = treatment with 5% of coconut coir with vacuum condition.

In this work has found that adsorption process using 5% of coconut coir in vacuum condition was the optimum ones for treating WCO. Hence, treatment using 5% of coconut coir was selected for further studies.

B. The Effect of Temperature Ttransesterification on Yield of Biodiesel

To investigate the effect of reaction temperature on yield of biodiesel, the experiments were conducted at several temperature levels of 35-65ºC with range of 10º C. The others condition such as: reaction time, molar ratio of treated WCO to methanol and amount of prepared CaO/KI/γ- Al_2O_3 catalyst kept constant at 5 h, 1:15 and 6% (wt. % to WCO), respectively. Fig. 4 shows the effect of reaction temperature on yield of biodiesel. As expected, that the reaction temperature of transesterification influenced the yield of biodiesel significantly. This is in accordance with the Arrhenius law, that the reaction rate constant is strongly influenced by the reaction temperature. The higher of the reaction temperature, the higher the reaction rate constant. The yields of biodiesel gradually increase from 3.50, 16.34 and 26.33% at 35, 45 and 55ºC, respectively. At 35ºC the yield of biodiesel obtained was very low due to the reaction was heterogeneous and the mass transfer was low, causing the low of rate reaction. However, yield of biodiesel drastically increased into 83.08% at 65ºC. This might be possible due to the methanol was completely boiled (boiling point of methanol 64.9ºC), resulting in increase of the kinetic energy and eventually increasing the rate of reaction. The similar results also reported by the previous researchers [4] [24] [25]. In contrary at 75ºC yield of biodiesel was decreased into 75.20% due to the evaporation of methanol.

Therefore, it can be stated that the optimum of reaction temperature was achieved at 65ºC with high yield of 83.08%. Meanwhile, the others conditions were fixed at 5h, 1:15 and 6% for reaction time, molar ratio WCO to methanol and amount of catalyst, respectively.

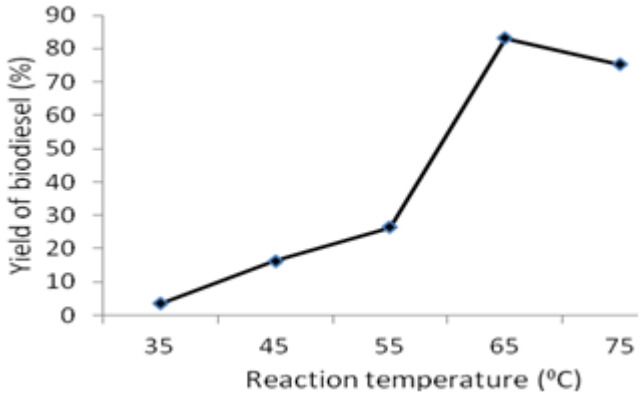

Fig. 4 Effect of reaction temperature on yield of biodiesel from CFC's WCO

V. CONCLUSION

Waste cooking oil used in this study was collected from California Fried Chicken restaurant in Surabaya. In order to reduced the FFA content of CFC's WCO, a series of treatment methods have been used include washing process, adsorption process and esterification or combined both of them. The result showed that the adsorption using 5% of coconut coir with vacuum process has given the best result with the FFA content of CFC's WCO reduced from 2.83 into 0.81%. The CFF's WCO was very promising to be transesterified using alumina supported base catalyst CaO/KI/γ-Al_2O_3. The optimum of transesterification reaction temperature was achieved at 65ºC with biodiesel yield of 83.08%. Therefore, instead of using refined vegetable oil, waste cooking oil can be used as raw material for biodiesel production.

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