Biodiesel production over potassium carbon-supported solid-based catalyst

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Abstract. Catalytic biodiesel production from vegetable oil and methanol using K₂O/C has been studied. The aim of this research was to investigate the performance of a K₂O/C catalyst for biodiesel production at varied of K₂CO₃ loading. The activated carbon-based from rice husk were fabricated via pyrolysis route. The catalyst were formed by depositing active phase K₂CO₃(10%; 20%; 30%; 40%; 50%) on the carbon surface for comparative purposes. To better understand of the impregnation effects, particle size, surface topography, and atom composition, the catalysts were analyzed by Scanning Electron Microscopy (SEM) and Energy Dispersive X-Ray (EDX). Biodiesel was produced via transesterification reaction. The reaction was carried out during 90 minutes at 65°C using 4wt% of the catalyst and methanol to oil molar ratio 10:1. The highest yield of biodiesel was reached of 96.62% at K₂O loading of 50% on carbon-based rice husk. The use of K₂O/C as catalysts enhance the purity of the product (carbon-based effect). The characterization of biodiesel, such as viscosity, density, and acid number were evaluated to confirm the purpose of catalyst function. The biodiesel characterization showed in the range with SNI standard.

1. Introduction
Nowadays, the development of renewable energy has gained importance since traditional fuel energy resource is decreased and terminated in finally. The high rate of traditional fuel energy consumption due to the rapid growth of the human population leads the pollution to the environment (greenhouse gas emissions) [1-4]. In this respect, to alleviate this problem, biomass energy has the advantages and potentials to substitute the traditional fuel energy cause it was biodegradable, cleaner, non-toxic, and environmentally-friendly [5-7]. Biodiesel is one of the renewable and green energies that has been chosen as a promising fuel (low emission impact). Biodiesel is commonly produced via esterification or the transesterification reaction using vegetable oils with methanol in the presence of homogeneous base catalysts [8-11]. Although it exhibited high catalytic activity, the homogeneous catalysts can’t be reused or recycled after the reaction [12-18]. Fortunately, it was inspired to develop the heterogeneous catalysts, which can be reusable and easy to separate after the biodiesel production.

Rice-husk is the major of agricultural waste which can be used and the potential to be heterogeneous catalysts. But, it has to concern in terms of the extraction of silica (SiO₂) component, which can act as sources of catalysts [19-20]. Few investigations have been made by rice-husk (ash) as a catalyst for...
biodiesel making. But, the use of rice-husk carbon (activated carbon) and the influence of impregnation loading for biodiesel production (using RH-carbon) rarely been reported.

So, this study aims to evaluate the catalytic activity in the transesterification of vegetable oil and methanol into biodiesel production under the different impregnation loading of K₂O to the RH-carbon (rice-husk) catalyst.

2. Experimental methods

2.1. Catalyst preparation
Rice-husk (RH) was collected from a rice mill in Aceh Besar, Indonesia. Vegetable oil (purchased on the market), methanol, and K₂CO₃ 99.9% (Merck, Germany) were used as received. Firstly, the rice-husk was washed with deionized water and then dried in the oven at 80°C for 24 hours. After that, a sample of rice-husk was converted into RH-carbon by pyrolysis process (at 450°C for 30 minutes) and followed with physical activation at 650°C for 1.5 hours [21]. The next process is impregnated the K₂CO₃ solution in different concentration (10%; 20%; 30%; 40%; 50%) into carbon. It was continued by stirring the mixtures for 6 hours, then placed in an oven for 12 hours. The prepared catalysts were stored and kept in the desiccator.

2.2. Catalyst characterization
The prepared catalysts were characterized using Scanning Electron Microscopy (SEM) and Energy Dispersive X-Ray Spectroscopy (EDX) to analyze the morphology and elements present near the surface of selected microscopic of catalysts.

2.3. Transesterification reaction
Biodiesel was synthesized by the transesterification reaction of vegetable oil and methanol using potassium carbon-based catalysts K₂O/C. The reaction was carried out by adding 30 grams of vegetable oil and 14.2 mL of methanol (molar ratio 10:1) using 4wt% of carbon-based catalysts in a batch reactor for 90 minutes at 65°C. After the complete end of the reaction, the generated biodiesel, glycerol, and catalysts were separated using funnel and filter paper to remove the catalysts and glycerol out of biodiesel, then dry-washed it (biodiesel) in an oven at 80°C for 12 hr. The yield of biodiesel was calculated according to Eq (1).

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\text{Biodiesel Yield (\%) = \frac{\text{Weight of biodiesel}}{\text{weight of used oil}} \times 100}\%
\]  

3. Results and discussions

3.1. Characterization of activated carbon-based (K₂O/C) catalyst

3.1.1. Scanning Electron Microscopy (SEM) and Energy Dispersive X-Ray spectroscopy (EDX). To investigate the impression of potassium (% K) in the impregnation. It can be seen using SEM-EDX analysis.
Figure 1. Scanning Electron Microscopy micrographs of the potassium-impregnated activated carbon.

Figure 2. Scanning Electron Microscopy micrographs of the potassium-impregnated activated carbon.

The surface structure and composition of catalysts were analyzed by SEM-EDX. Figure 1 and Figure 2 present information about the morphology and the elemental composition, respectively. As shown in Figure 1, it confirmed the amorphous carbon structure. The surface area of particles ranging from 595.8 – 1.650 nm which distributed in the catalysts. By comparing the images (Figure 1 and 2), it was observed that the coating on the catalysts due to the doping of K$_2$O. It was confirmed by the existence of C (22.10%), O (43.45%), Na (0.56%), Si (2.23%) and K (31.66%) on the catalysts.

3.2. Biodiesel production using activated carbon-based (K$_2$O/C) catalyst

3.2.1. Yield. The catalyst performance was investigated by the amount of biodiesel (yield). In this study, the yield of biodiesel was studied with the various concentration of impregnated K2O. The effect of impregnation loading can be seen in Figure 3.
As shown in Figure 3, the yield of biodiesel were found to be 84.96%; 86.42%; 90.53%; 93.02% and 96.62% respectively. The yield of biodiesel increased as the impregnation loading (K$_2$O concentration) increased from 10% to 50%. It is caused by the additional of K$_2$O will increase the contact of the catalysts to the reactant, and directly influencing the speed of reaction and the conversion [22-24].

3.2.2. Density and viscosity of biodiesel. Density and viscosity are other important parameters to measure the fuel quality cause of its influence on the combustion process and atomization in the engine. The density and viscosity measurement of biodiesel were carried out using pycnometer and viscometer canon-Fenske, respectively. The results of density and viscosity of biodiesel from transesterification in the varied K2O loading on the RH-carbon catalyst can be seen at Table 1.

| Impregnation Loading (%) | Density (gram/mL) | Viscosity (cSt) |
|--------------------------|------------------|----------------|
| 10                       | 862.54           | 4.47           |
| 20                       | 862.00           | 4.54           |
| 30                       | 861.72           | 4.54           |
| 40                       | 858.44           | 4.54           |
| 50                       | 859.97           | 4.54           |

As seen in Table 1, the overall result of biodiesel properties (density and viscosity) are in range according to the biodiesel SNI 7182:2015 (Density : 850 – 890 gr/mL ; Viscosity : 2.3 – 6.0 cSt).

4. Conclusions
A heterogeneous catalyst, K$_2$O/C, was successfully synthesized from rice-husk (carbon-based catalysts) through the impregnation method. The yield of biodiesel via transesterification over the potassium activated carbon (K$_2$O/C) catalysts reaches to 96.62% at impregnation loading of K$_2$O to C 50%. In conclusion, the fuel properties, according to biodiesel SNI 7182:2015 standards, indicate that the potassium rice-husk carbon catalyst can be used for biodiesel production (industrial scale) cause it has high catalytic performances.
Acknowledgments
The authors acknowledge the scholarship support from the Ministry of Education and Culture for the research funding under the Researcher Grant of Professor.

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