Optimization of Zeolite-X Catalysed Palm Oil Transesterification Using Response Surface Methodology

Diska Indah Alista[1], Kamisah Delilawati Pandiangan[2](✉), Khoirin Nisa[3], Wasinton Simanjuntak[2], Erika Noviana[4], and Selvia Anggraini Hasan[4]

[1] Graduate Program Student, Faculty of Mathematics and Natural Science, Lampung University, Bandar Lampung, Lampung 35145, Indonesia
[2] Department Chemistry, Faculty of Mathematics and Natural Science, Lampung University, Bandar Lampung, Lampung 35145, Indonesia
kamisah.delilawati@fmipa.unila.ac.id
[3] Department of Mathematics, Faculty of Mathematics and Natural Science, Lampung University, Bandar Lampung, Lampung 35145, Indonesia
[4] Undergraduate Program Student, Faculty of Mathematics and Natural Science, Lampung University, Bandar Lampung, Lampung 35145, Indonesia

Abstract. In this study, response surface methodology was applied to optimize the transesterification of palm oil using zeolite-X prepared from rice husk silica and aluminum foil as a catalyst. For this purpose, response surface methodology (RSM) with a 3-level-3 factor central composite design was applied to investigate the effect of three experimental factors on the percentage of conversion of the oil into methyl esters. A quadratic model was derived from the RSM with the aid of analysis of variance (ANOVA) and Design Expert 6.0.8 software to predict oil conversion and reveals that the mathematical model is Y = 53.38 + 14.4X1 + 15.2053 X2 + 2.3 X3, suggesting that the most influencing factor for transesterification of palm oil is the ratio of oil to methanol.

Keywords: palm oil · transesterification · zeolite-X · Response Surface Methodology (RSM)

1 Introduction

Renewable liquid fuels are fuels produced from non-fossil sources, including biomass derived fuels. One of the biomass-based liquid fuels that have been produced on an industrial scale is biodiesel (1). This renewable energy has been used by mixing with petrochemical diesel at a certain proportion. Along with the increasing role of biodiesel, production technology has been continuously developed and aimed to reduce production costs. In this development effort, two components that become the main focus are catalysts and raw materials (2).

Biodiesel is the result of a reaction between vegetable oil or animal fat with alcohol, which is also known as a transesterification reaction. The alcohol commonly used is

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methanol, and therefore biodiesel is a mixture of fatty acid methyl esters (FAME). Methanol and vegetable oils and animal fats are not reactive with each other, so the transesterification reaction requires a catalyst (3). Many types of catalyst now exist, one of them is zeolite, particularly synthetic zeolites such as zeolite-A and zeolite-X [4], and zeolite-Y [5].

Chemically, zeolites are aluminosilicate compounds and therefore for the manufacture of synthetic zeolites, amorphous silica and alumina sources are required which can be aluminum salts or aluminum metal. One source of amorphous silica that has been widely used is rice husk silica, and this silica has been used for the manufacture of several synthetic zeolites, including zeolite-A [6], zeolite-X [4], and ZSM-5 (7).

In this research, zeolite-X synthesized from rice husk silica and food grade aluminum foil was used as a catalyst for the transesterification of palm oil. In addition to the type of catalyst, the effectiveness of the transesterification reaction product is also influenced by reaction several other kinetic variables, including the ratio of alcohol to oil, catalyst load, temperature, and reaction time [8, 9]. The optimum conditions for conventional reactions are generally determined based on the influence of the single variable while the other variables are held constant [10], neglecting the possible effect of interaction between two variables or more. To overcome this drawback, the statistical method has been involved in developing experimental design which enables to prediction of the interaction effect of reaction variables.

One of the statistical methods that have been widely applied is known as Response Surface Methodology (RSM). This method has been applied to determine optimum conditions for transesterification of cooking oil [11, 12], Jatropha oil [13, 14], Jupati oil [4], rubber seed oil [15], palm oil [16, 17] and soybean oil [9]. Recognizing the advantage offered by RSM, in present study this statistical method was applied to optimize transesterification of palm oil using zeolite-X as a catalyst.

2 Research Method

2.1 Materials and Equipments

The materials used in this study included HNO_3 (68%, Brataco), NaOH (99%, Merck), rice husk, aluminum foil, and palm oil. The equipments used in this study include a lock n' lock container, hotplate stirrer Thermo SP88857105 Cimarec, magnetic stirrer, Jisico J-300S oven, Teflon magic com pan, thermometer, mortar and pastel, filter, 300 mesh sieve, furnace, autoclave, analytical balance, Scanning Electron Microscope (SEM) type ZEISS EVO MA 10, X-Ray Diffraction (XRD) PANalytical type XPert MPD diffractometer, X-Ray Fluorescence (XRF) PANalytical Epsilon 3, and Gas Chromatography-Mass Spectroscopy (GC-MS) type QP2010S SHIMADZU.

2.2 Zeolite-X Synthesize and Characterization

Sodium hydroxide (pa) was weighed to synthesize zeolite-X by the hydrothermal method, then 1.875 g food grade aluminum foil was weighed, and 10 g of rice husk silica was weighed. Next, the NaOH pellet was dissolved in distilled water and silica

Table 1. The Experimental design applied in this study

Run	Factor 1 A: catalyst load %	Factor 2 B: oil to methanol ratio	Factor 3 C: reaction time	Response Y Conversion %
1	A1	B1	C1	Y1
2	A2	B2	C2	Y2
…..	…	…	…	…
20	A20	B20	C20	Y20

was added, then stirred slowly. The mixture was heated with a hotplate stirrer at 70 °C for 3 h. After the silica has dissolved, the sodium silicate solution is filtered using filter paper to separate the solution from impurities. The sodium silicate solution was aged for 24 h [18]. Aluminum foil was added to the sodium silicate solution by spreading it evenly and stirring for 3 h using a stirrer to form a homogeneous gel. The next step is that the gel was poured into teflon and place in an autoclave then aged for 24 h. The sample was put in an oven and set the crystallization at 100 °C for 96 h. After the crystallization process is complete, the sample is filtered using filter paper slowly to separate the zeolite precursor formed from the remaining filtrate contained in the autoclave, and washed with distilled water to a pH 8. The sample was dried for 24 h in an oven at 100 °C. The zeolite precursor was calcined at 550 °C for 6 h to activate the zeolite active site. The next step is characterization of the samples with X-Ray Diffraction (XRD) and Scanning Electron Microscope (SEM) instruments.

2.3 Transesterification Experiments

A series of transesterification reactions were conducted following the experimental design as shown in Table 1.

After the reaction has taken place, the resulting product was cooled at room temperature and transferred to a separatory funnel, furthermore the product was left for 12 h to separate the mixture into two phases, the upper phase which is the biodiesel was collected. Excess alcohol was removed by evaporation and the volume of biodiesel was calculated for the percentage conversion calculation. The transesterification products were analyzed using GC-MS to identify the type of fatty acid ester.

3 Result and Discussion

3.1 Characterization of Zeolite

To ensure the formation of zeolite-X, the samples were characterized using XRD methods, and their diffraction patterns were compared with standard zeolite-X listed in the database of the International Zeolite Association (IZA), as shown in Fig. 1.

As can be seen in Fig. 1, the two chromatograms have very similar patterns, implying that the synthesized zeolite is zeolite-X as expected. For further confirmation, the sample

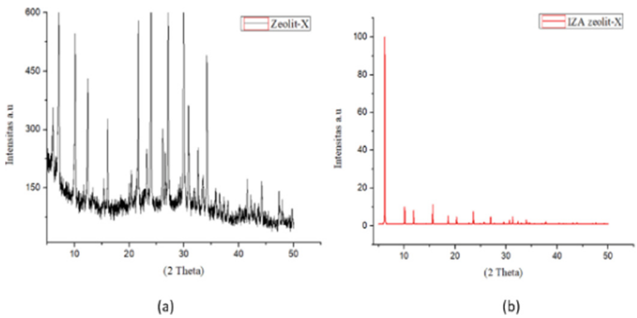

Fig. 1. Diffractogram of zeolite-X synthesized (a) and diffractogram of standard zeolite-X (b)

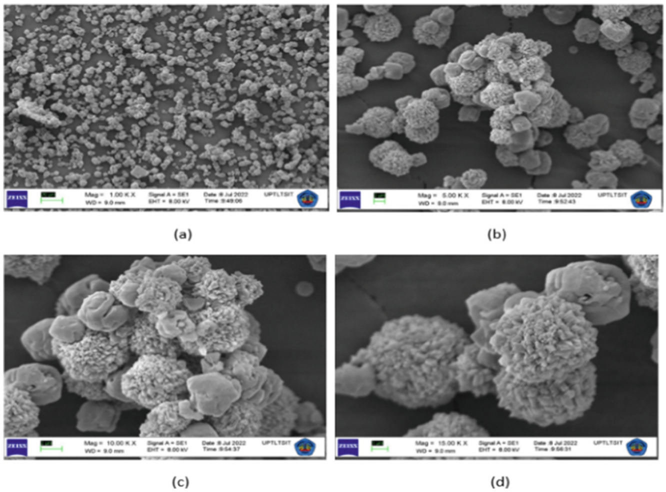

Fig. 2. Micrograph of zeolite-X synthesized with different magnifications: (a). 1000×, (b). 5000×, (c) 10,000× and (d). 15,000×

was also characterized using SEM technique, and the micrograph produced is shown in Fig. 2.

As can be seen, the surface morphology of the sample is characterized by the existence of a circular cluster of particles with the typical shape of faujasite, which is acknowledged as a typical shape of zeolite-X.

3.2 GC-MS Analysis

To justify the conversion of the palm oil into corresponding methyl esters, one of the samples was analysed using GC-MS. The GC-chromatogram of the sample is shown in Fig. 3, and the methyl ester compounds identified were listed in Table 2.

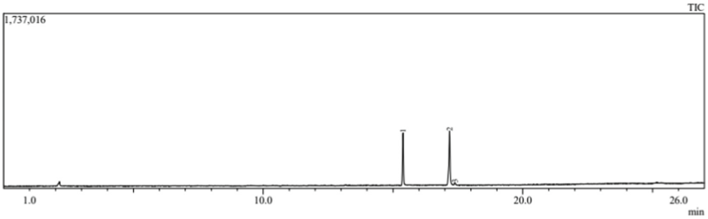

Fig. 3. Chromatogram of representative product of transesterification experiments

Table 2. List of compounds composing the product of transesterification

Peak No	Retention Time	% Relative	Component	Molecular formula
1	15.384	41.22	Methyl Palmitate	$C_{17}H_{34}O_2$
2	17.181	56.66	Methyl Oleate	$C_{19}H_{36}O_2$
3	17.381	2.12	Methyl Stearate	$C_{19}H_{38}O_2$

3.3 Mathematical Model of Transesterification

To obtain the mathematical model, the conversion data were analysed using central composite design, producing the graphs presented in Fig. 4.

Finally, from the RSM analysis, the following mathematical model is derived:

Y = 53.38 + 14.4X1 + 15.2053 X2 + 2.3 X3.

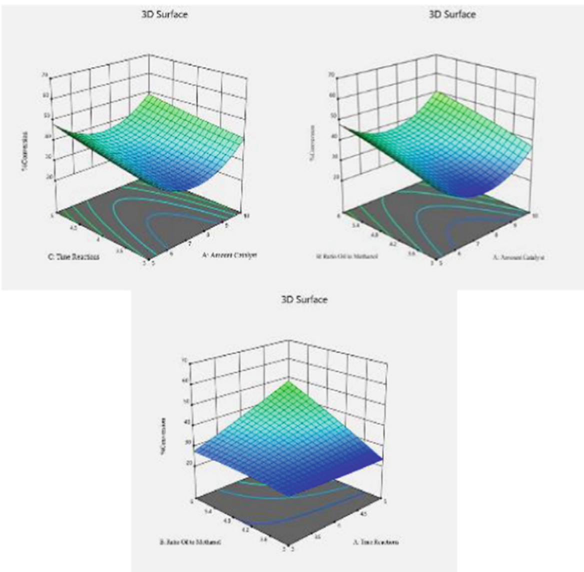

Fig. 4. Response surface area was drawn using the central composite design

According to the above equation, it can be seen that the most influencing factor for transesterification of palm oil is X2, which is the oil to methanol ratio.

4 Conclusion

The experimental results obtained indicate that zeolite-X was successfully produced from raw materials and the preparation method used. The zeolite was found to exhibit catalytic activity for the transesterification of palm oil to produce biodiesel. According to RSM results, the most influencing factor for the transesterification of palm oil is the ratio of oil to methanol.

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