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To cite this article: I Aziz *et al* 2019 *IOP Conf. Ser.: Earth Environ. Sci.* **299** 012065

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Catalytic cracking of *Jatropha curcas* oil using natural zeolite of Lampung as a catalyst

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Abstract. The scarcity of fossil fuel has led to the development of renewable energy sources, such as biofuel from plant oils. *Jatropha curcas* oil is one of the potential plant oil sources of biofuel. The conversion of *Jatropha* oil into biofuel can be achieved through catalytic cracking using the catalyst of Lampung natural zeolite. This study aimed to characterize the catalyst, to determine the optimum condition of the catalytic cracking process and to study the physicochemical characteristic of biofuel. The study was begun by activating Lampung natural zeolite, characterizing the catalyst, optimizing the catalytic cracking process and testing the biofuel. XRD result showed that the activated natural zeolite has a crystal structure, as suggested by the appearance of high-intensity peaks at 23.72°; 22.38° and 27.96°, which showed the generation of H-zeolite. Thermogravimetry analysis showed that H-zeolite started to decompose at 300 °C with 7.4% mass reduction and has thermal stability up until 700°C. The surface area of the zeolite was 53.2542 m²/g. The optimum condition for the cracking process was achieved at 375 °C for 2 hours and 5 % concentration of catalyst. The biofuel generated from this study consisted of 67.12 % gasoline (C₅-C₁₁), 11.87 % kerosene (C₁₂-C₁₅) and 21.01 % diesel (C₁₆-C₂₀). This biofuel has a density of 0.966 g/mL and pours point of 235°K.

1. Introduction

The increasing consumption rate of fossil-based fuel oils is not accompanied by the increasing rate of domestic fuel oils production, which can be contributed by the depletion of petroleum reserves. For this reason, alternative energy is needed to answer these challenges. Biofuel from *Jatropha curcas* oil could be one of the promising candidates of alternative energy sources. *Jatropha* oil can be produced at any time of the year and considered as a non-edible oil. Hence its use as a raw material for making biofuels does not compete with global food demands.

The production of biofuel from castor oil can be achieved by catalytic cracking method. This method only requires a catalyst to accelerate its reaction. Through this method, long chain unsaturated compounds in *Jatropha* oil are broken down into short-chain compounds such as gasoline, kerosene and diesel [1].

Studies on catalytic cracking of vegetable oils have been carried out by several researchers. Catalytic cracking use palm oil to produce biofuel using HZSM-5 alumina-silica [2]. Meanwhile, the cracking of Nyamplung oil using Zn-HZSM-5/ γ -alumina catalyst was carried out by [3]. In this study, we used



natural zeolite from Lampung due to its abundant availability. Before use, zeolite was activated and characterized. The objectives of this study are to characterize active zeolite, to find out the optimum condition for catalytic cracking and the composition of produced biofuel.

2. Materials and Methods

Jatropha curcas oil was obtained from Tokopedia. Natural zeolite was provided by PT. Winatama Lampung. AgNO₃ p.a and NH₄Cl p.a were purchased from Merck.

2.1. Preparation and Characterization of Catalyst

Natural zeolite was soaked in aquadest while stirred for three days at room temperature. It was filtered to get the precipitate, which was subsequently dried in an oven at a temperature of 120 °C for 2 hours. The precipitate was immersed in 1 M NH₄Cl solution for 24 hours at room temperature, filtered and washed with aquadest. The presence of chloride ion content (Cl⁻) was tested using a silver nitrate solution (AgNO₃). Chloride ion-free zeolite was then dried in an oven at a temperature of 120°C and calcined for 3 hours at a temperature of 450°C [4]. The resulting catalysts were characterized using X-Ray Diffraction Shimadzu type 7000. Diffraction patterns were measured at an angle of 2θ between 5-80° with CuKα as a radiation source and operating conditions at 40kV and 30mA.

Thermogravimetric analysis of catalyst was performed using LABSYS Evo TGA under nitrogen atmosphere with a flow rate of 40mL/minute and oxygen at a flow rate of 60mL/minutes. The temperature was raised 10°/minute for 100 minutes until it reached 900°C. Surface area analyzer was utilized to measure the surface area of the catalyst. N₂ and H₂ gasses were purged at 200 kPa and 138 kPa, respectively.

2.2. Optimization of Catalytic Cracking Process

A total of 50 ml of Jatropha oil and 2.5 grams of H-Zeolite were put into a catalytic cracking reactor. The reactor heater was run until the temperature reached 350°C. The process lasted for 1, 2 and 3 hours. The resulting gas was streamed to a condenser, which then turned into liquid and collected. The resulting product was calculated using the equation:

$$Yield = \frac{Biofuelmass}{Oilmass} \times 100\% \quad (1)$$

Catalytic cracking was also performed using different parameters: catalyst concentration (1, 5 and 7 %) and temperature (325, 350 and 375°C). The composition of the resulting biofuel was determined using GCMS Shimadzu. Operational conditions were as follows: injector temperature of 230°C, initial column temperature was 60° and raised at 10°C/minutes until it reached 250°C, and gas flow rate 1 mL/minute. Mass spectrometry used electron impact ionizer method with 70Ev energy. Other tests included density test (using ASTM D 1298 test method), titik tuang (using ASTM D 97 test method) and octan number (using ASTM D 2699 test method).

3. Results and Discussion

3.1. Characterization of Catalyst

XRD diffractogram from the catalyst (H-Zeolite) shows that the processed zeolite has a crystal structure, as marked by the presence of high-intensity peaks at 22.38°; 23.72°; 26.68°; 27.92°; and 30.09° (Figure 1). Based on JCPDS (Joint Committee on Powder Diffraction Standards) No. 71-1425 showed that the materials obtained were clinoptilolite with monoclinic crystal system, with hkl index at (131, 241, 222, 422, and 350) which 2θ region correspond to 22.28°; 23.68°; 26.01°; 28.12°; and 30.01°, respectively. Related to the previous work reported that the Lampung natural zeolite showed the main peaks at 22°, 28°, and 30° [5,6]. Moreover, [7,8] suggested that the region of 28° is a primary identifier of *clinoptilolite*. Hence, it can be inferred that the natural zeolite from Lampung is a *clinoptilolite* type.

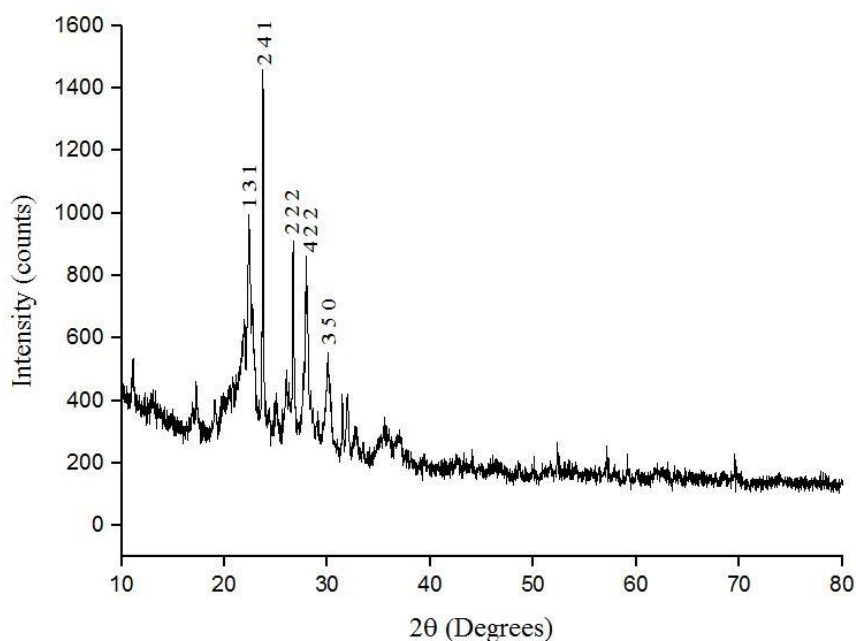


Figure 1. Diffractogram of H-zeolite catalyst.

Figure 2 exhibits the result of thermogravimetry (TG) and differential scanning calorimetry (DSC) analysis. It can be seen that there was a gradual decrease in weight until the temperature reached 700°C (TG analysis/green line). Generally, dehydration occurs at a temperature range of 30-500°C, while dehydroxylation takes place at temperatures above 500°C [9]. TG analysis suggested that there was a weight reduction of 7.40%.

The blue line of DSC result shows the existence of a sharp peak, namely the endothermic peak at a temperature range of 30-200°C, particularly at 131.75°C. This is due to a localized release of water inside the zeolite cavities and channels. Lampung natural zeolite was subjected to endothermic reaction at a temperature range of 30-630°C, where there is a fusion of phosphorus, potassium, and sulfur and phase transition [10]. Endothermic peaks also occurred at 700-750 °C, which can be caused by generally slow dehydroxylation of zeolite [9]. [11] also found that phase transition and endothermic release of Al and Ca occur above 658 °C.

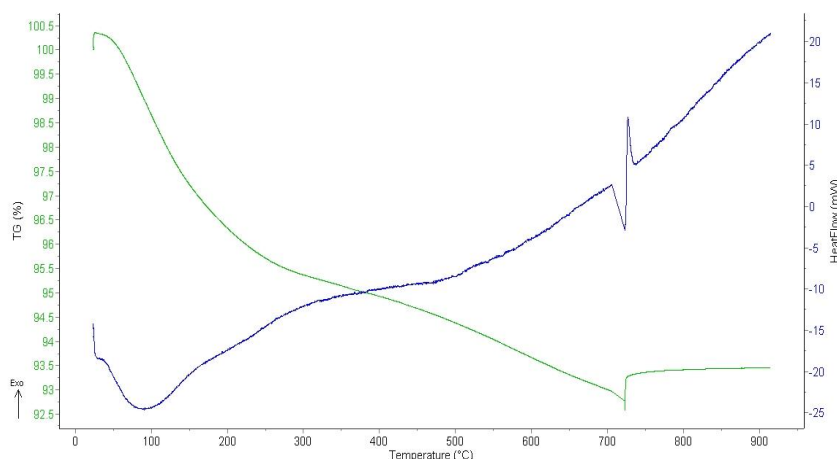


Figure 2. DSC and TGA results of H-zeolite catalyst.

The activation of natural zeolite with acid is beneficial to improve its performance. The large surface area allows reactants to enter the pores and increase the adsorbability of reactants onto the surface of the catalyst. These would lead to a faster reaction and more products. The surface area of H-Zeolite (activated zeolite) was 53.25 m²/g (Table 1). It increased by about 10% compared to the inactivated zeolite (48.61 m²/g). While pore volume, pore size and average particle size slightly increased. The growing in catalyst size was contributed by the acid soaking which dissolved impure oxide in the natural zeolite.

Table 1. Surface area analyzer results of activated and natural zeolite.

Sample	Surface area (m ² /g)	Pore volume (cm ³ /g)	Pore size (nm)
H-zeolit	53.25	9.59 x 10 ⁻³	20.88
Natural zeolit	48.61	9.95 x 10 ⁻³	20.92

3.2. The effect of reaction time on the catalytic cracking yield

Reactions were carried out for one, two and three hours. Three parameters were fixed: temperature (350°C), catalyst concentration (5%) and catalyst size (180 µm). The products consisted of gas, biofuel oil and water, whereas the remaining reactants were considered as residue. The yields of each product are displayed in Figure 3.

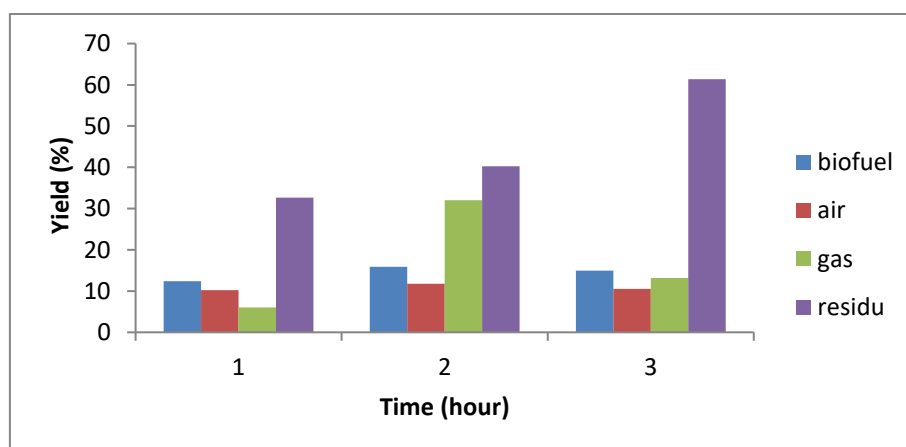


Figure 3. The effect of reaction time on the product yields.

The percentage of yield generated after one hour of the reaction was initially low (12.37%). It reached a peak after 2 hours of reaction (15.92%) before slightly declined after underwent a reaction for 3 hours (14.92%). Therefore, this study suggested that the optimum reaction time was 2 hours [11] stated that the percent yield of liquid product increase as the temperature and reaction time increase. However, it will decrease after achieving optimum reaction time. Longer cracking time could result in re-polymerization and condensation reaction [12].

Repolymerization is a reverse of cracking reaction which also involves carbonium ions, hence it can also accompany the cracking reaction. The result of this co-reaction is not only the decrease in biofuel yield but also in gas yield. More reactants turn into a heavier fraction (residue), which raises the amount of residue. Moreover, longer reaction time deactivates the catalyst, covering the active site of the catalyst [13]. These were proven by the increasing percent of residue from 40.29% (2 hours of reaction) to 61.37 % (3 hours of reaction).

3.3. The Effect of catalyst concentration on product yield

To study the effect of catalyst concentration on product yield, different concentrations of catalyst were applied (3, 5 and 7%). Reaction time was set at 350°C for 2 hours, using the catalyst size of 180 μm .

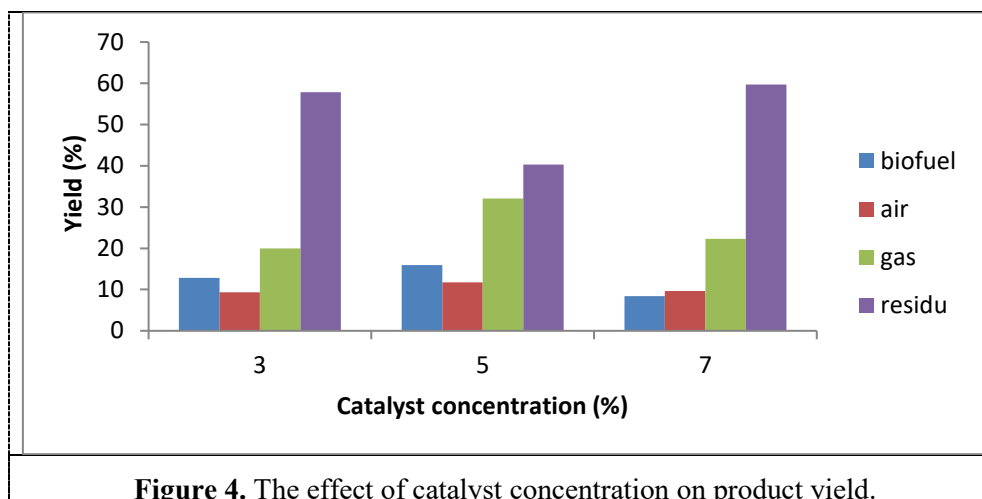
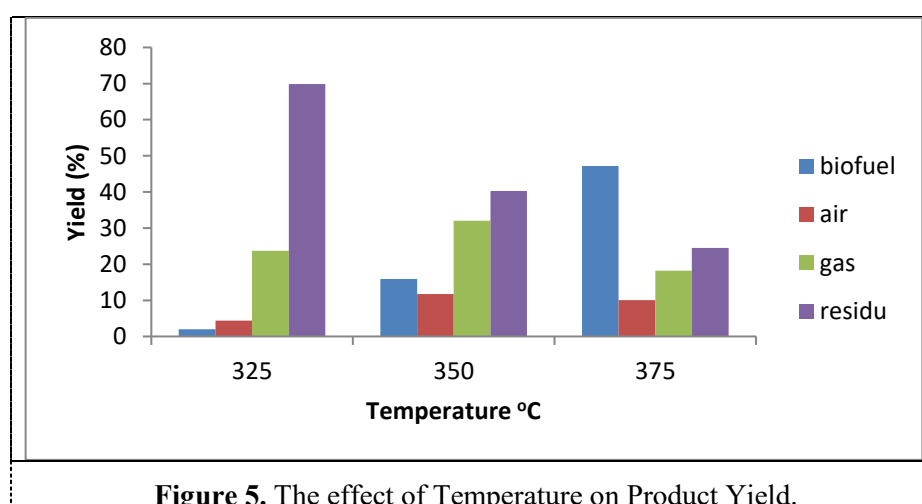


Figure 4 showed us that the addition of catalyst elevated biofuel yields. The highest yield was achieved at 5% catalyst concentration (15.92 %). However, it declined after the addition of 7% catalyst. This could be due to the overusing catalyst that will lead to excessive catalytic reaction and reduce the yield [13]. The decrease in product yield was also contributed by deactivation of the catalyst which is related to sintering (the loss of active surface due to crystal growth) and poisoning (the blocking of the active site due to strong chemisorption of impurities) [14].

3.4. The effect of temperature on product yield

To study the effect of temperature on product yield, three reactions were performed at a temperature of 325, 350, and 375°C. The reactions took place for 2 hours, using 5% catalyst concentration.



According to Figure 5, temperature influences biofuel yield. As the reaction temperature increased, the yield increased. The yield steadily increased among the three tested reactions.[11] stated that high temperature increases kinetic energy which means more collisions between molecules. However, the increasing yield leads to a decrease in the residue. These were supported by our research, where the

highest percentage of yield (47.20 %). was obtained at a temperature of 375 °C. Therefore, the optimum reaction temperature was 375 °C.

3.5. Physical and Chemical Characteristics of Biofuel

The chemical compositions of resulting biofuel were mainly C₆-C₁₈ (Figure 6), which can be categorized into gasoline (C₅-C₁₁; 34.52%), kerosene (C₁₂-C₁₅; 11.87%), diesel (C₁₆-C₂₀; 13.64%) and fatty acids. Therefore, gasoline was the largest fraction of the resulting biofuel.

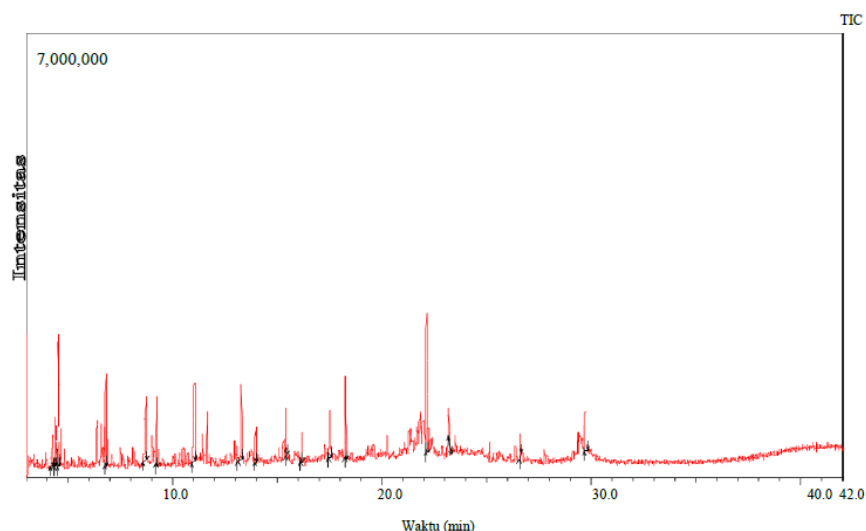


Figure 6. The GC-MS analysis of biofuel.

The density of the resulting biofuel was 0.96 g/mL, which is smaller than that of Jatropha oil (1.09 g/mL). The reduction of product density indicated that the catalytic cracking of Jatropha oil into shorter carbon chain compounds had been achieved successfully. However, the value did not meet the standard requirement for commercial gasoline, which falls within 0.7-0.8 g/mL [14]. The product density in this study was bigger than that of commercial gasoline due to the presence of other compounds besides gasoline, such as kerosene and diesel.

The observed pour point was 239 K, which came near to the pour point of gasoline (<273 K)[15]. Pour point is influenced by the length of the carbon chain; the longer the carbon chain, the higher the pour point. This pour point is an important factor because it shows the lowest temperature where oil samples can still flow.

The research octane number (RON) for the resulting product was 78.96, which is lower than the quality standard for gasoline(88). This is because there were still many heavy fractions contained in the product.

4. Conclusions

H-zeolite utilized in this study was of *clinoptilolite* category. Thermal analysis revealed that there was 7.40% weight reduction at 85-700°C. The surface area of the catalyst was 53.25 m²/g. The highest yield for the catalytic cracking of Jatropha oil was obtained at a temperature of 375 °C, catalyst concentration of 5%, catalyst size of 180 µm and reaction time of 2 hours. Chemical constituents of the biofuel were gasoline (C₅-C₁₁), kerosene (C₁₂-C₁₅) and diesel (C₁₆-C₂₀). The density, pour point and RON of the biofuel were 0.96 g/mL, 235 K and 78.96, respectively.

Acknowledgments

The authors would like to thank Universitas Islam Negeri Syarif Hidayatullah Jakarta for funding the research through Program Pembiayaan Penelitian, Kluster Penelitian Pengembangan Ilmu Pengetahuan (Research Fund Program, Research for The Development of Science Cluster) obtained from BOPTN DIPA APBN year 2017.

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