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Converting Catalytic Palm Oil (MEPO) to Produce Biogasoline Using Zeolite Faujasite Catalyst From Fly Ash with Nickel Impregnation (Ni)

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ABSTRACT - The process hydrocracking methyl ester of palm oil into fractions biogasoline by faujasite catalyst of fly ash impregnated with nickel have been made. Preparation for faujasite synthesis of fly ash can be done by removing organic compound and refluxing HCl. Synthesis do by melting the fly ash which has been prepared with NaOH 1: 1.2 and in aging for 8 hours and in the hydrothermal autoclave for 24 hours. The character faujasite using XRD and Si / Al ratio produces crystallinity main peak of 67% and Si / Al ratio of 1.65. Hydrocracking process using a variety of 4 catalyst used fly ash leaching results, faujasite, Ni-Faujasite 2%, and Ni-Faujasite 4%. Test the activity and selectivity of the catalyst to produce liquid product analyzed by GC-MS with the best catalyst was Ni-Faujasite 4% to yield 42.34% of the activity and selectivity of biogasoline fraction of 7.12%. The impregnation of the nickel catalyst is made by soaking in salt of nickel and then oxidation using O2 gas and reduction using H2 gas. The impregnation of nickel will affect the character of the catalyst so that the activity and selectivity of the catalyst is changed. The impregnation of nickel 4% on faujasite successfully done with nickel content of 3.71%, increasing Si / Al ratio of 2.27 and an acidity of 0.0035 mol/g.

Keywords: faujasite, impregnation, nickel, hydrocracking, biogasoline.

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INTRODUCTION

Human needs in the field of fuel oil are increasing. Efforts to find alternatives to obtain renewable fuel sources must be carried out. One of the renewable energy sources is the production of biofuels obtained from palm oil. The composition of palm oil consists of long hydrocarbon chains with oleic acid and palmitic acid content exceeding 80% which is a great potential for palm oil as a raw material (Farouq et al, 2009). Palm oil conversion process can be done in various ways. Yusman (2012) carried out catalytic cracking using methyl ester palm oil with a modified natural zeolite (modernite) catalyst impregnated with 3% nickel which was carried out with a weight ratio of 2:1 oil/catalyst for 30 minutes at 450 oC to produce a biogasoline fraction of 12.06 %. The hydrocracking process of palm oil methyl esters using a modified natural zeolite (modernite) catalyst impregnated with 2% nickel carried out with an oil/catalyst ratio of 2:1 for 30 minutes at a temperature of 450 oC has been carried out by Junaidi (2011) by producing a biogasoline fraction of 19 ,62%.

The hydrocracking process produces a larger biogasoline fraction, so this research uses the hydrocracking process. The hydrocracking process in this study uses a faujasite zeolite synthesized from fly ash. Natural zeolites (modernites) are usually found in sediments as a result of alteration of volcanic ash. The sedimentation process is also mixed with other minerals such as feldspar and quartz to form a non-uniform and irregular structure. Irregularity is what makes mineralogy researchers make zeolite with zeolite stability with other minerals (Las, 2004).

Synthetic zeolites can be made under hydrothermal conditions. Hydrothermal system is a closed system by using water as a solvent to produce water vapor pressure on the system (Rodhie, 2006). The zeolite that will be synthesized this time is the faujasite zeolite. Faujasite zeolite is a synthetic zeolite from alumina and silica sources in the environment of sodium hydroxide solution at 100 ° C (Tovina, 2009). This zeolite has a 3-dimensional axial structure with pores perpendicular to each other and is constructed from construction units 4-6 and 6-6 with a Si/Al ratio of faujasite of 1-3 (Rodhie, 2006).

Previous researchers have synthesized faujasite from fly ash. Sutarno (2004) used fly ash from PLTU Suralaya with a Si/Al ratio of 2.14 to produce faujasite which had a Si/Al ratio of 2.79 with the crystallinity of XRD results showing 70% (main peak) of the standard faujasite. Faujasite synthesis was also carried out by Somerset (2004) using fly ash from South Africa with a Si/Al ratio of 1.95 but zeolite A and sodalite were also found. The synthesis of faujasite zeolite in this study came from fly ash of the Paiton Probolinggo PLTU with the largest content of SiO2, which is 30.25% and Al2O3 which is 14.52% (PJB Paiton, 2002). This content has the potential to be used as a faujasite zeolite.

Hydrocracking in this study used a faujasite catalyst synthesized from fly ash impregnated with Ni metal to produce Ni-Faujasite. Nickel metal impregnated onto a zeolite carrier will increase the acidity and specific surface area of the catalyst if the Ni metal is evenly spread on the surface of the zeolite (Wulansari, 2004). Hayat (2007) explained that the distribution of Ni2+ metal is better than that of Co2+ which is based on a smaller Ni metal radius, thereby increasing its distribution inside and outside the pore cavity. Yusro (2012) used natural zeolite (modernite) impregnated with 3% nickel with a hydrocracking process at a temperature of 450 oC for 30 minutes in methyl palmitate to produce a biogasoline fraction of 12.01%. Sutarno (2007) impregnated Ni metal at a weight ratio of 4% of the faujasite catalyst synthesized from fly ash of the Suralaya PLTU with hydrocracking in petroleum distillate, the ratio of oil:catalyst weight was 2:1 with a temperature condition of 320 oC for 45 minutes producing a gasoline fraction of 8 %.

The use of a catalyst from fly ash that has been impregnated by Ni has the potential to be developed as an alternative to biogasoline production from palm oil using the hydrocracking method. This research uses a faujasite zeolite catalyst from fly ash PLTU Paiton Probolinggo. Faujasite zeolite catalyst from fly ash was then impregnated with active metal Ni. Sutarno (2007) revealed that changes in the catalytic properties of a metal can occur with variations in the composition of the carrier from the carrier preparation, therefore, in this study, variations in the levels of Ni metal were carried out on the catalyst. The hydrocracking results will be characterized using GC-MS.

METHODOLOGY

A. Fly ash preparation

50 grams of fly ash from PLTU Paiton-Probolinggo was soaked and stirred in water at 100oC for 2 hours to extract water-soluble organic matter.

B. Leaching Fly Ash

Reflux 10 g of fly ash (procedure A) and 100 mL of 5 M HCl solution. Reflux was carried out at 90 oC for 1 hour. The refluxed fly ash was then washed with distilled water until the pH of the washing filtrate was neutral and the solids were dried in an oven. Smelting was carried out on fly ash resulting from reflux of 5 M HCl with solid NaOH (weight ratio of NaOH: fly ash = 1,2) at a temperature of 550 oC for 1 hour.

C. Hydrothermal

The smelting result (Procedure B) was then dissolved in distilled water, stirred and allowed to ferment for 8 hours. The results of dissolving and aging are then reacted hydrothermally in an autoclave for 24 hours at a temperature of 100 oC. The results of the hydrothermal reaction were then washed with distilled water until neutral and in an oven at 100 oC (Somerset, 2004).

D. Ni Impregnation

The impregnation of Ni on faujasite (Procedure C) was carried out by the wet impregnation method. Ni(SO4).6H2O salt with variations of 2% and 4% was dissolved into 100 mL of distilled water while stirring until homogeneous. The result of the dissolution is added with 10 g of the extraction result then heated and evaporated at a temperature of 80 oC to 90 oC while stirring until the water evaporates. The result of evaporation is continued by oxidation process with oxygen gas 20 mL/minute at a temperature of 500 oC for 2 hours and reduction at a temperature of 500 oC with hydrogen gas 20 mL/minute for 2 hours (Handoko, 2001).

E. Faujasite Crystallinity

The obtained solid (Procedure C) was then characterized by X-ray diffraction method. X-ray diffraction measurements were carried out using an Xpert MPD X-ray diffractometer. The measurement conditions are, target = Cu, voltage 40 kV, current 30 mA, differgen slit = 0.25 degrees, and receiving slit = 12.7500. The crystallinity of the synthesized Faujasite was determined from the ratio of the height or intensity of the 8 main peaks of the synthesized Faujasite with standard Faujasite.

F. Determination of Total Acid

The total acidity of the catalyst can be determined gravimetrically on the basis of the adsorption of ammonia gas on the catalyst surface. The porcelain dish was then heated in an oven at a temperature of 120 oC for 2 hours, after which it was cooled and the mass was weighed. The acidity of the catalyst represents the number of moles of NH3 adsorbed to the catalyst per gram by weight. The measurement of zeolite acidity was carried out by the method of adsorption of ammonia base on the surface of the catalyst. Ammonia will be adsorbed on the surface of the Bronsted acid site and the Lewis acid site (Handoko et al, 2009).

G. MEPO Hydrocracking Process

Palm oil methyl ester (MEPO) as much as 10 grams was placed in the feed tank and 3 grams of catalyst from each catalyst variation. The heat of the reactor system is set at 450oC and H2 gas with a gas flow rate of 20 mL/min in Figure 1. The reaction product in the form of gas is passed into a condenser (coolant) until it turns into a liquid. Hydrocracking of palm oil methyl ester (MEPO) was carried out for 60 minutes. The reaction products from each catalyst variation were analyzed using GC-MS (Haliq, 2012).

H. Calculation of Activity and Selectivity

The cracking product of palm oil methyl ester was analyzed using GC-MS. The data from the GC-MS analysis was used to test the activity and selectivity of the catalyst. The sample was inserted into the GC-MS tool set with an FID detector. Column temperature at 60 oC. Flow rate 0.51 mL/min. The detector temperature is 280 oC. Injector temperature 310 oC. Analysis time 21 minutes. Injection Volume 0.20 mL. %Selectivity=Amount of % area of hydrocarbon compounds (Haliq, 2012)

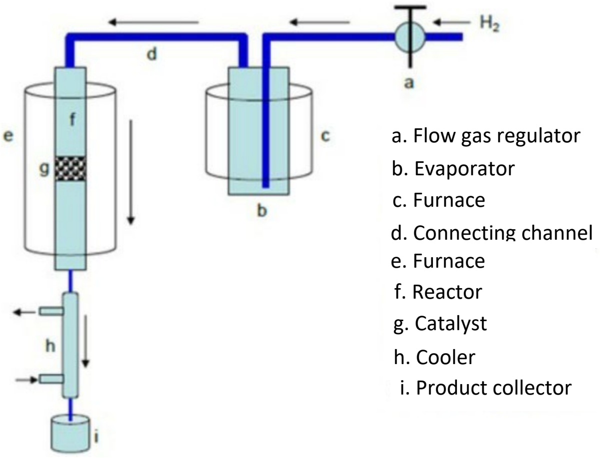


Figure 1
Fixed bed reactor (Handoko, 2009)

RESULTS AND DISCUSSION

A. Synthesis of Faujasite Zeolite From Fly Ash

- **Fly Ash Preparation Results**

Faujasite synthesis begins with the preparation of fly ash from the Paiton-Probolinggo PLTU. The result of the preparation has a grayish color due to the loss of impurities. The carbon in fly ash is separated based on differences in density. The nature of carbon with a large surface area and high absorption will absorb alkaline solutions so that it can reduce its concentration (Fansuri, 2015). Reducing the concentration of the alkaline solution will reduce the ability to dissolve Si and Al in the next faujasite synthesis process. The results of the fly ash immersion were then refluxed using 5M HCl solution for 1 hour. The result of the reflux process is that fly ash changes color from gray to brownish black. The reflux process ended by neutralizing the fly ash with distilled water to remove excess acid species. Reflux with HCl increases the ratio of Si/Al because the dissolving process of Al species is easier than Si species. Reflux can also remove metal impurities such as iron and calcium. Calcium ions in the hydrothermal solution system result in the deposition and stabilization of silicate species so that they do not form a Faujasite framework structure (Sutarno, 2004).

- **Hydrothermal**

Hydrothermal in the synthesis of faujasite is done by melting the prepared fly ash with NaOH. Hydrothermal is a heating process at a certain temperature with the balance of steam and water maintained. The presence of water here is needed because it is to determine the crystallinity (of the zeolite). The result of hydrothermal and aging is the formation of faujasite with a bright white color and solid form as shown in Figure 2.

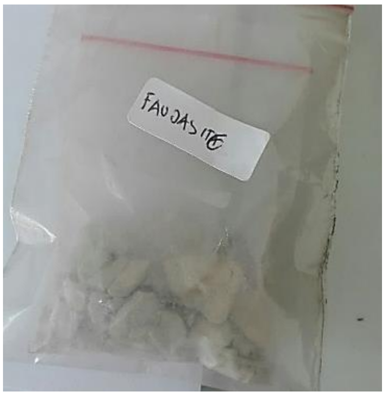


Figure 2
Faujasite crystal

The prepared fly ash was added with alkaline NaOH so that it was able to attract protons and resulted in the deposition of Al3+ cations to produce aluminate metal deposits. Alkali metal Na also accelerates the nucleation process because it reduces the effect of protonic acid sites (Bronsted acid) and Na is smaller in size so that it is easier to exchange (Ulfah, 2006). Sodium aluminate and sodium silicate are formed by the reaction:

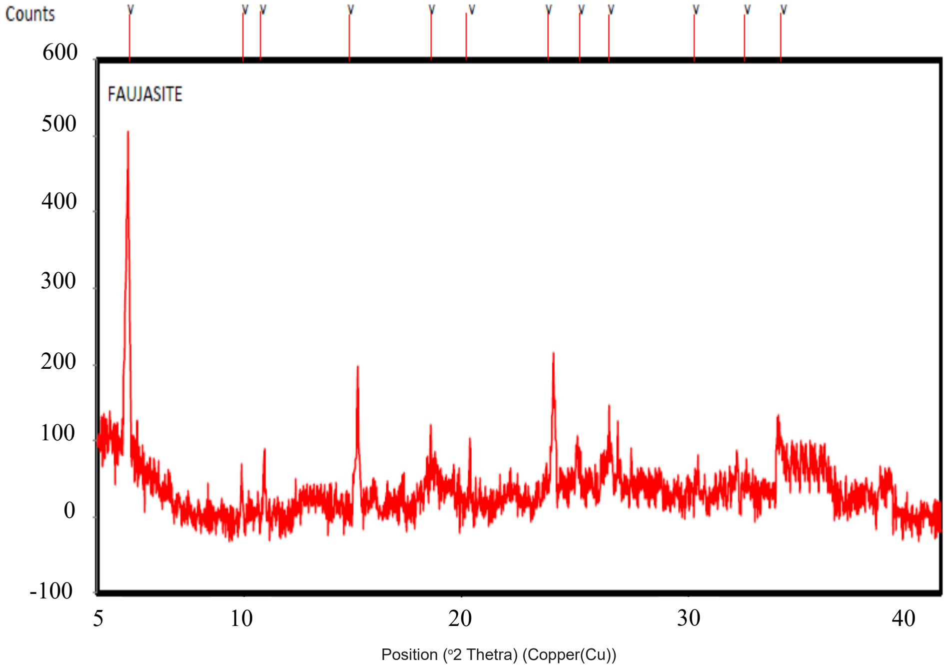
$Al_2O_3 + 2\,OH^- + 3\,H_2O \rightarrow 2\,[Al(OH)_4]^-$
$SiO_2 + 2\,OH^- \rightarrow Si(OH)_3^{2-} + H_2O$

B. Characteristics of Faujasite Zeolite

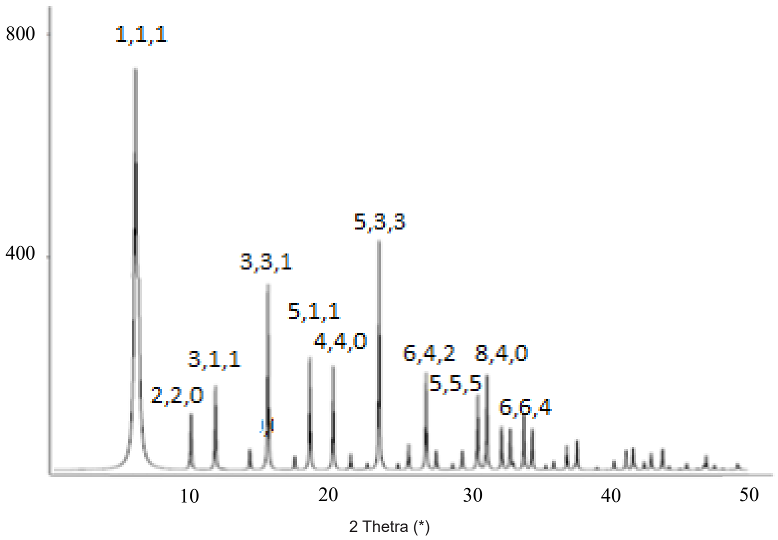
- **Characteristics of XRD Faujasite**

The resulting Faujasite was analyzed using XRD. Comparison of the 2 thetha angle from the synthetic faujasite from fly ash was matched with the standard faujasite from Nino (2013). The XRD results on the x axis indicate the 2 thetha angle while on the y axis it is the intensity. Figure 4.3 shows the XRD diffractogram of synthetic faujasite and standard faujasite from Nino (2013).

XRD results show a similar pattern between synthetic faujasite and standard faujasite (Nino, 2013) in other words, the synthesis of faujasite from Paiton PLTU fly ash this time has been successfully carried out. The characteristic peak at 2 thetha angle of the faujasite is 8 then matched with the synthesized faujasite listed in Table 1.



(a)



(b)

Figure 3
(a) Synthesis faujasite XRD diffractogram, (b) standard faujasite (Nino, 2013)

Table 1
Comparison of peak characteristics of 2 thetha

No.	2 Thetha (degrees) Standard Faujasite (Nino, 2013)	2 Thetha (degrees) Faujasite Synthesis
1.	6.19	6.2
2.	10.11	10.9
3.	15.61	15.8
4.	20.30	20.4
5.	23.58	23.9
6.	26.97	27.1
7.	30.66	30.2
8.	33.99	33.2

The main peak of the faujasite is at 2 thetha angle of 6.2°. The main peak crystallinity of synthetic faujasite was 67%. The appearance of another peak is Hydroxysodalite. The Si/Al ratio of leached fly ash is only 1.28 causing a very high amount of dissolved alumina causing the crystal growth rate to be fast so that zeolite structures that are easier to form are Hydroxysodalite (Sutarno, 2004). The time required for hydrothermal and aging also affects the crystallinity as done by Sutarno (2004) has a main peak crystallinity of 70%, but when compared to synthetic faujasite from Paiton PLTU fly ash this time it has a slightly lower crystallinity but the method used is quite efficient because shorter synthesis time.

- **Characteristics of Faujasite Si/Al Ratio**

Faujasite synthesized by Paiton fly ash was analyzed using AAS to determine the ratio of Si to Al. The results of the analysis showed that the Si/Al ratio of faujasite was 1.65. Faujasite generally has a Si/Al ratio between 1-3. The results of this synthesis have entered the range. Faujasite synthesized this time has similarities with the ratio of Si/Al from faujasite Y. Faujasite type Y is a zeolite with more silicon content than aluminum (Rodhie, 2006). Y type Faujasite has better thermal stability and crystallinity.

The synthesized Faujasite was then compared with Type X and Y type. Table 2 shows the crystallographic data of X and Y faujasite.

Table 2
Differences between faujasite X and faujasite Y
(Baerlocher, 2007)

Differentiator	Faujasite X	Faujasite Y
Unit Cell	Cube	Cube
Unit Cell Constant	25,009	24,2576
Framework	D6R	D6R
Si/Al . Ratio	1-1,5	1,5-3

Faujasite synthesized this time when viewed from the XRD data between type X and type Y, this time the faujasite has a tendency similar to that of type Y with a unit cell constant of 24,503. A review of the Si/Al ratio also shows that the faujasite synthesized this time is similar to type Y faujasite. From the data above, it can be concluded that the faujasite synthesized from fly ash this time is more similar to type Y faujasite zeolite.

• Hydrocracking Activity and Selectivity

Catalyst Activity

Catalyst activity is the ability of a catalyst to produce new compounds. The new compound is a conversion of 2 dominant products from MEPO, namely methyl palmitate and methyl oleate by producing a mixture of new compounds between types of paraffins, olefins, naphthenes, carboxylic acids, aromatics, methyl esters, aldehydes, and ketones. The activity value can be seen from the concentration of the number of new products produced after the hydrocracking process with each catalyst divided by the total concentration of MEPO before hydrocracking shown in Figure 4.

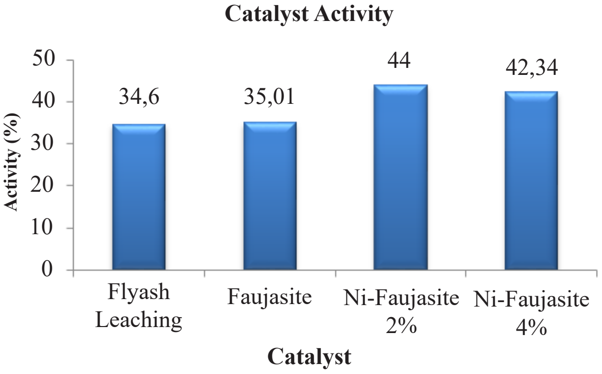


Figure 4
Catalyst activity

The activity of the catalyst increases with each catalyst used. Leached fly ash resulted in 34% activity. The ability of leached fly ash for hydrocracking is still not good because the acidity and Si/Al ratio of the leached fly ash catalyst is still low. Faujasite catalyst that has not been impregnated experienced a slight increase in activity from the leached fly ash catalyst. The acid site formed caused the hydrocracking activity of the faujasite catalyst to increase slightly to 35.01%. The slight increase was caused by the not yet active faujasite zeolite as a catalyst. Catalysts that have not been activated so that they do not have an active site to activate them are given acid treatment (HF, HCl, and NH4Cl) to open the pores on the surface of the zeolite (Satterfield, 1980). The impregnation process on the catalyst showed that the nickel content increased the activity of the catalyst. Faujasite catalyst impregnated with nickel as much as 2% had the largest activity of 44% and then decreased to 42.34 on catalyst impregnated with 4% Ni.

I. Catalyst Selectivity

Catalyst selectivity is the ability of a catalyst to produce certain products. The selectivity of the catalyst was grouped into 3 groups, namely C6-C12, namely the biogasoline fraction, C13-C18, the biodiesel fraction, and C19-C24, the biokerosene fraction. The percentage of selectivity can be determined by the total % area of the GC-MS results obtained and then presented in Table 3.

Table 3
Catalyst selectivity of various fractions

Fraction	*Fly ash* Leaching	Faujasite	Ni-Faujasite 2%	Ni-Faujasite 4%
C_6-C_{12}	0,27	1,23	3,68	7,12
C_{13}-C_{18}	7,57	6,84	11,0	2,25
C_{19}-C_{24}	0	1,7	1,04	0

The catalyst selectivity process for each catalyst was dominated by products with medium fraction liquid hydrocarbons (C13-C18). Hydrocracking using 4% Ni-Faujasite catalyst has the highest selectivity value in cracking producing short fractional liquid hydrocarbon compounds (C6-C12) which is 7.21%. The higher nickel content increases the selectivity of the catalyst due to the increasing number of Lewis acid sites. The hydrocracking results for the short fraction (C6-C12) consisted of a mixture of 1-hexene, 5 methyl hexene, methyl cyclopentene, benzene, 1-heptene, n-heptane, toluene, 1-octene, 2,4-dimethyl hexane, 1-nonene, 1-decena, 1-undecana, 1-dodecena.

Determination of the activity and selectivity of the catalyst for the formation of short fraction liquid hydrocarbons cannot be separated from the characteristics of the catalyst, such as acidity, Si/Al ratio and the amount of nickel metal impregnated on the catalyst from the results of research conducted by Yusman (2012). The results of the characterization of the 4% faujasite catalyst have higher acidity, Si/Al ratio, and impregnated nickel than other catalysts. This research contributes to the conversion process from palm oil methyl esters to short fraction liquid hydrocarbons with activity of 42.34% and selectivity of 7.12%.

- **The Effect of Nickel Impregnation Variations on Catalyst Selectivity and Activity**

The impregnation in this study used Ni metal. Impregnation was carried out with variations of 2% and 4% of the weight of the faujasite. Comparative data is shown from the percentage of nickel content of leached fly ash and synthetic faujasite. Leached fly ash has a nickel content of 0.09%. Fly ash from Paiton-Probolinggo does contain nickel but in low levels. Faujasite decreased nickel content to 0.083%. The existence of a hydrothermal process which is a process of forming faujasite crystals so that there may be a decrease in nickel content but not significantly in the formed faujasite crystals. The addition of 2% and 4% salt showed an increase in nickel content to 2.05% and 3.7147%, respectively. The relationship between the amount of impregnated nickel with the activity and selectivity of the catalyst has a different trend. The graph in Figure 5 shows that the total nickel content for the cracking process with optimum activity is 2% nickel impregnated catalyst. Ni-Faujasite 4% catalyst decreased the activity of the catalyst against the hydrocracking process.

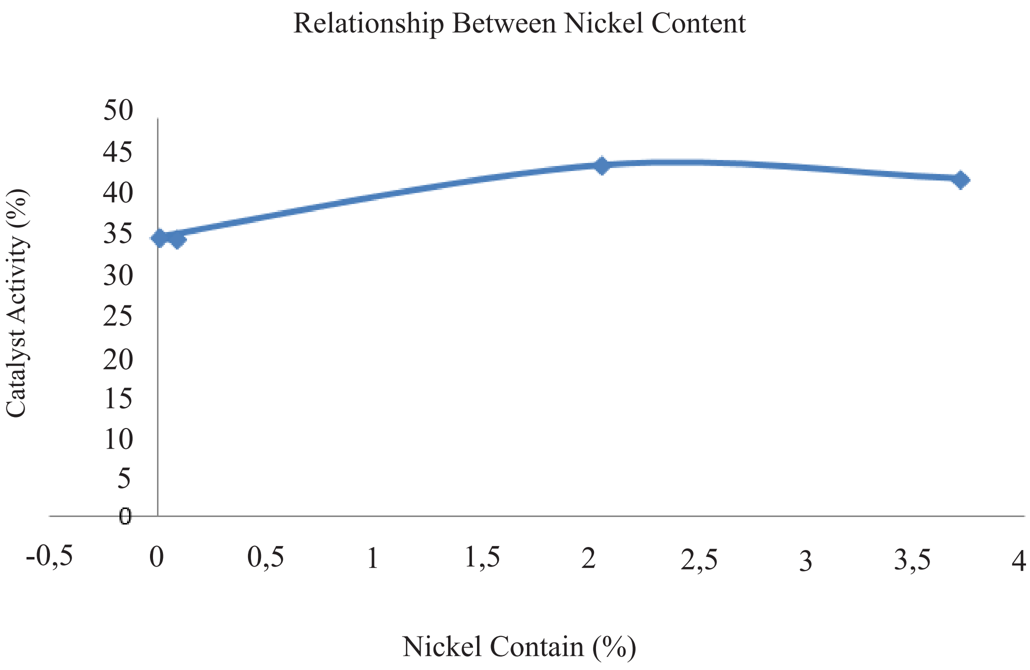


Figure 5
The relationship between nickel levels with catalyst activity

The increasing concentration of active metal impregnated onto the surface of the carrier may result in a decrease in the specific surface area of the catalyst. This phenomenon can be explained that the impregnated active metal is distributed unevenly or accumulates at the pore mouth (sintering) and closes the pore mouth (Figure 6). This situation occurs because the cohesive interaction between the active metal and the active metal is stronger than the adhesive interaction between the metal and the zeolite carrier. This situation should be analyzed using the BET method to provide information on the decrease in the specific surface area of the zeolite (catalyst) even though the impregnated metal showed an increase (Bartholhomew, 2006).

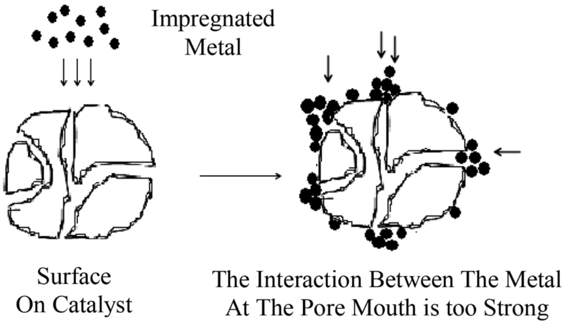


Figure 6
Accumulation of impregnated active metals in the pore mouth which causes a decrease in surface area (Bartholhomew, 2006).

The relationship between nickel content and catalyst selectivity can be seen in the graph in Figure 7 that the value of catalyst selectivity is increasing. The selectivity of using leaching and faujasite fly ash catalysts has a small value because nickel has not had a significant effect on nickel impregnated catalysts. The use of nickel during impregnation showed an increase in both the 2% Ni-Faujasite and 4% Ni-Faujasite catalysts. The highest nickel content was in the 4% Ni-Faujasite catalyst which produced the maximum biogasoline product (Figure 7).

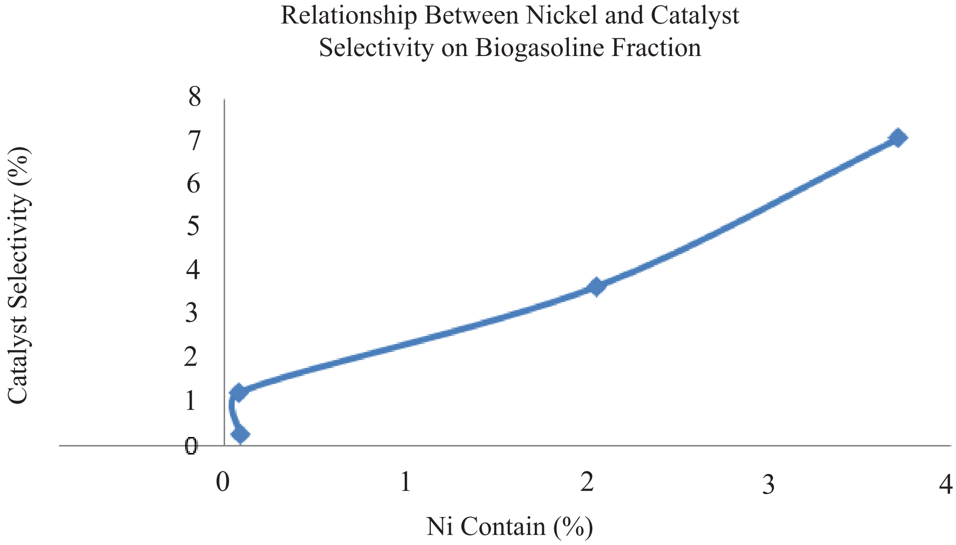


Figure 7
Correlation of nickel levels with catalyst selectivity to biogasoline fraction (C6-C12)

The nickel content is very influential on the selectivity of the catalyst to obtain short carbon fractions (C6-C12). Haliq (2011) has conducted research using nickel metal for the catalytic cracking process, so that alkane and alkene products have a fairly large concentration compared to other products produced. The development of Ni metal causes an increase in the number of products, especially in the form of paraffins and olefins. This study also showed that the highest use of nickel metal was in Ni-Faujasite 4% with a nickel content of 3.7147% resulting in a selectivity of the biogasoline fraction of 7.12% (Figure 7).

II. Relationship Between Si/Al Ratio With Catalyst Activity And Selectivity

The measurement results with AAS show that from various processes the trend is increasing. Leached fly ash has a low Si/Al ratio of 1.27. Faujasite has increased the ratio to 1.64. Faujasite has increased the ratio of Si/Al due to the hydrothermal process (Hamdan, 1992). This stage is a crystal growth stage, so it is possible that under these conditions there will be more silica reacting than alumina, causing the Si/Al ratio of the faujasite to increase. Faujasite impregnated with 2% and 4% nickel had Si/Al ratios of 2.080 and 2.272, respectively. Impregnation of the faujasite catalyst increases the Si/Al ratio. The increase in the Si/Al ratio is due to dealumination events during oxidation and reduction at high temperatures (500 oC) (Handoko, 2009).

The relationship between the Si/Al ratio with activity and selectivity has a different trend as well. Similarly, the trend of nickel content with catalyst activity with optimum activity on 2% Ni-Faujasite catalyst. The graph in Figure 8 shows the relationship between the Si/Al ratio and the catalyst activity.

The biggest catalyst activity using 2% Ni-Faujasite catalyst. Faujasite zeolite synthesized this time has similarities with type Y faujasite. Type Y faujasite has a higher Si/Al ratio than type X faujasite. The large Si/Al ratio makes this faujasite has smaller pores so that the structure is more stable for catalyst activity. . The characteristics of the catalyst in terms of nickel content also affect the effect of the Si/Al ratio with catalyst activity as previously discussed, that as much as 2% nickel content allows a more even distribution (Bartholomew, 2006) so that the optimum catalyst activity is 2% Ni-Faujasite catalyst. It is the same as the trend of the relationship between

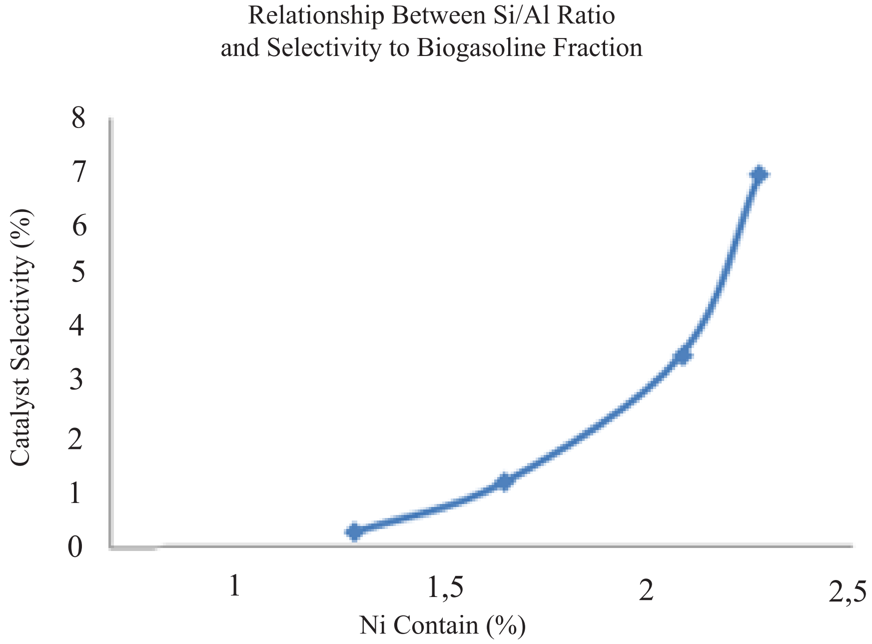


Figure 8
The relationship between Si/Al ratio and catalyst activity

nickel content and the selectivity of the catalyst with the trend of the relationship between the ratio of Si/Al to the selectivity of the catalyst. The graph of the relationship between the Si/Al ratio and the selectivity of the catalyst is shown in Figure 8.

The trend of the relationship between the Si/Al ratio on the selectivity of the catalyst shows that the higher the Si/Al ratio, the greater the selectivity of the catalyst. Catalysts with a high Si/Al ratio will have high thermal stability (Hamdan, 1992) and are effective in cracking non-polar compounds (Sutarti and Rahmawati, 1994). Catalysts containing more silicon then the affinity for non-polar molecules will increase because the Si-O (silicate) group is neutral. Neutral silicate zeolite will have minimal polarity so that it gives preference to the adsorption of non-polar reactants, namely non-polar compounds (Sutarti and Rahmawati, 1994). 4% Ni-Faujasite catalyst with a Si/Al ratio of 2.22 resulted in an optimum catalyst selectivity of 7.12%.

III. The Relationship Between Acidity With Catalyst Activity And Selectivity

The acidity of the leached fly ash showed the smallest value, which was 0.00012 mol/g. Fly ash from leaching has not yet formed a zeolite framework so that the Bronsted acid site has the smallest value for the acidity of this catalyst. The increase in acidity was seen in the faujasite of 0.000353 mol/g. The increase is caused by the hydrothermal process that forms the zeolite framework, the framework can cause the appearance of Bronsted acid sites. Nickel impregnation also increases the acidity of the catalyst. The more nickel contained in the catalyst, the greater the acidity value of the catalyst. The acidity of 2% and 4% nickel impregnation were 0.003071 mol/g and 0.003494 mol/g, respectively.

The increase in acidity of the nickel impregnated faujasite has a significant increase. This phenomenon occurs because the metal Ni distributed on the catalyst as a carrier matrix has d orbitals that are not fully filled so that it can be used by NH_3 molecules to form complexes with NH_3 as ligands. In this case, Ni metal acts as a Lewis acid site contributor and contributes to the increase in the acidity of the catalyst in the 2% Ni-Faujasite and 4% Ni-Faujasite catalysts. The more Ni metal is deposited, the more Lewis acid is formed and the more NH gas that can be bound by a nickel impregnated catalyst (Satterfield, 1980). Faujasite catalyst impregnated with nickel as much as 2% and 4% did not experience an increase in acidity so rapidly.

The more active metal impregnated onto the surface of the zeolite, the smaller the amount of impregnated Ni metal but relatively allows an increase in the specific surface area of the zeolite solid, although the increase in surface area is not too large. This phenomenon can be explained that the higher the amount of active metal impregnated on the catalyst, the competition between the active metals occurs so that nickel will block each other. Furthermore, accumulation occurs in one place even though it is not at the pore mouth, so that it is not evenly dispersed onto the surface of the zeolite. The possibility that occurs in this situation is a multilayer layer of impregnated active metal, so that the presence of active metal in the second layer and so on will be easily released again when thermal treatment (Bartholomew, 2006). This situation may cause the Lewis acid site of 2% Ni-Faujasite and 4% Ni-Faujasite not to be significantly different.

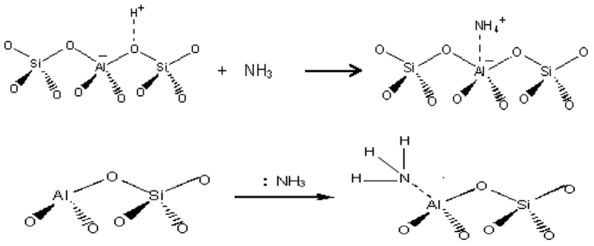


Figure 9
Chemisorption of ammonia on zeolite surfaces in determining catalyst acidity in bronsted acid
(a) Lewis Acid (b) (Satterfield, 1980).

The acidity of the catalyst is strongly influenced by the acid site of the catalyst. The ability of the catalyst is seen from the ability to absorb NH_3 base. Satterfield (1980) stated that the acidity of a catalyst is defined as the ability of the catalyst to adsorb ammonia bases due to the presence of both Bronsted acid sites and Lewis acid sites on the catalyst surface. The reaction that occurs is shown in Figure 9 by chemisorption. The more acid sites on the catalyst allow the more NH_3 to absorb, which means the acidity value of the catalyst is getting bigger. The increase in the acidity of the catalyst will be proportional to the amount of NH_3 gas bound to the catalyst. The relationship between acidity with activity and with catalyst selectivity is also different from other catalyst characteristics. The catalyst activity has the optimum condition on 2% Ni-Faujasite catalyst, while the selectivity of the catalyst has a selectivity for the biogasoline fraction using 4% Ni-Faujasite catalyst with the highest acidity of 0.0035 mol/g. The graph in Figure 10 shows the relationship between catalyst acidity and catalyst activity.

The trend between catalyst acidity and optimum activity indicates the use of 2% Ni-Faujasite catalyst. Ni-Faujasite 4% catalyst does have a higher acidity but its activity decreases. The higher the acidity value of the catalyst, the more acid sites available. The available acid sites are expected to make the catalyst more active in the cracking process. The

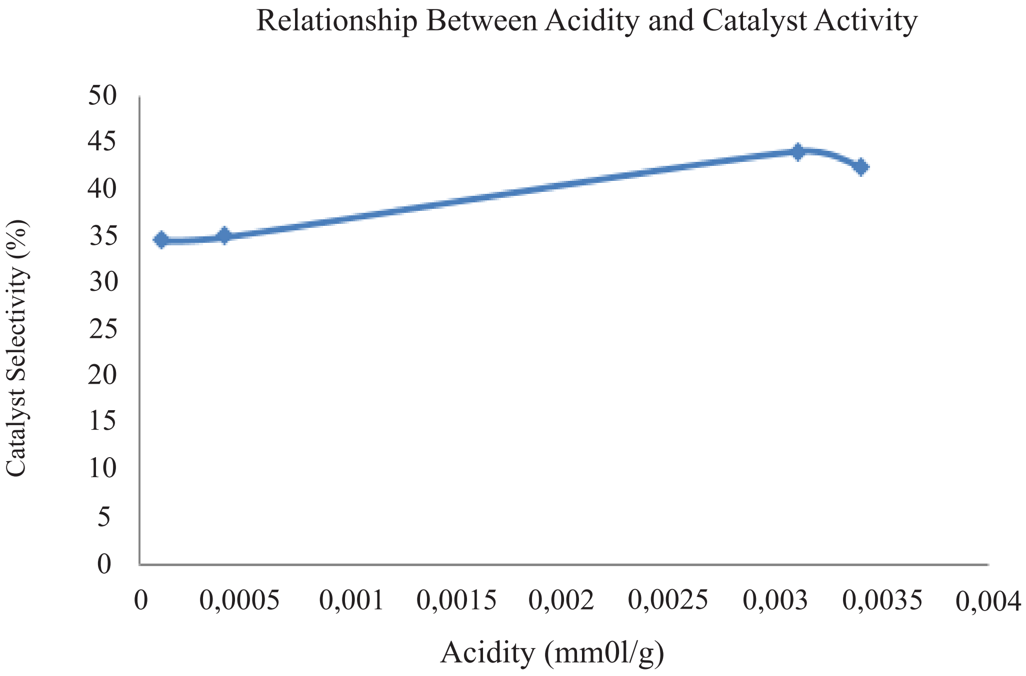


Figure 10
The relationship between acidity and catalyst activity

acidity characteristic of the catalyst is still related to the nickel content which may be distributed less evenly (Bartholomew, 2006) causing its activity to decrease compared to the 2% Ni-Faujasite catalyst, although the acidity is lower but not significantly different from the 4% Ni-Faujasite catalyst.

The trend of the relationship between acidity and selectivity of the catalyst to the biogasoline fraction shows that the higher the acidity of the catalyst, the greater the selectivity to the biogasoline fraction. Figure 13 shows the relationship between acidity and catalyst selectivity for the biogasoline fraction.

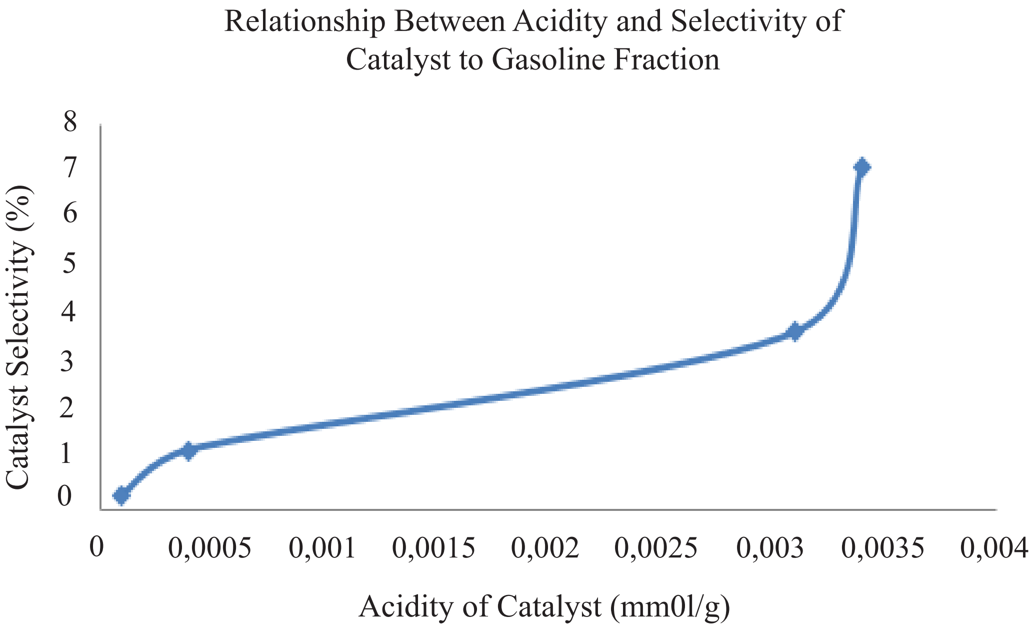


Figure 11
Relationship between acidity and catalyst selectivity to biogasoline fraction.

Catalysts with high acidity have more acid sites. The many acid sites will later affect the catalytic cracking process in producing the product. The increase in selectivity starting from the leached fly ash catalyst with the lowest acidity has the lowest selectivity as well. The graph in Figure 11 shows that the higher the acidity of the catalyst, the greater the selectivity for the biogasoline fraction. In this study, the highest acidity was 0.0035 mol/g on 4% Ni-Faujasite catalyst which was able to convert into liquid hydrocarbon products with the largest short fraction of 7.12%.

CONCLUSIONS

The conclusion in this study is that the characteristics of faujasite synthesis from fly ash are the main peak crystallinity of 67% and the Si/Al ratio of 1.64. Impregnation of Ni increases the ability of the catalyst in hydrocracking methyl ester palm oil to produce biogasoline fraction with activity yield of 42.34%. and a selectivity of 7.12% using a 4% Ni-Faujasite catalyst.

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GOSSARY OF TERMS

Symbol	Definition	Unit
XRD	X Ray Difraction	
GC-MS	Gas Chromatography-Mass Spectroscopy	
MEPO	Methyl Ester Palm Oil	
AAS	Atomic Absorption Sepectroscopy	
PLTU	Pusat Listrik Tenaga Uap	

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