REVIEW JOURNAL: THE EFFECT OF HETEROGENEOUS ZEOLITE CATALYST AND MODIFIED ZEOLITE CATALYSTS ON BIOGASELINE YIELDS

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Abstract
Increased fuel consumption of oil has an impact on the energy crisis. Biogasolin is an alternative fuel from vegetable oil that has the potential to overcome this problem. One of the methods used to produce biogasoline is the catalytic string method. Such methods generally use a catalyst to help speed up reactions. The use of catalysts in the cracking process is increasingly diverse. In this review article will compare the biogasoline percent value between several types of catalysts focused on zeolite and zeolite catalysts that are modified with metal, including the general picture of the arrest of the material, type and characteristics of the zeolite catalyst, operating conditions, and ways of performance of each catalyst. Zeolite catalyst which is a heterogeneous catalyst is considered to have good ability in selectivity, activity, and ease of modification so that it is very influential on the cracking results. Some zeolite catalysts modified with metals such as nimo/zeolite; Zn/HZSM-5; Cu/HZSM-5; Ni/HZSM-5; MO/HZSM-5; HZSM-5; PD/HZSM-5; PT/HZSM-5; Zn/Na-ZSM-5; Ni/ZSM-5; Ni/mnz; Co-NI/HZSM-5 and each biogasoline is obtained at 11.93%; 28.38%; 17.55%; 32-37%; 9-23%; 30.2%; 32-41%; 11.73% are reportedly able to provide performance to improve biogasoline results and certainly become a new catalyst that is useful in the scientific world

1. Introduction

Alternative fuel from vegetable oil has long been used, even various studies to make this fuel have been carried out in various countries even in the world and continue to grow until now [1]. Alternative fuels created are very environmentally friendly, derived from renewable energy sources, used efficiently and affordable prices. Very many renewable alternative energy sources, especially from vegetable oils such as oil palm oil, soybean oil, castor oil, sunflower oil and others [2]. Taking into account the availability that is easy enough to obtain it, is used efficiently and has a good chemical composition and characteristics, vegetable oils can be selected for vegetable fuel [3].

Based on a review of several literature, most of them are focused on making biodiesel, while the manufacture of biogasoline is not as much as making biodiesel [4]. However, from this biodiesel research that began the formation of a small amount of biogasoline so that research to produce biogasoline developed by N. Le-Phuc et., al [5]. Wargadalam, V. J et., al [6], Yoshimura, T., et., al [7] and Inaloo, E. B., et., al [8]. There are several processes of making biogasoline from vegetable oils that have been carried out by researchers and can be a reference for further research with a more innovative process both technically and economically. Especially considering there are still weaknesses in previous studies. The process used is dilution, microemulsion, thermal decomposition, cracking, and transesterification[9]. The cracking process is very appropriate to produce biofuels, especially biogasoline without catalysts or catalysts [10]. The catalytic cracking process has been carried out by several researchers as reported in the review written by N. Le-Phuc et., al [5] and Wargadalam, V. J et., al [6]. Generally the catalyst used is heterogeneous catalyst[11]. One heterogeneous catalyst is zeolite. Zeolite has a regular crystal shape with cavities connected in all directions causing the surface area of zeolite to be very large so it is best used as an adsorbent and catalyst [12]. Several studies that produce biogasoline have been carried out with an arrest using a zeolite catalyst [6],[5].

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Furthermore, research with zeolite catalysts has developed where zeolite is modified with metals such as Ni, Cu, Zn, Pd, AU and Pt have shown good results for the catalytic arrest process. Most catalyst making is done using methods such as impregnation of Planck methods and methods [13]. As for the catalyst characteristics which include the specific surface area, the distribution of the catalyst pore size, the cumulative pore volume, the nature of the catalyst texture is carried out using a number of analyzes such as XRD (X-ray diffraction), N_2 adsorption by Bet (Brunauer-Emmet-Teller) Method, Tem, SEM, BJH, and others[14]. In this article, the process of catalytic arrest with various zeolite and zeolite catalysts is modified with metal will be reviewed. The influence and performance of heterogeneous catalysts on biogasoline results is important to be discussed in this article.

2. Method

In this study using a review method from various process methods, understanding what the core of the theme or topic listed in the journal is. In scientific journals, reviews are conducted to understand the process and results of research listed therein. This journal review method is carried out by searching for previous journals regarding biogasoline production using raw materials Palm oil, Bio-oil, Coconut Oil.

3. Result and Discussion

The cracking process is the process of breaking down large hydrocarbon molecules into small molecules with the help of heat and/or without using a catalyst.

1. Cracking Process

The cracking process is the process of breaking down large hydrocarbon molecules into small molecules with the help of heat and/or without using a catalyst [15]. This process is often called pyrolysis which is basically thermal. Pyrolysis itself is a high temperature process that causes molecules to break and can crack. For vegetable oils this process begins at a temperature of 593 K [16]. Furthermore, cracking produces molecules and deoxygenation occurs simultaneously under the influence of heat. This process has been studied extensively and has been reported in several reviews[17].

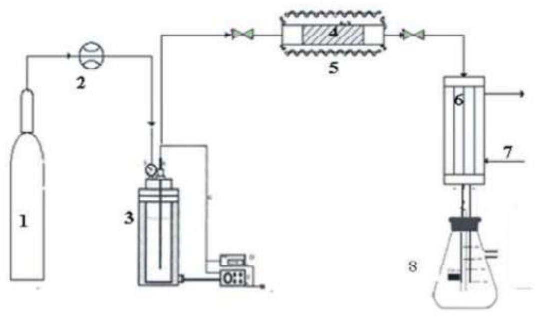

Figure 1. Experimental setup of catalytic cracking

The company is trying to find alternative fuels from vegetable oil through a process similar to cracking petroleum with temperatures ranging between 718 - 758 K and a pressure of 931.36 kPa [9]. From this process, biodiesel fuel and a small amount of biogasoline and biokerosene are produced. The formation of biogasoline is the beginning of the interest of researchers to focus on biogasoline production.

2. Catalytic Cracking Using Zeolite Catalyst

In its development, the process of cracking vegetable oil is carried out using a catalyst (catalytic cracking) which takes place at high temperatures and pressures, where the breakdown of fatty acids can be directed and changed with different reaction pathways. The purpose of using a catalyst is to reduce the temperature and speed up the reaction time, in addition to the presence of a catalyst can make the process selective for the desired reaction [18]. Vegetable oil used in the manufacture of biogasoline with the catalytic cracking process is vegetable oil. In addition, the raw materials of bio-oil have also been studied [7],[9]. In general, triglycerides are raw materials used for thermal and/or catalytic cracking processes. The use of triglycerides for thermal cracking processes is less beneficial because it produces acrolein and triolein, which are very toxic in the product. In addition, it produces coke which results in high viscosity in oil. Pre-treatment of raw materials by converting them into methyl esters through the transesterification process will prevent the formation of toxic materials in the reaction products [11]. For the selection of catalysts, the performance of zeolite catalysts is quite effective and promising in the process of converting triglycerides into biogasoline. The use of this catalyst is considered more beneficial because it does not require additional materials such as ethanol and methanol [19]. Table 2 shows some studies on biogasoline production using zeolite catalyst.

Table 2. Shows some studies on biogasoline production using zeolite catalyst.

Raw material	Catalyst	Temperature °C	Time (Hour)	% Biogasoline Yield	Reference
Palm oil	HZSM-5	360-420	2- 4 t	Biogasoline (40– 70%)	[20]
Palm oil	HZSM5, β-zeolite,	350-450	1-4 t	Biogasoline (28%)	[21]
Palm oil	REY Zeolite	400 – 500	10s, 20s, 30s	Biogasoline (59,1% wt)	[22]
Bio-oil	Zeolite, ZSM-5	450-500	15 min	Biogasoline (91,67%)	[23]
Coconut oil	HZSM-5, HY	450	30 – 30 t	Biogasoline (26,9 – 40,1%)	[24]

HZSM-5 catalyst is a synthetic zeolite, generally used in catalytic cracking processes to produce biogasoline [25]. The active catalyst HZSM-5 and capable of producing more gasoline than zeolite has been reported by H. K. Gurdeep[26]. But unfortunately this catalyst supports the formation of many gases, thus reducing the liquid fraction. Other catalysts such as ZSM-5, REY zeolite, HZSM5, β-zeolite, ZSM-5 zeolite, and natural zeolite are the catalyst options for further researchers. For the ZSM-5 catalyst, it is able to provide very good results with a biogasoline yield reaching 91.67[23]. The process of converting palm oil into fuel and other chemical compounds with the HZSM-5 catalyst at operating temperatures ranging from 360oC to 420oC with results showing that 40 - 70% of palm oil can be converted into aromatic compounds and hydrocarbons such as biogasoline [20]. In the same year, K. Wijaya et., al made liquid fuel which was still based on palm oil using the HZSM-5 β-Zeolite catalyst, the resulting product had a biogasoline yield of 28% through a cracking process at a temperature of 350 - 450°C [27]. Zarkoni Azis et.,al [22]conducted an in-depth study of the relatively new REY zeolite catalyst at that time in producing biogasoline. The catalytic cracking process used a transport riser reactor (TRR) with a biogasoline yield of 59.1%. An interesting catalytic cracking process was carried out by Hew et al using bio-oil as the raw material. The Taguchi L9 method found the optimal operating conditions for the catalytic cracking process of the ZSM-5 Catalyst and zeolite at a temperature of 400oC, a reaction time of 15 minutes and a catalyst of 30 grams. In this condition, the biogasoline yield was 91.67%[23]. Research using two-stage processes (transesterification and thermal cracking processes) has also been carried out using sunflower oil as the raw material [28]. Their research stated that the transesterification process as a pretreatment stage before carrying out the cracking process, provides better final results. The use of triglycerides for thermal cracking without pretreatment is less beneficial because it produces acrolein and triolein, which are very toxic in the product. It also produces coke which produces high viscosity in the oil. With this pretreatment, undesirable things can be prevented. The properties of fuel after cracking sunflower oil and its methyl ester have been compared to determine the advantages of the transesterification process as a pretreatment stage for triglycerides. The transesterification process took place for 3 hours with a reaction temperature of 60oC using methanol and KOH catalyst. The two stages of the process above can be used as a solution to increase biogasoline yield, which is still low until now. Good pretreatment will produce methyl ester optimally, of course it will be beneficial to be used as raw material for the next process.

3. Catalytic Cracking Using Zeolite Catalyst Modified With Metal

In the use of zeolite catalysts, several factors must be considered, including activity, selectivity, use time and ease of regeneration. Zeolite catalysts in the cracking process can control activity and selectivity, but a number of biogasoline yields produced are low. Its performance can be improved through modification with metals such as Cu, Ni, Zn. Biogasoline production using zeolite catalysts modified with metals has not been studied too much when compared to studies using zeolite. Table 3 shows some of these studies. Haswin Kaur Gurdeep Singh et al [26]. compared the effect of HZSM-5 catalyst without and with impregnation of Cu, Ni and Zn metals on biogasoline yield. Zn/HZSM catalyst gave the highest yield compared to other catalysts, which was 29.38% at a temperature of 450°C. However, they recommended Ni/HZSM catalyst for biogasoline selectivity in palm oil cracking. In line with the recommendations above [30], biogasoline yield of 32 - 41%[30], Zeolite catalyst with Ni and Mo metal or NiMo/zeolite has been used in several studies even though the biogasoline yield is still low.

Cracking process with this catalyst produces biogasoline yield of 4 to 15%. Later, other updates of catalyst in biofuel production were also found by Rafiani, A [31]. studied the effect of HZSM-5 Catalyst with and without Pt and Pd impregnation in biodiesel production. His research turned out to produce biogasoline yield of 29.38%. RBD (refining, bleaching, deodorizing) process was carried out first, then continued with cracking process. After RBD process, 55% oleic acid and 30% palmitic acid were obtained, which will then be cracked using HZSM-5, Pt/HZSM-5 and Pd/HZSM-5 Catalyst. The results showed that the amount of biogasoline yield increased with the reduction of biodiesel amount and biokerosene yield.

Table 3. Biogasoline Production from Vegetable Oil with Catalytic Cracking Process Using Zeolite Modified with Metal

Raw material	Catalyst	Temperature T°C	Time t (hour)	% Biogasoline Yield	Reference
Palm oil	NiMo/Zeolite	300-320	1, 1.5, 2	11,93%	[4]
Palm oil	Zn/HZSM-5 Cu/HZSM-5	350-500	2	28,38% 17,55%	[29]
Palm oil	Ni/HZSM-5	550	1-3	32-37%	[30]
Palm oil	HZSM-5, Pd/HZSM-5, Pt/HZSM-5	350-550	2 min	9-23% 30-35% 47%	[31]
Coconut Oil	Zn/Na-ZSM-5	450- 500	0,2;0,6; 1	30,2%	[32]
Palm oil	Ni/ZSM-5	550	1-3	32-41%	
Coconut Oil	Ni/MNZ	360-410	-	11,73 %	[33]
Coconut Oil	Co-Ni/ HZSM-5	300-375	2	1,97%	[34]

Pt and Pd are noble metals which are transition metals. They have many electrons that are easily delocalized. This can affect the reaction especially to break covalent bonds during cracking. Therefore, their use is considered to increase the amount of yield and have better selectivity [31]. Meanwhile, to crack Carinata oil using Zn/Na-ZSM-5 catalyst at various temperatures of 400, 450 and 500oC. Kongparakul, S et al[32] also investigated the catalytic cracking process of camelina oil with Zn/ZSM-5 Catalyst. Both studies produced hydrocarbon fuels. The frequency of oil extraction and the liquid space velocity per hour were considered as important factors for producing liquid hydrocarbons. The optimum conditions were achieved at 550oC, space velocity at 1.0-hour and extraction frequency of pressure was 15 Hz. The Zn/HZSM-5 catalyst was also used by Widayat in palm oil cracking which produced a biogasoline yield of 2.54% [32]. Furthermore, research by Singh, H. K. G et., al used Callophyllumnophyllum L. oil as raw material [30] which not only contains palmitic, oleic and linoleic acids, but some long chain fatty acids C_{20}, C_{22}, C_{23} are also present in this oil. The cracking process takes place in an autocluster batch reactor (ABR) and uses a NiMo/zeolite catalyst. From here, the research obtained a biogasoline yield of <15%.

4. Conclusion

Biogasoline production can be done by catalytic cracking using zeolite and zeolite catalyst modified with metal. Zeolite catalyst which is a heterogeneous catalyst, in its use must be considered several factors including activity, selectivity, usage time and ease of regeneration. In the cracking process, zeolite catalyst can control activity and selectivity where activity and selectivity are influenced by catalyst characteristics. Therefore, the right catalyst preparation method and active metal content are needed so that the catalyst characteristics are good and have a good effect on the cracking results. Biogasoline production through the direct catalytic cracking process of triglycerides is not very appropriate because it forms acrolein and triolein compounds (toxic compounds) and its formation from coke. With a two-stage process where the first stage of the process as a pretreatment followed by the catalytic cracking process will provide better results. The use of the ZSM-5 catalyst is able to provide very good results in the cracking process with a biogasoline yield reaching 91.67% and 70%.

The catalytic cracking process can produce biogasoline at temperatures ranging from 300-550^{o}C. Catalytic cracking of bio-oil into biogasoline is recommended at a temperature of 400^{o}C, a reaction time of 15 minutes and a catalyst of 30 grams. With these conditions, the biogasoline yield was 91.67%. Meanwhile, the reaction temperature at 450^{o}C is likely to produce high-octane gasoline, low-heavy oil and gas. However, the problem is the formation of coke on the catalyst, which results in the deactivation of the catalyst by the accumulation of coke on the catalyst surface. Therefore, residence time is needed as a solution to increase the biogasoline yield and reduce coke formation.

Review of several biogasoline production studies in this article is expected to provide an overview of the development of biogasoline production through the catalytic cracking process using zeolite and zeolite catalysts modified with metal. In addition, it can provide information on the types of zeolite catalysts that have good performance in the cracking process and also the right operating conditions for producing biogasoline so that optimal biogasoline results are obtained.

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