Upgrading Gasoline Yield and Octane Quality in Fluid Catalytic Cracking by Coprocessing of Vacuum Gasoil with Palm Triglyceride Fatty Acid using REY-Type Zeolite Catalysts

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Abstract

Gasoline fuel produced by fluid catalytic cracking unit is mostly influenced by characteristics of catalyst used and process condition, eventhough feed nature determines the product's yield and quality. Coprocessing of vacuum gasoil (VGO) with palm triglyceride fatty acid has been performed experimentally using fluid-bed reactor of ACE unit over equilibrium catalyst of single REY-type zeolite at reaction temperature 530°C and catalyst-oil ratio of 5.5 g/g. Three kind of cracking feeds were tested and evaluated including neat VGO, mixture of VGO with 0-15% refined bleached deodorized palm oil (RBDPO) and VGO mixed with 0-15% of RBDPO and palm fatty acid distillate (PFAD) at ratio 9:1.

It was found that cracking feeds containing palm triglyceride fatty acid influenced significantly the product yield and conversion. The change of gasoline yield from 42.9 to 48.3% and octane number from 92.4 to 97.7 was found from coprocessing of VGO with 5% RBDPO-PFAD. The role of palm triglyceride fatty acid along with VGO feed was suppossed to be dominantly in strengthening the cracking ability as indicated by increase of strong acid sites of catalyst from action of hydrogen transfer reaction. The special nature of feeds prepared by mixture of vacuum gasoil with a few palm triglyceride fatty acids would be good potentials of alternative route to upgrade gasoline in fluid catalytic cracking process.

Keywords: Catalytic cracking, vacuum gasoil, gasoline, palm triglyceride fatty acid, catalyst acidity

I. INTRODUCTION

Gasoline is liquid hydrocarbon fuel composed of C_5 to C_{12} carbon atoms per-molecule that's mostly needed for transportation fleets. One of vital units in petroleum oil refinery to produce a lot of gasoline is fuid catalytic cracking (FCC) that processes less valuable feeds of heavy oil fractions like vacuum gasoil (VGO) and oil residue. The necessary of gasoline nowdays has exceeded the production capacity of existing units, for example in Indonesia the consumption of gasoline was about 33 million kiloliters in 2017 while the production capacity was about 14 million kiloliters [1], so it is crucial to search the alternative potential to cover the gap. A common effort to upgrade gasoline in fluid catalytic cracking was catalyst selection as well as process optimization,

however feed nature affected the resulting yield and quality [2]. Vacuum gasoil is generally used as standard feedstock for FCC unit in oil refinery. Chemically, VGO is a complex combination of hydrocarbons containing a broad range of different polarities compounds such as paraffinic, aromatic and heteroatomic molecules [3] as shown in figure 1.



Figure 1. General structure of VGO

The long chain hydrocarbon molecules of VGO under FCC condition will be broken into shorter hydrocarbon molecules like gasoline fraction and other lighter products. The properties of VGO will give reliable information for prediction of conversion and product yields [4]. The performance of cracking reaction is mainly influenced by catalyst properties especially its acidity representing the active sites of catalyst [5]. Catalytic cracking reaction is basically an acid-catalysed reaction, which the number of catalyst active sites affects the reaction process. Cracking reaction over zeolite catalyst will cause the longer hydrocarbon molecules in oil feed leading to smaller hydrocarbons as a result of multiple reactions starting from primary and secondary cracking, hydrogen producing reactions, hydrogen consuming reactions, production of some molecules by C-C bond formation such as aldol condensation or Diels-Alder reactions and dehydration [6]. The acid distribution of REY-type zeolite catalyst is more concentrated in the range of intermediate and strong acidity, improving the hydrogen transfer activity. The reduction of strong acid decreases the formation of coke [7].

The molecular structure of vegetable oil commonly contain a large amount of long-chain fatty acids, the hydrocarbyl group of which is similar to the paraffinic VGO. It is well-known that paraffinic VGO is the ideal feedstock to produce light olefins through catalytic cracking process [8]. The main difference between vacuum gas oil and vegetable oil is the presence of oxygenate compound of carboxyl groups wherein the general structure of triglyceride in vegetable oil as shown in figure 2. The conversion process of vegetable oil is influenced by feed properties and fatty acid composition,

operating condition parameter and characteristics of catalyst used [9]



Figure 2. Main structure of triglycerides [10]

Compared to bio-oil with poorer properties, upgrading vegetable oil seems to be the initial step towards higher yield of biofuel [11]. Crude palm oil (CPO) is produced abundantly in Indonesia from extracting the ripened fruit mesocarp of the oil palm tree [12]. This vegetable oil is composed of triglycerides about 94–98%, diglycerides 5–8%, monoglyceride 0.21–0.34% and free fatty acid 2–5% added with few of moisture and impurities, trace metals and minor carotene content [13].

Refined bleached deodorized (RBD) palm oil is pure palm oil derived from the process of refining, bleaching and deodorizing of crude palm oil. RBD palm oil is composed mainly of triglycerides, few diglycerides and minor free fatty acid below 0.1% wherein both free fatty acid and moisture & impurities is limited at 0.1% maximum. The fatty acid composition of palm oil generally includes myristic acid (14:0), palmitic acid (16:0), stearic acid (18:0), oleic acid (18:1) and linoleic acid (18:2), wherein the amount of saturated fatty acids is almost equal to the unsaturated fatty acids [14]. Palm fatty acid distillate (PFAD) is byproduct of CPO during RBD process which is used in many industries such as laundry soap, animal feed and as raw material for oleo-chemical industry. PFAD is also often considered as a valuable and low cost raw material for biodiesel production. Free fatty acids, phospholipids or gums and lipid oxidation products are the major undesirable compounds of crude palm oil which can be removed during refining [15]. The main component of PFAD is free fatty acids above 80% and few of triglyceride, partial glyceride and unsaponifiable matters like vitamin E, sterols, squalenes and volatile substances [16]. PFADs from several palm oil refineries had slightly different oxidation level and free fatty acid content [17]. Main usage of palm oil is for edible purposes after refining, and some was also used for energy purpose by turning it into biodiesel after trans-esterification process of RBD palm oil with methanol resulting in methyl ester or biodiesel. Due to excess of palm oil production, it has high potential for domestic biofuel precursor in addition to food-grade and other non-food applications. As petroleum gasoil cracking, palm oil can be converted into gasoline fraction because its molecular chain is longer than gasoline that enables to be cracked further into shorter hydrocarbon molecules. Coprocessing of palm oil with conventional feedstock in existing FCC unit is a promising option to produce biofuel.

II. RELATED WORK

Extensive researchs regarding catalytic cracking of palm oils solely or by coprocessing using small scale of fixed and fluid

bed reactors have been done by some authors previously. Palm oil added in steps of 20% up to 100% into conventional feed of VGO was cracked in the pilot scale of fluid catalytic cracking unit over dual-zeolite catalyst of REY-ZSM5. The result of cracking reaction showed declining in conversion and gasoline yield, besides increase of water content formed at higher level of palm oils above 20% [10]. Abbasov et al (2016) investigated blends of cottonseed and sunflower oils with vacuum gasoil which were catalytically cracked in fixed bed microreactor using commercial high alumina zeolite FCC catalyst and halloysite nanotubes as a catalyst. The number of unsaturated fatty acids in cottonseed and sunflower oils showed more than content of saturated fatty acids. Yield of gasoline resulted from cracking reaction was increased by addition of cottonseed and sunflower oils within VGO feed by 4% to give 50% of gasoline [18].

Doronin et al (2019) studied experimentally that catalytic cracking of vacuum gas oil mixed with palm oil at concentration of 5-10 wt% over dual-zeolite catalyst using micro reactor had allowed increasing conversion and yields of gasoline as well as light olefins. They reported that effect of olefin to the distribution of cracking products from coprocessing feeds could be affected by the number of double bonds in triglyceride oils as represented by unsaturation index. The addition of palm oil to vacuum gas oil limited the absolute increment of gasoline yield due to lower unsaturation index compared to sunflower oil [19].

The comparison study of product yield distribution from palm oil with those from conventional petroleum hydrocarbon feedstocks showed that palm oil has a unique advantage in the production of light aromatic hydrocarbons because of its double-bonded straight chain structure. The molecular structure of palm oil is beneficial to be treated over the shapeselective zeolite, since the yield of light aromatics is higher than using hydrocarbon feedstocks, especially in the formation of more benzene and toluene [8]. In the cocracking of vacuum gas oils with palm oil using dual-zeolite catalysts of HREEY-ZSM5, the amount of vegetable oil with relatively low unsaturation index of palm oil should be in the range of 5-10 wt% since increasing the oil content in the mixture enhances the coking of the dual-zeolite catalyst and simultaneously reduces yield of desired products. Increase of gasoline yield was obtained at 10 wt% concentration of palm oil from 38.7 wt% of neat VGO to be 40.6 wt% after coprocessing along with increase of coke yield from 8 to 9.4 wt% while LPG yield decreased [20].

The aim of current research work is to find out the effect of feed nature prepared by vacuum gasoil mixed with palm triglyceride fatty acids on the cracking performance in order to upgrade gasoline yield and octane quality. RBD palm oil and palm fatty acid distillate as primary source of triglyceride fatty acid was used for coprocessing with VGO feed. Single REY-type zeolite of commercially FCC catalysts was set at co-cracking of three kind of feeds run in the confined fluidbed reactor of ACE unit at certain process condition. The change of cracking performance caused by feed reactivity was evaluated from the alteration of catalyst acidity before and after cracking reaction that significantly influenced to the product yield distribution.

III. METHODOLOGY

3.1 Feedstock and catalyst

Vacuum gasoil used for standard cracking feed was supplied from petroleum oil refinery. Refined bleached deodorized palm oil and palm fatty acid distillate used for coprocessing was technical grades and supplied from the palm oil products industry. The physical dan chemical properties of VGO is shown in table 1, whereas the properties of RBDPO and PFAD is shown in table 2.

Test Parameter, unit	Method	Value
Density @20°C, gr/cm ³	Pycnometric	0.913
Sulfur content, wt%	ASTM D-2622	0.140
Carbon Residue, wt%	ASTM D-4530	0.260
Total Acid Number, mgKOH/g	ASTM D-664	0.175
Distillation, °C	ASTM D-2887	
IBP		251.2
10%		330.6
30%		381.6
50%		414.8
70%		438.4
90%		471.0
FBP		558.2
Metal content, mg/kg:	AAS	
Fe		1.34
Na		1.42
V		0.96
Ni		0.11

Table 1. Main properties of VGO

Coprocessing feeds were prepared for cracking reaction namely VGO, mixture of VGO with 0-15% RBDPO and VGO mixed with 0-15% of RBDPO-PFAD (9:1), respectively. Each feed was pretreated before entered into reactor by means of heating at temperature 90°C and then it was filtered using small size of sieve to eliminate particle contaminants in oil. In each cycle of run, hot feed is maintained at temperature 180°C for 2 hours in the feed injector line of ACE unit before flowing and entering into fluid-bed reactor.

Tabel 2. Properties of RBDPO and PFAD

Test Parameter, unit	Method	RBDPO	PFAD
Free Fatty Acid, %	ASTM D-664	0.09	89.9
Water & Impurities, %	ASTM D-2709	0.05	0.94
Total Acid Number, mgKOH/g	ASTM D-664	0.18	179.8
Iodine Value, g/100g	AOCS	52.01	55.14
Peroxide Value, meq/kg	AOCS	0.45	3.11
Carbon Residue, wt%	ASTM D-4530	0.42	0.51

Testing iodine value is intended to determine the level of unsaturation fatty acid in palm oil fraction. Iodine value refers to the percentage of iodine absorbed by a substance of vegetable oil that is an indication of a its potential to be oxidized as it measures the reaction of iodine with the double bonds of unsaturated fatty acids. The greater number of double bonds in PFAD provide more sites for oxidation than RBDPO as shown in tabel 2. Oxidation level of palm oil fraction was detected from peroxide value indicating the primary oxidation products formed. An alternative indicator representing the level of double bonds in vegetable oil can be calculated from unsaturation index based on Doronin formula as the percentage of double C=C bonds corresponding to a carbon atom in the molecules of triglyceride fatty acids [19].

Catalyst used for cracking reaction in the research work was single REY-type zeolite catalyst coming from the commercial FCC unit in form of equilibrium catalyst or E-cats having chemical and physical properties as shown in table 3.

Test Parameter, unit	Method	Value
Total Surface Area, m ² /g	ASTM D3663	156.1
Zeolite surface area, m^2/g	ASTM D4365	114.3
Matrix surface area, m ² /g	ASTM D4365	41.8
Total Pore Volume, cc/g	ASTM D4222	0.201
Average Pore Radius, Å	ASTM D4222	25.72
Composition, % Cons:		
Al_2O_3		37.53
SiO_2		42.04
SiO ₂ / Al ₂ O ₃	XRF	1.12
Rare earth (La)		0.76
Na		0.27
Ni		0.028
Loss on Ignition		5.91
@750°C, %wt	UOP-954	5.71
Average Particle Size, µm	Malvern	106.3

Table 3. Main characteristics of catalyst

Catalysts used for cracking rection before entering into reactor was pretreated namely sieving of catalyst to know the particle size distribution of catalyst in range of 38 – 212 μ m. The catalyst was dryed at 225°C for 1 hour and then calcined at 600°C for 1 hour to eliminate some catalyst contaminants such as moisture, hydrocarbon residue and coke.

The changes of catalyst acidity before and after cracking reaction was analyzed by ammonia temperature-programmed desorption (NH₃-TPD) test method using Micromeritic Chemisorb 2750 automated system. At first step catalyst was pretreated by heating at 400°C for 1 hour under inert gas of helium. NH₃ adsorption (5% v/v in helium) was performed at 100°C for 30 minute, then it was purged by helium gas at the same temperature for 30 minute. NH₃ desorption was performed at temperature increment of 100 up to 600°C by heating rate of 10°C/minute, then holding it at 600°C for 30 minute.

3.2 Cracking process and products analysis

The experimental cracking process was carried out in the confined fluid-bed reactor of advanced cracking evaluator (ACE) unit using commercially FCC equilibrium catalyst of REY-type zeolite. The reaction condition was set at temperature 530°C, feed rate 1.2 grams/minute and catalyst load 9 grams wherein feed injection time is varied from 112 to 69 second to get approximately 4 to 6.5 of catalyst-oil ratio. The general scheme of reactor is shown in figure 3.



Figure 3. Simplified fluid-bed reactor

The result of cracking reaction in form of gaseous product was collected in a water displacement receiver and analyzed by an on-line gas chromatograph to obtain gaseous composition of C₁, C₂, C₃ and C₄ hydrocarbons, including H₂, CO and CO₂. The liquid product was collected in a chilled glass receiver and analyzed the boiling point distribution using an off-line gas chromatograph of simulated distillation based on test method of ASTM 2887 to obtain gasoline, light cycle oil (LCO) and bottom oil. Composition of individual hydrocarbons in gasoline fraction including group type of hydrocarbons and calculated octane number was analyzed using gas chromatograph of detailed hydrocarbon analyzer (DHA). The technique of DHA is based on the separation of individual hydrocarbon components by utilizing ultra-high resolution capillary column of gas chromatography and reference component databases based on retention time. Analysis of group type of hydrocarbons in gasoline included normal paraffins, isoparaffins, olefins, napthenes and aromatics.

Coke on catalyst was determined by in-situ air calcination of the spent catalyst at fluid-bed regeneration temperature 715°C for 240 second of reduction time and the measurement of product carbon dioxide with an infrared analyzer to obtain carbon content or coke. Few of water collected in liquid product was detected by volumetric karl-fichser test method. Mass balances of reation process were calculated and only runs with balances >95 wt% were accepted for the data correlations. The product yields were calculated and normalised (excluded coke yield) into main groups of dry gas ($C_1 \& C_2$ hydrocarbons, H_2 , CO and CO₂), liquefied petroleum gas (LPG) consisting of C_3 and C_4 hydrocarbons, gasoline fraction (boiling point range of C_5 –215°C), light cycle oil (LCO) having boiling point range of 216-370°C, bottoms oil with boiling point range above 370°C and coke, including water if formed. All product yields were expressed on a weight percent of feed basis.

The yield of each product fraction (Y) was calculated by the following equiation:

$$Y = mass of fractioni / total mass of feed x 100\%$$
(1)

Conversion is defined as the sum of dry gas, LPG, gasoline, coke and water if any, or it can be expressed in the equation:

$$X = (Dry gas + LPG + Gasoline + Coke + water), \%$$
$$= (100 - LCO - Bottoms), \%$$
(2)

where X is conversion in wt%, Y is yield of productⁱ in wt%

IV. RESULTS AND DISCUSSIONS

4.1 Acidic property of feeds

Track of fatty acid in the coprocessing feeds can be detected from testing parameter of total acid number as shown in figure 4. Few acidity of VGO feed generally has a source from naphtenic acid deliverance of crude oil as primary feedstock of the oil refinery. Mixture of VGO with RBDPO has low acidity because of less free fatty acid and major triglycerides in RBDPO.



Figure 4. Acid number of each feed

Mixture of VGO with RBDPO-PFAD increased the acidity of feeds caused by major content of free fatty acid in PFAD although it was added in small concentration. The high acidity of PFAD let the blending ratio of RBDPO-PFAD was maintained at 9:1 giving the acidity level of feed about 1 mgKOH/g so that it meets in safe condition for metallic equipments from corrosion effects.

4.2 Conversion and products yield distribution

Co-processing of VGO with RBDPO and RBDPO-PFAD at reaction temperature 530°C and catalyst-oil ratio 5.5 g/g each resulted in products distribution which's similar to neat VGO namely dry gas, LPG, gasoline, light cycle oil, bottom oil and coke, except for insignificant CO and CO_2 in dry gas, and trace of water collected in liquid product from oxygenate reaction.

Conversion and gasoline yield resulted from catalytic cracking of VGO with 0-15% RBDPO are shown in figure 5 & 6, wherein the highest conversion and gasoline yield was obtained at concentration of 5% RBDPO.



Figure 5. Conversion against %RBDPO in VGO



Figure 6. Gasoline yield against %RBDPO in VGO

Increasing yield of gasoline and conversion might be effect of triglycerides of RBDPO in feed that's under cracking condition was converted into fatty acids and further cracked to be hydrocarbon molecules. The detailed mechanism of vegetable oil cracking had not been well explained by some previous researchers, but most of them agreed to the phenomenon of primary cracking involving thermal decomposition of triglycerides to result in oxygenated hydrocarbons by free radical mechanism. These of highmolecular compounds then undergo secondary cracking reaction resulted in several hydrocarbon fractions due to role of catalysts acidity [21].

Cracking of VGO mixed with 0-15% of RBDPO-PFAD (9:1) gave significant change of conversion and gasoline yield as shown in figure 7 - 8.



Figure 7. Conversion against %RBDPO-PFAD in VGO



Figure 8. Gasoline yield against %RBDPO-PFAD in VGO

Increase of gasoline yield from 42.9% to 46.8% was found at coprocessing of VGO with 5% RBDPO. The highest gasoline yield of 48.3% was obtained from mixture of VGO with 5% RBDPO-PFAD (9:1) under reaction temperature of 530°C and catalyst-oil ratio of 5.5 g/g.

The overall products yield distribution obtained from cracking of three co-processing feeds is shown in figure 9.



Figure 9. Product yields against co-feeds

Coprocessing of VGO with 5% of RBDPO and 5% of RBDPO-PFAD respectively did not only affect to raise gasoline yield, but also affected to change LPG, LCO and bottom oil yields to be slightly lower. Dry gas was almost unchanged, but coke yield was slightly increased.

There was not oxygenate compounds found from analysis resulut of gasoline composition. In addition to gas oline yield, the coprocessing feeds improved the octane quality gasoline as shown in figure 10.



Figure 10. Gasoline octane against feed types

The octane quality of gasoline as expressed by reseach octane number (RON) has increased from 92.4 to 96.2 after adding VGO feed with 5% RBDPO. The highest RON of gasoline at 97.7 was achieved at coprocessing of VGO with 5% RBDPO-PFAD (9:1). The higher iodine value of PFAD meaning more unsaturated fatty acid than that of RBDPO may be possible a trigger to enlarge hydrogen transfer toward zeolite surface as indicated by increase of strong acid sites of catalyst.

Since VGO and palm oil fractions have different molecular structure, they undergo different reactions. The triglycerides of RBDPO decomposed in a fast thermal cracking step to free fatty acids and an allyl-carbenium ion. The fatty acids then enter to the catalyst pores and undergo further reactions to form hydrocarbon molecules. Vu (2014) reported that cracking of vegetable oil in the primary step involved initial thermal decomposition of triglycerides to yield fatty acid in form of heavy oxygenated hydrocarbons by means of free radical mechanism, which is independent of catalyst characteristics. The oxygenated compounds with highmolecular weight then will undergo secondary cracking reactions resulting in different products where the acidity of catalysts play an important role [21]. Combining PFAD and RBDPO with VGO feed was useful to enlarge fatty acids and forming hydrocarbons during cracking reaction. The presence of unsaturated fatty acid in feeds would be also possible factor in the alkylation, cyclization and dehydrogenation reactions to form gasoline having higher octane [10]. The phenomenon of hydrogen transfer influenced by feed reactivity can be evaluated from the change of catalyst acidity before and after cracking reaction as shown in figure 11 and 12.



Figure 11. NH₃-TPD spectra of catalysts

where

- (a). Initial catalyst before cracking (E-cats)
- (b). Catalyst after cracking of VGO
- (c). Catalyst after cracking of VGO+5%RBDPO
- (d). Catalyst after cracking of VGO+5% RBDPO-PFAD

Based on NH₃-TPD spectra analysis displayed in figure 11, there is one peak at $175 - 200^{\circ}$ C and one peak above 500°C,

that corresponds to weak acid sites and strong acid sites of catalyst, respectively.



Figure 12. Catalyst acidity before and after cracking where (a), (b), (c), (d) as noted previously

The acid sites of catalysts obtained from NH₃-TPD analysis was boken down into strong acid sites and weak acid sites as shown in figure 12. The strong acid sites of catalyst represents the number of active sites of zeolite increased significantly when coprocessing VGO with 5% RBDPO-PFAD. Weak acid site of catalyst indicating the physical desorption of NH₃ from matrix of catalyst tends to be lower along with decrease of bottoms product.

The increase of strong acid sites of catalyst had played important role to upgrade gasoline through transfer mechanism of proton from reactive feed to zeolite surface of catalyst that resulting in paraffinic and aromatic hydrocarbons as shown in figure 13.



Figure 13. A proposed scheme of hydrogen transfer mechanism of palm triglyceride fatty acid and petroleum hydrocarbon [19]

The higher carbon residue in RBDPO and PFAD than that of VGO feed could also contribute to keep gasoline yield and conversion at certain level of coprocessing feed although its reactivity increased with increase of fatty acid content or feed acidity. The enhancement of gasoline octane was supposedly due to formation of more iso-paraffinic and aromatic hydrocarbons in the gasoline fraction after coprocessing.

LPG product consisting of C_3 and C_4 hydrocarbons had tendency to decrease because of lower aromatic hydrocabon in the coprocessing feeds. The increase of catalyst acidity also affected to raise the conversion and coke slightly, meanwhile dry gas kept almost unchanged. Coprocessing of VGO with palm triglyceride fatty acid could contribute to olefin polymerization from unsaturated fatty acid that increased slightly of coke yield. The co-cracking conditions should give insignificantly amount of CO and CO₂ in dry gas and least water in liquid product. Light cylce oil as diesel fraction and bottoms oil associated with unconverted products showed

tendency to decrease after coprocessing due to the effect of hydrogen transfer reactions moving faster than primary cracking.

V. CONCLUSION

Coprocessing of vacuum gasoil with palm triglyceride fatty acids in fluid-bed reactor using REY type zeolite catalyst under certain process condition have shown significant results both of reactive conversion and product yield distribution. The addition of 5% RBDPO and 5% RBDPO-PFAD (9:1) into VGO feed influenced significantly the results of catalytic cracking towards enhancement of gasoline vield and octane quality. The improvement of gasoline occurred not only by accumulation effect of selective products originally from cracking of VGO, RBDPO and RBDPO-PFAD respectively, but also caused by increase of catalyst acidity having higher strong acid sites from hydrogen transfer reaction. The special nature of feeds prepared by combining vacuum gasoil with a few palm triglyceride fatty acid would be good potentials of alternative route to upgrade gasoline in fluid catalytic cracking process.

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