

Recent Catalyst Technology Development in Residue Catalytic Cracking Unit: A Mini Review

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ABSTRACT

The use of fuel oil from year to year until now has experienced a rapid increase, due to increasing population growth in vehicle operations and the development of industries that require fuel. The position of renewable energy has not been able to shift the position of petroleum fuels to the needs of the world market. The RCC unit is secondary processing that converts heavy fraction hydrocarbons to light fraction hydrocarbons with the help of using catalysts into high-value petroleum products such as Naphtha, LPG, and Light Cycle Oil. RCC unit has three main parts of the process, namely riser, stripper, and regenerator, with the use of the main raw material of residue and using a catalyst. This paper aims at reviewing recent journals concerning the catalyst applied in the RCC unit. The basis of the RCC unit to be reviewed is from an existing RCC unit in Indonesia. The development of research on catalysts and regeneration technology in the RCC unit has experienced rapid development until now. It is expected that this paper can contribute to the future development and application of catalysts for the RCC unit, both for national and international levels.

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1. Introduction

Fuels consist of petroleum fuels and renewable energy. The need for petroleum fuels has increased rapidly along with population growth. And the position of renewable energy is still not able to shift the position of fuel oil which becomes an energy source and still dominates the availability in the world market [1]. Figure 1 shows the estimate consumption world's energy needed.

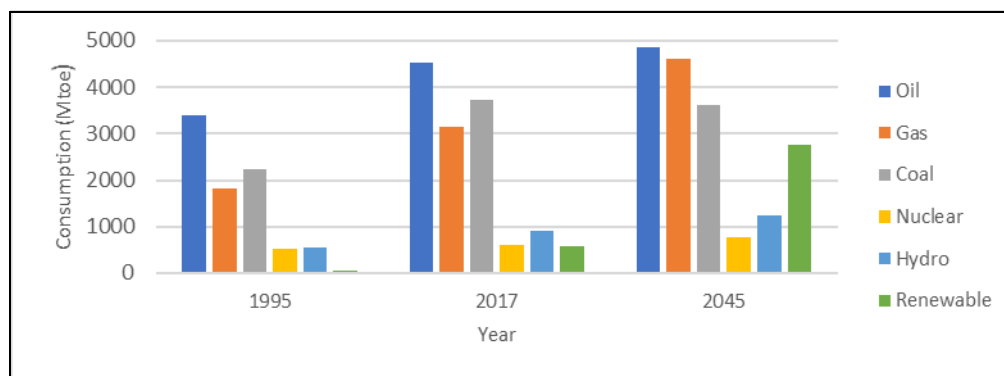


Fig. 1. Estimation of the world's energy needs according to BP-Energy Outlook 2019

Referring to Figure 1, fuels consist of petroleum fuels and renewable energy. The need for fuel oil in Indonesia will continue to increase until 2050 with an estimated increase of 40.1% from 4,569 million BOE [2]. Therefore, to optimize and maximize the production of petroleum fuels, one of them is by utilizing residue, which is a by-product into a more valuable petroleum product by using the Residue Catalytic Cracking Unit [3].

Residue Catalytic Cracking (RCC) is secondary industrial processing that converts heavy fraction hydrocarbons to lighter fraction hydrocarbons into more valuable petroleum products. The RCC unit consists of three main parts namely riser, stripper, and regenerator. In the RCC process, heavy oil experiences direct contact with the catalyst at high temperatures. Then, the output from the reactor is further processed in the column to be separated according to the type of oil product based on its boiling point, which will produce kind of light oil and heavy oil. Then catalyst after used will be fell into the stripper and after this flowed to the top of the regenerator with the aim to reactivate the catalyst by removing coke attached to the catalyst surface and regenerated by combustion of coke attached to the catalyst [4].

Until now the development of residue catalytic cracking technology continues, especially the development of catalyst types and regeneration technology. Catalysts used in the RCC process are various, but generally use zeolite catalysts [5], synthetic silica-alumina, natural composites or aluminosilicates, and silica aluminate [6]. However, most of the commercial catalytic cracking processes use catalysts of silica-alumina and zeolite [7]. Then catalyst will be recycling with regeneration technology using a regenerator, with the aim to reactivate the catalyst by removing coke attached to the catalyst surface [8].

2. Thermodynamic Study of Catalytic Cracking Reaction

In this paper, the thermodynamic study of catalytic cracking reaction is based on an available theory of quantum chemical calculation. Quantum chemical calculation. is a method that is generally used in the field of chemical research, with high accuracy results for chemical reactions [9]. Quantum chemical calculations through Density Functional Theory (DFT) are used to estimate the thermodynamic reaction parameters of the refinery oil process, modeling of the structure of catalyst materials, investigating the mechanism of substance formation on the catalyst surface and the stage of forming transition parts (which describe the adsorption hydrocarbons in different natural catalysts) various pore geometries, etc.) [10]. The reference data shows that the value calculated by the DFT method is more reliable for the reaction of isoparaffin cracking, isomerization of paraffin, olefin, and aromatics cracking, and naphthene dealkylation. The molecular model created in the Gauss View software and presented in Figure 2 (as an example of the paraffin hydrocarbon cracking reaction).

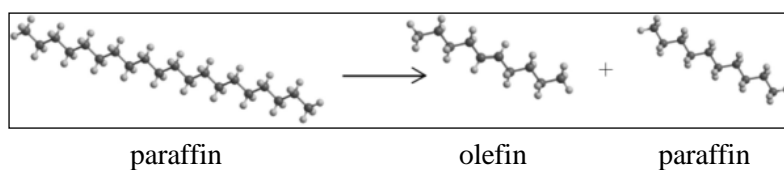


Fig. 2. The reaction of paraffin cracking Gauss View

Table 1 shows the thermodynamic parameters that have the possibility of catalytic cracking reactions of thermodynamics according to the results calculated using the DFT method. Paraffin reactivity in an endothermic cracking reaction is increased by increasing the molecular weight of n-paraffin. Paraffin reactivity in an endothermic cracking reaction is increased by increasing the molecular weight of n-paraffin. The value of Energy Gibbs change is $\Delta G = -42.8$ kJ/mol for cracking C₅-C₁₁ paraffin and $\Delta G = -77.4$ kJ/mol for cracking from C₁₄-C₄₀ paraffin. C₅-C₁₁+ isoparaffin cracking reaction from a thermodynamic point of view ($\Delta G = -56.7$ kJ/mol), then the normal hydrocarbon cracking structure is ($\Delta G = -42.8$ kJ/mol).

The endothermic reaction of dealkylation and cracking of naphthenes is characterized by high thermodynamic probabilities ($\Delta G = -145.5$ kJ/mol and $\Delta G = -186.5$ kJ/mol) comparison with dealkylation of aromatic hydrocarbons ($\Delta G = -91.6$ kJ/mol). At the same time, the dealkylation of aromatic hydrocarbons with less molecular weight (aromatic from the gasoline fraction) is ($\Delta G = -$

84.0 kJ/mol). The reaction of hydrogen transfers and aromatization ($\Delta G = -162.0$ kJ/mol and $\Delta G = -137.0$ kJ/mol) possible thermodynamics under process conditions and hydrogen transfer of the reaction are written through the next few steps. The aromatic hydrocarbon phase and the formation of isoparaffins from olefins and naphthene are the most likely thermodynamic, because of the naphthenic structure due to their high aroma [11].

Table 1. Thermodynamic Parameters of Catalytic Cracking Reaction

Reaction	ΔH , kJ/mol	ΔG , kJ/mol
Primary Cracking		
Cracking of n-paraffins C₁₄-C₄₀		
$C_{20}H_{44} \rightarrow n-C_{10}H_{22} + C_{10}H_{20}$	64.2	-77.4
$C_{20}H_{44} \rightarrow i-C_{10}H_{22} + C_{10}H_{20}$	64.8	-66.4
Naphthene Dealkylation		
$(C_{10}H_{21})_2-C_6H_4 \rightarrow C_6H_{10} + 2 C_{10}H_{20}$	142.4	-145.5
Aromatic Dealkylation		
$(C_{10}H_{21})_2-C_6H_4 \rightarrow C_6H_6 + 2 C_{10}H_{20}$	155.9	-91.6
Naphthene Cracking		
$(C_7H_{15})_2-C_{10}H_{16} \rightarrow C_8H_{17}-C_6H_5 + 2C_{10}H_{20} + 2H_2$	248.3	-186.5
Secondary Cracking		
Cracking of n-paraffins C₅-C₁₁₊		
$n-C_6H_{14} \rightarrow C_3H_6 + C_3H_8$	76.6	-42.8
Isomerization of n-paraffin C₅-C₁₁₊		
$n-C_7H_{16} \leftrightarrow i-C_7H_{16}$	-1.9	-2.4
Cracking of Isoparaffin C₅-C₁₁₊		
$i-C_6H_{14} \rightarrow i-C_4H_8 + C_2H_6$	70.1	-56.7
Cracking of Olefin C₅-C₁₁₊		
$C_7H_{14} \leftrightarrow C_2H_4 + C_5H_{10}$	93.4	-31.1
Hydrogen Transfer		
$C_6H_{10} + C_6H_{12} \leftrightarrow C_6H_6 + i-C_6H_{14}$	-85.5	-38.2
$C_6H_{10} + C_6H_{12} \rightarrow C_6H_6 + i-C_6H_{14}$	-169.7	-162.0
Condensation		
$C_2H_3-C_6H_5 + C_8H_{16} \rightarrow C_{10}H_6-(C_3H_7)_2 + 2.H_2$	-5.0	-77.3
$C_2H_3-C_6H_5 + C_8H_{16} \rightarrow C_{10}H_6-(C_3H_7)_2 + 2.H_2$	-28.7	-120.4
Coke		
$C_{10}H_8 \rightarrow 5.C_{24}H_{12} + 18.H_2$	114.0	-623.7

3. Brief Description of RCC Plant Process

Figure 3 shows the process scheme for the existing RCC units in Indonesia. Referring to Figure 3, the RCC feeds at stream 1, namely atmospheric residue (AR) and vacuum residue that will be contacted to the reactor, using lift steam and lift gas. Cracking reaction occurs in the reactor riser with operating conditions 485 °C - 540 °C (900 °F - 1000 °F) and pressures up to 100 psi [6].

A stream 2, the catalyst contacts with oil and accelerates the cracking reaction, besides the catalyst will provide heat to the hydrocarbons so that it accelerates the cracking reaction that occurs. The catalyst and hydrocarbons rise to the top of the riser with the help of a steam elevator and gas lift. After the saturation reaction occurs at the top of the riser (reactor) it will produce long-chain paraffin, the catalyst must be separated from the hydrocarbons to reduce the thermal cracking

process so that the hydrocarbon chains become smaller and eventually will cause the formation of coke. The product of the cracking reaction in the form of hydrocarbons will then separate from the catalyst and fall into the stripping section.

A stream 4, the results of the top product in the form of hydrocarbons will go out into the fractionation column to be separated. Separating hydrocarbons into overhead vapor, which will produce a light fraction fuel in the form of naphtha, LCO, HCO, and DCO, based on its boiling point. A stream 5, take advantage of the material to be refluxed and separated again, while stream 6 will produce a light oil fraction in the form of naphtha [12].

A stream 7, the catalyst collected in the stripper will be flowed to the top of the regenerator with the aim to reactivate the catalyst by removing coke attached to the catalyst surface and regenerated by combustion of coke attached to the catalyst by contacting stream 8 in the form of air. In the upper regenerator, partial combustion occurs where the coke will be burned to produce CO to CO₂. A stream 9, the CO-containing fuel gas exits the upper regenerator through the cyclone to be separated from the remnants of the catalyst [8].

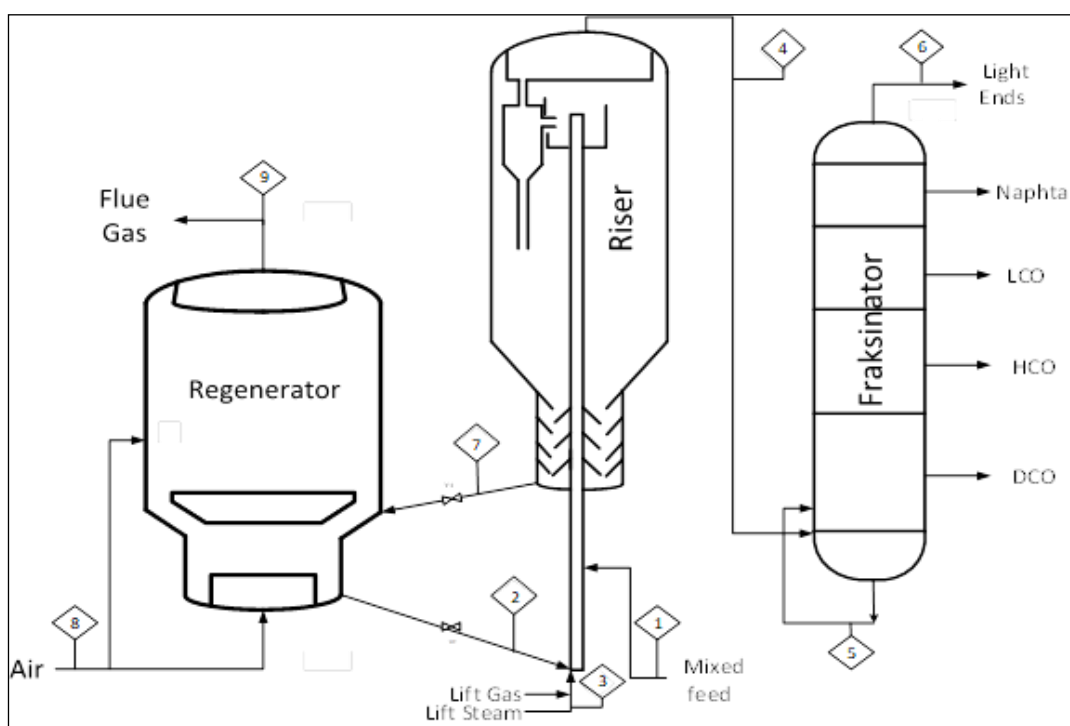


Fig. 3. RCC Plant Unit

4. The Catalyst of Residue Catalytic Cracking

One of the challenges in choosing a catalyst is that the metal contained in the feed is continuously deposited on the catalyst, causing the deactivation of the catalyst. To prevent deactivation, the catalyst must have a high tolerance to deposited metals. Another challenge is the high level of coke formed by high boiling residual feeds, so the regenerator must be well designed and the catalyst must have high hydrothermal stability and the ability to withstand high regenerator temperatures [13]. Therefore, catalysts that have suitable activities, high gasoline selectivity, and catalyst stability are very important in this process [14, 15]. The catalyst used is usually in the form of microspheres, spheres, and other performed forms [7].

The catalyst used in the RCC process generally uses zeolite catalysts [5], synthetic silica-alumina, natural composites or aluminosilicates, and silica aluminate [6]. But most of the commercial catalytic cracking process is carried out using silica alumina and zeolite catalysts type [7]. Even so, research on catalysts in the process of catalytic cracking residues continues to grow until now such as the development of zeolite catalysts [5, 16], catalysts of metal oxide nanoparticles [17], zirconia catalysts impregnated in red mud [18, 19], and rare earth catalyst [20, 21]. Table 2

shows the variates of catalysts and feeds for the RCC unit equipped with the operation condition and conversion value.

Table 2. Residue Catalytic Cracking Catalyst

Catalyst	T	P	Feed	Reactor	Conversion	Reference
E-Cat/ZSM-5	650 °C	N/A	Crude Oil	Fixed-Bed MAT Unit	60.8%	[22]
ZrO ₂ /MaRM	500 °C	Ambeint Atmospheric Pressure	Vacuum Residue	Fixed-Bed Reactor	79.6%	[19]
Ni/K	440 °C	300 psig	Deasphalted Vacuum Residue	Bench-Scale Pilot Plant Equipped with an Up-Flow Open Tubular Reactor	51.4%	[23]
NiK/MaAl ₂ O ₃	500 °C	N/A	Vacuum Residue	Fixed-Bed Reactor	79.37%	[24]
NiK/10Ce _x Zr1-xO ₂ -MaAl ₂ O ₃					78.66%	
Ni/Activated Carbon	400 °C	30,1 MPa	Oil Sand Bitumen	Batch Auto Clave Reactor	97.7%	[25]
ZrO ₂ /ARM	470 °C	3 MPa	Vacuum Residue	Batch Type Autoclave Fitted with a Stirrer	85.2%	[18]
Fe-Char	800 °C	N/A	Oily Sludge	Two Stage Fixed-Bed Reactor	95.8%	[26]
NiK/CeZr-Al	500 °C	Atmospheric Pressure	Vacuum Residue	Fixed-Bed Reactor	88.7%	[27]
USY/ZSM-5	500 °C	N/A	Residue Oil	Fixed Fluid Bed	76.11%	[28]
Sandstone based drill (Silica Alumina)	420 °C	Atmospheric Pressure	Vacuum Residue	Batch Parr Reactor	77%	[29]
			Atmospheric Residue		62%	
Silica Alumina/ β Zeolite	500 °C	0,45 MPa	Vacuum Gas Oil	Curie Point Pyrolyzer	46%	[30]
Ca/Al	650 °C	N/A	Vacuum Residue	Fluidized Bed Reactor	97.4%	[31]

5. Catalyst Regenerator Technology

Table 3 shows the methods of regeneration process equipped with gas composition. In the catalyst regeneration of the RCC process that occurs in the regenerator, the aim is to reactivate the catalyst by removing coke attached to the surface of the catalyst, and regenerated by burning coke attached to the catalyst [32]. Conventionally, the spent FCC catalyst is reactivated via coke combustion in the fluidized bed regenerator. It will generate excessive heat in the system because of its high coke yield when treating heavy oil. External catalyst cooler or boiler should be applied to extract the heat and maintain the heat balance of the operation, which leads to a great waste of carbon resources and high SO_x and NO_x emissions during coke combustion. On the other hand,

hydrogen is usually of great deficiency in the refinery, especially processing heavy oil into light oil products [33]. Based on Table 3 about the comparison of output gas composition in regeneration technology, the Oxy-Combustion method is profitable in the catalyst regeneration process. The basic concept is to use oxygen to burn coke contained in the catalyst which will produce lower emissions, and higher efficiency and produces high CO₂ flow [34]. The feasibility of an oxy-combustion step for the catalyst regeneration has been studied with reference to a spent catalyst due to carbon deposition during the methane thermo-catalytic decomposition [35].

Table 3. Gas Composition of Regeneration Process

Method	Gas Composition	% Mol	Reference
Oxy – Combustion	CO ₂	94.5	[36, 37]
	O ₂	3.1	
	N ₂	2.1	
	CO	0.01	
Multi-Phase Particle-In-Cell	CO ₂	12	[38, 39]
	O ₂	4.4	
	N ₂	75.3	
	CO	0.69	
FCC Alliance R2R Resid FCC unit	CO ₂	7.2	[40]
	O ₂	8.6	
	N ₂	74.8	
	CO	3.6	

6. Conclusion

One of fuel processing technology, namely the RCC unit, RCC unit is secondary processing that converts heavy fraction hydrocarbons into light fraction hydrocarbons with the help of the use of catalysts into high-value petroleum products such as naphtha, LPG, and Light Cycle Oil. The RCC unit has three main parts for this process, namely the riser, stripper, and regenerator. Based on the review of catalyst development in the RCC unit in this paper, important considerations for choosing a catalyst consists of catalysts must have a high tolerance to deposited metals, have high hydrothermal stability, suitable activities, high gasoline selectivity, and catalyst stability. Ca/Al type Catalyst with Vacuum Residue feed on Fluidized Bed Reactor produces 97.4% conversion. Ca/Al type catalyst the most match for RCC unit, cause use feed from vacuum residue and have the high conversion although greater than Ni/Activated Carbon. Ni/Activated Carbon Type catalyst with oil sand Bitumen Clave feed on Auto Clave Reactor Batch Reactor produces 97.7% conversion. The two types of catalysts are suitable for the RCC unit because they have the highest conversion with different feeds.

In addition, regeneration technology should be made based on the comparison of the output gas composition, and then the Oxy-Combustion method is profitable in the regeneration process of the catalyst because it uses oxygen to burn coke which will produce lower emissions, and higher efficiency and produces high CO₂ flow

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