

Investigating the effect of additive ZSM-5 ratio on propylene production via simulation method at the catalytic cracking unit of Dung Quat Refinery

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ABSTRACT

The Dung Quat refinery is exploring solutions to increase the yields of light olefins, especially propylene, to meet the rising demand for petrochemical industry materials, specifically for the Polypropylene plant. This study also investigates the yields of other products, such as gasoline and LCO. Although various methods can be used to address this issue, such as changing operating parameters or using catalysts with high Si/Al ratios, adjusting the ZSM-5 additive ratio has been found to be the most effective and flexible approach. Because an experimental study of this kind would require a Residue Fluid Catalytic Cracking (RFCC) pilot plant and would be both time-consuming and expensive, a steady-state model of the R2R (Reactor - two Regenerators) configuration of the RFCC unit at Dung Quat refinery was developed in Aspen Hysys, using the built-in FCC module together with the Peng-Robinson fluid package. The model was set up for the atmospheric residue of Bach Ho crude oil and was calibrated against plant operating data: the deviations in the flow rates of light ends, naphtha, LCO and bottoms, as well as in the riser outlet temperature, all remained below 5.5%, which confirms that the model is reliable for parametric investigation. The ZSM-5 content in the catalyst inventory was then varied from 0 to 20 wt% and the resulting product yield structure was analysed. The simulation results from the Hysys program indicate that adding less than 2.5% ZSM-5 additive can make the mass flow of LPG in the outlet gas stream increase by nearly 7% higher than initial capacity without changing the configuration of the related unit in the refinery. By adding 20% ZSM-5 additive leads to nearly triple the propylene yields and increase the yield of LPG to 31.15 wt%. However, gasoline yields decreased by 15 wt%. The gasoline composition was also analyzed, revealing that the amount of paraffins and olefins decreased while cyclic components, such as naphthene and aromatics, increased. Furthermore, the addition of ZSM-5 additive increased olefin branching, improving gasoline quality with a higher octane number. Quantitatively, the propylene yield rises from 3.80 wt% to 10.74 wt% and the octane number of the cracked gasoline increases from 89.82 to 98.64 when the ZSM-5 content is raised from 0 to 20 wt%. These results provide a quantitative basis for selecting a suitable ZSM-5 dosing strategy at Dung Quat refinery, balancing the propylene demand of the downstream Polypropylene plant against the loss of gasoline yield and the capacity limit of the Propylene Recovery Unit.

Key words: RFCC, ZSM-5, additive, propylene

INTRODUCTION

Demand of Propylene

Propylene serves as a crucial primary ingredient in petrochemicals, playing a vital role in the production of various derivatives like propylene oxide, acrylonitrile, and polypropylene. The utilization of propylene¹ is visually represented in Figure 1.

Among the end products derived from propylene, polypropylene (PP) holds the largest share, accounting for 71.1% of the total, thereby driving the demand for polypropylene as a key determining factor. In Vietnam, industries such as packaging and medical device manufacturing heavily rely on PP. Consequently, refineries like Dung Quat are actively seeking

solutions to enhance the production of light olefins, particularly propylene. Various methods have been proposed to address this challenge, including adjustments to operating parameters and the utilization of catalysts with high Si/Al ratios. However, none have proven to be as versatile and efficient as the implementation of ZSM-5 additive in RFCC catalysts.

RFCC unit at Dung Quat refinery

The Residue Fluid Catalytic Cracking (RFCC) unit at Dung Quat refinery is specifically designed to process two types of crude oil residues obtained from the atmospheric distillation of Bach Ho crude oil and the Bach Ho/Dubai oil mixture.

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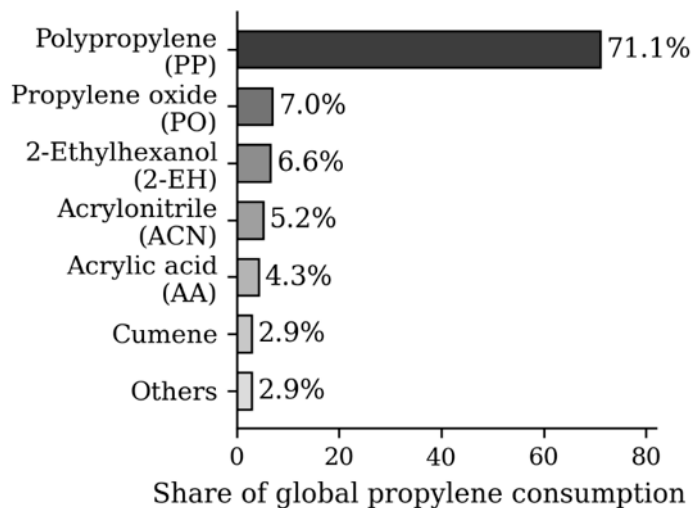


Figure 1: Distribution of propylene in various industries [Source: The authors, drawn from data in ¹]

With a design capacity of 3,256,000 tons of Bach Ho crude oil distillate residue per year, equivalent to a volumetric flow rate of 69,700 BPSD², the RFCC operates for 8,000 hours annually. The unit is also equipped for residue treatment resulting from the sour crude oil mixture processed in the Crude Distillation unit (CDU), with a ratio of 1 million tons of Dubai crude oil to 5.5 million tons of Bach Ho crude oil. The capacity for blending sour residue is also identical to the design capacity when processing residue from Bach Ho crude oil. The RFCC unit operates in two modes, catering to both Bach Ho oil residue and Bach Ho/Dubai mixed residue: RFCC Naphtha maximum or LCO maximum. Additionally, the RFCC is designed to directly handle 100% of the hot feed from the direct distillation and accommodate up to 100% of the cold supply from storage. The RFCC gas plant is also capable of processing streams such as off-gas flow from the rear crude distillation unit and LPG-rich gas flow from the CDU.

Originally, the plant's design focused on stable operation using Bach Ho oil or the Bach Ho/Dubai oil mixture, which are rich in paraffin and have low impurities. Therefore, the Vacuum Distillation Unit (VDU), which treats residue from the CDU, was not initially included in the design. Instead, all residue is treated within the RFCC unit. The unique feature of this unit lies in its utilization of two Regenerators, as opposed to a single Regenerator commonly found in FCC configurations. Consequently, it is referred to as R2R (Reactor-2-Regenerator). Diagram in Figure 2³ illustrates its configuration.

The RFCC's feedstock consists of residue from the CDU, which contains various metal impurities, particularly Ni, Na, and V. In the first stage, only 30-70% of the coke and most of the hydrogen within the coke are burned. While steam is produced, incomplete combustion prevents the formation of V₂O₅ and consequently avoids the production of Vanadic acid, thereby preventing catalyst toxicity. The remaining coke is fully burned in the second regenerator. However, the atmosphere is dry³, meaning no water vapor is generated. Although V₂O₅ is produced due to high temperatures, there is no available water for the production of Vanadic acid. As a result, the catalyst remains unpoisoned.

The unit currently employs Zeolite Y as the catalyst, but there is a possibility of enhancing the production of light olefins by incorporating ZSM-5 into the mix.

Role of ZSM-5 additive in RFCC

ZSM-5 is invented by Mobil Oil in 1972 with formula: $\text{NaAlSiO}_96\text{-nO}192 \cdot \sim 16\text{H}_2\text{O}$ ($\text{n} < 27$). ZSM-5 has a basic orthorhombic crystal structure, belonging to the Pnma symmetry group with the corresponding side lengths: $a=20.1 \text{ \AA}$, $b=19.9 \text{ \AA}$, $c=13.4 \text{ \AA}$. The lattice of ZSM-5 contains TO₄ tetrahedra where T is the element in the center of the tetrahedron, which can be Al or Si. The tetrahedra are linked together into a 5-sided SBU (secondary unit building) structural unit: 5-1. These SBUs are connected to each other to create two intersecting capillary channels, with a 10-sided capillary window.

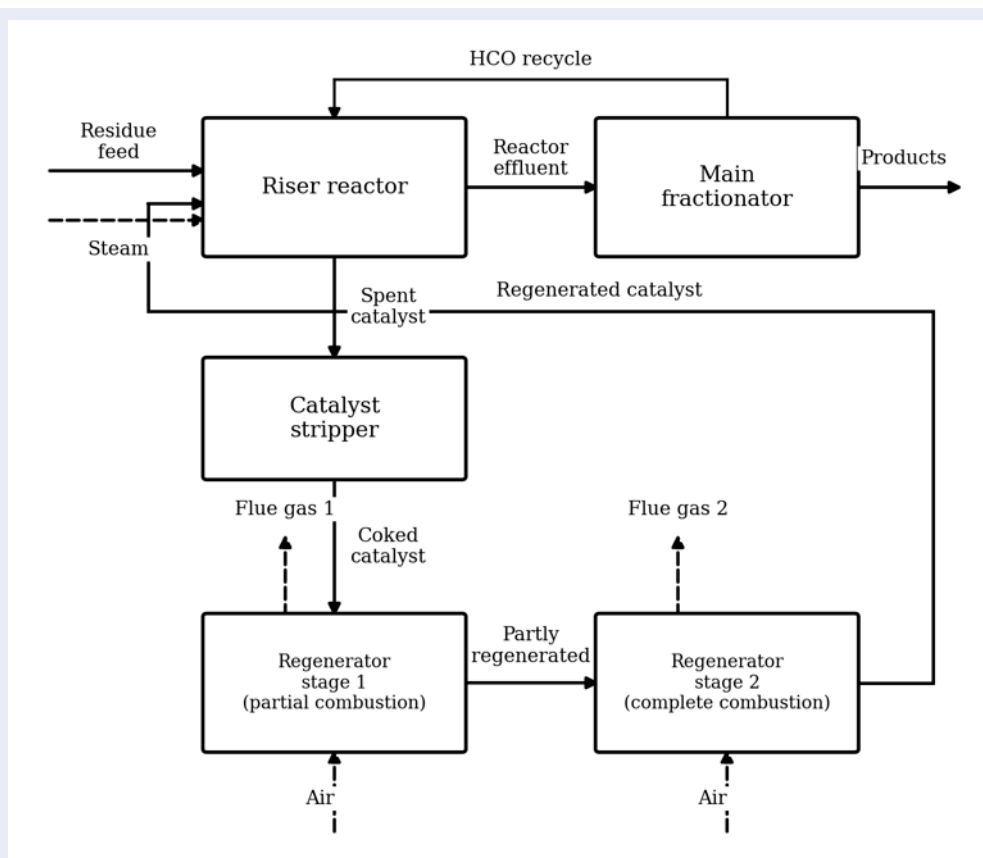


Figure 2: Diagram of R2R [Source: The authors, adapted from ³]

A capillary channel system parallel to the a-axis of the orthorhombic crystal, has a zigzag shape with a nearly circular window size (5.4 – 5.6 Å). Another channel system, straight and parallel to the b-axis, has the form of an elliptical window (5.1 – 5.7 Å). These two channel systems intersect to create a large cavity of size 9 Å and are the settlement of strong acid centers that characterize the catalytic activity of ZSM-5 zeolite. Figure 3. illustrates the capillary system of ZSM-5.

The critical factor for enhancing propylene yield and the octane number of gasoline, when used as additives with FCC catalyst, lies in shape selectivity. This desirable attribute stems from the unique capability of ZSM-5 to control the size and shape of molecules as they diffuse within the capillary channels, thereby maximizing efficiency.

The shape selectivity can be described in detail as follows: a) according to the reactant, that is, to remove the molecules participating in the reaction whose size is larger than the capillary size of the zeolite; b) by reaction product, that is, only the reaction product molecules formed must be smaller than the zeolite

pore size to escape (diffuse) out of the zeolite crystal; c) in the intermediate state, i.e. reactions that take place through bulky intermediate states, having dimensions larger than the capillary size, do not occur, only reactions occurring through small-size intermediate state.

Based on the capillary channel size of the FCC catalyst (0.74 nm), ZSM-5 (0.53 nm) and the kinetic diameter distribution diagram, shown in Figure 4⁴, it is clear that only straight-chain and branched-chain hydrocarbons can enter the capillary channels of ZSM-5 easily.

The straight chain olefins from C7 to C10 will be broken down by ZSM-5 to light olefins (C3, C4 and C5) and isomerize the straight chain olefins to branch olefins with higher octane number. As a result, the olefin content of which is mainly propylene and the octane number of the gasoline product will be significantly increased.

On the other hand, the Si/Al ratio of ZSM-5 is much larger than that of conventional Y zeolite, ZSM-5 crystals are often synthesized with Si/Al ratios from

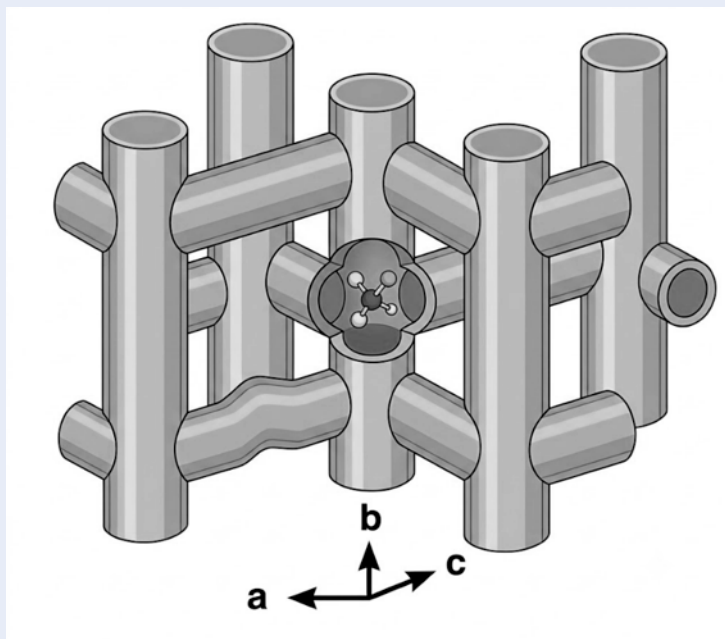


Figure 3: Capillary system of ZSM-5 [Source: The authors]

Pore diameter / kinetic diameter of molecules (nm)

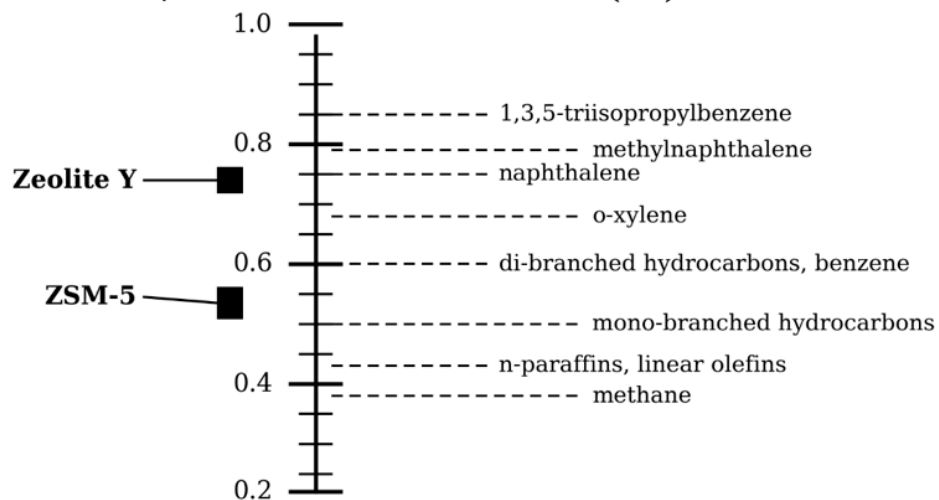


Figure 4: Comparison of capillary channels between Zeolite and ZSM-5 [Source: The authors, adapted from⁴]

20 to 50⁵. The Si-O bond is stronger than the Al-O bond, so when the Si/Al ratio increases, the zeolite crystal structure is more stable under the influence of temperature and steam⁶. Therefore, ZSM-5 has higher thermal and hydrothermal stability than conventional FCC catalysts (containing mostly Y zeolite). It is also because of the large Si/Al ratio that the number of acid aluminum centers is significantly reduced, along with the small pore size, it is possible that the ZSM-5 additive will have the effect of reducing the Hydrogen transfer reaction, the amphiphilic reaction⁷. Therefore, it is possible to limit the amount of paraffin in gasoline and increase the olefin content in LPG.

The implementation of experiments to investigate the effects of ZSM-5 additive on product yield structure at RFCC unit in reality is complicated. It requires building a RFCC pilot plant including small reactor/regenerator, distillation column and other necessary auxiliary equipments which would be time-consuming and even expensive. However, Hysys software with available FCC module as well as integrated ZSM-5 can help with this. The additive concentration in catalyst is changed from 0-20wt% in this study.

BUILDING A SIMULATION MODEL OF RFCC USING ZSM-5

The simulation of RFCC unit at Dung Quat refinery is carried out by using Hysys software version 12.1, which includes an FCC module containing a readily available reactor/regenerator and other necessary equipments. The calculation formulas for this process have been configured in Hysys, and the results will be instantly available once the simulation converges. Fluid package used in this simulation model is Peng-Robinson. Figure 5. illustrates the simulation model diagram.

Materials

This study utilizes the crude oil residue extracted from Bach Ho oil. The characteristics of the feed oil⁸ are outlined in Table 1.

Bach Ho crude oil is a light and sweet oil with high API gravity of 30.09 and low purities. Specifically, residue from this oil contains 0.062 wt% of sulfur and 0.29 ppmwt of vanadium. Besides, the flowrate of feed stream is 461.7 m3/h with temperature of 290°C and pressure at reactor is 1.427 kg/cm2g. All these parameters are essential in order to describe the feed stream in simulation.

Table 2: Composition of catalyst [Source:⁸]

Composition	Unit	Value
Zeolite	wt%	48
Alumina	wt%	50
Rare Earth	wt%	1.14

Catalyst

The percentages of components in RFCC catalyst are shown in Table 2. below.

Besides, there are many catalytic coefficients for each component presented in the attached simulation file.

Simulation of R2R

The R2R configuration comprises a single riser and a two-stage regenerator, with separate flue gas handling. A stream of HCO (heavy cycle oil) will be reintroduced into the riser for recycling purposes. The dimensions of the reactor and regenerator⁹ can be found in Table 3.

Table 3: Geometric parameters of R2R [Source:⁹]

Specifications	Value	
Riser		
Total Length (m)	43.7	
Diameter (m)	1.6	
Riser Termination Zone		
Length (m)	1	
Diameter (m)	4.371	
Catalyst stripper		
Height (m)	8	
Diameter (m)	3	
Annulus Diameter (m)	1.3	
Regenerator		
	Stage 1	Stage 2
Dense bed Height (m)	4.572	4.572
Dense bed Diameter (m)	8.9	8.0
Dillute Phase Diameter (m)	10.9	10
Interface Diameter (m)	10.9	10
Cyclone Inlet Height (m)	18.5	18.5
Cyclone Inlet Diameter (m)	2.286	2.286
Cyclone Outlet Diameter (m)	1.219	1.219

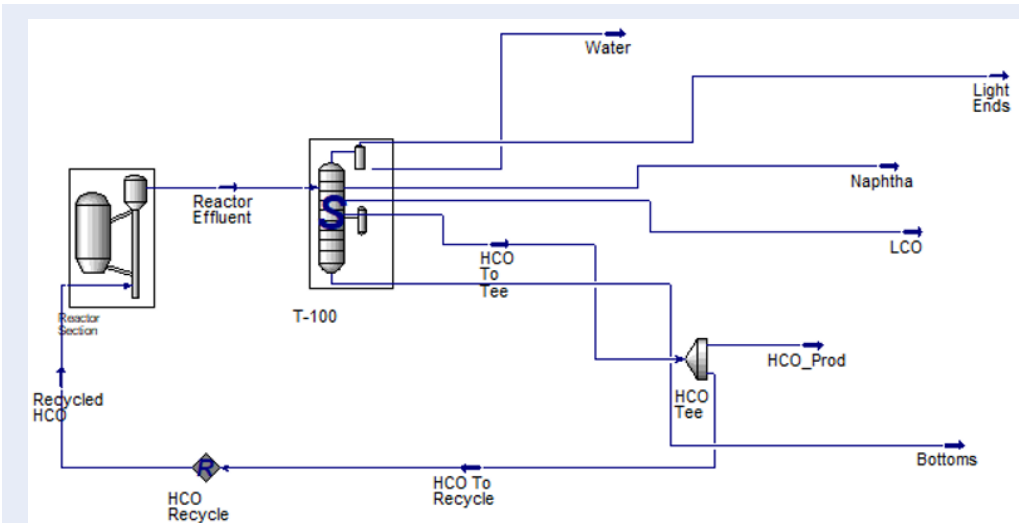


Figure 5: RFCC simulation model diagram [Source: The authors, exported from Aspen Hysys]

Table 1: Properties of feed oil [Source: 8]

Feature	Unit	Value
Specific Gravity 60F/60F	-	0.8757
°API	-	30.09
Conradson Carbon Residue	wt%	1.8
Total Nitrogen	ppmwt	1434
Sulfur Content	wt%	0.062
Vanadium	ppmwt	0.29
Nickel	ppmwt	4.02
5% TBP	°C	340.2
50% TBP	°C	469.4
90% TBP	°C	610.9
Operating parameters		
Volumetric Flowrate	m3/h	461.7
Temperature	°C	290
Pressure	kg/cm2g	1.427

Accuracy of simulation model

Once the simulation model reached convergence, the objective was to compare the simulation results with the real operating conditions of the RFCC unit. This involved analyzing the flow rates of various products such as light ends, naphtha, LCO (Light Cycle Oil), and Bottoms. To ensure the accuracy of the technological parameters, the author adjusted the simulation data to closely match the actual operating parameters. Table 4 presents a comparison between the actual and

simulated data for evaluation. The differences between extracted streams as well as reaction temperature compared to plant data were low and reliable for investigation when changing the ratio of ZSM-5 in the catalyst afterwards.

RESULTS AND DISCUSSION

Table 5 provides an overview of the impact of ZSM-5 addition on the product yield structure. Figures 6, 7, 8, 9, 10, 11 and 12 depict the trends in product yield

Table 4: The difference between simulation and reality [Source: The authors, from Aspen Hysys and plant data ^{8]}

Parameters		Light Ends	Naphtha	LCO	Bottoms
Extracted flow rate, 103 kg/h	Simulation	236.3	21.28	84.65	1.236
	Actual data	250	21.54	88.69	1.293
	Difference (%)	5.48	1.2	4.55	4.41
ROT, °C	Simulation		491.6		
	Actual data		503		
	Difference (%)		2.27		

changes as the proportion of ZSM-5 additives in the catalyst increases.

The findings demonstrate a notable rise in propylene yield, increasing from 3.8wt% to 10.74wt% when using 20wt% ZSM-5 additive. The observed increase in propylene is attributed not only to the cracking of light olefins within the ZSM-5 capillaries but also to the higher Si/Al ratio of the ZSM-5 additive compared to the base catalyst. The higher Si/Al ratio leads to a reduction in the number of acid centers, which significantly decreases the Hydrogen transfer reaction, ultimately boosting the content of propylene.

The LPG yield also experiences a significant increase, rising by 14wt% to 31.15wt% with the addition of 20wt% ZSM-5 in the catalyst. This can be attributed to the shape selectivity of ZSM-5¹⁰. Specifically, straight-chain olefins in the C6-C10 range present in gasoline undergo cracking, transforming into C3 and C4 straight-chain olefins, consequently boosting the LPG yield. Additionally, when incorporating a 2.5wt% ZSM-5 additive, the mass flow of LPG reaches 82,610 kg/h, representing an almost 7% increase compared to the initial capacity of the Propylene Recovery Unit (PRU). Despite this substantial increase, the unit is capable of handling the additional LPG flow effectively.

Conversely, gasoline production experiences a decline of 15wt% to 38.41wt% when the ZSM-5 additive is introduced at 20wt%. The addition of ZSM-5 reduces the ratio of the base catalyst used, leading to a decrease in pre-cracking reactions of larger residue hydrocarbons. As a result, there is a shortage of raw materials available for the cracking process to produce components for the gasoline fraction. Furthermore, due to the smaller capillary size of ZSM-5 compared to the base catalyst, newly formed C5+ components in the base catalyst's capillary, as well as those already present in the material, enter the ZSM-5 capillary and continue to crack into C3 and C4. Consequently, this causes a loss of constituents intended for the gasoline

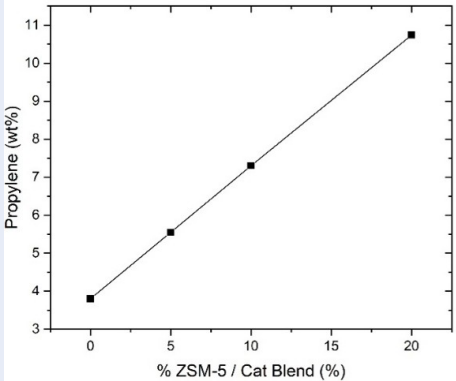


Figure 6: Effect of ZSM-5 additive on the yield of propylene [Source: The authors]

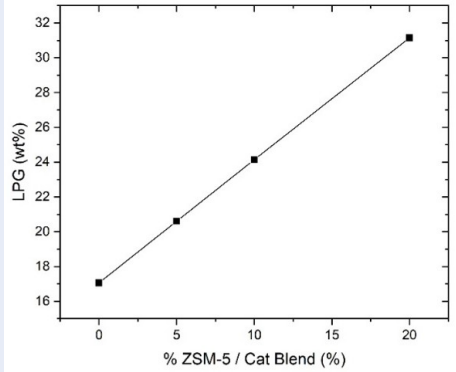


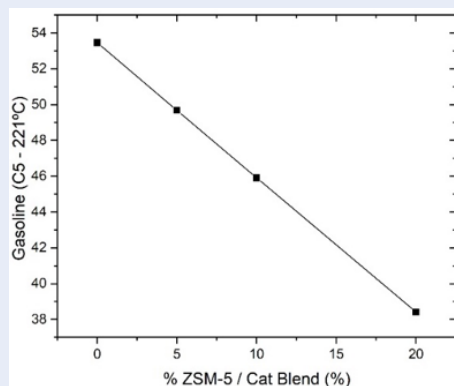
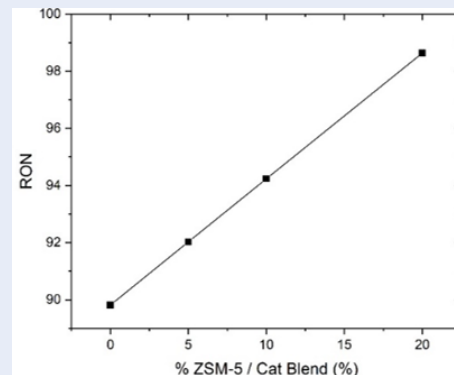
Figure 7: Effect of ZSM-5 additive on the yield of LPG [Source: The authors]

fraction, resulting in a reduction of the final gasoline content.

The rise in the octane number of gasoline with increasing ZSM-5 additive is advantageous for blending with lower octane number gasoline sources from other units at the refinery. When long-chain olefins

Table 5: Product yield at 0-20wt% additive [Source: The authors, from Aspen Hysys]

	Additive content			
	0	5.0	10.0	20.0
Catalyst/oil ratio	9.078	9.172	9.26	9.422
Product yields (wt%)				
Dry gas	2.69	2.71	2.73	2.77
LPG	17.06	20.61	24.14	31.15
Propylene	3.8	5.55	7.3	10.74
Gasoline (C5 - 221°C)	53.46	49.68	45.91	38.41
Paraffins in gasoline (vol%)	63.21	61.34	59.46	55.7
Olefins in gasoline (vol%)	28.92	29.1	29.2	29.48
Olefins in gasoline (wt%)	28.43	28.4	28.33	27.93
Naphthene in gasoline (vol%)	5.373	5.6	5.81	6.25
Aromatics in gasoline (vol%)	2.498	4.2	5.54	8.57
Octane number	89.82	92.03	94.24	98.64
LCO (221-343°C)	12.48	12.46	12.44	12.39
Bottoms (+343 °C)	8.05	8.28	8.53	9.02

**Figure 8:** Effect of ZSM-5 additive on the yield of gasoline [Source: The authors]**Figure 9:** Effect of ZSM-5 additive to Octane number [Source: The authors]

enter the ZSM-5 capillary, they undergo cracking, resulting in the formation of short-chain olefins. Moreover, these long-chain olefins also experience isomerization, leading to the creation of multi-branched olefins, which contributes to the improved octane number of the gasoline. Furthermore, ZSM-5 contains numerous Bronsted acid sites¹¹ that facilitate the aromatization of olefins, leading to the generation of gasoline fractions rich in aromatics. This aromatic-rich gasoline fraction further adds to the overall increase in the octane number of the gasoline product.

The investigation into the distribution of PONA composition in gasoline reveals notable changes. The volume percent of paraffins decreases, whereas the volume percent of naphthenes and aromatics increases. As for olefins, their weight percent decreases as they are cracked into smaller olefins. However, despite the decrease in weight percent, the volume percent of olefins in gasoline actually increases.

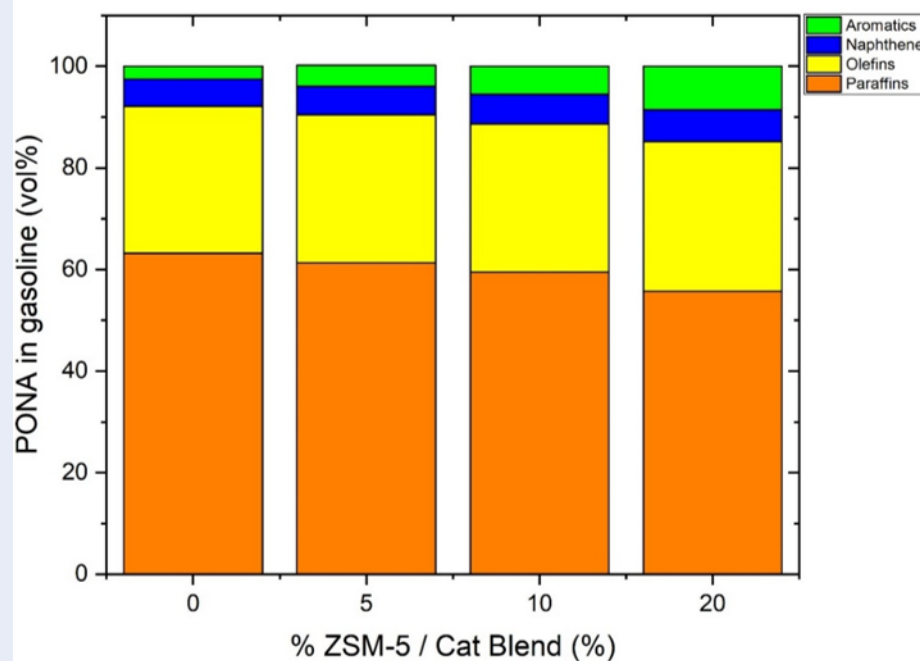


Figure 10: Distribution of PONA composition in gasoline [Source: The authors]

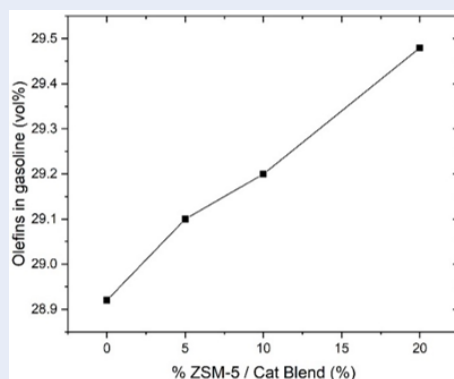


Figure 11: Effect of ZSM-5 additive on the volume percent of olefins in gasoline [Source:The authors]

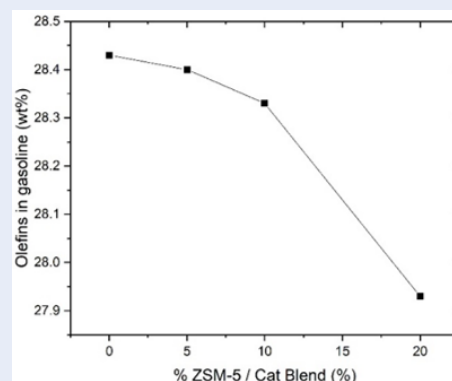


Figure 12: Effect of ZSM-5 additive on the weight percent of olefins in gasoline [Source:The authors]

CONCLUSION

The investigation into the effects of ZSM-5 additive on product yield distribution through RFCC simulation at Dung Quat refinery demonstrates that incorporating ZSM-5 is a highly effective method to increase the yields of light olefins, especially propylene. With the addition of 20wt% ZSM-5, the propylene yield significantly rises to 10.74wt%, almost three times higher than without the additive. This also leads to an increase in LPG yield, reaching 31.15wt%. At 2.5wt%

ZSM-5, the mass flow of LPG is expected to be 82,610 kg/h, which is nearly 7% higher than the initial capacity of PRU, without impacting its configuration. However, one trade-off observed is a decrease in gasoline yield by 15wt%. The gasoline's composition undergoes changes, with a decrease in paraffin content and an increase in cyclic components such as naphthenes and aromatics. Interestingly, the volume percent of olefins in gasoline rises, while the weight percent experiences the opposite trend. Furthermore, the addition of ZSM-5 enhances gasoline quality through iso-

merization, resulting in a higher octane number.

LIST OF ABBREVIATIONS

API American Petroleum Institute gravity
BPSD Barrels Per Stream Day
CDU Crude Distillation Unit
FCC Fluid Catalytic Cracking
HCO Heavy Cycle Oil
LCO Light Cycle Oil
LPG Liquefied Petroleum Gas
PONA Paraffins, Olefins, Naphthenes and Aromatics
 Polypropylene
PRU Propylene Recovery Unit
R2R Reactor - two Regenerators configuration
RFCC Residue Fluid Catalytic Cracking
ROT Riser Outlet Temperature
SBU Secondary Building Unit
TBP True Boiling Point
VDU Vacuum Distillation Unit
ZSM-5 Zeolite Socony Mobil - 5
vol% percent by volume
wt% percent by weight

CONFLICT OF INTEREST

The authors declare that there is no conflict of interest regarding the publication of this article.

AUTHOR CONTRIBUTION

Phuc Hong Nguyen: built the RFCC simulation model in Aspen Hysys, carried out the simulation runs at different ZSM-5 additive ratios, processed the data and prepared the first draft of the manuscript.

Khang Vi Chau Ngo: collected the design and plant operating data of the RFCC unit, validated the simulation model against plant data, prepared the figures and tables.

Trung Chi Duong: proposed the research idea, supervised the work, interpreted and discussed the results, revised and finalised the manuscript, and acted as corresponding author.

All authors have read and approved the final version of the manuscript.

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Nghiên cứu ảnh hưởng của tỷ lệ phụ gia ZSM-5 đến sản lượng Propylene thông qua phương pháp mô phỏng tại phân xưởng Cracking xúc tác Nhà máy lọc dầu Dung Quất

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TÓM TẮT

Nhà máy lọc dầu Dung Quất đang nghiên cứu các giải pháp nhằm nâng cao hiệu suất sản xuất olefin nhẹ, đặc biệt là propylene, nhằm đáp ứng nhu cầu ngày càng tăng về nguyên liệu công nghiệp hóa dầu, đặc biệt cho nhà máy Polypropylene. Nghiên cứu này cũng đánh giá sản lượng của các sản phẩm khác, chẳng hạn như xăng và LCO. Mặc dù có thể sử dụng nhiều phương pháp khác nhau để giải quyết vấn đề này, chẳng hạn như thay đổi thông số vận hành hoặc sử dụng chất xúc tác có tỷ lệ Si/Al cao, việc điều chỉnh tỷ lệ phụ gia ZSM-5 được cho là phương pháp hiệu quả và linh hoạt nhất. Do việc triển khai thực nghiệm đòi hỏi phải xây dựng hệ pilot RFCC, vừa tốn kém vừa mất nhiều thời gian, nghiên cứu này xây dựng mô hình mô phỏng ở trạng thái dừng cho cấu hình R2R (một thiết bị phản ứng - hai thiết bị tái sinh) của phân xưởng RFCC Nhà máy lọc dầu Dung Quất, sử dụng module FCC có sẵn và hệ nhiệt động Peng-Robinson. Mô hình được hiệu chỉnh theo số liệu vận hành thực tế với nguyên liệu là cặn chưng cất khí quyển dầu thô Bạch Hổ: sai lệch về lưu lượng các dòng khí nhẹ, naphtha, LCO, cặn đáy và nhiệt độ đầu ra ống phản ứng đều dưới 5,5%, cho thấy mô hình đủ tin cậy để khảo sát. Hàm lượng ZSM-5 trong xúc tác được thay đổi từ 0 đến 20% khối lượng. Kết quả mô phỏng cho thấy việc bổ sung ít hơn 2,5% phụ gia ZSM-5 có thể làm cho lưu lượng khối LPG trong dòng khí đầu ra tăng cao hơn gần 7% so với công suất ban đầu mà không làm thay đổi cấu hình của các thiết bị liên quan trong nhà máy lọc dầu. Bằng cách thêm 20% phụ gia ZSM-5 dẫn đến sản lượng propylene tăng gần gấp ba lần và tăng sản lượng LPG lên 31,15% khối lượng. Tuy nhiên, sản lượng xăng giảm 15% khối lượng. Thành phần xăng cũng được phân tích, cho thấy lượng parafin và olefin giảm trong khi các thành phần có vòng, chẳng hạn như naphthen và chất thơm, lại tăng lên. Hơn nữa, việc bổ sung phụ gia ZSM-5 làm tăng khả năng phân nhánh olefin, cải thiện chất lượng xăng có trị số octan cao hơn. Cụ thể, hiệu suất propylene tăng từ 3,80% lên 10,74% khối lượng và trị số octan của xăng tăng từ 89,82 lên 98,64. Kết quả này là cơ sở định lượng để lựa chọn tỷ lệ phụ gia ZSM-5 phù hợp cho Nhà máy lọc dầu Dung Quất, cân đối giữa nhu cầu propylene của nhà máy Polypropylene với mức giảm hiệu suất xăng và giới hạn công suất của cụm thu hồi propylene (PRU).

Từ khoá: RFCC, ZSM-5, phụ gia, propylene

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