Effect of Hydrocarbon Partial Pressure on Propylene Production in the FCC

Effective management of hydrocarbon (HC) partial pressure is crucial for many FCC units operating at high feed rate while targeting high octane gasoline and petrochemical yields. In the article "Effect of Hydrocarbon Partial Pressure on Propylene Production in the FCC," published in *Catalagram* No. 103 in 2008, results of comprehensive laboratory work are discussed to better understand this relationship. There is a clear correlation between HC partial pressure and hydrogen transfer (HT), explained by the increase of bimolecular reaction rate relative to monomolecular reactions such as cracking. This higher HT has a detrimental impact on gasoline octane and LPG olefin yield, as well as the efficiency of ZSM-5 based additives. The results of this study help to build more realistic simulation models for optimizing both operating conditions and catalyst technologies. High-activity ZSM-5 additives like ZAVANTI™ enables refiners to overcome high HC partial pressure and maintain required propylene yields. Grace also has a wide portfolio of ultra-low HT catalysts that provide improved high unit HC partial pressure management. Ask your Grace representative how to extract the most value of your unit's operation.

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Many refiners have continually revamped and debottlenecked their FCC units to increase feed throughput and improve profitability Most FCC units are running at a significantly higher feed rate than the original design. With higher throughput, in order to maintain catalyst and vapor velocity in the riser and cyclones, the unit pressure and consequently the hydrocarbon partial pressure need to be increased. Current laboratory methods for evaluating FCC catalysts and additives cannot match hydrocarbon partial pressures in commercial FCC units. One reason is that available laboratory testing equipment, such as ACE and MAT typically operate at atmospheric pressure. The Davison Circulating Riser (DCR), a pilot plantscale testing unit, is regularly operated under total pressure similar to commercial FCC units[1]. However, due to the small diameter of the DCR riser, a relatively large amount of nitrogen is needed to lift the catalyst, thus decreasing the hydrocarbon partial pressure. Studies documenting the effect of hydrocarbon partial pressure on FCC yields are scarce.

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It is generally expected that an increase in hydrocarbon partial pressure will increase the rate of all bimolecular reactions, including hydrogen transfer, relative to cracking, which is unimolecular. An increase in the rate of hydrogen transfer will result in a reduction of olefins in both gasoline and LPG and an increase in gasoline range aromatics and paraffins. The change in the rate of hydrogen transfer could also affect gasoline sulfur concentration and the effectiveness of gasoline sulfur reduction catalysts and additives. Moreover, the effectiveness of ZSM-5 additives, which are used to produce light olefins, especially propylene, could be affected by hydrocarbon partial pressure. Since ZSM-5 works by cracking gasoline range olefin molecules, changing the rate of hydrogen transfer could have a profound impact on propylene yield.

This paper will discuss the results of a series of cracking experiments in the DCR, where the hydrocarbon partial pressure was varied by varying the total reactor pressure, the feed rate and the amount of lift gas. The effect of changing hydrocarbon partial pressure on hydrocarbon yields, especially that of light olefin, and gasoline sulfur will be discussed.

Experimental

At the right is a schematic diagram of the standard DCR setup. The range of operating conditions in the DCR is shown in Table X. Operation of the DCR has been described previously[1]. Similar to commercial FCC units, the DCR is operated in adiabatic mode. In typical DCR operation, the regenerator temperature, the riser outlet temperature and the feed rate are set. The catalyst circulation rate and thus, the catalyst to oil ratio, is changed by varying the feed pre-heat temperature. During operation of the DCR, a metering pump precisely controls the feed rate as feed is pumped from the load cell through a preheater. Nitrogen and steam, injected through

a separate preheater/vaporizer, are used as a feed dispersant. Catalyst and product pass from the riser to the stripper overhead disengager. Products exit the disengager through a refrigerated stabilizer column to a control valve which maintains unit pressure at the desired level. A section of the stripper-regenerator spent catalyst transfer line consists of a shell and tube heat exchanger. The rate of heat transfer across this exchanger provides a precise and

reliable method to calculate the catalyst circulation rate. The stabilizer column, also called the debutanizer column, is operated to separate C_4 minus from the liquid product, which is condensed and collected. The collected liquid is analyzed by GC (SIMDIS – simulated distillation) to provide gasoline (ibp - 430°F), LCO (430-700°F), and 700°F + bottoms fractions. The gaseous products are metered and batch collected for subsequent analysis by GC.
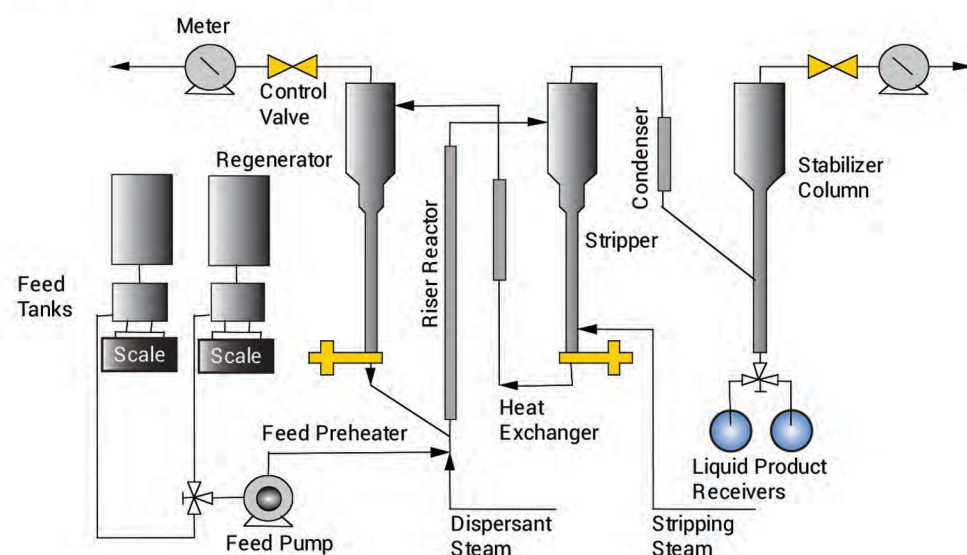
Schematic Diagram of Grace Davison DCR

Control Parameter	Range
System Pressure	< 45 psig
Catalyst Charge	1500-4000 g
Catalyst Circulation Rate	2500-15000 g/h
Feed Rate	350-2000 g/h
Feed Types	GO, VGO, Resid
Feed Preheater Temperature	120-400°C (250-750°F)
Riser Temperature	<590°C (<1100°F)
Disengager Temperature	<746°C (800-1100°F)
Stripper Temperature	427-593°C (800-1100°F)
Stabilizer Column Temperature	-34°C (-30°F)

Table X. DCR Operating Ranges

	DCR			ACE	Commercial FCCU
	Condition 1	Condition 2	Condition 3		
Reactor Top Temp (°F)	970	970	970		
Regenerator Temp (°F)	1300	1300	1300		
Unit Pressure (psig)	40	25	25		
Reactor Delta P (in H_2O)	5.24	4.11	3.45		
Feed Temp (°F)	575	575	300		
Feed Rate (g/h)	1500	1000	1000	180	
Reactor Water Rate (g/h)	30	30	30		
Reactor N_2 (L/h)	25	31	131	7.8	
Catalyst Cir. Rate (g/H)	8820	6160	6790		
C/O RATIO	5.9	6.1	6.9	6.0	6 to 9
Conversion wt%	72.0	72.2	72.7		
Molar Expansion	4.3	4.3	4.2		
Gas Residence time (s)	2.4	2.3	1.7	2.0	2 to 4
Slip Factor	2.0	2.0	1.7	Infinite	1 to 1.3
Catalyst Contact Time (s)	4.9	4.6	2.9	30 to 150	2 to 5
Catalyst Hold Up (g)	11.9	7.9	5.4	9	
WHSV (h-1)	125	126	183	20	100 to 250
HC Partial Pressure 1/3					
inlet + 2/3 outlet (psia)	44	28	20	12	20 to 50

Table XI. Operating Conditions in the DCR

We investigated two methods of changing hydrocarbon partial pressure. The first method involved keeping the total pressure, feed rate, and steam injection rate constant while reducing the nitrogen lift gas. The second method involved keeping the nitrogen lift gas and steam injection rate constant while increasing the total pressure and feed rate. The latter case is similar to some commercial FCC unit revamps where the total pressure of a FCC unit is increased to accommodate higher feed and catalyst circulation rate.

Table XI shows the three DCR operation conditions. Condition 3 is a commonly used DCR operating condition, while Conditions 1 and 2 are modifications to raise the hydrocarbon partial pressure closer to the value in commercial

FCC operations. Since cracking is a molecular weight reduction process, the hydrocarbon mole fraction and, therefore, partial pressure increase along the riser. The molar expansion (moles of product/moles of feed) in a typical FCC unit is between four and five. For the purpose of engineering calculations, it is common to approximate the hydrocarbon mole fraction as equal to 1/3 of the mole fraction at the inlet and 2/3 of the mole fraction at the outlet of the riser. The total moles of the hydrocarbon products are calculated by using GC analyses of the light gases and gasoline PIONA and assuming average molecular weight values of 220 and 350 for LCO and bottoms, respectively.

In varying hydrocarbon partial pressure, we chose operating conditions so

as not to greatly change the weight hourly space velocity (WHSV), as that in itself could change the selectivity and complicate the interpretation of the results. The slip factor in the riser (ratio of the gas velocity to catalyst velocity), estimated by the correlation of Pugsley and Berruti[2], varied from 1.7 to 2. These values were consistent with those reported by Bollas et al.[3]. Once the slip factor was determined, the catalyst holdup (the amount of catalyst in the riser), catalyst contact time and WHSV were readily calculated (Table XI). The catalyst holdup values followed the trend of pressure drop measurements across the riser. Conditions 1 and 2 varied in hydrocarbon partial pressure by a factor of 1.55. However, the values of the WHSV, catalyst-to-oil ratio and

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Analysis	Catalyst A	Catalyst B
Al_2O_3, wt.%	40.8	46.9
RE_2O_3, wt.%	1.16	3.05
Na_2O, wt.%	0.39	0.28
Ni, ppm	537	523
V, ppm	520	510
Surface Area, m^2/g	219	146
ZSA, m^2/g	179	113
MSA, m^2/g	40	33
Unit Cell Size, Å	24.24	24.32

Table XII. Properties of Catalysts Deactivated at 500ppm Ni/500ppm V CPS-3/1480ºF

conversion were essentially identical. Therefore, the changes in selectivity could be attributed principally to the change in hydrocarbon partial pressure. Compared to Conditions 1 and 2, the hydrocarbon partial pressure of Condition 3 was 2.3 times lower. However, its WHSV was also somewhat greater, due to the higher level of lift nitrogen used. In this case, we would need to rationalize the contribution of hydrocarbon partial pressure to the selectivity shifts.

Table XI also compares the current DCR operating conditions with that of commercial FCC units and ACE. Compared to the earlier operating conditions (Condition 3), Conditions 1 and 2 are closer to the commercial units, especially in hydrocarbon partial pressure. Furthermore, compared to the operating conditions of ACE, the operating conditions of the DCR are much closer to those of the commercial unit.

API Gravity	25.5
Specific Gravity, g/cm³	0.9012
K Factor	11.94
Refractive Index	1.5026
Sulfur, wt%	0.369
Basic Nitrogen, wt%	0.05
Total Nitrogen, wt%	0.12
Conradson Carbon, wt%	0.68
ndm Analysis	
Arom Ring Carbons Ca, wt%	18.9
Naphthenic Ring Carbons Cn, wt%	17.4
Paraffinic Carbons Cp, wt%	63.6

Average MW	406
Ni, ppm	0.4
V, ppm	0.2
Similated Distillation	
IBP	307
10%	607
30%	740
50%	818
70%	904
90%	1034
End Point	1257

Table XIII. Properties of VGO Feedstock

Two Davison commercial FCC catalysts, labeled Catalyst A and Catalyst B, containing 1.2 and 3.1% RE_2O_3, respectively, were used in this study. Both catalysts were steam deactivated according to CPS-3 protocol[4] at 1480ºF with 500 ppm nickel and 500 ppm vanadium. The chemical and physical properties of the two catalysts are listed on Table XII. The deactivated unit cell size measurements of the low and high RE_2O_3 catalysts are 24.24Å and 24.32Å, respectively. Catalyst A was also blended with 20% OlefinsUltra® additive, a commercially available ZSM-5 additive and deactivated according to CPS-3 protocol at 1480ºF with 500 ppm nickel and 500 ppm vanadium. A Gulf Coast vacuum gas oil feed was used in this study. The properties of the feedstock are shown on Table XIII.

Results and Discussion

Case I

In this example, Catalyst A was tested in the DCR under both Conditions 1 and 2. Under Condition 1, the unit pressure was 40 psig, the feed rate was 1500 g/h, the dispersing steam was 30 g/h, and 25 l/h nitrogen was injected to help disperse the feed as well as to lift the catalyst. Based on the above discussion, the time-averaged (1/3 inlet + 2/3 outlet) hydrocarbon partial pressure under this condition was 44 psia. Under Condition 2, the unit pressure was 25 psig, the feed rate was 1000 g/h, whereas the steam and nitrogen flow rates were the same as that of Condition 1. The hydrocarbon partial pressure under this condition is 28 psia. The above comparison is very similar to a common revamp of a commercial FCC unit where in order to increase feed rate and catalyst circulation the total pressure of the unit has to be increased to maintain velocity.

The plots of catalyst to oil ratio, total C_3, total C_4, gasoline, LCO, and coke yields against conversion are shown in Figure 32. Increasing HC partial pressure increases dry gas and coke at the expense of gasoline. The yields of total C_3, C_4 and LCO remain about the same. The higher coke yield may be attributed to a higher rate of oligomerization, which is a bimolecular reaction and favored at high pressure. The higher dry gas could be the result of oligomerization/recracking.

Figure 33 shows that increasing the HC partial pressure decreases the yields of propylene, butenes, and gasoline olefins, while increasing the yield of gasoline isoparaffins. Increasing HC partial pressure substantially lowers the C_3 and C_4 olefinicities. These yield shifts suggest that the rate of hydrogen transfer increases with HC partial pressure, as would be expected for a bimolecular reaction.
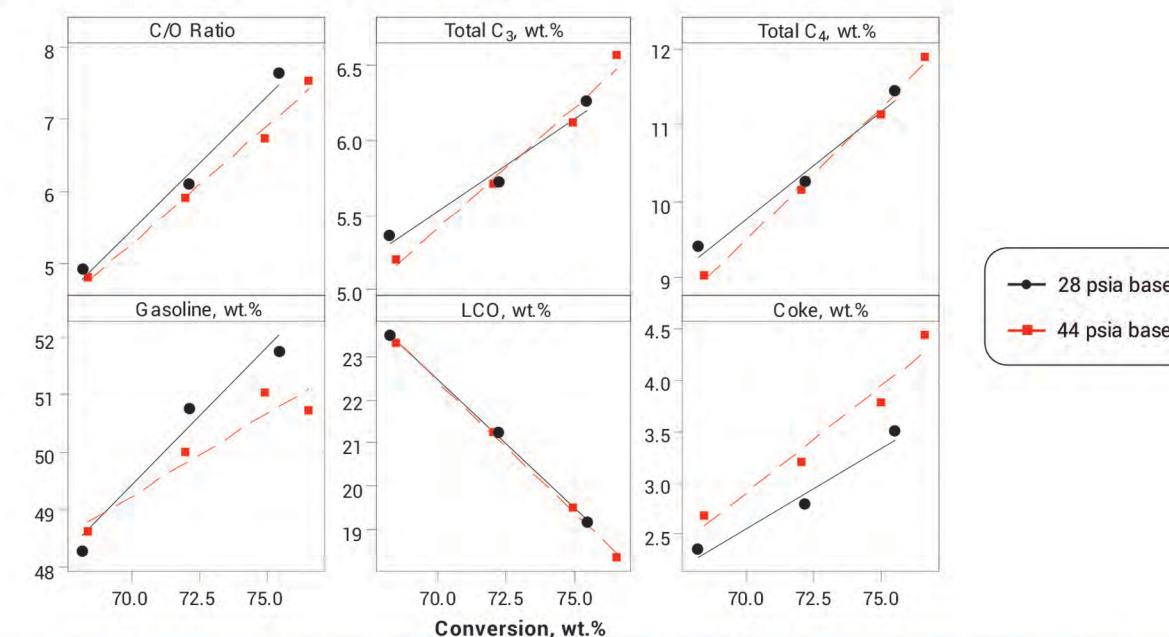

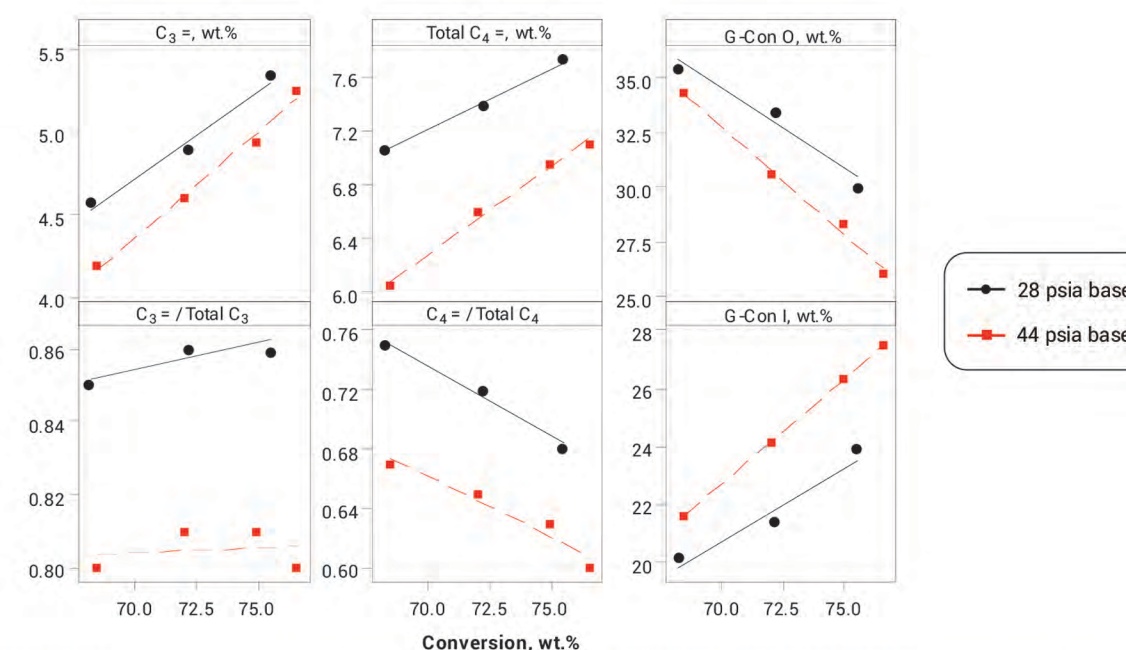**Figure 32.** Effect of DCR Operating Conditions on the Yields of Catalyst A

Figure 33. Effect of DCR Operating Conditions on the Olefins Yield and Olefinicity of Catalyst A

Continued from Page 38

The interpolated yields at 73 wt% conversion are listed on Table XIV. A convenient way to gauge the hydrogen transfer rate is to look at the paraffins to olefins ratio of C_3, as well as linear and branched C_4 compounds[5]. The hydrogen transfer indices are defined as the ratios of propane/propylene, n-butane/(1-butylene + trans-2-butylene + cis-2- butylene) and isobutane/isobutylene. These ratios are shown in Table XIV. In this analysis we are assuming that the C_3 and C_4 alkanes are the product of hydrogen transfer from their parent alkenes and ignoring the alkanes formed by thermal or protolytic cracking.

The hydrogen transfer reaction of isobutene proceeds via a tertiary carbenium ion intermediate and thus occurs at a much faster rate than the hydrogen transfer reactions of propylene and linear butenes, which proceed through a less stable secondary carbenium ion intermediate. All of the hydrogen transfer indices increase by a factor of 1.5, as the HC partial pressure increases almost proportionally by a factor of 1.6 from 28 to 44 psia. Thus, all the yield shifts are consistent with an increase in the rate of hydrogen transfer with the increase in HC partial pressure.

	Condition 1	Condition 2	
HC Partial Pressure, psia	44	28	Ratio of HC Pressure 1.6
Cat to Oil	6.1	6.5	
H_2 Yield, wt%	0.05	0.05	
$C_1 + C_2$'s, wt%	2.8	2.4	
C_2=, wt%	0.8	0.8	
Total C_3, wt%	5.9	5.9	
C_3=, wt%	4.7	5.0	
Total C_4, wt%	10.5	10.6	
iC_4, wt%	3.0	2.5	
nC_4, wt%	0.8	0.6	
Total C_4=, wt%	6.7	7.5	
iC_4=, wt%	1.9	2.4	
Gasoline, wt%	50.1	50.8	
G-Con P, wt%	4.0	3.7	
G-Con I, wt%	24.8	22.2	
G-Con A, wt%	30.5	30.2	
G-Con N, wt%	10.9	11.5	
G-Con O, wt%	29.6	32.2	
G-Con RON EST	92.2	92.0	
G-Con MON EST	79.8	79.2	
LCO, wt%	20.5	20.7	
Bottoms, wt%	6.3	6.2	
Coke, wt%	3.5	3.0	
Hydrogen Transfer Index			Ratio of HT Index
C_3 / C_3=	0.24	0.17	1.4
$nC_4 /(1C_4$= $+ t2 C_4$= $+c_2 C_4$=)	0.18	0.12	1.5
iC_4 /iC_4=	1.56	1.05	1.5

Table XIV. Interpolated Yields at 73 wt% Conversion Over Catalyst A

Case II

Catalyst A was blended with 20% OlefinsUltra® additive and tested in the DCR under Conditions 1 and 2. The catalyst to oil ratio, total C_3, total C_4, gasoline, LCO, and coke yields against conversion plots are shown in Figure 34. The yields of C_3=, C_4=, gasoline olefins and gasoline isoparaffins, as well as the olefinicities of C_3, C_4, and gasoline are shown in Figure 35. As in the case without OlefinsUltra® additive (Case I), increasing HC partial pressure increases coke and dry gas and dramatically decreases gasoline and LPG olefinicity. The C_3 olefinicity of ca. 0.84 at the higher HC partial pressure is much more realistic and close to the commercially observed values.

The interpolated yields at constant conversion of 73 wt% are shown on Table XV. Remarkably, increasing the HC partial pressure from 28 to 44 psia decreases the propylene yield by 1 wt% absolute and decreases the butylenes yield by 0.6 wt% absolute. It is known that the addition of ZSM-5 increases LPG olefins by cracking gasoline range olefins[6-8]. Increasing the rate of hydrogen transfer, by increasing the HC partial pressure, depletes the gasoline range olefins and decreases the effectiveness of ZSM-5. The rate of hydrogen transfer, as estimated by the hydrogen transfer indices, described above, increases approximately proportionally to HC partial pressure.
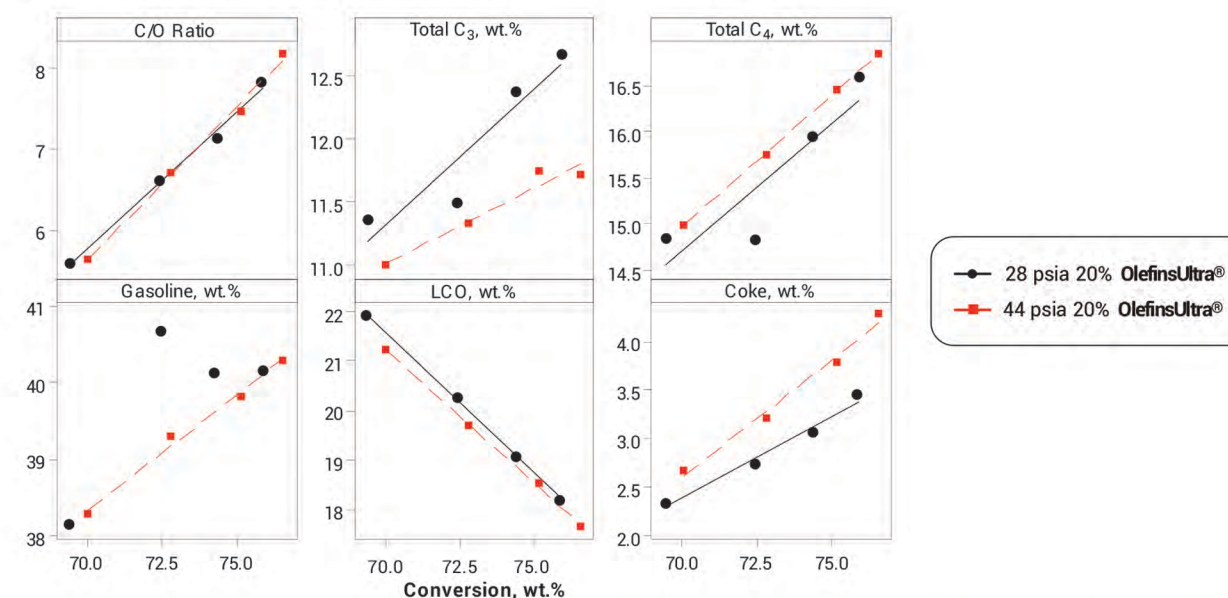

Figure 34. Effect of DCR Operating Conditions on the Yields of Catalyst A with 20% OlefinsUltra® Additive
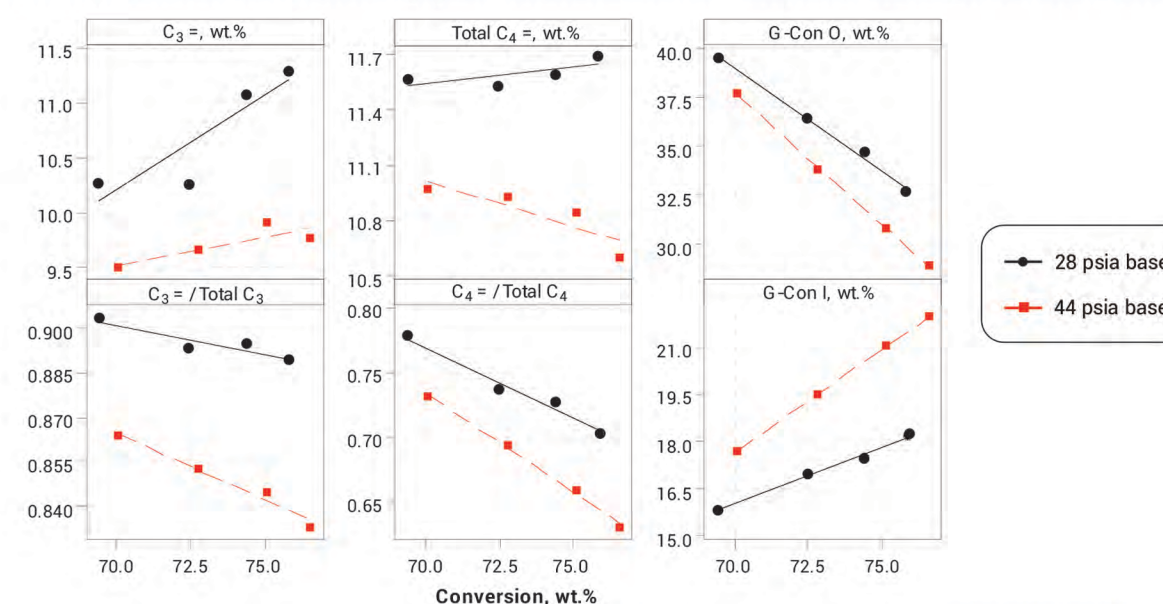

Figure 35. Effect of DCR Operating Conditions on the Olefins Yield and Olefinicity of Catalyst A with 20% OlefinsUltra® Additive

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	Condition 1	Condition 2	
			Ratio of HC Pressure
HC Partial Pressure, psia	44	28	1.6
Cat to Oil	6.7	6.7	
H_2 Yield, wt%	0.04	0.05	
$C_1 + C_2$'s, wt%	3.0	2.6	
$C_2=$, wt%	1.4	1.3	
Total C_3 , wt%	11.4	12.0	
$C_3=$, wt%	9.7	10.7	
Total C_4 , wt%	15.8	15.5	
iC_4 , wt%	3.9	3.3	
nC_4 , wt%	1.0	0.8	
Total $C_4=$, wt%	10.9	11.4	
$iC_4=$, wt%	4.1	4.3	
Gasoline, wt%	39.2	39.8	
G-Con P, wt%	4.0	3.8	
G-Con I , wt%	19.6	17.1	
G-Con A , wt%	33.7	33.9	
G-Con N , wt%	8.9	9.2	
G-Con O , wt%	33.5	35.8	
G-Con RON EST	95.2	94.9	
G-Con MON EST	81.5	80.9	
LCO , wt%	19.6	19.8	
Bottoms , wt%	7.3	7.1	
Coke, wt%	3.3	2.9	
Hydrogen Transfer Index			**Ratio of HT Index**
C_3 /$C_3=$	0.17	0.12	1.5
nC_4 /($1C_4=$ + t_2 $C4=$ +C_2 $C_4=$)	0.15	0.11	1.4
iC_4 /$iC_4=$	0.95	0.76	1.2

Table XV. Interpolated Yields at 73 wt% Conversion Over Catalyst A with 20% OlefinsUltra® Additive

Figure 36. Variation of Yields with Unit Cell Size and DCR Operating Conditions
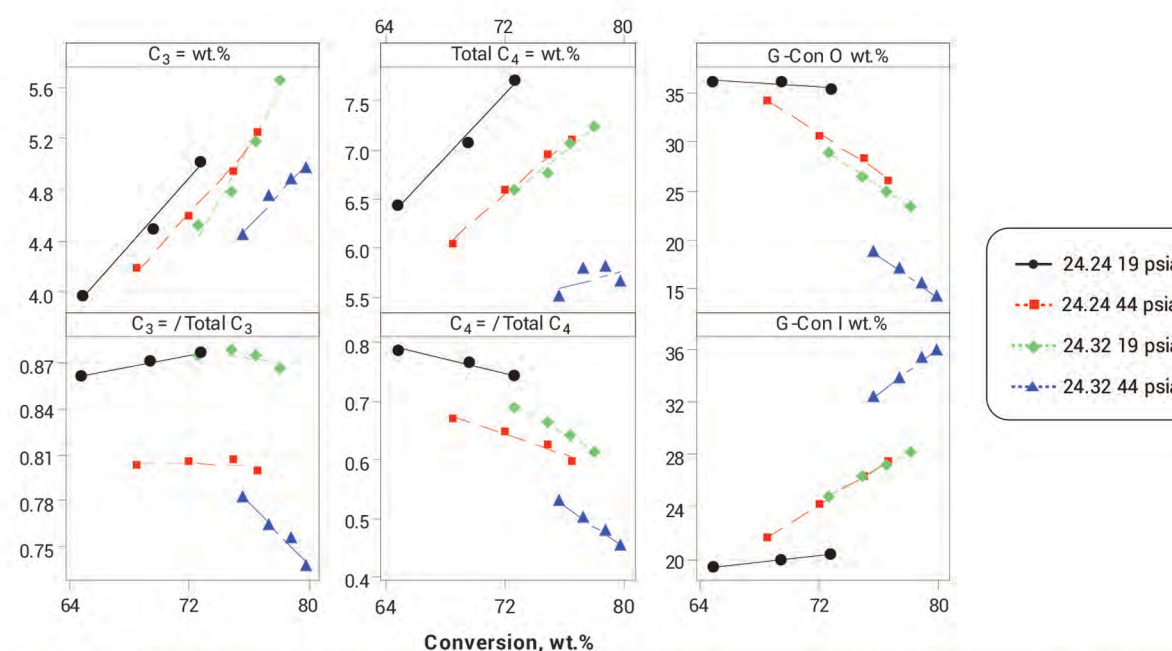
Figure 37. Variation of Olefins Yield and Olefinicity with Unit Cell Size and DCR Operating Conditions

Case III

In this example, Catalysts A and B, having unit cells size values of 24.24Å and 24.31Å, respectively, were tested under DCR Conditions 1 and 3. Condition 3 featured a unit pressure of 25 psig, 1000 g/h feed rate, 30 g/hour steam, and 128 l/h nitrogen. The main difference between Condition 1 and Condition 3 was the greater amount of nitrogen lift gas used in Condition 3, which not only decreased the HC partial pressure by a factor of 2.3, from 44 to 19 psia, but also increased the WHSV by a factor of 1.4. The effect of the change in WHSV will be discussed.

The main yields are shown in Figure 36, while the LPG and gasoline composition are shown in Figure 37. For both the high and low unit cell size catalysts, the response of the LPG and gasoline olefin yields to the changes in DCR conditions are very similar to that observed in Case 1, namely increasing HC partial pressure decreases LPG and gasoline olefins and olefinicity. The hydrogen transfer indices increased by a factor of two as HC partial pressure increased by a factor of 2.3 (Table XVI). Thus, as in Case I, the change in the hydrogen transfer indices are nearly proportional to the change in HC partial pressure. This suggests that the change in HC partial pressure is mainly responsible for the yield changes while the shifts in WHSV may be responsible for the shifts in conversion at a given catalyst to oil ratio.
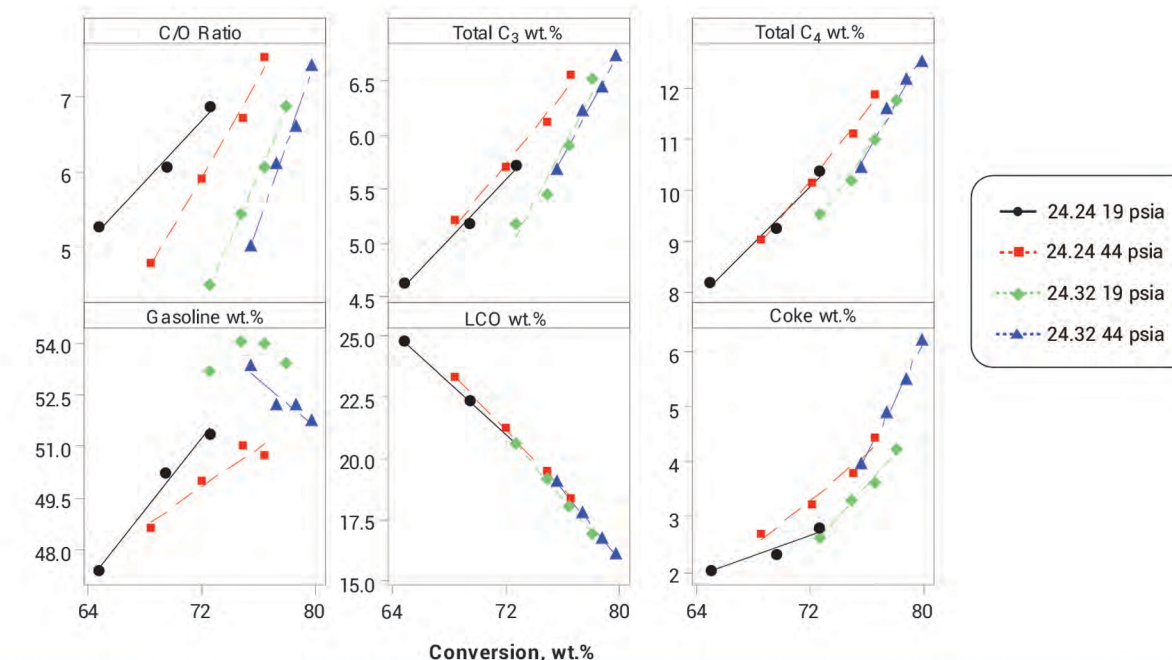
The yields shifts due to changing unit cell size are consistent with what has been reported in the literature[9,10], namely that the higher UCS catalyst makes higher gasoline, lower octane, lower LPG and gasoline olefins. These trends are observed at both DCR conditions. The effect of unit cell size and HC partial pressure on the rate of hydrogen transfer appears to be simply additive.

The rate of bimolecular reactions can be increased by increasing acid site density as well as increasing HC partial pressure. The ratios of the hydrogen transfer indices of Catalyst B to Catalyst A are about the same at both low and high HC partial pressure (Table XVI).

	Catalyst A		Catalyst B	
	Condition 3	Condition 1	Condition 3	Condition 1
HC Partial Pressure, psia	19	44	19	44
Cat to Oil	7	7	5	5
H_2 Yield, wt%	0.04	0.05	0.03	0.03
$C_1 + C_2$'s, wt%	2.2	2.7	1.9	2.2
$C_2=$, wt%	0.7	0.8	0.6	0.7
Total C_3, wt%	5.9	6.2	5.6	5.6
$C_3=$, wt%	5.2	5.0	4.9	4.4
Total C_4, wt%	10.8	11.2	10.4	10.3
iC_4, wt%	2.4	3.4	2.9	3.7
nC_4, wt%	0.5	0.9	0.6	1.0
Total $C_4=$, wt%	8.0	6.9	6.8	5.5
$iC_4=$, wt%	2.7	1.9	1.8	1.4
Gasoline, wt%	53.6	50.7	53.6	53.4
G-Con P, wt%	3.4	4.0	4.0	4.6
G-Con I, wt%	20.8	26.3	26.3	31.8
G-Con A, wt%	29.1	31.3	30.8	32.3
G-Con N, wt%	11.4	10.6	12.5	12.1
G-Con O, wt%	35.4	27.7	26.3	19.5
G-Con RON EST	92.4	92.1	89.9	88.8
G-Con MON EST	79.2	80.0	78.4	78.6
LCO, wt%	20.0	19.3	19.0	19.5
Bottoms, wt%	6.0	5.6	5.9	5.6
Coke, wt%	2.9	3.9	3.2	3.7
Hydrogen Transfer Index				
$C_3/C_3=$	0.13	0.24	0.14	0.27
$nC_4/(1C_4= + t_2C_4= + c_2C_4=)$	0.09	0.18	0.12	0.24
$iC_4/iC_4=$	0.88	1.79	1.57	2.69

Table XVI. Interpolated Yields at 75 wt% Conversion Over Catalyst A and B
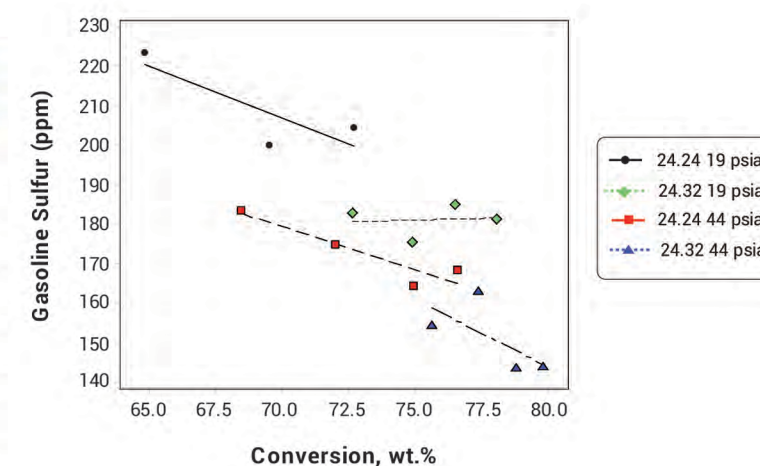

Figure 38. Effect of UCS and DCR Operating Conditions on Gasoline Sulfur Concentration

Figure 38 shows the concentration of gasoline sulfur (including all thiophene species with a boiling point below 430ºF, tetrahydrothiophene, and benzothiophene) for both Catalysts A and B under the two DCR operation conditions. Gasoline sulfur concentration decreases with increasing unit cell size and with increasing HC partial pressure. These results suggest that the reduction of gasoline sulfur follows the trend of increase hydrogen transfer activity and are consistent with the previously proposed mechanism, shown below[11,12].

Scheme 1

High rate of hydrogen transfer speeds up this reaction by promoting the formation of the reaction intermediate, tetrahydrothiophene.
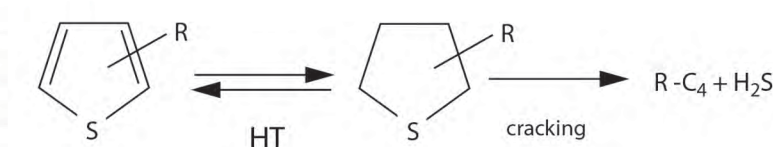
Scheme 1

Conclusions

By varying the operating conditions of the DCR, we have been able to conduct cracking experiments over a wide range of hydrocarbon partial pressure. The results indicate that increasing the hydrocarbon partial pressure decreases the olefinicity and olefins yield of LPG and gasoline. This observation is consistent with the notion that hydrogen transfer reactions, being bimolecular in nature, increase with increasing HC partial pressure. The hydrogen transfer index, defined as the paraffin/olefin ratio of C_3, linear C_4 and branched C_4 species increase almost linearly with HC partial pressure. It has been demonstrated that the effectiveness of ZSM-5 additives is lessened at high HC partial pressure due to the depletion of gasoline range olefins via hydrogen transfer reactions. The concentration of gasoline sulfur species decreases at higher HC pressure, again due to higher rate of hydrogen transfer. Recent advancements in DCR operation enable more realistic simulation of commercial FCCU operation.

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