

KBR Catalytic Olefins Technologies Provide Refinery/Petrochemical Balance

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Abstract:

Many refiners find themselves with underutilized FCC capacity while at the same time they must cope with stranded streams such as coker naphtha and FCC C4's and C5's which have little value as fuel blending stock due to olefin content and octane issues. This problem will be aggravated as more cokers come on-stream to convert additional resid material and as FCC units increase severity to maximize propylene production. It is possible, however, to upgrade such olefins-containing streams to high-value petrochemicals using SUPERFLEX™, a commercialized fluid catalytic cracking process, which converts olefins-containing C4-C8 streams predominantly to propylene, with large quantities of ethylene and aromatics also produced. In addition, elements of high severity C4 and naphtha cracking can be integrated into an FCC unit co-processing traditional FCC feedstocks such as VGO using the MAXOFIN™ FCC Process.

This paper provides a general overview of the KBR SUPERFLEX, ACO and MAXOFIN FCC processes, including discussion of their similarities to, and differences from, refinery FCC's. Information is included regarding the yield performance of cracking streams such as coker naphtha and FCC C4/C5's and the implications regarding product octane and olefin content.

Introduction

The naphtha byproducts from cokers have low octane, high olefins content as well as high levels of unstable diolefins and sulfur that make them unsuitable for direct blending into the gasoline pool without hydrotreating. Many refiners are gravitating to the processing of heavier crudes to conserve feed costs. In addition to the installation of more residue hydrocracking and solvent deasphalting capacity to handle the increased amount of heavy residues^{1,2}, the shift to heavier crudes is also expected to result in added coker capacity and increased coker naphtha production.

Situations also arise where the potential motor gasoline supply exceeds demand and refiners would

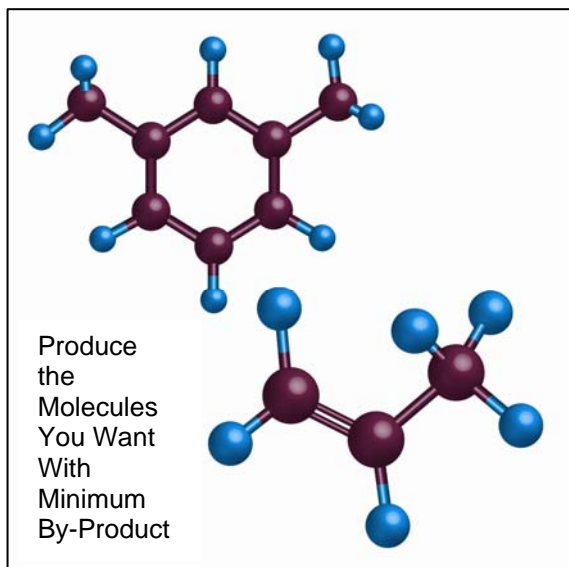


Figure 1 – m-Xylene and Propylene

rather produce petrochemicals such as propylene and BTX instead of motor gasoline from their FCC units. The influence of propylene in the refinery economic picture has driven a "paradigm shift" over the last several decades. Propylene has gradually moved from a byproduct of refining to a co-product. That may seem like a trivial distinction, but it signifies that today's refiner must consider petrochemical economics, as well as refining economics, when making operating decisions or planning new facilities³.

Today, C4 and C5 olefins also offer little utility as a blendstock in a modern gasoline motor fuel because of their high vapor pressure and high olefinicity. The refiner's best choices may be to convert these materials into either higher or lower carbon number hydrocarbons that better meet the market demand.

Refiners looking to increase the value of refinery propylene can also choose to increase the purity of propylene produced relative to a traditional refinery grade propylene product. Higher purity options include chemical grade propylene and polymer grade propylene⁴.

Refiners looking to increase the value of refinery propylene can also choose to increase the purity

KBR Catalytic Processes for Propylene

KBR offers three catalytic olefin cracking processes which are FCC processes designed to produce propylene and other high value products from readily available C4 and naphtha feed sources. From among these processes, the process selection is based on the degree of saturation in the feed and each process uses a different catalyst.

Table 1

KBR Catalytic Olefin Processes

Process	Feedstocks	Products
SUPERFLEX	Olefinic streams, such as C4's and light naphthas from cokers and FCC units ⁵	50-60% total C2/C3 olefins along with a significant quantity of BTX. Typical P/E ratios are about 2/1
Advanced Catalytic Olefins (ACO™)	Paraffinic naphtha streams which already are the steam cracker feed source for more than half of the world's ethylene ⁶	Propylene, Ethylene and other high value Products. ACO produces more total olefins than pyrolysis, with P/E ratios up to 1/1
MAXOFIN	Typical FCC feeds such as VGO's and atmospheric residues along with imported C4's and naphthas ⁷	More than 20% propylene along with significant quantities of BTX and ethylene

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SUPERFLEX

SUPERFLEX was discovered and patented by LyondellBasell and it is licensed exclusively by KBR. The technology is built around the high selectivity of ZSM-5 catalysts and the optimal process conditions for high yield of propylene and ethylene. KBR provided fluid solids technology, as well as engineering, procurement and management services for construction of the world's first SUPERFLEX unit for SASOL; this included additional patented improvements and customized catalyst development to facilitate the application of the required process conditions. Since then, additional projects have been initiated to leverage this technology for meeting the increasing worldwide propylene demand.

SUPERFLEX technology builds on over 70 years of experience in fluidized bed catalytic cracking (FCC) and over 50 years of cost-effective purification units for ethylene and propylene plants at KBR. This expertise, combined with KBR engineering excellence, strategic procurement, expert construction and rigorous project management, provides the right processes to meet the specific needs of petrochemical and refinery clients.

Feeds for SUPERFLEX can be sourced from nearly any cracking process, steam cracking or various refinery cracking processes. Following are some examples of feedstocks that can be effectively upgraded:

- Mixed C4s from refineries and conventional steam crackers
- Amylenes, TAME feed and mixed C5s
- Cracked naphtha from FCCs, steam crackers, cokers and visbreakers
- Other low-value olefinic streams.

SUPERFLEX technology provides 50 to 60 percent ultimate yield of propylene plus ethylene and an aromatic rich gasoline stream. This is achieved due to the high single pass conversion to valuable products and recycle of higher olefins to extinction.

Efficient Plant Design for Flexible Olefins Production

Like the refinery FCC, the SUPERFLEX reaction system is comprised of four sections:

- Riser/reactor, where the cracking reactions take place in the presence of catalyst
- Disengager, where catalyst is separated from product gas through the use of cyclones
- Stripper, where cracked gas contained in catalyst pores is removed via stripping with steam and routed with the other product gas
- Regenerator, where coke formed on the catalyst during the cracking process is removed by combustion with oxygen, and where heat is generated to support operation of the process.

Although special mechanical modifications to KBR's FCC system are made to accommodate the particular operating conditions, the functionality is not changed. As with other KBR FCC technologies, SUPERFLEX utilizes the KBR Orthoflow™ configuration as shown in Figure 2. In this configuration, the disengager is stacked above the regenerator, rather than side-by-side as with other designs. The Orthoflow configuration has a number of advantages. The plot space required is much smaller for this configuration compared to the side-by-side type of unit. Foundation and

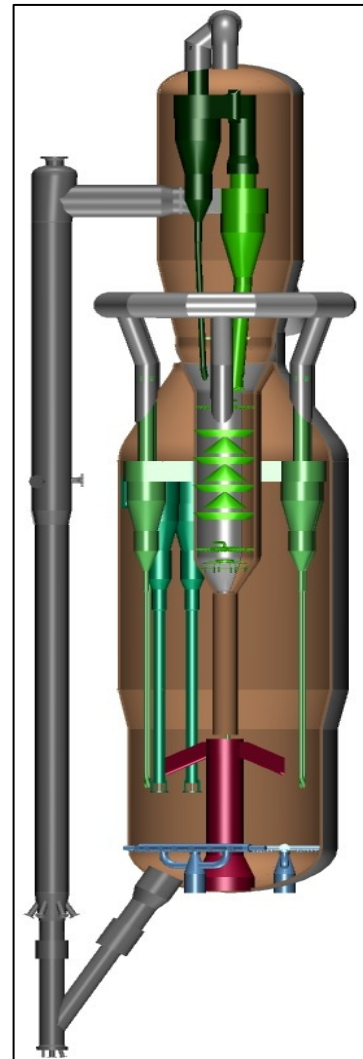


Figure 2 – Orthoflow™ FCC Converter

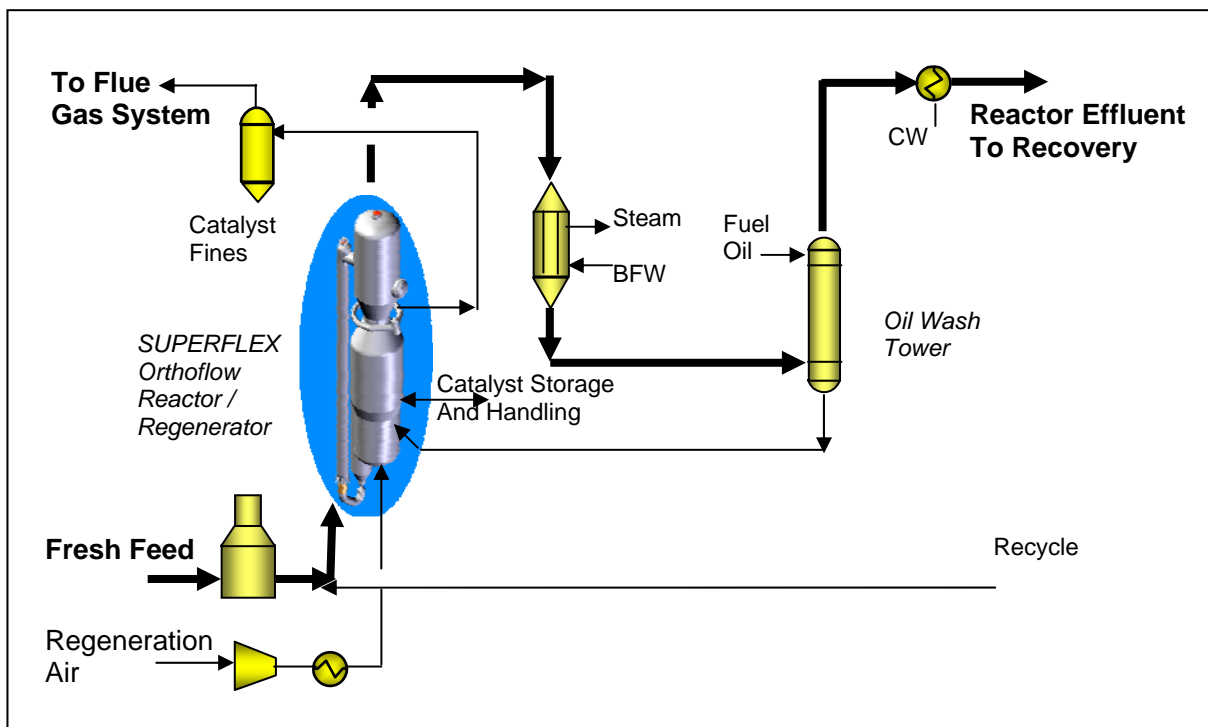
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structural costs are also reduced. The Orthoflow configuration allows the unit to be fabricated and dressed off-site and set into place with one lift, saving on construction costs and with less required welding in the field. The stripper is submerged within the regenerator, reducing the vertical height of the unit and thus the cost.

The typical SUPERFLEX reactor section is shown in Figure 3. Reactor overheads are cooled (generating high pressure steam) and washed to recover entrained catalyst, which is recycled back to the reaction system. The net overhead product is further cooled and then routed to product recovery, either integrated with an olefins plant or in a stand-alone recovery section.

No feed pretreatment is required because of the robust catalyst system employed. Accessory systems for the reactor are standard FCC systems and include catalyst storage, air supply, flue gas handling and heat recovery.

Figure 3
SUPERFLEX Reactor System



Although in many ways the SUPERFLEX reactor system is very similar to the refinery FCC unit, there are some unique and innovative features that differentiate it from a heavy oil refinery FCC unit. Some of these features, all of which are commercially demonstrated, include:

- **Heat Balance** – Since the feeds are light, the amount of coke produced on the catalyst is insufficient to supply the necessary process heat. As such, to maintain heat balance, fuel must be imported into the regenerator. KBR's patented catalyst well design with continuous fuel firing has now been commercially proven⁸.
- **Catalyst/Hydrocarbon Separation** – Although the cyclones are quite efficient, invariably some catalyst fines will carry over with the reactor effluent cracked gas. KBR's patented catalyst fines removal system has now been commercially demonstrated and is applicable to SUPERFLEX technology⁹. The catalyst fines are concentrated in the bottoms product from the quench tower and returned to the regenerator with the fuel oil that is needed to support the process heat balance. There is no net bottoms product production.

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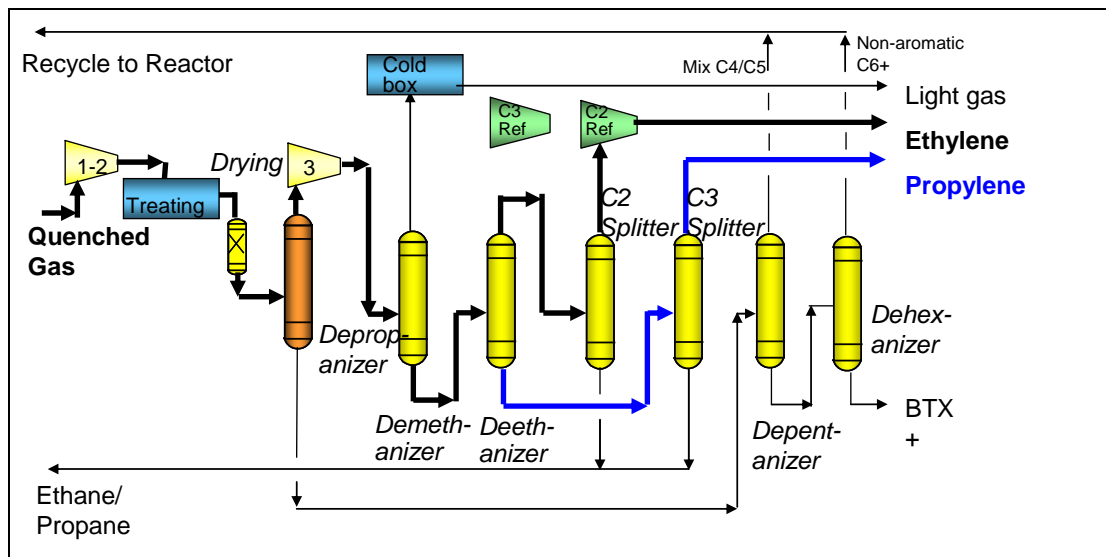
- **Reactor Overhead Heat Exchange** – Because the quench tower bottoms operates at a relatively low temperature, higher level heat recovery is achieved with a steam generating heat exchanger located in the reactor overhead line.
- **Feed Vaporization and Superheat** – To minimize the requirement for burning fuel in the regenerator and to maintain the catalyst-to-oil ratio in the desired range, the feed is vaporized and superheated before being charged to the riser.

The SUPERFLEX process produces both polymer grade ethylene and propylene. Much of the process flow scheme is similar to typical olefins plant recovery sections, but there are some unique features. For example, there are contaminants such as nitrogen oxides, oxygen, and other trace impurities by virtue of the FCC-type reactor that must be removed. These and other issues are addressed in the SUPERFLEX process flow scheme, which features a front end Depropanizer as shown in Figure 4:

Design of a vapor recovery unit to produce polymer grade ethylene and propylene products includes consideration of several factors not typically addressed in FCC VRU design.

- The cold fractionation train begins with a Depropanizer system, followed by a Demethanizer, Deethanizer and ethylene-ethane Splitter. Polymerization fouling is minimized in both columns as a consequence of the low operating temperatures.
- Facilities are required to remove impurities from the process gas and to prevent freezing and hydrate formation in low temperature operations.
- In situations where propylene has a high value but ethylene will be routed to refinery fuel, an absorption oil recovery system will be more economic. If the refinery also wants to recover and produce a petrochemical ethylene product, then cryogenic recovery would be indicated.

Figure 4
Typical SUPERFLEX Recovery Flow Scheme



SUPERFLEX Commercialization

The first commercial SUPERFLEX unit started up at Sasol in South Africa in 2006. This unit converts a highly olefinic C6/C7 stream to propylene and ethylene, with a reactor propylene capacity of approximately 250 KTA. The feed also contains a high concentration of oxygenates, but is processed directly in the SUPERFLEX converter with no pretreatment. The recovery section is designed to also process various olefins-rich streams from the complex in addition to the SUPERFLEX reactor products with total polymer-grade production rates of approximately 500 KTA of propylene and 200 KTA of ethylene.

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JiHua is the second licensee, located in Jilin City, China. The capacity of that unit will be 200 KTA of propylene from C4 and C5 feeds. KBR has completed basic engineering for that plant.

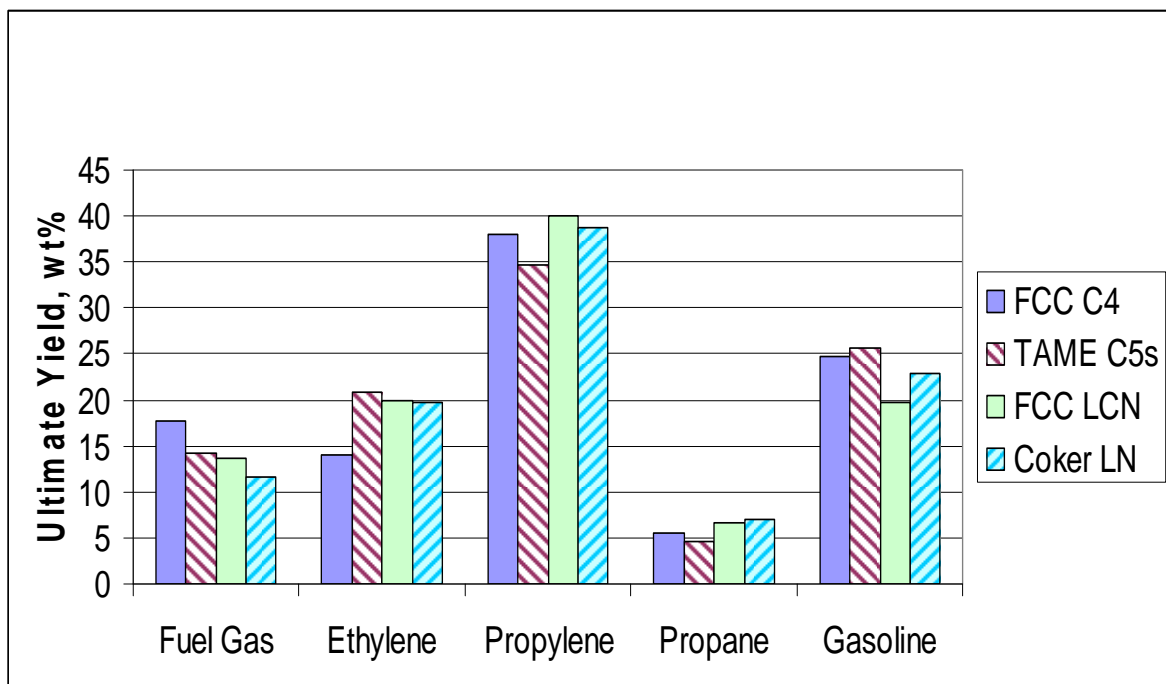
KBR has licensed a third unit, also in China, for which the process design package is just being completed by KBR.

SUPERFLEX Pilot Test Yields For Typical Refinery Streams

An FCC pilot plant at KBR's Technology Development Center located in Houston, Texas, USA has been used to determine yields from numerous SUPERFLEX feeds at various process conditions.

KBR has tested refinery streams and some of these results are summarized in Figure 5. Typical ultimate yields are given on a recycle-to-extinction basis (i.e. including recycle to extinction of the C4 and C5 products from the SUPERFLEX reactor).

**Figure 5
SUPERFLEX Yields**



Feedstocks for SUPERFLEX are comprised mainly of C4 through C8 olefins and typically contain various quantities of paraffins, aromatics, and naphthenes. Potential sources of SUPERFLEX feeds can be found at distressed values in many locales due to either lack of demand or perhaps due to changing product markets. In both the olefin plant and refinery, many of the olefin containing streams (pyrolysis gasoline; FCC C4's and Light FCC naphtha; coker naphthas and visbreaker naphthas) are used in gasoline blending. Gasoline regulations are changing throughout the world in response to environmental pressures. Reductions in vapor pressure, olefins, and oxygenate specifications and removal of lead are reducing the blend value of these components.

As another example of potential SUPERFLEX feed, consider the case where steam cracker butadiene and C5 diolefins are not extracted; these components may currently be hydrogenated to paraffins and recycle cracked consuming quantities of ever more valuable hydrogen. If these components are not completely hydrogenated prior to steam cracking, furnace yields are reduced and run lengths are shortened due to increased tube coking rates. Alternate routing of these streams to a SUPERFLEX unit to make propylene can provide substantial returns.

In addition to propylene production, valued byproducts are produced including ethylene and high octane aromatic rich gasoline. Ethylene production varies with feed composition, ranging in

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propylene to ethylene ratio (P/E ratio) from 1.5 to 2.0 P/E. A SUPERFLEX addition to an existing olefins plant could thus, under a constant ethylene scenario, reduce the consumption of expensive naphtha while simultaneously increasing the ultimate plant P/E ratio. No other propylene-on-purpose process produces ethylene co-product in these quantities; in contrast there are some which consume higher valued ethylene to make propylene. SUPERFLEX is unique in the robustness of the process with respect to the allowed variability in feedstock composition. There is no operating limit on aromatics or diene content. There is a point where the economics lead the operation to selective hydrogenation of dienes to olefins for more optimum return on installed capacity. Paraffins are partially converted with each pass through the reactor, contributing to ultimate light olefins yield and allowing recycle to extinction operation.

SUPERFLEX effluent can be readily accommodated in the olefin recovery plant, applicable to all flow schemes employed. Trace quantities of acetylene and MAPD are produced so there is no change to existing converter loads. Insertion of an oxygenate guard bed upstream of the caustic/amine system may be necessary to meet product specifications and for protection of downstream catalyst systems. This is due to a small quantity of regenerator flue gas carried with catalyst into the reaction riser.

Installation of a SUPERFLEX reaction system in the refinery can be implemented, with the product purification undertaken as dictated by economics. Processing SUPERFLEX effluent can be configured as one of the following: stand alone recovery section; compression and partial recovery (up to Depropanizer) to allow ISBL recycle and OSBL product purification in a neighboring olefins plant; or processing in an existing FCC gas concentration unit. Refinery based economics will be affected by the credit for coproduced ethylene, which would require modification of the absorber/stripper to refrigerated distillation for recovery of high purity product in the standard unsaturated gas concentration unit. The dilute ethylene stream is a very good feed for ethylbenzene production as an alternate to recovery of polymer grade product.

Commercial Naphtha Composition and Octane Improvements

The first commercial SUPERFLEX unit commissioned for Sasol in Secunda, South Africa provides a vivid example of how catalytic olefins processes can create valuable naphtha products from naphtha streams having limited value in traditional motor gasoline manufacture. At Sasol's site, the coal derived synfuels operations yields a very olefinic, low octane C6-C7 naphtha by-product (with about a 70 RON) that makes an excellent feedstock for a catalytic olefins facility. Table 2 provides a summary of some of the major components of the SUPERFLEX unit feedstock along with the corresponding octane values for reference.

Table 2
April 2008 SUPERFLEX Feed Composition

Component	C6C7 Fresh Feed (Wt %)	RON	MON
iso-Hexenes	17.1	96	81
Cyclo-Heptanes	12.4	84	63
1-Heptene	10.9	55	64
iso-Heptenes	10.8	92	80
Cyclo-Octanes	9.6	70	59
n-heptane	3.0	0	0
n-hexane	1.8	25	26
Sub-Total	60.8		

Design of the Sasol unit was based on pilot plant tests conducted by KBR on feed samples supplied by Sasol. Actual plant operation shows that propylene yields are slightly better than those

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predicted in the pilot tests, while ethylene yields are about the same as predicted. Product purities for polymer grade propylene and ethylene are very high.

While Sasol recovers the C2 and C3 olefins for petrochemical use, the SUPERFLEX naphtha product is used as a high octane motor gasoline component with reduced olefin content and increased octane. In this case, the C5-C9 Gasoline product is typically RON 97-98 and MON 82-84 as shown in Figure 6 below.

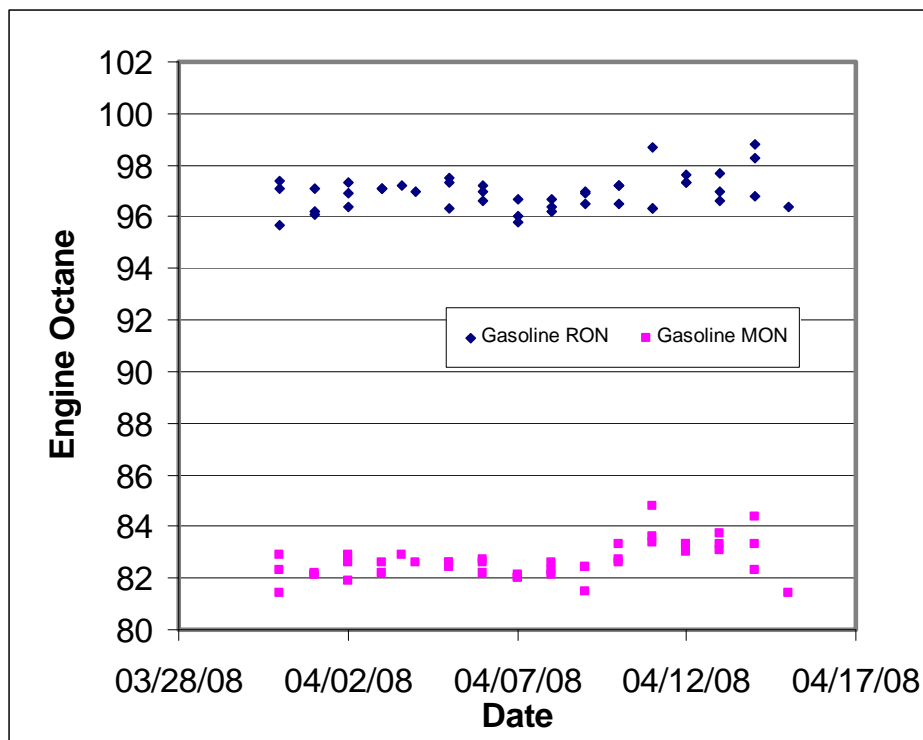


Figure 6 - Octane Numbers of Commercial Gasoline Product

Detailed Analysis of Naphtha from SUPERFLEX Pilot Plant

KBR took a further step to better understand the sensitivity of gasoline distillation on octane number. A SUPERFLEX pilot plant naphtha sample created from a Sasol feedstock was analyzed using gas chromatography to determine its molecular composition. Then a KBR proprietary database and correlations were applied to the molecular composition to estimate effect of different boiling point ranges on gasoline octane number.

The estimated Research Octane Number (RON) of the full range 410 °F (210 °C) end point sample was 97.6. This provides reasonable agreement with a 99.3 RON value measured in a test engine for the sample.

Figure 7 gives the molecular type distribution and octane numbers in the different boiling ranges. Above the figure, the percentage of each cut in the total sample is also provided for reference.

The boiling point range of IBP-160°F (IBP-71°C) has lower octane numbers compared to the cuts in the higher boiling point ranges mainly because about 22% percent of the paraffins in the IBP to 160°F (71°C) cut are low octane n-hexane along with some other unconverted feed components. Benzene is the only aromatic in the 160 to 200°F (71 to 93°C) range. It is clear that aromatic compounds are dominant species above a boiling point of 200°F (93°C). Toluene is the only aromatic in the 200 to 250°F (93 to 121°C) range, and the aromatics in the 250 to 300°F (121 to 149°C) range are 85% mixed xylenes. A total of just 7.3 wt% of total gasoline sample is in the boiling point range of 300 – 410°F (149 - 210°C). These higher boiling aromatic cuts have lower

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octanes number compared to the toluene and xylene rich boiling point ranges of 200 - 300 °F (93 - 149°C).

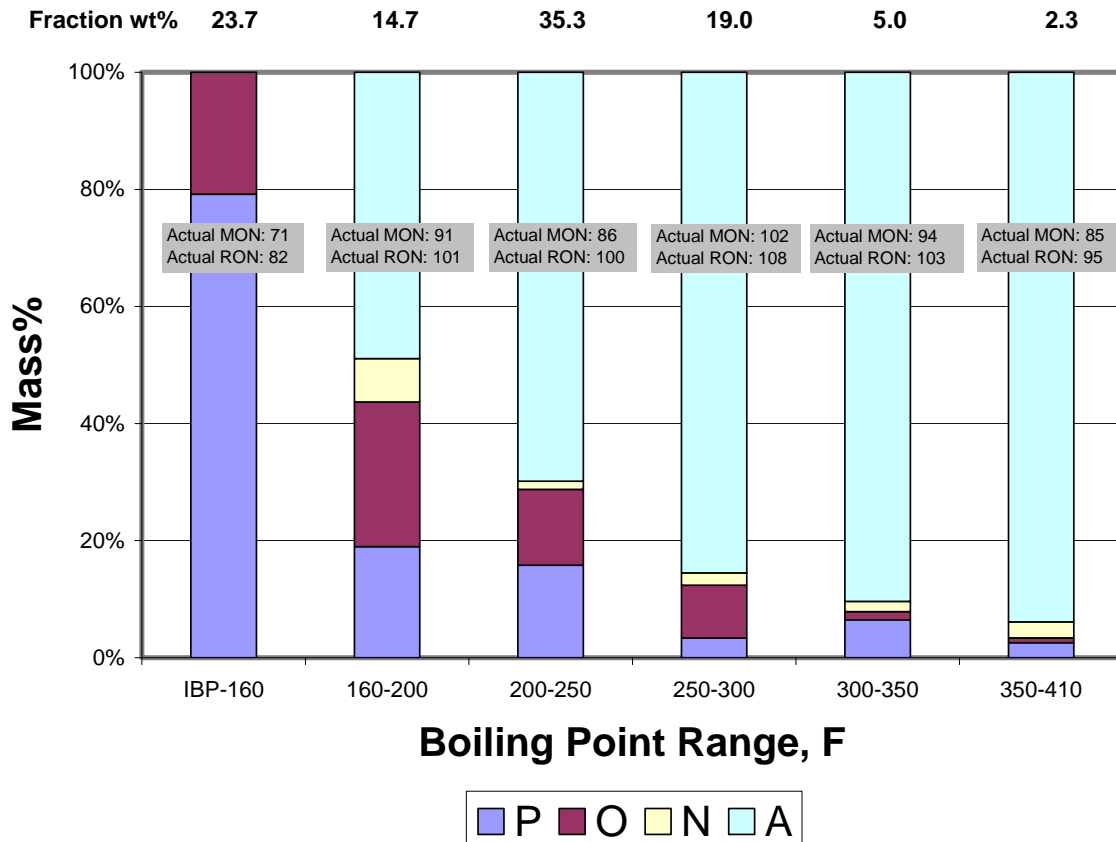


Figure 7 - Species Profile of Different Boiling Point Ranges of Gasoline Sample

The gasoline octane numbers for different boiling point ranges are shown in Figure 8 and Figure 9. The corresponding cumulative mass percentages for different boiling ranges are also shown in the figures. All sample points in both figures are weight average octane numbers for different boiling point ranges.

All boiling point ranges in Figure 8 have the same end point, while all boiling point ranges in Figure 9 have the same initial boiling point. Figure 8 shows what might be expected by removing the front end gasoline while Figure 9 demonstrates what might be expected by changing the end point of the gasoline.

Figure 8 shows that the octane number of the remaining gasoline increases as light components are excluded from the sample. However, the octane number reaches a peak at the boiling point range of 250 – 410 °F (121 - 210 °C) and then declines as the xylenes become excluded from the higher boiling ranges.

Figure 9 shows the percentage and low octane of the IBP to 160 °F (71 °C) boiling range fraction that depresses the octane of the full range gasoline sample.

Because of the very low octane and relatively high concentration of n-hexane (B.P. = 156 F (69 °C)) in the IBP to 160 °F (71 °C) boiling range and because of the relatively low mass percent of the naphtha boiling above 300 °F (149 °C), the figures show that adjusting the front end temperature of the gasoline distillation would be expected to have a larger impact on octane and yield than would be achieved by adjusting the gasoline end point.

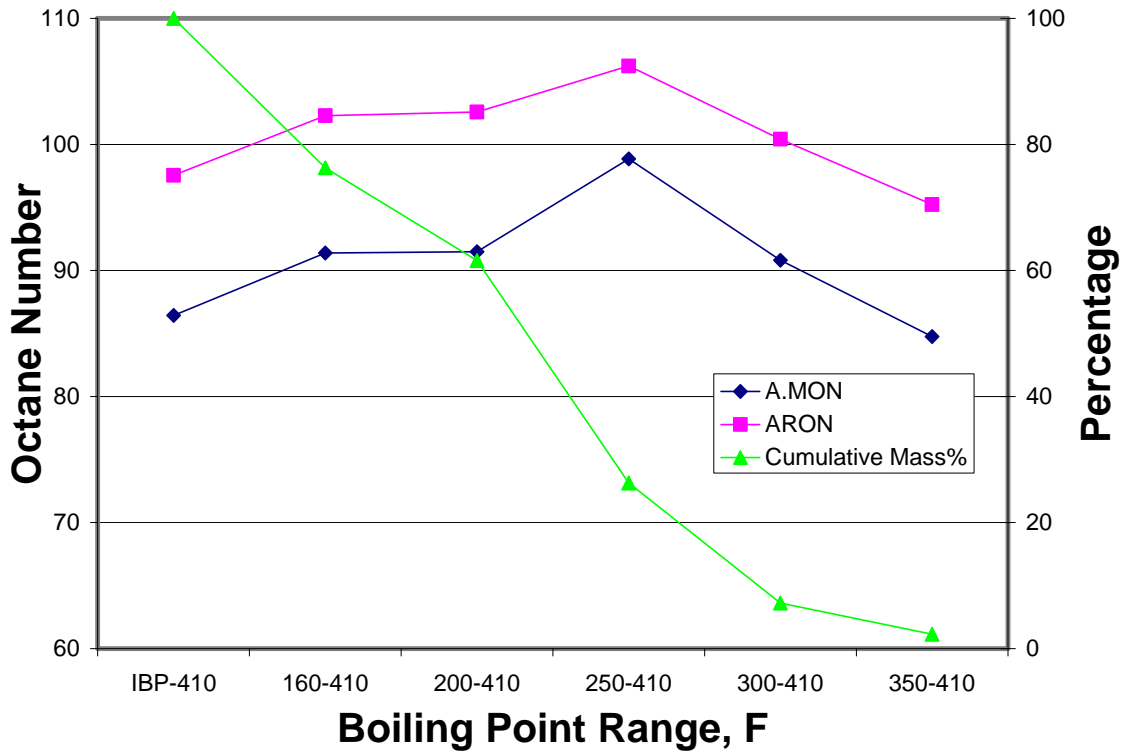


Figure 8 - Octane Number Profile of Different Gasoline Initial Boiling Points

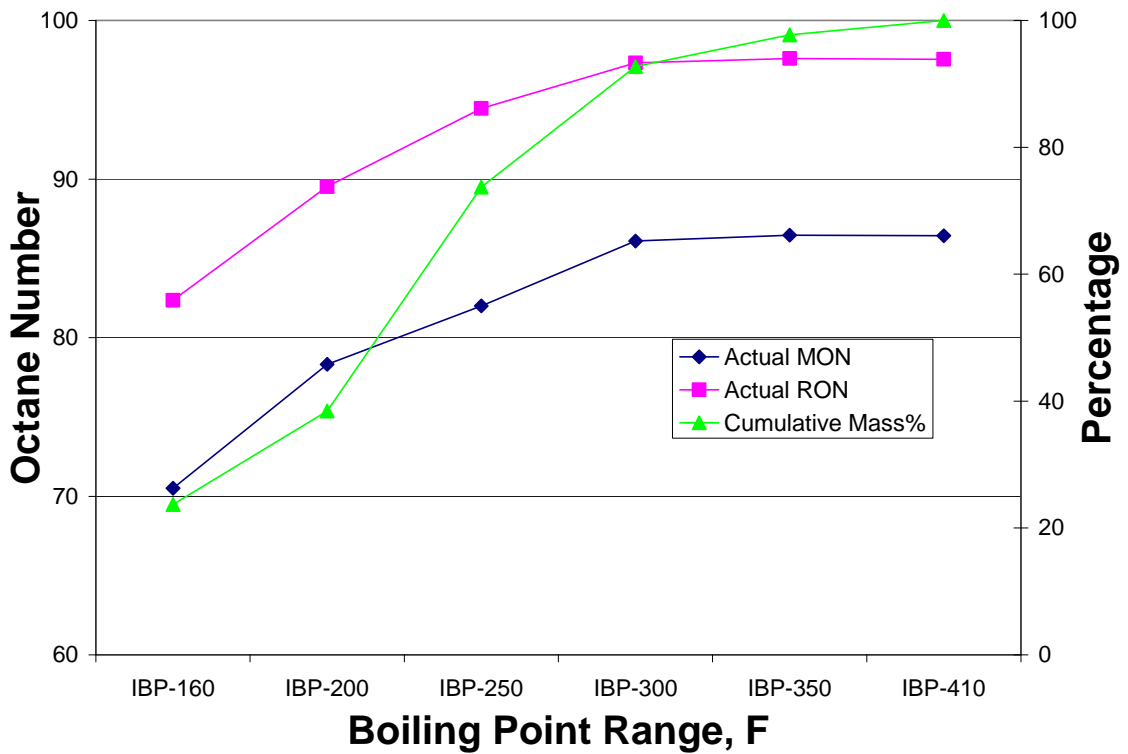


Figure 9 - Octane Number Profile of Different Gasoline End Boiling Points

MAXOFIN FCC

Figure 10: MAXOFIN™ FCC UNIT
Alternate Operating Modes

	Max C ₃ = (MAXOFIN)	Inter- mediate	Max Fuels (Traditional FCC)
Recycle ?	Yes	No	No
ZSM-5 ?	Yes	Yes	No
Riser Temp, °C	538 / 593	538	538
YIELDS, wt%			
C ₂ -minus	7.6	2.3	2.2
Ethylene	4.3	2.0	0.9
Propylene	18.4	14.4	6.2
Butylenes	12.9	12.3	7.3
Gasoline	18.8	35.5	49.8
Conversion	86.4	87.7	85.4
Coke	8.3	6.4	5.9



The proprietary MAXOFIN™ FCC Process licensed by KBR and ExxonMobil is depicted in Figure 10. The process is designed to maximize the production of propylene, ethylene, and aromatics from traditional FCC feedstocks and selected naphthas. The process allows increased production of petrochemical feedstocks relative to that produced by conventional FCC units while also allowing operation in a mode of maximizing FCC gasoline (motor fuel)

production. This is achieved by combining the effects of FCC catalyst, ZSM-5 catalyst additive and proprietary hardware, including a second high severity riser designed to crack surplus naphtha and C₄'s into incremental light olefins and aromatic naphtha¹⁰.

The higher severity second riser of a MAXOFIN FCC unit can process recycled light naphtha and C₄'s as well as C₄'s and low valued naphtha from elsewhere in the refinery complex, such as coker naphtha streams, and upgrade these streams into additional light olefins and aromatics. Products produced from both risers merge in the disengager vessel and travel as a common stream to the main fractionator⁹.

Olefinic streams such as coker naphtha convert most readily with the MAXOFIN FCC process. Paraffinic naphtha such as light straight run naphtha can also be upgraded in the MAXOFIN FCC unit, but to a lesser extent than olefinic feedstocks.

Refiners today require a technology that can respond to market demands. KBR's MAXOFIN can produce 20 wt% or more propylene from FCC feedstocks, yet still maintain the flexibility to produce fuels when market demand shifts. When the margin between propylene and fuels is low, the MAXOFIN FCC can operate as a conventional FCC to produce mostly gasoline, and when the demand of propylene is relatively high, the process can maximize propylene at the expense of gasoline. If markets require distillate and propylene, the MAXOFIN FCC Process allows refiners to run the primary riser at low conversion to preserve the distillate yield. Light naphtha and C₄s formed in the primary riser can be recycled at more severe conditions in the second riser to maximize conversion of these components to propylene.

While naphtha yield is limited in the high severity propylene operating mode, octane and BTX content of the MAXOFIN FCC naphtha product are improved greatly relative to that from a conventional FCC unit. In addition to the BTX, refiners can economically recover ethylene produced by the MAXOFIN FCC process for use as a high value petrochemical.

While the MAXOFIN FCC Process produces a highly aromatic gasoline product, light gasoline from the lower severity primary riser is rich in olefins and paraffins. Table 3 shows the composition by hydrocarbon type of a light naphtha stream produced by cracking VGO in a lower severity FCC pilot plant riser. When recycled to the second, higher severity riser, the olefins and paraffins in this light gasoline are largely converted to light olefins and additional BTX product.

Table 3
MAXOFIN FCC Circulating Pilot Plant Data

Light Naphtha Recycle Properties

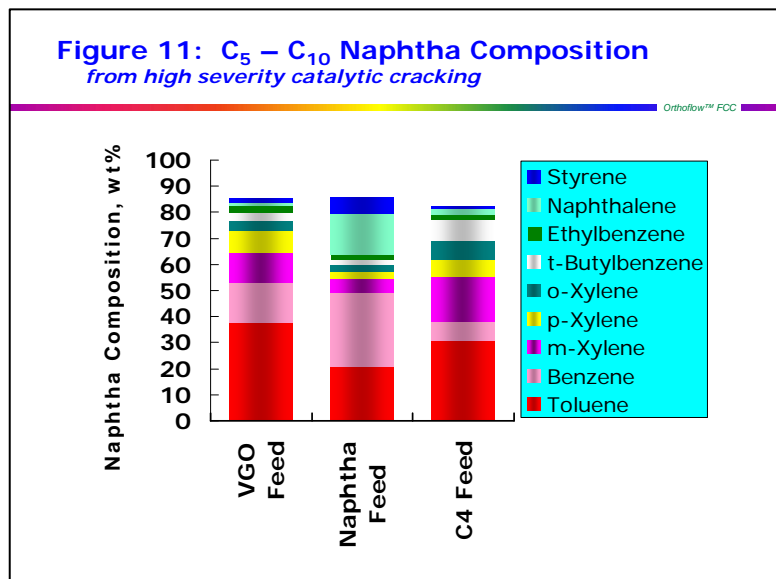
Distillation End Point 210 °F (99 °C) TBP

Hydrocarbon Type, wt%

Paraffins	5.6
Isoparaffins	44.9
Aromatics	2.8
Naphthenes	1.2
Olefins	45.6

What are the species that make-up most of the MAXOFIN FCC naphtha product? Referring to Figure 11 showing naphtha compositions produced in the pilot plant run at the higher severity second riser conditions, the simple answer is BTX¹¹. Benzene, toluene and xylenes constitute over 60 percent of any of these MAXOFIN FCC naphthas. With the economics of separating the naphtha into its higher value constituents becoming much more tenable, the Maxofin FCC Process may be considered an efficient means of BTX production. MAXOFIN is also expected to increasingly compete with catalytic reforming for supply of BTX to the petrochemical industry. The MAXOFIN FCC Process is also making inroads into the production of ethylene, a domain until now dominated by thermal cracking.

A traditional FCC unit produces a naphtha product suitable for blending into motor fuel. It is a mixture of so many different molecular species in small concentration that separating the FCC naphtha into discrete components for petrochemical use is typically not economic. It is better to simply burn the FCC naphtha as motor fuel. The MAXOFIN FCC Process changes this paradigm by producing a naphtha product that is very different from traditional FCC naphtha.



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Conclusions

Of all the "propylene-on-purpose" processes available, only the catalytic olefin cracking technologies accept low-value streams that are typically found in refineries.

Low-valued olefinic and/or low octane refinery streams, such as FCC C4's/C5's and light FCC and coker naphthas, can be upgraded to high-value propylene, ethylene, and aromatics by the SUPERFLEX process. Hydrotreatment for diene and/or nitrogen removal is not required, which is an important advantage. In addition to the generation of BTX for chemical manufacture, SUPEFLEX can be used to produce a naphtha product with superior value as a motor fuel relative to the olefinic, low octane feedstock.



Figure 12 – KBR Orthoflow™ FCC Unit

The SUPERFLEX reactor uses the same state-of-the-art KBR fluidized catalytic cracking technology that has been proven in many operating FCC installations, and uses the cost-effective KBR Orthoflow configuration. FCC operation is familiar to refiners, and thus SUPERFLEX is an on-purpose propylene technology that will have a very short learning curve for refinery operators.

Low octane paraffinic naphtha feeds can likewise be upgraded to produce C2 and C3 olefins and a higher octane naphtha product using the ACO process offered by KBR in association with SK Corporation.

A new generation fluid catalytic process, the MAXOFIN FCC Process, is expected to increase refinery propylene production for petrochemical applications while also becoming a major supplier of both ethylene and aromatics to the petrochemical industry.

MAXOFIN FCC offers refiners the flexibility to operate their FCC units in a traditional FCC mode for gasoline production or in a maximum petrochemical mode. The MAXOFIN FCC can therefore adapt to changing market conditions. In addition to the recycle of FCC C4 and light naphtha products, the second riser of the MAXOFIN FCC can accept low valued naphtha streams from elsewhere in the refinery.

In the integrated refinery/petrochemical complex, the use of KBR's SUPERFLEX, ACO and MAXOFIN FCC Process provides an internal supply of light olefins and aromatics, basic building blocks for the manufacture of today's petrochemicals.

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