Reactivity and Product Selectivity in FCC and Hydrocracking

Influence of Feedstock

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There is a wide variation in the concentration distribution of various compound types (paraffins, mono- and poly-cycloparaffins, mono- and poly-cyclic aromatics) in vacuum gas oils from various crude oils. This variation in the compound type distribution has a significant influence on the design and operation of secondary catalytic conversion processes such as FCC and Hydrocracking. Specifically, the reactivities of various compound types differ significantly from one another. The consequences of differing reactivities is highlighted by a kinetic analysis of available pilot plant data for FCC and Hydrocracking, both with amorphous catalysts. Paraffins, especially straight chain compounds, are shown to have a significantly lower reactivity than say cyclo-paraffins and, therefore, the overall reactivity of highly paraffinic feedstocks is lower than that for feedstocks with a low concentration of paraffins. The influence of lower reactivity of paraffinic feedstocks is highlighted by kinetic computer simulations of the product yields in a hydrocracking reactor processing Kuwait and Bombay High Vacuum Gas Oils. To achieve maximum middle distillates, operation with Bombay High (highly paraffinic) will require operation at higher severity (say with increased catalyst holdup) than with Kuwait VGO (lower paraffin content).
The rationale for using zeolite based catalysts for maximizing middle distillates, in both FCC and Hydrocracking operations, to cater for highly paraffinic feedstocks is underlined in the discussions.

Die Konzentrationsverteilung der verschiedenen Verbindungstypen (Paraffine, Mono- und Poly-Cycloparaffin, Mono- und polycyclischen Aromaten) in den Vakuumgasölen aus diversen Rohölen zeigt eine große Bandbreite. Diese Unterschiede sind bedeutend für die Konstruktion und den Betrieb von katalytischen Konversionsverfahren wie FCC und Hydrocracking. Die Reaktivität der Verbindungen unterscheidet sich stark voneinander. Die Konsequenzen der unterschiedlichen Reaktivitäten sind erläutert durch eine kinetische Analyse der Daten einer Pilotanlage für FCC und Hydrocracking bei Verwendung eines amorphen Katalysators. Da Paraffine (insbesondere geradkettige Paraffine) beträchtlich geringere Reaktivität aufweisen als Cycloparaffine, ist die Gesamtreaktivität der paraffinreicheren Feedstocks entsprechend geringer. Die Auswirkung geringerer Reaktivität der paraffinischen Feedstocks ist anhand einer kinetischen Computer-Simulation der Produktausbeute bei FCC und Hydrocracking der Kuwait- und Bombay-High-Vakuumgasöle. Für eine maximale Mitteldestillat-Ausbeute erfordert das Vakuumgasöl aus Bombay High (hochparaffinisch) höhere Strenge (severity) als das aus Kuwait (geringere Paraffingehalte). Die Überlegungen zur Verwendung von Zeolith-Katalysatoren zur Maximierung der Mitteldestillate aus hochparaffinischen Feedstocks sind in der Diskussion unterstrichen.

Introduction

The increasing demand for middle distillates in India places greater emphasis in refineries to convert residues to middle distillates. Currently, there are nine Fluid Catalytic Cracking (FCC) units in operation in India with a total intake capacity of 6.3 million tpa. A Hydrocracker project is underway at IOC Gujarat and the proposed new refineries at Karnal and Mangalore will also incorporate hydrocracking technology for middle distillates maximization. The optimization of existing FCC units, and the optimum design of future hydrocrackers, for achieving the best selectivities to middle distillates requires *inter alia* a proper realization of the influence of feedstock **type** on product selectivity. This feedstock influence is emphasized by a kinetic analysis of available pilot plant data on FCC and hydrocracking, both using amorphous catalysts.

Concentration Distribution of Compound Types in Vacuum Gas Oils

The feedstocks for FCC and hydrocracking are Vaccum Gas Oils (VGO) typically boiling in the range 371–538 °C (i.e. 700–1000 °F). *Figure 1* presents a summary of data, culled from various sources, on the compound type distributions in VGO's from different crude oils. The concentration of paraffins (normal and branched) varies from 28% for Lt Arab VGO to about 52% for Bombay High VGO. A further remarkable feature of Bombay High VGO is that the paraffins are predominantly (about 90%) of the straight chain type. The information in *Fig. 1* will be discussed later.

Kinetic Analysis of Fluid Catalytic Cracking

In a particularly instructive paper *White* [1] has presented some very useful data on the crackability (or reactivity) of various compound types in an FCC pilot plant reactor using amorphous catalyst. Various compound types were isolated and individually used as feedstocks. *White* was interested in a gasoline maximization mode of operation i.e. maximization of products boiling in the range C_5 – 221 °C (C_5 – 430 °F). He has presented data on the conversions obtained to the fractions C_5 – 430 °F and C_4 –; no kinetic interpretation was presented. The authors have reanalysed the data of *White* to obtain rate constants for cracking of various compound types. The percent unconverted 430 F+ fraction for various compound types are presented in *Figure 2* as a function of the reaction severity; the reaction severity is defined as:

$$\text{Severity} = \frac{\text{Surface Area, m}^2/\text{g}}{100\ \text{WHSV}} \tag{1}$$

where WHSV is the Weight Hourly Space Velocity. For a catalyst with a surface area equal to 100 m^2/g the severity is simply the space time, measured in hours. From the various straight lines for different compound types, the first order reaction rate constant, for conversion of 430 F+ material into 430 F– material, can be obtained; these data are presented in *Figure 3.* The rate constants are for the reaction sequence:

$$\text{Compound Type 430 F}^+ \xrightarrow{k_1} \text{430 F}^- \text{ product} \tag{2}$$

and the values of k_1 have been calculated for a catalyst with a surface area of 100 m^2/g. It is evident that the rate constants for the different compound types vary greatly; the normal paraffins have a rate constant k_1 = 0.33/h while the rate constant for polycyclicparaffins is 1.7/h, about *five times* higher than normal paraffins. A further point to note is that polycyclic aromatics are even less reactive than normal paraffins and such condensed structures end up either in clarified oil or form coke.

A closer examination of data on the distribution of C_5–430 F and C_4– products shows that cracking of normal paraffins yields a proportionately higher amount of lighter C_4– material. It may be concluded that highly paraffinic stocks such as Bombay High require a higher severity for cracking of the sluggishly reactive normal paraffins than would be required for cracking of other feedstocks with a lower paraffinic content. The wide differences in the reactivities of compound types in the feedstock, portrayed by the rate constants in *Fig. 3,* poses special problems if middle distillates should be maximized. At the level of severity required to crack the normal paraffins, the highly reactive compound types such as cycloparaffins and monocyclicaromatics will suffer *over*cracking to undesirable lighter products. The problem is thus one of choosing the optimum operation level (i.e. severity) to achieve the best product selectivities. If all the compound types were equally reactive this particular problem of selection of the optimum severity level would not arise. The above kinetic analysis provides a background for the observed high bottoms make (CLO) when processing Bombay High VGO in FCC operations geared towards middle distillates [2]. With Bombay High VGO a much higher clarified oil (CLO) yield was observed in operations, at the expense of total cycle oils (TCO) *(Figure 4).* More interestingly, due to the sluggish reactivity of n-paraffins present in the BH VGO, these compounds find their way, largely unreacted, in the CLO. Consequently the CLO density is very low (hydrogen rich compounds present in CLO) and has a high pour. See *Figure 5* for verification of this in actual operations.

The rationale for incorporating a proportion of zeolite crystals in the catalyst follows logically from the foregoing discussion. Due to the small channel dimensions of the zeolite catalysts the bulkier molecules such as polycycloparaffins are excluded from entry and the cracking rate constant for "narrow" molecules such as normal paraffins is *higher* than for such bulky structures [3]. Incorporation of zeolites into FCC catalysts will therefore enhance the reactivity of the normal paraffins with respect to bulky structures. It must, however, be emphasized that the role of the matrix is particularly important in middle distillates maximization because the bulkier molecules such as cycloparaffins and cyclic aromatics are to be "catered" for by the matrix; these molecules cannot enter the zeolite cages. Adequate matrix activity must be ensured in order to ensure good bottoms conversion. This is highlighted in the data presented by *Maselli* and *Peters* [4] (see *Figure 7).*

Zeolite based catalysts have been used in Indian refineries and the results with BH VGO, for example at Madras Refineries Limited [5] and Mathura Refinery [6], show improved bottoms conversion when switching over from amorphous catalysts, even partly. *Figure 8* summarizes the results of switchover to zeolite catalysts at Mathura Refinery [6]. Significantly improved cycle oil yield at the expense of CLO may be observed.

Kinetic Analysis of Hydrocracking

With the increasing importance of hydrocracking in the Indian context it is worthwhile examining whether. We should expect a parallel experience to FCC in processing a highly paraffinic stock like Bombay High. There is little published information on this subject which can be used directly for the purpose at hand. One very useful source of information is the paper of *Bennet* and *Bourne* [7], who have published pilot plant data with Kuwait Vacuum Gas Oil. Two special features of their work are that product yields at four different space velocities are given and that the 700 F+ unconverted "residue" was characterised for obtaining the compound type distributions. They did not attempt, at least in the paper as published, to obtain a kinetic interpretation of their own data. The authors of this present paper have reanalysed the data of *Bennet* and *Bourne* [7] with the objective of obtaining a usable kinetic model for hydrocracking with a view to selection of optimum conditions for design and operation.

From the data on the concentrations of various compound types in the unconverted 700 F+ residue, the plots were constructed as shown in *Figure 9,* which must be considered as a parallel to the one presented in *Figure 2* for FCC. It is heartening to note the straight lines on the semi-logarithmic plot suggesting a pseudo-first order reaction rate constants for the various compound types. These rate constants, obtained by a regression analysis, are shown in *Figure 10.* The paraffins (here both

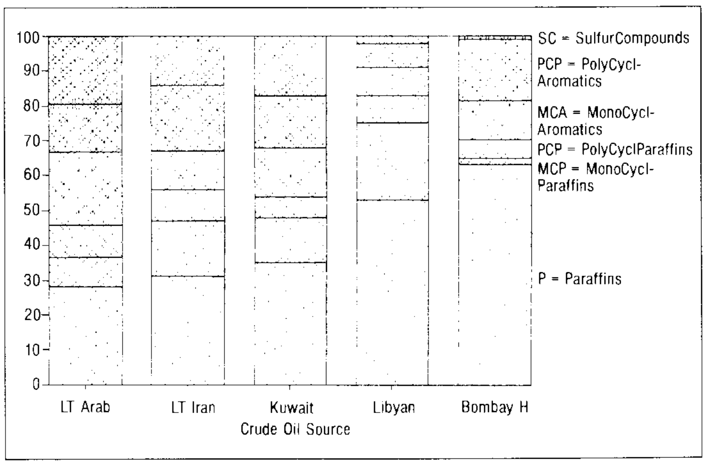


Figure 1: Compound Type Composition Distributions (in %wt) in Vacuum Gas Oils from various Crude Oils.

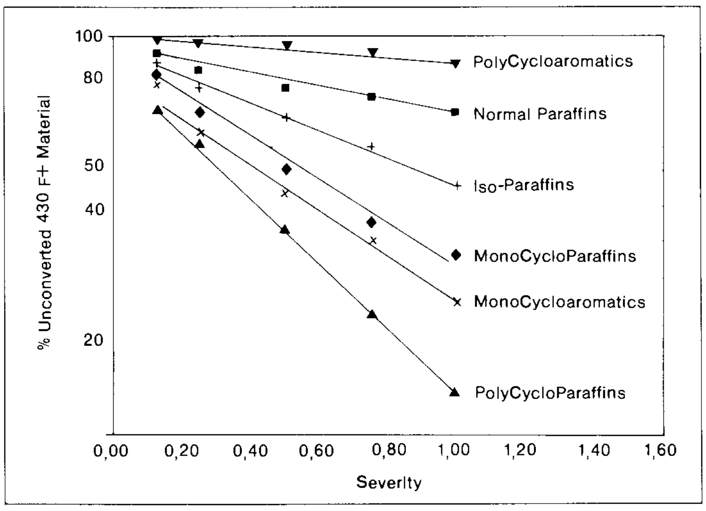


Figure 2: Effect of various compound types on conversion into 430 F– material. Data taken from White, 1968. Severity is defined as [(Surface Area, m^2/g/100]/WHSV. The vertical axis (log scale!) is the %wt of unconverted 430 F+ material.

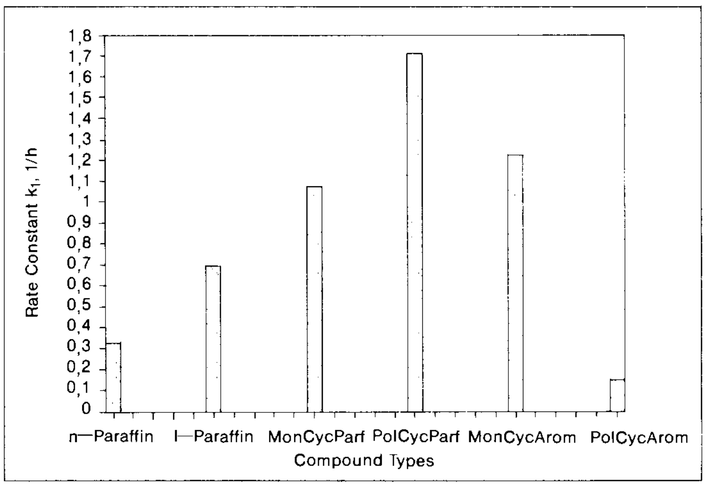


Figure 3: First-Order Rate Constants, k_1, $_1h^{-1}$, for various compound types. Calculated from data in Fig. 2. Surface area is assumed to be 100 m^2/g for the calculations presented in this Figure.

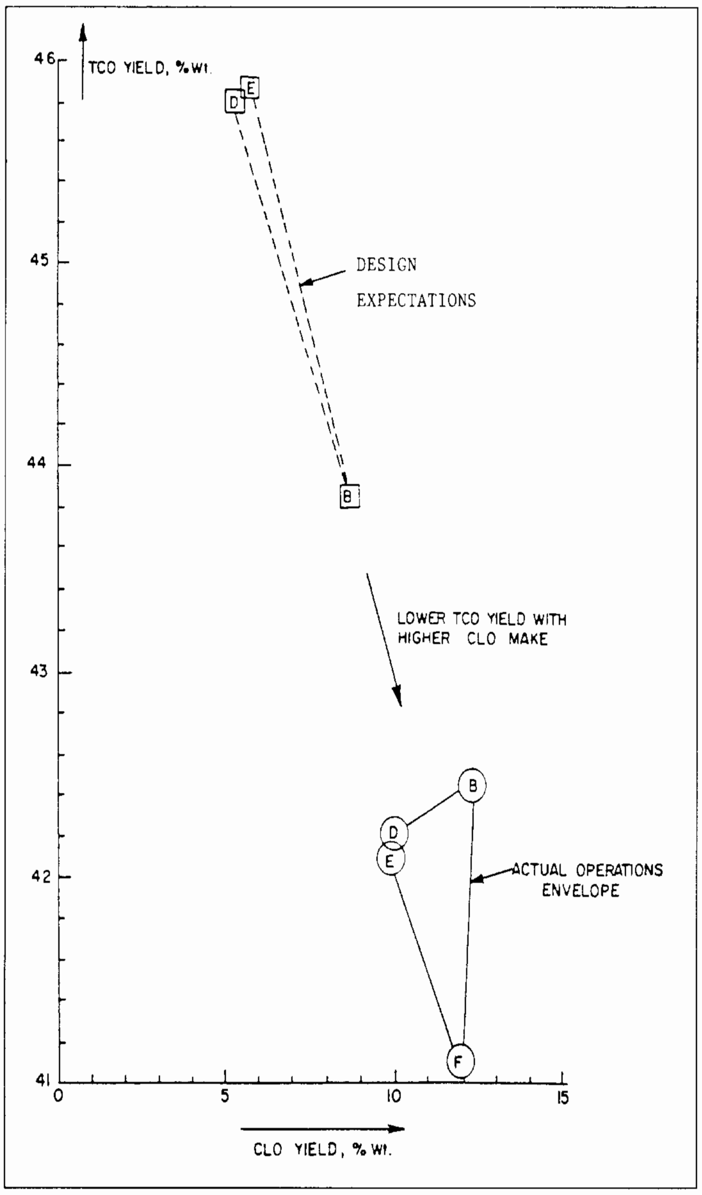


Figure 4: Comparison of actual operations vs design expectations for FCC operations with BH VGO with amorphous catalysts. Letters B, D, E, F denote various refinery units in India. TCO = Total Cycle Oils; CLO = Clarified Oil.

normal and branched compounds have been lumped) are seen to have a rate constant of 0.52/h, about *four* times smaller than the rate constant for cracking of polycycloparaffins. This is in line with the results for FCC shown in *Figure 3*. An important difference between the reactivities of FCC and Hydrocracking is noticeable from a comparison of *Figures 3* and *10*. In FCC operations, polycyclic aromatics are difficult to crack; not so in hydrocracking where these compound types can be hydrogenated without difficulty, and further cracked. This is reflected in the fact that the rate constant for hydrocracking of polycyclicaromatics is about the same for polycycloparaffins and monocyclic aromatics. Also, in hydrocracking, cleavage of the C-S bonds takes place quite easily and this is reflected in the fact that the rate constant for hydrocracking of sulphur compounds is 2.92/h, the highest for any compounds type. In a trickle bed hydrocracker the sequence of reactions taking place down the bed will reflect the relative reactivities and, for an amorphous catalyst, this sequence is shown schematically in *Figure 11*. Reactions involving paraffins take place at the very bottom of the reactor, as is to be expected. If the reactor is, for some reason, underdesigned with respect to catalyst volume, the hydrocracking of paraffins will not be effected, leading to loss of potential middle distillates yield.

We now develop a usable model kinetic model for hydrocracking. Let R_i represent the %wt of compound type i in the 700 F+ unconverted material. The first order rate constants for each of the individual compound types is given in *Figure 10*. The decay of each of these species is will follow the first order process

$$R_i = R_{io} \exp(-k_{1i} t) \tag{3}$$

where k_{1i} is the first order rate constant for species i and t is the space time. Summing Eq. (3) over all the various compound types we obtain the following expression:

$$\begin{aligned} R &= \sum_i R_i = [\sum_i R_{io} \exp(-k_{1i} t)] \\ &= [\sum_i R_{io}] \exp(-k_1 t) = R_o \exp(-k_1 t) \end{aligned} \tag{4}$$

where k_1 is the lumped first order decay constant for the 700 F+ material as a whole; this rate constant can be calculated from the rate constants of the individual compound types; cf. Eqs. (3) and (4):

$$k_1 = -\ln\{\sum_i [R_{io} \exp(-k_{1i} t)]/R_o\}/t \tag{5}$$

which expression shows that the lumped rate constant for the 700 F+ material, as a whole, depends on the space time. For a space time t = 2 h, the lumped rate constant works out to be 0.96/h and has been portrayed in *Figure 10* along with the rate constants for the individual compound types. The advantage of knowing the individual rate constants for the various compound types is that we will be able to *calculate* the decay constant for the 700 F+ material for any feedstock for which we know the compound type distribution; we shall return to this point later in the paper.

Having considered the (hydro)cracking kinetics the 700 F+ material, we now turn our attention to the focal point of the paper: maximization of middle distillates, i.e. compounds boiling in the range 300–700 °F. Middle distillates formed from cracking of 700 F+ have the possibility to (over)crack to lighter materials boiling below 300 °F, i.e. gasoline and gases. We may set up the following kinetic expression:

$$\begin{array}{ccccc} 700\ \text{F+} & \xrightarrow{k_1} & 300\text{–}700\ \text{F} & \xrightarrow{k_2} & 300\ \text{F–} \\ \text{VGO} & & \text{middle distillates} & & \text{gasoline + gases} \end{array} \tag{6}$$

where k_2 represents the first order rate constant for over cracking of the middle distillates to gasoline and gases. *Bennet and Bourne* [7] have also given the yields of the products 300–700 F and 300 F– at the four different space velocity conditions for Kuwait VGO *(Figure 12)*. *Figure 12* also shows the kinetic (computer) model predictions of the product yields taking $k_1 = 0.96$/h, as calculated above from the rate constants of the individual compound types, and with the overcracking rate constant ratio: $k_2/k_1 = 0.26$. The overcracking rate constant ratio k_2/k_1 was estimated by statistical data fitting. The unconverted 700 F+ "residue" yield was calculated from Eq. (4) and the product yield of 300–700 F was calculated from the following expression:

$$\text{Yield 300–700 F} = \frac{R_o}{(k_2/k_1 - 1)} [\exp(k_1 t) - \exp(-k_2 t)] \tag{7}$$

The 300 F– Yield can be calculated from:

$$[\text{Yield 300 F–}] = [R_o] - [\text{Yield 700 F+}] - [\text{Yield 300–700 F}] \tag{8}$$

It can be seen from *Figure 12* that the kinetic model predictions of the yields, especially for the middle distillates production, are very good. In particular the maximum in the middle distillates production at a space time t = 1.7 h is anticipated quite well by the simple kinetic model developed here. The results also validate, if indirectly, the lumping procedure we have proposed to calculate the rate constant of the 700 F+ material from the compound type distributions.

Emboldened by the success we have achieved with the simulation of the behaviour of Kuwait VGO, let us attempt to predict the expected performance with Bombay High VGO. We calculated the decay constant k_1 for the 700 F+ Bombay High VGO

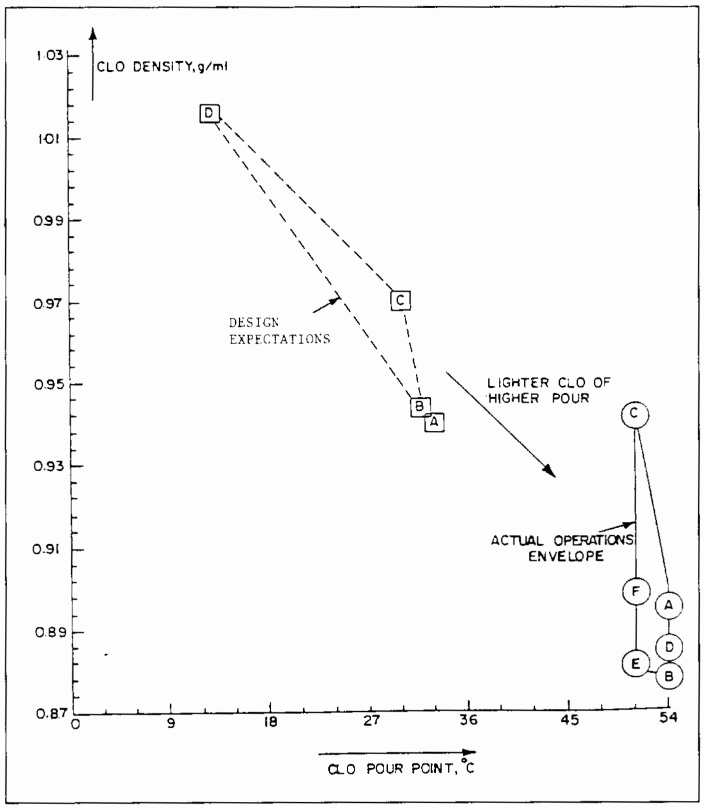


Figure 5: Comparison of actual operations vs design expectations for FCC operations with BH VGO with amorphous catalysts. Letters A, B, C, D, E, F denote various refinery units in India.

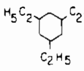
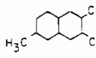
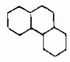
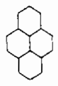
REACTANT HYDROCARBON	$SiO_2-Al_2O_3$ RATE CONSTANT	REHX RATE CONSTANT	RATIO $K_{REHX}/K_{SiO_2-Al_2O_3}$
$n\text{-}C_{16}H_{34}$	60	1000	17
	140	2370	17
	190	2430	13
	205	953	4·7
	210	513	2·4

Figure 6: Comparison of reactivities on amorphous and zeolite catalysts Cracking Rate Constants at 482 °C (units: h^{-1}) for both amorphous $SiO_2 \cdot Al_2O_3$ and Zeolite REHX catalysts for various compounds types. Data from *Nace* [3].

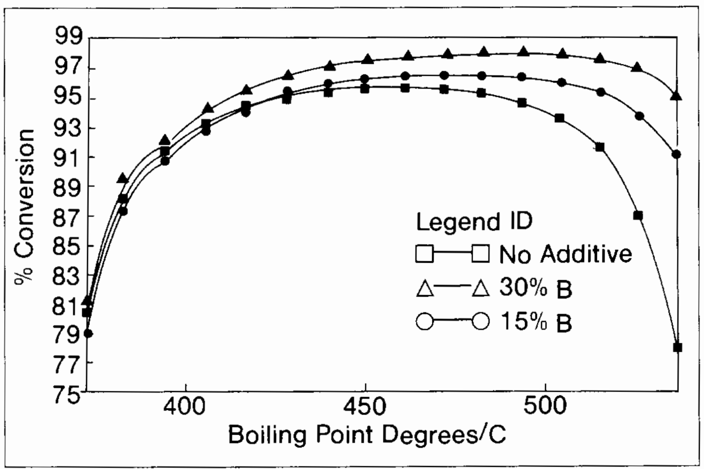


Figure 7: Influence of activity of matrix on bottoms conversion. Data from *Masel-li* and *Peters* [4].

Sl. No.	*Parameter*	*Unit*	*Winter*		*Summer*	
			Amorphous	*Zeolite*	*Amorphous*	*Zeolite*
			8.12.84	*14.5.87*	*7.8.85*	*15.5.87*
1.0	**Yield Pattern**	Wt%				
1.1	Gas		3.20	4.52	4.47	4.65
1.2	LPG		10.59	10.35	10.89	10.32
1.3	Gasoline		19.52	19.77	17.86	20.63
1.4	TCO		45.41	48.14	47.58	51.90
1.5	CLO		15.19	12.32	12.61	7.94
1.6	Coke		4.27	4.58	4.48	4.65
1.7	Balance		1.82	0.31	2.11	(−)0.09
2.0	**Operating Condition**					
2.1	Feed Rate	M^3/Hr	180.1	172.5	164.3	166.0
2.2	Comb. Feed Ratio	—	1.28	1.38	1.27	1.42
2.3	Reactor Temp.	°C	493	492	489	492
2.4	Comb Feed Temp.	°C	328	338	337	337
2.5	Heater Inlet/Outlet	°C	292/335	282/336	287/347	290/336
2.6	Regen. Dense Temp.	°C	652	653	663	647
3.0	**Feed/Product Quality**					
3.1	Feed, FBP	°C	535	540	552	550
3.2	TCO, Flash	°C	31.5	31.0	32.0	32.5
	Pour	°C	(+)3	(+)3	(+)12	(+)12
3.3	Gasoline, RON	—	92.5	91.9	—	91.6
4.0	**Economic Benefit**		Considering 7 months summer and 5 months winter operation.			
	(Per Annum)		*Amorphous*		*Zeolite*	
4.1	Cost of catalyst	Rs.	Base		3,145,200	
4.2	**Operating Cost**					
4.2.1	Catalyst loss	Rs.	Base		(−) 38,812	
4.2.2	Heater duty	Rs.	Base		143,336	
4.2.3	Total	Rs.	Base		104,526	
4.3	**Benefits**					
4.3.1	Yield improvement	Rs.	Base		37,600,000	
4.3.2	Octane tons	Rs.	Base		13,923,185	
4.3.3	Total	Rs.	Base		51,523,185	
4.4	**Net Benefit** (4.3–4.2)	Rs.	Base		51,418,659	

Figure 8: Benefit of switchover from amorphous to zeolite catalysts at Mathura Refinery [6].

feed from the compound type distributions given in *Figure 1*; this first order decay constant works out to be 0.73/h (compared to 0.96/h for Kuwait VGO). Further if we assume that the overcracking ratio k_2/k_1 remains the same for both feedstocks (= 0.26 as estimated for Kuwait VGO), then we are in a position to compare the product yields from Bombay High and Kuwait *(Figure 13)* for the computer simulation results. It shows that for achieving maximum middle distillates with Bombay High VGO we need to operate with a space time t = 2.4 h, as compared to 1.7 h required for Kuwait VGO. For a given throughput, the catalyst holdup required with Bombay High VGO, will be about 40% higher than for Kuwait VGO. If a reactor designed for Kuwait VGO is used to process Bombay High VGO, the yield of middle distillates would be less than potentially achievable by operating at a higher severity (meaning catalyst holdup); this situation can be rectified, at least to some extent, by operating at a higher temperature with Bombay High VGO feedstock.

Rationale for Zeolite Based Catalysts for Hydrochracking

Exactly parallel to the discussions for FCC, we can conclude that to overcome the problem posed by the different reactivities

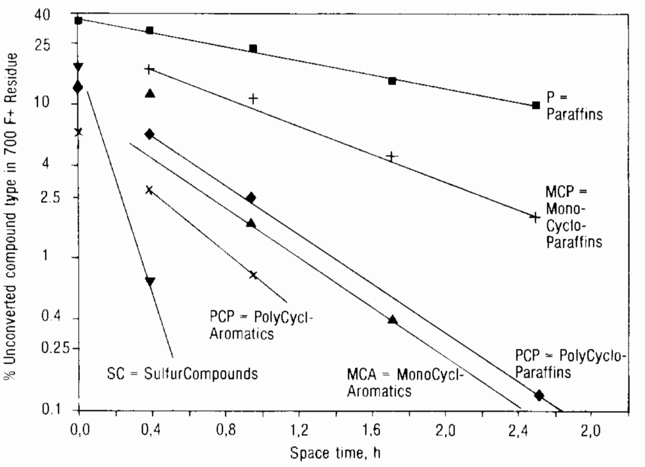


Figure 9: Hydrocracking of Kuwait Vacuum Gas Oil – % unconverted 700 F+ residue as a function of space time for various compound types (Pilot data from British Petroleum).

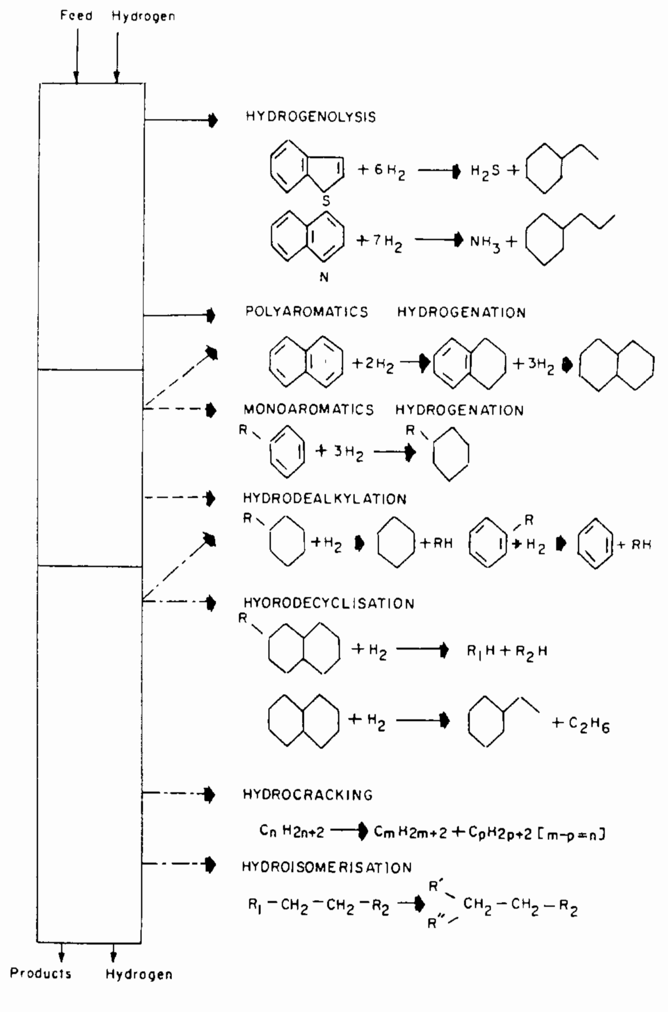


Figure 11: Sequence of reactions taking place down the height of a trickle bed hydrocracker employing amorphous catalyst.

of compound types in VGO, particularly the lesser reactive paraffins, we need to incorporate zeolite crystals in the hydrocracking catalyst formulation. These zeolites will "cater" for the "narrow" molecules, such as normal paraffins, whereas the matrix should be able to cope with the bulkier, but easier to crack, structures. Y-Zeolite is most appropriate for hydrocracking, and FCC. There are two aspects of zeolite design which are important for middle distillates maximization:

(i) there is an optimum number of acid centres *(Figure 14)*, and (ii) the size of the zeolite crystals. To prevent overcracking to gasoline and gases, the size of the crystals has to be small. There is, however, a lower limit to the crystal size because of the danger of thermal degradability (breakdown of structure). The Y-zeolite for hydrocracking catalyst needs to be "modified" to reduce the acid site density to a level optimum for middle distillates maximization *(Figure 15)*. This modification, or dealumination, will also create a secondary pore structure *(Figure 16)* which is extremely helpful in allowing bulkier molecules to enter the pores of the zeolite. One may argue, of course, that the function of the secondary pore structure could just as well be

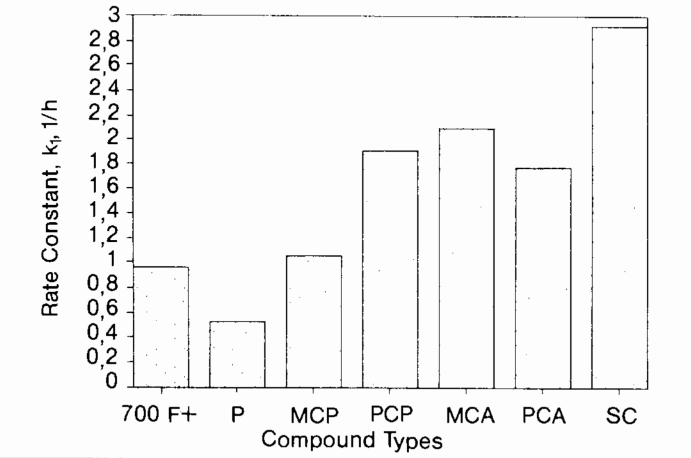


Figure 10: First Order Rate Constants in Hydrocracking, k_1, h^{-1}, for various compound types calculated from data in Fig. 4. The rate constant for the 700 F+ residue has been calculated using the lumping technique described in the text (Pilot data from British Petroleum).

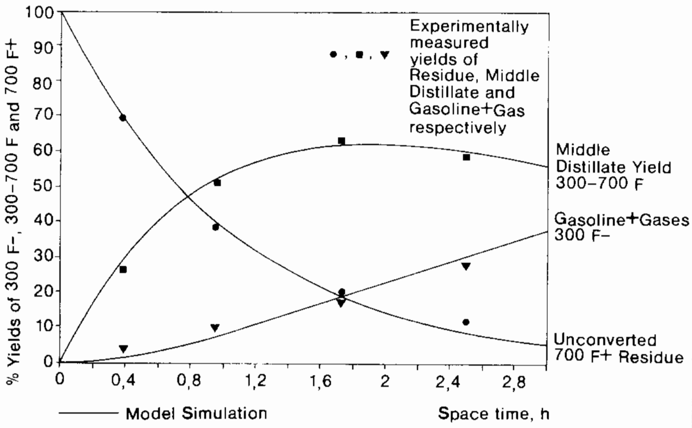


Figure 12: Comparison of hydrocracking model simulations (k_1 = 0.96 h^{-1}, k_2/k_1 = 0.26) with measured pilot plant data (from British Petroleum) with Kuwait Vacuum Gas Oil.

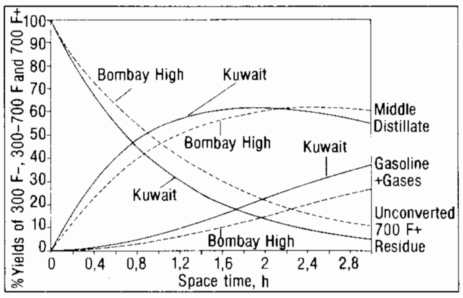


Figure 13: Simulations of product distributions expected in hydrocracking (with amorphous catalyst) of Kuwait and Bombay High Vacuum Gas Oils.

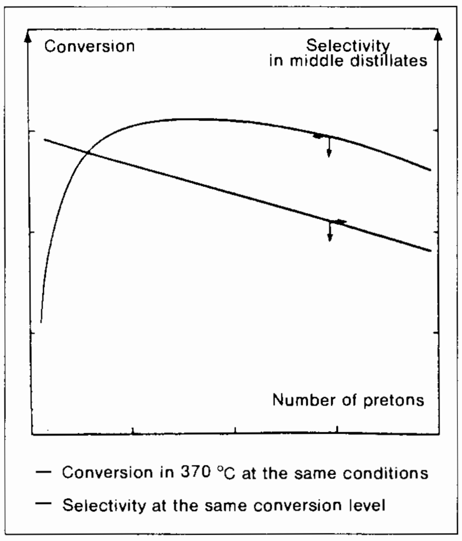


Figure 14: Showing that there is an optimum acidity level for zeolites in hydrocracking for maximum middle distillates.

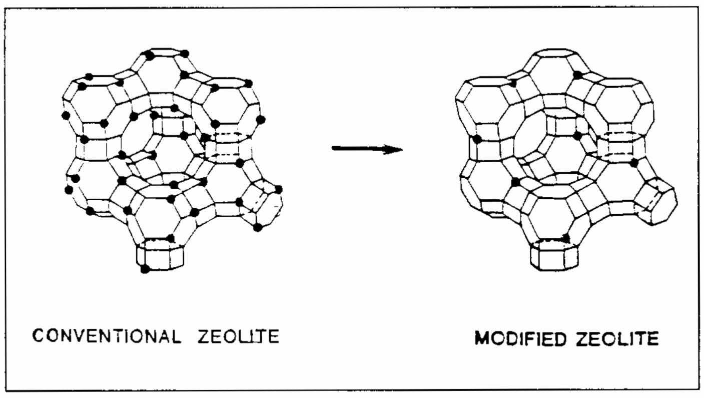


Figure 15: Reduction of acid site density in zeolites ("modifications") by dealumination.

undertaken by the (amorphous) matrix but the advantage of incorporation of a secondary pore structure within the zeolite crystals is that this would allow the use of larger zeolite crystals which would otherwise (i.e. in absence of the secondary pore structures) offer undesirable diffusion resistance. The creation of these secondary pore structures in the zeolite crystals while maintaining structural integrity under actual reaction conditions offers many challenges to be hydrocracking catalyst developper.

Concluding Remarks

1. The concentration distribution of various compound types have a significant influence in the reactivity and selectivity (to middle distillates) in FCC and hydrocracking with different feedstocks.

2. Paraffins, particularly straight chain compounds, have a significantly lower reactivity than say cycloparaffins or monocyclic-aromatics in both FCC and hydrocracking reactions, with amorphous catalysts. This fact is particularly important in the Indian context because Bombay High has an unusually high concentration of normal and iso-paraffins in the VGO range. This leads to poor reactivity, as already experienced by FCC's operating with amorphous catalysts.

3. To counter the problem posed by the low reactivity of paraffins, zeolite crystals, of the Y-type (suitably modified by dealumination to reduce the acid site density), need to be incorporated into both FCC and hydrocracking catalysts.

4. A simple kinetic model analysis of the hydrocracking reactor shows the importance of proper choice of the operating severity (e.g. space time, temperature), depending on the nature of the feedstock. There is much to be gained, or lost, in middle distillates yield by the choice of the operating conditions.

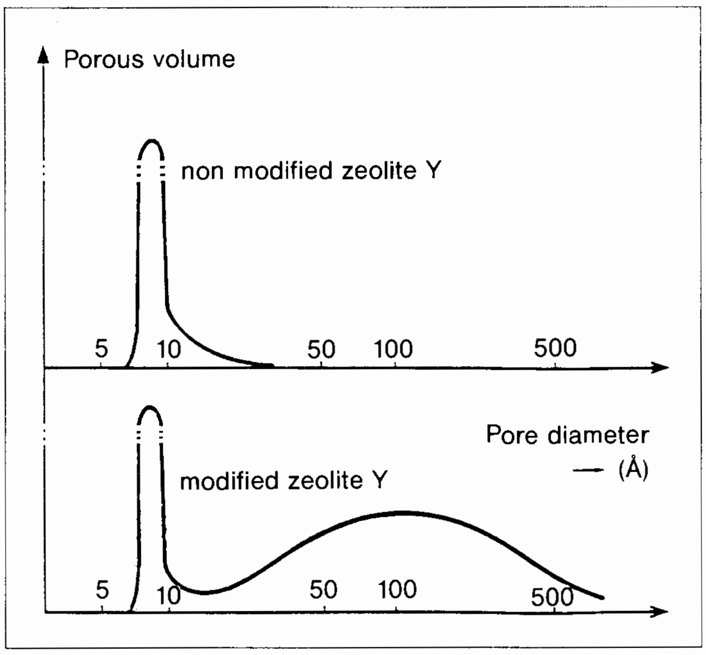


Figure 16: Creation of a secondary pore structure as a result of desalumination.

Nomenclature

k_1	First order rate constant for cracking of feedstock [1/h]
k_{1i}	First order decay rate constant for cracking of compound type i in 700 F+ VGO
k_2	Rate constant for overcracking of middle distillates to lighter products: gasoline + gases [1/h]
LHSV	Liquid hourly space velocity [vol Feed/vol Catalyst/h]
R_i	weight of compound type i as %wt of R_o
R_{io}	Initial % wt of compound type i in 700 F+ material
R_o	Initial % weight of 700 F+ material = 100%
R	Weight of unconverted 700 F+ as % of R_o
WHSV	Weight hourly space velocity [Wt Feed/Wt Catalyst/h]

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