

Recycling Hydrocarbon Cuts into FCC Units

María L. Fernández, Ainhoa Lacalle, Javier Bilbao, and José M. Arandes*

*Departamento de Ingeniería Química, Universidad del País Vasco,
Apartado 644, 48080 Bilbao, Spain*

Gabriela de la Puente and Ulises Sedran

INCAPE (FIQ, UNL-CONICET), Santiago del Estero 2654, (3000) Santa Fe, Argentina

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A bench scale reactor, which is a simulator of FCC riser units, has been used to study the upgrading of low value hydrocarbon cuts (a heavy coker naphtha and a visbreaker naphtha) by catalytic cracking of a mixture of these cuts (20 wt %) with the conventional feedstock (vacuum gas oil). The results of yield and composition of gasoline have been compared with those obtained by catalytic cracking of vacuum gas oil and with those obtained following other upgrading strategies for these cuts, such as their direct catalytic cracking or their blending with the gasoline stream of the FCCU. In general terms, increases in gasoline yields oppose to losses of its quality. The results show that the cracking of the heavy paraffins contained in these naphthas is inhibited by the adsorption of heavy olefins present in these cuts. Coke yields are not higher than the ones obtained when cracking standard VGO, and then the impact on units' heat balances would be negligible.

Introduction

One of the reasons for the continuous modifications that are introduced in the operation of FCC units (process conditions, catalyst or feedstock composition) are the requirements imposed on product quality, particularly of motor fuels.¹ The new regulations dictate the modification of gasoline composition. Changes in feedstock composition are very important because they have an impact on the gasoline formulation and on the overall operation of the refinery, which has consequences on the commercial viability.²

Those changes in the composition of the feed to FCC units might also be imposed by the need to increase refinery profits by processing low value hydrocarbon cuts, like coker or visbreaker naphthas, or various column bottoms.³ These cuts are not usually a good choice for direct blending into the gasoline pool because of their very low octane number or unsuitable composition (e.g., high proportion of *n*-paraffins or olefins), and their upgrading may entail high costs.^{4,5} On the other hand, increases in the yields of important products such as gasoline can be envisaged, but in a situation in which

a trade-off between yields and quality should be reached. Certain other factors may lead to the use of new feedstocks in FCC, such as the possibility of incorporating products of the pyrolysis of waste plastics,^{6–9} or even their direct dissolution into conventional VGO feeds.^{10–12}

The modification in yields and selectivities produced by the incorporation of a heavy coker naphtha and a visbreaker naphtha in a typical VGO feedstock (standard operation) was studied in this work.

The present state of FCC research requires a proper experimental methodology in order to obtain, quickly and accurately, the information needed for the simulation of commercial units for operating with new feedstocks. Rather than the more conventional micro-activity test, MAT,^{13,14} which is widely used and subjected to modifications in order to correct certain original deficiencies, the laboratory Riser Simulator reactor^{15,16} was used in this work.

* Author to whom all correspondence should be addressed. Telephone: 34-94-6012511. Fax: 34-94-4648500. E-mail: iqparej@lgdx04.lg.ehu.es.

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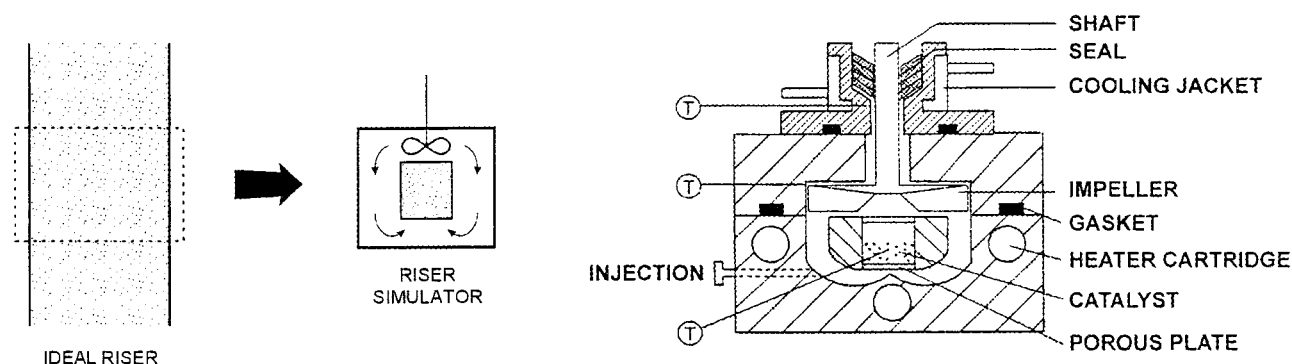


Figure 1. Schematic representation of the Riser Simulator Reactor.

Experimental Section

Reaction Equipment and Product Analysis. The Riser Simulator reactor is a batch unit with internal gas recirculation, where the catalyst bed is fluidized.¹⁶ Its basic design concept (Figure 1) considers that a slice of the environment of an ideal riser reactor, involving certain mass of catalyst and gases and moving along it during a given residence time, is located into this batch reactor; then, reaction time evolved in the Riser Simulator is equivalent to contact time, and motion, along the ideal riser. These characteristics avoid coke profiles and gas shortcuts. Furthermore, the reactor reproduces the high bed voidage of the riser in the FCC unit. The catalyst basket, a cylinder of 1.85 cm radius and 1.57 cm height, is located in the lower block of the reactor, where the catalyst is placed between two porous plates, which are the bases of the cylinder. The cavity where the basket is located is designed to facilitate the circulation of the gaseous phase in an annulus during the reaction, and through the catalyst basket, thus inducing the fluidization of the catalyst bed. The temperature in the bed is measured by means of a thermocouple, which is also used by the temperature control system.

The reactant is injected into the reactor with a glass syringe and the sample is instantaneously vaporized. Simultaneously, a timer is activated which, once the preset reaction time has elapsed (between 3 and 12 s in this case), actuates a four-way valve to let the gaseous phase in the reactor be evacuated into the analysis system. This system consists of a vacuum chamber (528 cm³) and a gas chromatograph (Hewlett-Packard 6890 provided with a FID detector and a high-resolution capillary column of methyl silicone stationary phase). The identification of the products was performed by two methods: (1) injection of pure standards (Alphagaz PIANO calibration standards from Air Liquide), and (2) mass spectrometry and gas chromatography, MS-GC (5989B from Hewlett-Packard). RON numbers were calculated according to the method of Anderson et al.¹⁷

The reaction conditions were the following: temperature, 500 and 550 °C; catalyst/feed ratio (C/O), 6; and reaction time, 3, 6, 9, and 12 s.

The riser simulator has been used in previous papers in which its capacity for both reproducing the conditions of industrial FCC reactors and obtaining the cracking kinetic parameters was proven.^{15,18–21}

Table 1. Characteristics of the Feedstocks.

	vacuum gas oil	heavy coker naphtha	visbreaker naphtha
density at 15 °C, g/cm ³	0.9082	0.7737	0.7307
simulated distillation, °C			
initial point, °C		66.8	32
10 wt %	354	108.6	70
30 wt %	412	133.9	100
50 wt %	447	152.0	120
70 wt %	476	165.8	136
90 wt %	539	183.3	153
end point		200.8	163
sulfur, wt %	0.69	1.22	
Rambsbottom C, wt %	0.28		
aniline point, °C	85		
paraffins, wt %	12.5	40.2	46.0
naphthenes, wt %	26.6	9.1	12.4
aromatics, wt %	60.9	30.1	14.5
olefins, wt %		20.6	27.1

Feedstocks. The base feed is a refinery vacuum gas oil, VGO, which is the usual feed of a Spanish FCC unit. The new feedstocks to be added are a heavy coker naphtha (HCN) and a visbreaker naphtha (VN). The cuts were characterized by simulated distillation. For the VGO, the aromatic and non-aromatic fractions were separated following the ASTM D 2549-85 standard, and subsequently the two fractions were analyzed following ASTM D 2786-86 and ASTM D 3239-86 standards, respectively. The results of the analysis are shown in Table 1.

Experiments were performed under the aforementioned conditions by feeding VGO, each of the naphthas (pure), and mixtures with 80 wt % VGO and 20 wt % naphthas.

Catalyst. The catalyst used (an equilibrated OCTYDINE 1169 BR catalyst from Engelhard) is a commercial one, sampled from a Spanish FCC unit. Catalyst properties are shown in Table 2. The porous structure was studied from isotherms of adsorption-desorption of N₂ (mesopores) and of Ar (micropores) in a Micromeritics ASAP 2000 apparatus.

Results

Cracking of Pure Naphthas and Pure VGO. Considering the compounds originally present in the naphthas, Table 3, it can be appreciated that in the case of the HCN, aromatics (30.1 wt %), *n*-paraffins (26.4 wt %), and olefins (20.6 wt %) are the most important groups, which are concentrated in the C₈–C₁₁ range. In the case of the visbreaker naphtha, olefins (27.1 wt %), *n*-paraffins (25.8 wt %), and *i*-paraffins (20.3 wt %) are the most important groups, concentrated in the C₇–C₉ range. The composition of the naphthas are compared in Table 3 against the composition of the gasoline cuts obtained by cracking these naphthas and the VGO feedstock (6 s was chosen as a representative reaction time).

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Table 2. Equilibrated Catalyst Properties

Physical Properties			
particle size, μm			
0–20	0	wt %	
0–40	6	wt %	
0–80	60	wt %	
average particle diameter, μm	75		
bulk density, cm^3/g	0.94		
BET surface area, m^2/g	205		
zeolite surface area, m^2/g	40		
matrix surface area, m^2/g	165		
UCS, Å	24.31		
Composition			
Cu	10	ppm	
Ni	270	ppm	
V	1200	ppm	
Fe	0.35	wt %	
Na	0.17	wt %	
C	0.10	wt %	
Al_2O_3	48.0	wt %	
ReO	1.26	wt %	
Catalytic Properties			
MAT activity	71	wt %	
CF (Carbon Factor)	0.64		
GF (Gas Factor)	0.93		

The effect of reaction time at 500 and 550 °C on the yields of dry gas, LPG, and gasoline is shown in Figure 2a,b for the conversion of both pure naphthas and VGO. It has to be mentioned that it is not possible to assess the conversion in the usual way, since there exist conversions of reactants in the feedstock leading to products in the same range of molecular weights as the feedstock itself. Then, the reaction time is the parameter that allowed us to obtain a wide range of compositions and to follow the evolution of cracking reactions. It can be seen in the cases of both naphthas that the yield of gasoline decreases and the yields of dry gas and LPG increase up to certain values. It can also be seen that the higher the temperature, the lower the yield of gasoline (which also means higher conversion of the

naphthas) and the higher the yields of LPG. It has to be considered that the gasoline yield indeed is composed of two main fractions: the fraction of the original naphtha that remains unconverted and the products from the naphtha cracking that fall within that molecular range.

For the case of using VGO as the feedstock, it can be seen in Figure 2c that the yield of gasoline increases with time, and in a more pronounced way at 500 °C than at 550 °C. Consequently, for times higher than 7 s the yield corresponding to 500 °C is higher.

As temperature is increased from 500 °C to 550 °C, the yields of LPG and of dry gas increase and, as a result, the yield of LCO decreases. The effect of time on the yield of lumps differs. Thus, whereas the yield of dry gas remains constant, the yield of LPG increases, for the whole experimental period at 500 °C but up to the 9 s mark at 550 °C. The yield of LCO decreases all the time at 500 °C and reaches a smooth maximum at around the 8 s mark at 550 °C.

In Figure 2, due to the characteristics of the feeds under study, contact time has been taken as a variable instead of conversion for monitoring the reaction. Conversion cannot be defined in the classic mode in the case of the naphtha feedstock, since there exist conversions of its compounds leading to products in the same range of its molecular weights. Even though an index could be used to represent conversion (e.g., the yield of light gases), it would not be possible to compare this information with the conversion obtained in the case of VGO feedstock. The problem would also be present in the case of naphtha–VGO mixtures. These results of yields vs contact time will be useful for developing kinetic models for cracking and catalyst deactivation in the future, which will allow for simulating the industrial reactor for these feeds.

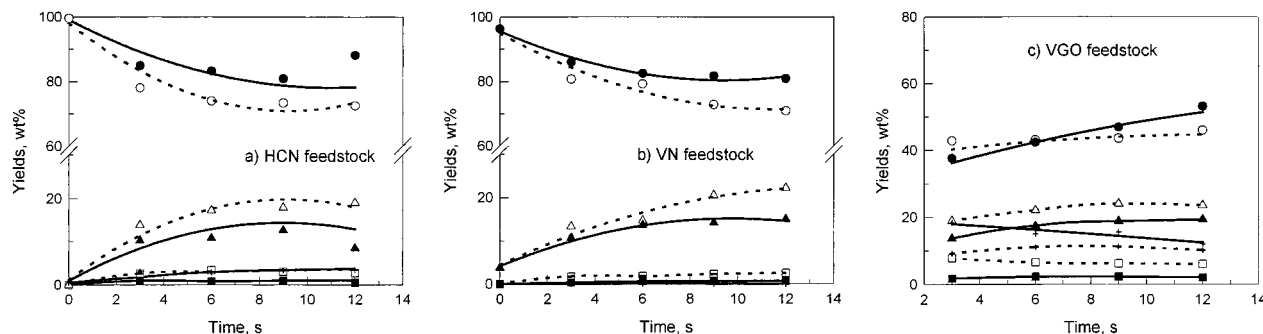


Figure 2. Effect of reaction time on the yields of dry gas(□), LPG(Δ), gasoline(○), and LCO(+) at 500 °C (solid lines) and 550 °C (dashed lines). (a) heavy coker naphtha catalytic cracking; (b) visbreaker naphtha catalytic cracking; (c) VGO catalytic cracking.

Table 3. Yields and Characteristics of the Gasoline Cut in the Catalytic Cracking of VGO, Heavy Coker Naphtha, and Visbreaker Naphtha^a

	VGO		heavy coker naphtha			visbreaker naphtha		
reaction temp, °C	500	550	unconverted	500	550	unconverted	500	550
coke yield, wt %	4.7	6.7		2.3	2.6		3.5	4.5
gasoline yield, wt %	42.3	43.0	99.5	83.2	73.9	96.2	82.4	79.1
RON	95.1	99.1	84.1	82.7	84.3	78.4	76.3	77.5
<i>n</i> -paraffins, wt %	3.6	4.0	26.4	25.7	24.4	25.8	26.5	26.3
<i>i</i> -paraffins, wt %	34.9	22.1	13.8	21.5	19.4	20.3	27.6	27.1
aromatics, wt %	35.0	36.1	30.1	31.6	34.9	14.5	15.9	19.0
naphthenes, wt %		5.5	9.1	8.3	7.2	12.4	10.6	9.4
olefins, wt %	19.0	32.3	20.6	12.9	14.1	27.1	19.5	18.2

^a C/O: 6; reaction time: 6 s.

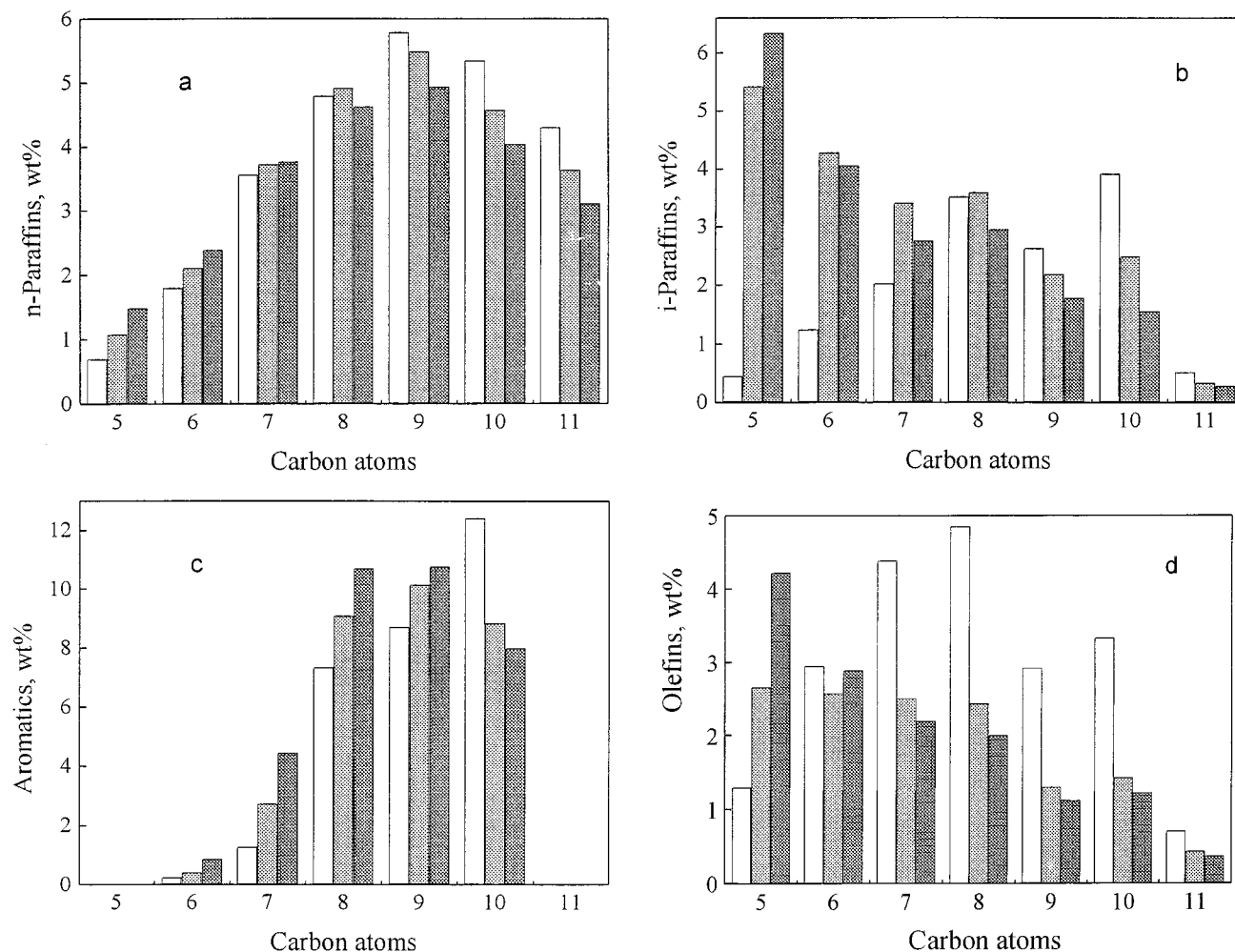


Figure 3. Gasoline composition. Carbon atom distribution in the cracking of heavy coker naphtha at 500 (light grey) and 550 °C (dark grey) as compared with the unconverted feedstock (□). Reaction time, 6 s. (a) *n*-paraffins; (b) *i*-paraffins; (c) aromatics; (d) olefins.

The analysis of the yields of the various groups (total aromatics, olefins, *n*- and *i*-paraffins, naphthenics, refer to Table 3) in the conversion of both pure naphthas shows that they are quite stable; however, it is clear that individual components indeed change in some cases. Results in Table 3 complement those of Figures 3 and 4, which show the effects of catalytic cracking on both naphthas over the composition of the gasoline cuts at the same reaction time of 6 s. In both cases an important yield of isoparaffins (C_5 – C_6) was observed, probably as a consequence of the cracking of high molecular weight olefins, following the usual mechanism for catalytic cracking which leads to a high proportion of saturated iso compounds. An analysis of the yields of gasoline components based on a fresh feed basis (data can be obtained from Figures 3 and 4 and Table 3) shows that the cracking of heavier paraffins, those of the C_8 – C_{12} range, was also moderate, probably due to the preferential cracking of olefins. Moreover, an increase in the yield of aromatics benzene, toluene, and xylenes was observed, leading to an increase in their proportions inside the gasoline cut.

These changes in composition in the gasoline range impact on its quality. Thus, (refer to Table 3), the unconverted naphthas have very low octane numbers (HCN, 84.1; VN, 78.4), a fact that is well-known and that makes them inappropriate for direct blending in

the gasoline pool. On the other hand, FCC gasoline from VGO has a very high RON which, as expected, increases with temperature (95.1 at 500 °C and 99.1 at 550 °C). The RON of the gasoline cuts of the naphthas does not change significantly by cracking it at 550 °C and it decreases slightly at 500 °C. Two facts can be pointed out which could help to make a decision whether to add these naphthas to VGO, even in these borderline cases. First, the proportion of olefins in the gasoline cuts of the reaction products of both naphthas is much lower than in the unreacted naphthas and, second, the opposite is obvious in the case of isoparaffins.

Cracking of VGO–Naphtha Mixtures. The cases studied correspond to converting both the VGO and the naphthas mixed in the proportions 80 wt % VGO–20 wt % naphthas, feeding them at the usual injection point of riser units, that is, at the bottom of the reactor.

It can be seen in Table 4 (results corresponding to 6 s were considered representative, because yield curves do not change significantly at 9 or 12 s) that the yields of gasoline obtained by cracking VGO–naphtha mixtures are significantly higher than those obtained with VGO alone. In the case of VGO–HCN, the yield of gasoline decreases with the temperature, whereas in the case of VGO–VN, it is constant. It can also be observed that the yields of coke in the case of cracking pure naphthas (refers to Table 3) are lower than in the case

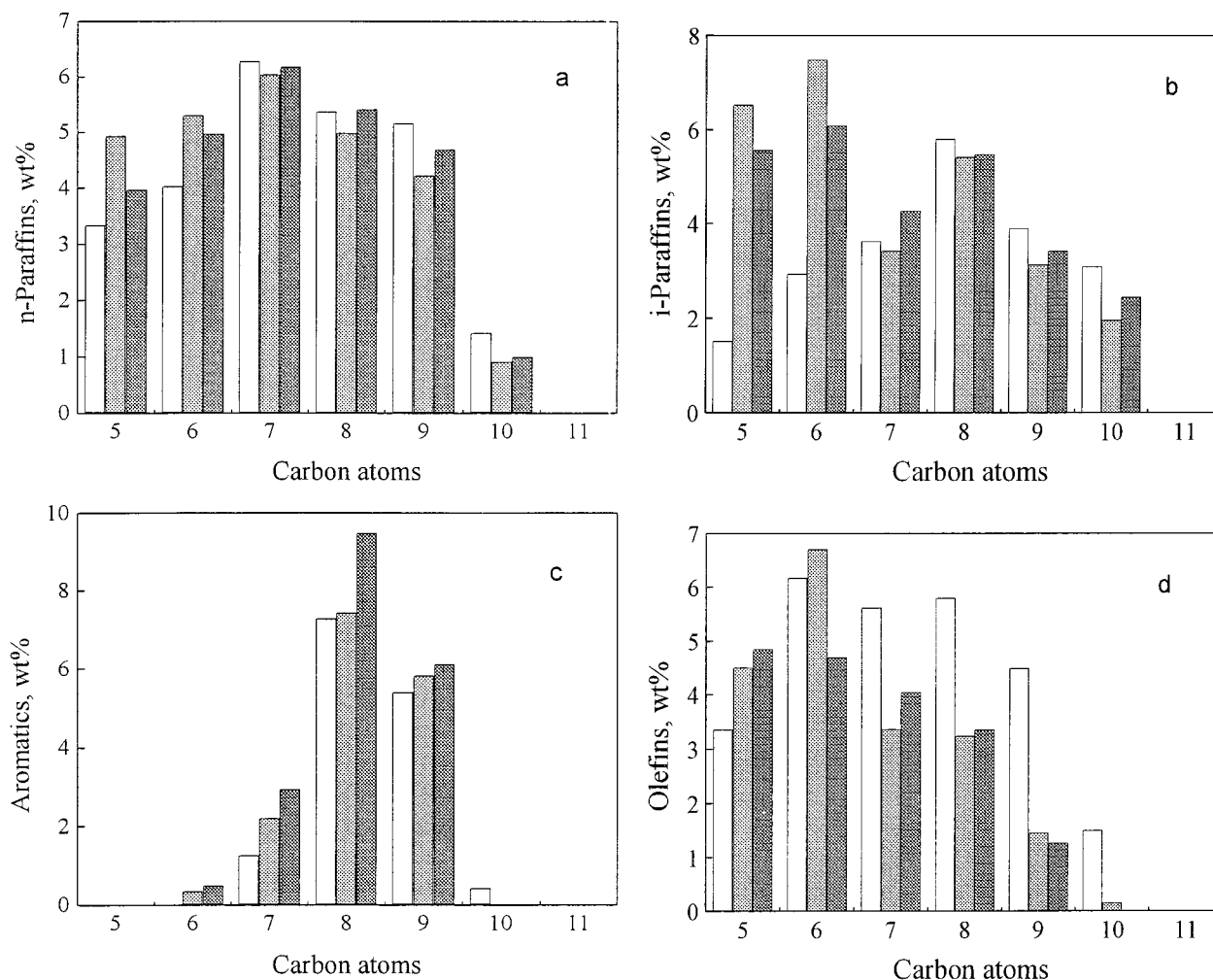


Figure 4. Gasoline composition. Carbon atom distribution in the cracking of visbreaker naphtha at 500 (light grey) and 550 °C (dark grey) as compared with the unconverted feedstock (□). Reaction time, 6 s. (a) *n*-paraffins; (b) *i*-paraffins; (c) aromatics; (d) olefins.

Table 4. Yields of Hydrocarbon Groups Obtained by Catalytic Cracking of VGO and of 80 wt % VGO–20 wt % Naphtha Mixtures, and Characteristics of the Gasoline Cut Obtained Comparison with Values from the Simulations of Strategies #2 and #3^a

temp, °C	Experimental, S#1						Simulated, S#2				Simulated, S#3			
	VGO		80 wt % VGO–20 wt % HCN		80 wt % VGO–20 wt % VN		80 wt % VGO–20 wt % HCN		80 wt % VGO–20 wt % VN		80 wt % VGO–20 wt % HCN		80 wt % VGO–20 wt % VN	
	500	550	500	550	500	550	500	550	500	550	500	550	500	550
dry gas, wt %	2.19	6.53	1.44	4.00	3.50	6.44	1.75	5.22	1.75	5.22	1.91	5.89	1.86	5.57
LPG, wt %	17.25	22.01	14.06	18.59	13.81	20.81	13.80	17.61	14.52	18.35	15.94	21.04	16.52	20.54
LCO, wt %	14.49	10.82	10.9 ^b	8.5 ^b	11.0 ^b	8.2 ^b	11.59	8.66	11.59	8.66	12.18	9.27	11.59	8.66
coke, wt %	4.68	6.72	4.32	4.68	5.40	6.66	3.74	5.38	3.74	5.38	4.21	5.89	4.45	6.28
gasoline, wt %	42.32	43.00	61.81	56.52	50.14	49.56	53.76	54.28	53.11	53.63	50.50	49.15	50.34	50.23
RON	95.1	99.1	88.50	90.60	84.90	95.00	91.0	93.6	89.0	91.7	91.0	94.7	88.9	92.3
<i>n</i> -paraffins, wt %	3.60	4.00	16.70	16.00	16.30	8.00	12.04	12.24	11.63	11.83	10.88	10.16	11.08	11.02
<i>i</i> -paraffins, wt %	34.90	22.10	27.30	22.30	32.00	23.20	27.08	19.04	29.59	21.42	30.47	21.28	32.50	23.66
aromatics, wt %	35.00	36.10	33.60	37.20	29.00	36.90	33.19	33.90	27.55	28.32	33.88	35.74	28.74	30.72
naphthenes, wt %	7.60	5.50	8.00	6.20	8.50	7.20	8.12	6.80	9.30	7.95	7.79	5.98	8.54	6.71
olefins, wt %	19.00	32.30	14.40	18.30	14.20	24.70	19.54	28.01	21.91	30.46	16.95	26.85	19.13	27.89

^a C/O: 6; reaction time: 6s. ^b According to mass balance calculations.

of standard VGO feedstock and then, as expected, coke yield is lower when cracking mixtures.

Coke content for all the feeds increases when temperature is raised, which is evidence that this increase favors the evolution of coke precursors (by means of oligomerization, cyclization, and dehydration) to a greater degree than their cracking. The yield of coke through

the cracking of VGO–VN is similar to that corresponding to the cracking of VGO at 550 °C and greater than its cracking at 500 °C. This yield of coke for the VGO–VN mixture is notably higher than that of the VGO–HCN mixture. These results are explained by the composition of the feeds. Thus, the high light olefin content of the VN contributes to a greater degree to the

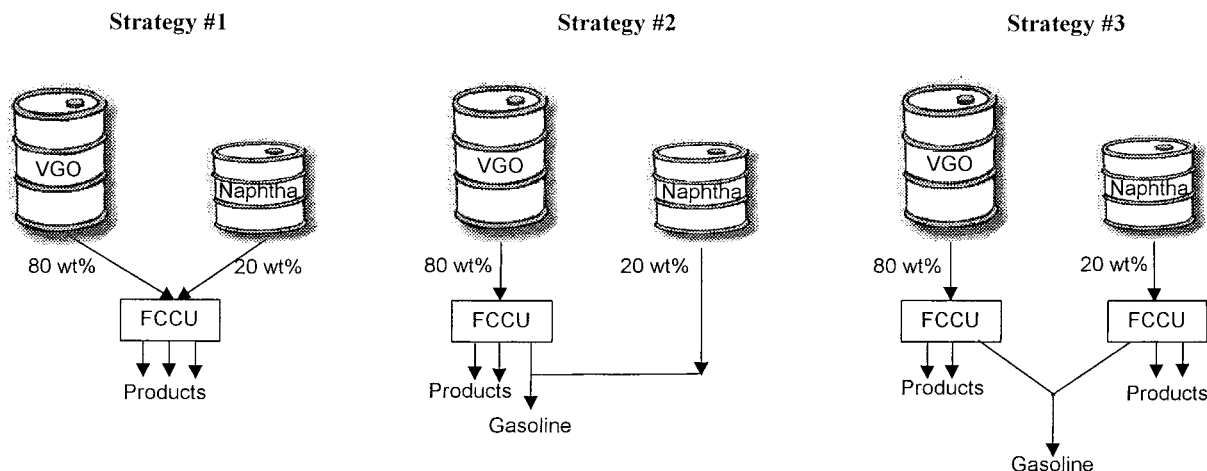


Figure 5. Strategies for naphtha upgrading by feeding to a FCC unit.

formation of coke by oligomerization and cyclization (in order to generate polyaromatic structures) than a high monoaromatic content, as is the case of a feed with HCN.

However, the drawback of cracking the mixtures lies in the resulting decrease in the quality of the gasoline. Only in the case of cracking the VGO–VN mixture at 550 °C, the RON is close to that of VGO cracking (Table 4). The reasons for these drops are mainly based on the lower yield of olefins and on the higher yield of undesired *n*-paraffins (they do not convert extensively under these conditions). The other important hydrocarbon group that controls gasoline RON, the aromatics, does not change significantly as compared to the VGO feedstock.

The proportion of *n*-paraffins, especially those in the C₉–C₁₁ fraction, is higher than the one expected when the feeds are cracked separately, or even when the naphthas are added directly to the gasoline fraction from VGO cracking. These results suggest that the naphthas play an inhibiting role in the cracking of these heavy paraffins when added to the VGO, which could be assigned to the high molecular weight olefins initially present in the naphthas; these olefins can compete successfully for the acid sites in the catalyst, since they adsorb strongly (note that their proportion is even lower than the one expected according to the conversion of “pure” compounds). It is well-known that the cracking mechanism of olefins and paraffins is different and the results show that the adsorption of olefins on Bronsted sites to form carbenium ions is easier than the adsorption of paraffins on these sites to form oxonium ions.²²

A final comparison can be established among at least three different strategies to plan the recycling of these particular naphthas in terms of gasoline yields and quality, which are schematized in Figure 5. Strategy number 1 (S#1) is the one already described, that is, the addition of naphthas to VGO and further cracking of the mixtures; strategy number 2 (S#2) is the direct addition of the gasoline cut of the naphthas to the gasoline pool; and strategy number 3 (S#3, which is certainly less viable) consists of the separate conversion of VGO and naphthas feedstocks and the later addition of the gasoline cuts obtained. Comparisons between

strategies were done on the basis of simple mass balance calculations performed on S#2 and S#3, in which the ratio 80/20 for VGO/naphtha mixtures and the experimental conditions were kept constant, but these conditions and ratios could certainly be changed in order to obtain better results.

Results in Table 4 corresponding to S#2 and S#3 were estimated from mass balance calculations. For example, the simulated results for gasoline yields in S#2 were obtained as follows: amount of VGO multiplied by gasoline yield in VGO experiments, plus amount of naphtha. In simulating results for S#3, results from “pure” VGO and “pure” naphtha experiments were taken into account. For example, the simulated results for gasoline yields were obtained as follows: amount of VGO multiplied by gasoline yield in VGO experiments, plus amount of naphtha multiplied by gasoline yield in naphtha experiments.

Results in Table 4 show that for the case of recycling HCN, S#1 is the strategy leading to much higher yields of gasoline, although RON is very much decreased; S#2 shows smaller gains in gasoline yields but the loss of quality is also smaller; S#3 would be intermediate. Then, under a general approach, strategy S#1, especially at high temperature, appears as the most interesting.

Results are somewhat different in the case of recycling VN. Thus, the increase in the yield of gasoline when VN is converted simultaneously with VGO (S#1) is smaller than in the case of HCN but, as stated, the loss of RON numbers is smaller. S#2 (direct addition to FCC naphtha) shows higher gains in gasoline yields, but RONs are lower, and S#3 shows yield gains similar to those of S#1, but lower RON numbers. Thus, the option of recycling visbreaker naphtha together with VGO and converting them at high temperature appears to be interesting, because the trade-off between yield and RON seems more attractive.

The changes in product distribution, particularly in gasoline and LPG cuts, deserve more study. It is also interesting to expand this study to situations where the feedstocks can be injected into the riser units at different heights, to benefit from the different crackabilities of the various cuts. In the case of these naphthas, their injection at a lower point than the VGO feedstock could provide a more severe environment leading to a better

product distribution, even though the VGO is in contact with a catalyst of lower temperature.

Conclusions

The simultaneous conversion of heavy coker or vis-breaker naphthas together with the usual VGO feedstock in FCC units is an interesting approach to increase the yield of gasoline, although a trade-off with product quality has to be considered. High temperatures seem to be the best choice. This option enhances the value of these naphthas, and it seems to be more convenient than cracking them separately or adding them directly to the FCC naphtha in the gasoline pool. Olefins in the naphthas are the more crackable group, which leads to an important increase in the yield of

isoparaffins. Coke yields are not higher than the ones obtained when cracking standard VGO, and then the impact on units' heat balances would be negligible.

The results of this work suggest that the injection of the naphthas at a lower point than the VGO in the FCC riser might yield better results, and it is an interesting option for controlling this process. The riser simulator laboratory reactor proved to be suitable for these types of FCC studies.

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