

Reconversion of Olefinic Cuts from Fluidized Catalytic Cracking Naphthas

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The feasibility of reconversion of a highly olefinic cut (OLEF; 60–110 °C), obtained from the bottoms of depentanizer columns used to separate the C₅ fraction from fluidized catalytic cracking (FCC) naphtha, was studied under realistic FCC conditions over two equilibrium commercial catalysts. A riser simulator reactor was used at 500 and 550 °C, a catalyst-to-oil ratio of 5.6, and short reaction times of up to 15 s to assess (a) the crackability and the products of the conversion of the cut OLEF, (b) the conversion of a standard vacuum gas oil feed (VGO) to be used as a reference, and (c) the conversion of a mixture with a mass ratio of 20:80 OLEF–VGO. The gas fraction in the conversion of OLEF showed high yields of propene and isobutane, while aromatics and *i*-paraffins appeared among products with the same range of molecular weights as the feedstock, thus determining a research octane number value in the gasoline cut that is higher than the feedstock's. Olefins showed to be converted selectively. The conversion of the mixture OLEF–VGO showed the following main characteristics that differ from the standard operation (VGO feedstock): (i) an increase in the yield of gasoline, which is higher than the one expected from the separate conversion of equivalent masses of the individual feedstocks, (ii) an increase in the yield of liquified petroleum gas and some individual hydrocarbons, like propene or isoamylenes, and (iii) a better octane-barrel balance in gasoline. The particular characteristics of each catalyst (activity and hydrogen transfer capability) reflected clearly on the product distributions obtained in the conversion of the various feedstocks. This recycling option appears as very interesting because it could contribute to improve the refinery's economy through the improvement of different issues.

Introduction

Gasolines are composed in the refineries of the addition of various hydrocarbon streams from different processes, with wide ranges of composition and fuel quality. Among them, the catalytic cracking of hydrocarbons (FCC) and reforming processes are the most important contributors, but depending on availability and particular conditions, butanes, light straight run naphtha, isomerate, alkylate, naphthas from thermal processes, and/or oxygenates such as methyl *tert*-butyl (MTBE) or *tert*-amyl methyl (TAME) ethers are also incorporated into the gasoline blending operations.¹ Some of the low-value cuts in the refineries, such as coker or visbreaker naphthas, can be upgraded in the FCC unit.^{2,3}

MTBE and TAME are gasoline blending components that increase octanes and help in improving fuel combustion and decreasing contaminant emissions. They are produced from methanol and the corresponding C₄ or C₅ isoolefins (isobutylenes or isoamylenes), which are also produced by the FCC units.¹ In the case of TAME, reactant isoamylenes are included in the C₅ fraction that is separated from FCC naphtha in distillation columns,⁴ with the bottoms being usually directed at gasoline blending. However, because these units operate under conditions aimed at maximizing olefin yields, the concentrations of these hydrocarbons in their naphthas could be so high that, after blending, the olefin maximum allowances in the gasoline in some markets could

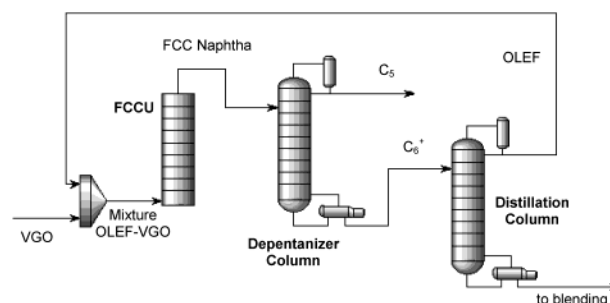


Figure 1. Configuration proposed to recycle an OLEF to the FCC process.

be compromised. Interestingly, most of the olefins occur in a given fraction (the lighter fraction) of the depentanizer bottom stream, which could be separated from the main cut. This fraction then could be upgraded by recycling it back to the FCC process,⁵ either by addition to the usual feedstock or injection at another point (height) of the riser reactor, according to its reactivity, the catalyst used, and the operative conditions. The configuration proposed is shown in Figure 1. Depending on the particular conditions of the refinery, the recycle ratio (olefinic cut, OLEF, versus vacuum gas oil feed, VGO) could amount to about 10–20%. A similar approach can be based on the utilization of C₅ raffinate from petrochemical networks.⁶

It is the objective of this work to study the feasibility of recycling an olefinic fraction from the bottom stream of depentanizer columns in refineries that produce TAME. The crackability of OLEF, the products observed from its conversion, and the results of feeding a mixture

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Table 1. Properties of the Mother FCC Naphtha and OLEF

Distillation of Cut OLEF			
vol (%)	<i>T</i> (°C)	vol (%)	<i>T</i> (°C)
10	60	70	92
30	70	90	102
50	81		

hydrocarbon group	FCC naphtha (%)	cut OLEF (%)
aromatics	26.5	7.7
olefins	29.4	38.5
<i>i</i> -paraffins	24.1	26.9
<i>n</i> -paraffins	7.1	7.6
naphthenics	12.9	19.3
RON	94.6	84.8

Table 2. Properties of the VGO Used

density (g/cm ³)	0.9240	ramsbottom carbon (%)	0.55
sulfur (%)	0.57	aniline point (°C)	96.40

Distillation			
vol (%)	<i>T</i> (°C)	vol (%)	<i>T</i> (°C)
10	399	70	498
30	441	90	544
50	470		

of this cut with VGO are studied in a riser simulator laboratory reactor with equilibrated commercial catalysts under typical FCC conditions.

Experimental Section

Two equilibrated commercial catalysts were used, namely, E-Cat A (specific surface area, 139 m² g⁻¹; zeolite content, 16.9%; unit cell size, 24.23 Å; rare-earth oxides, 1.26%) and E-Cat B (specific surface area, 151 m² g⁻¹; zeolite content, 15.9%; unit cell size, 24.24 Å; rare-earth oxides, 0%). E-Cat B was taken from a refinery operating under the "light olefins" mode, maximizing their yields. Three feedstocks were used: (a) an OLEF, which was generated by distillation from an typical FCC naphtha, having a range of boiling points between approximately 60 and 110 °C (the compositions of both cuts are shown in Table 1); (b) a standard VGO, with properties that are shown in Table 2; (c) a 20:80 mixture (mass ratio) of OLEF and VGO.

A laboratory-batch fluidized-bed reactor that is specially suited for FCC-related studies, the riser simulator unit,^{7,8} was used in the conversion experiments. The reactor was previously used in catalyst evaluation,⁹ kinetic and diffusive modeling,^{10,11} and the study of new operative modes.^{3,12,13} Detailed descriptions and performance of the unit can be found in the publications mentioned.

Experimental conditions were as follows: reaction temperatures, 500 and 550 °C; catalyst-to-oil ratio, 5.6; reaction times, 3, 6, 9, 12, and 15 s. Mass balance calculations showed agreements over 96% in all cases. Reactants and reaction products were analyzed by standard capillary gas chromatography (GC). Coke yields were assessed by means of a method with temperature-programmed oxidation and further methanation of the carbonaceous deposits on the catalysts. The research octane numbers (RONs) of the various feedstocks and products were assessed by means of a modified Anderson's GC method.¹⁴

Results and Discussion

Conversion of Feedstock OLEF. The cut with a high olefin content in this study represented about 10%

Table 3. Conversion of Cut OLEF: Example Yields of Some Hydrocarbon Groups or Compounds and Gasoline Composition at 9 s of Reaction Time and a Catalyst-to-Oil Ratio of 5.6, as Compared to Those of the Cut

compound or hydrocarbon group	OLEF	temperature (°C)			
		E-Cat A		E-Cat B	
		500	550	500	550
<i>i</i> -C ₅ =	0.0	0.4	0.9	0.8	0.4
<i>i</i> -C ₅	0.0	3.5	4.1	1.5	2.6
C ₆ =	26.9	7.4	4.3	12.7	4.4
<i>i</i> -C ₆	9.7	18.3	16.5	17.6	17.1
C ₇ =	9.8	1.8	0.8	3.0	0.7
benzene	4.4	2.1	2.4	2.4	3.8
toluene	2.5	4.2	5.5	3.7	5.7
C ₅ -C ₁₂ groups					
aromatics	7.7	15.6	24.8	10.1	19.7
olefins	38.5	13.7	9.8	20.2	10.5
<i>i</i> -paraffins	26.9	45.1	43.1	41.4	42.6
<i>n</i> -paraffins	7.6	6.4	6.6	6.3	6.1
naphthenics	19.3	19.2	15.7	22.0	21.1
RON	84.8	87.5	89.5	87.0	89.0

of the "standard" FCC naphtha, from which it was separated by distillation. It can be seen in Table 1, which shows its composition and distillation curve, that the fraction ranging from about 60 to 110 °C (named cut OLEF) is in effect more concentrated in olefins than its mother FCC naphtha (38.5% versus 29.4%). It is expected that this difference will be even higher in the case of FCC naphthas coming from units aimed at the production of light olefins. For example, Cortés and Ruiz⁵ reported 44.8% of olefins in the 60–110 °C cut and 34.7% in the FCC naphtha in one of those refineries. Standard values for olefins in FCC naphthas are approximately 30%.¹⁵ Moreover, it is also confirmed that the olefins are essentially concentrated in the C₆ and C₇ fractions in OLEF because they amount respectively to 71.8 and 26.1% of the olefins in the cut. Consistently with the final boiling point of the cut, the concentration of aromatics in OLEF is lower than that in the naphtha. It is interesting to observe that the RON of the cut OLEF (84.8) is significantly lower than that of the mother FCC naphtha (94.6) because of the much lower concentration of aromatic compounds, which is not compensated for by the higher proportion of olefins.

The conversion of the cut OLEF over both catalysts produces a wide distribution of products that include gases C₁–C₄, hydrocarbons in the same range of molecular weights as the feedstock, and heavier hydrocarbons up to C₁₂. Because of the interconversion observed, that is, the emergence of products with the same range of molecular weights as the cut, which can be exemplified with an increase in C₆ *i*-paraffins or toluene (see Table 3), it is not possible to define the feedstock conversions in the usual way. However, results in this work can be analyzed as a function of the reaction time, which is directly related to conversion.

It can be seen in Figure 2 that the conversion of the cut OLEF, as shown by gas yields, increases as a function of the reaction time for both catalysts. This is also reflected by the evolutions in gasoline and coke yields because gasoline, where the feedstock cut OLEF is included, decreases while coke increases as a function of time. The highest conversions were observed at 550 °C and 12 s of reaction time over both catalysts, with gasoline losses to gases and coke of about 32% in the case of E-Cat A and 23% in the case of E-Cat B. At 500 °C, the highest conversions (gases plus coke yields) were 20.7 and 13.4%, respectively. While many of the cata-

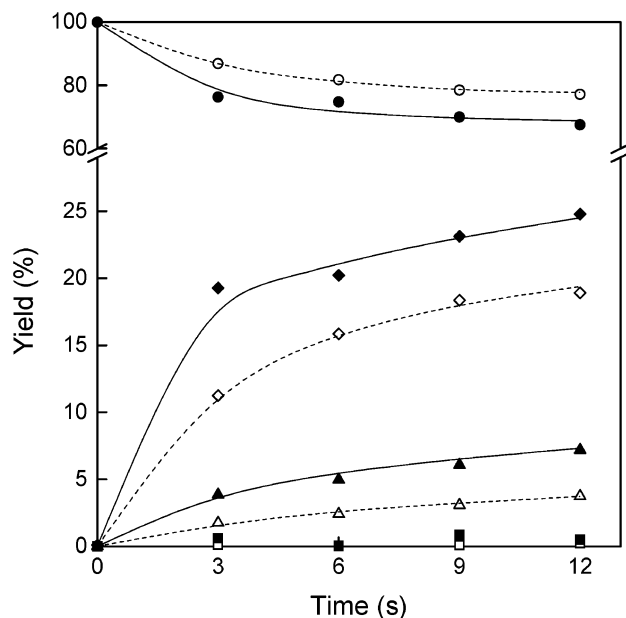


Figure 2. Yields of gases (◆, ◇), gasoline (●, ○), LCO (■, □), and coke (▲, △) in the conversion of the cut OLEF at 550 °C as a function of the reaction time: E-Cat A (closed symbols, full lines); E-Cat B (open symbols, dashed lines).

lysts' properties are similar, it is clear that E-Cat A is more active than E-Cat B at all conditions, a fact that can be explained based on its rare-earth content. The yields of the most important hydrocarbon groups for a particular condition are shown in Table 3.

Among gases, a high proportion of olefins can be observed in the C₂ and C₃ fractions (from 60 to 70%), and isobutane accounts for about 59% (E-Cat A) and 43% (E-Cat B) of the C₄ fraction. Olefins, particularly those in the C₆ and C₇ fractions, were the hydrocarbons most sensitive to conversion, as can be seen in the examples in Table 3. In the gasoline fraction, olefins decreased from 38.5% to values of around 10% at 550 °C and 16% at 500 °C. It can also be noticed that benzene, originally present at 4.4% in the cut, does not represent an additional problem, because it decreases to average values of 3.2% in the gasoline fraction. Even though coke yield values in E-Cat A are somewhat higher than those in the standard FCC operation (a maximum value of 7.2% was observed at 550 °C and 12 s), the coking trend of the cut OLEF seems not to be particularly high (refer to Figure 2).

The gasoline fractions in the products of the conversion of the cut OLEF on both catalysts show much higher RONs, as compared to the feedstock. In effect, while the feedstock has a RON of 84.8, the C₅–C₁₂ liquid products have average values of about 87 at 500 °C and 89 at 550 °C. It is to be noted, however, that these RON values are still lower than that of the mother FCC naphtha (94.6). These observations can be explained in light of changes in the composition of the resulting gasoline cuts as compared with OLEF (see Table 3): the strong decrease in olefins is largely compensated for by the increase in aromatics (particularly toluene) and C₆ *i*-paraffins. The differences in the distributions for each catalyst, which are associated with the higher activity and hydrogen-transfer capacity of E-Cat A because it yields more aromatics and paraffins and fewer olefins and naphthenics than E-Cat B, compensate each other to reflect on similar calculated RONs. While most of the properties are similar in both catalysts, E-Cat A has a

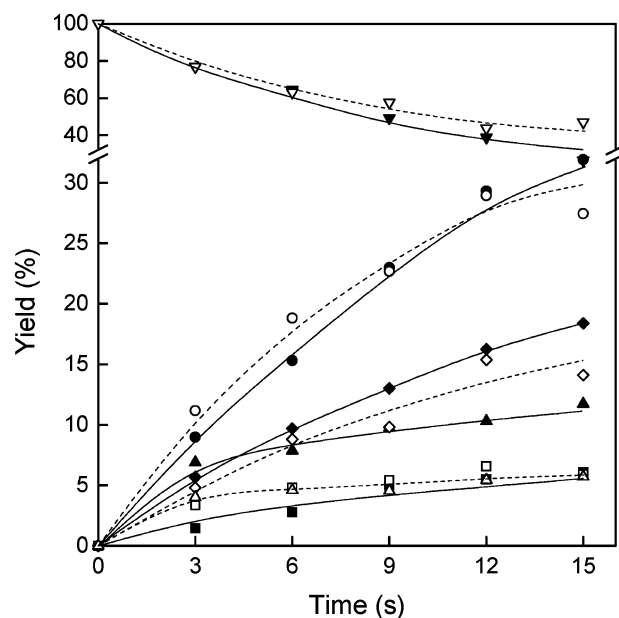


Figure 3. Yields of gases (◆, ◇), gasoline (●, ○), LCO (■, □), VGO (▼, ▽), and coke (▲, △) in the conversion of VGO at 550 °C as a function of the reaction time: E-Cat A (closed symbols, full lines); E-Cat B (open symbols, dashed lines).

standard load of rare earths and E-Cat B has not. The effect of the reaction temperature on the product distribution is as expected¹⁶ because lower yields of paraffins and olefins and higher yields of aromatics are observed at the higher reaction temperature.

All of these results confirm that the cut OLEF has a high degree of crackability, which mainly affects olefins, and that the products from its sole conversion will not induce problems on the operation of FCC units.

Conversion of Feedstock VGO. The conversion of a typical VGO (refer to Table 2) was used as a reference for the standard operation of a FCC unit. It can be seen in Figure 3 that the performance of both catalysts in the conversion of VGO is similar in terms of activities and global yields of gases, gasoline, and light cycle oil (LCO), with some differences in coke yields, where E-Cat A forms more coke. Coke on catalyst has maximum values of 1.92 and 0.94% at 550 °C and 15 s of reaction time, with conversions being 68 and 53% over catalysts E-Cat A and E-Cat B, respectively.

The compositions of the gasoline cuts, however, show strong differences between the catalysts (see Figure 4). In effect, in the whole range of reaction times, E-Cat A yields higher amounts of aromatics and *i*-paraffins than E-Cat B, while the opposite applies to olefins; *n*-paraffin and naphthene yields are very similar. As a direct consequence of these particularities, which can be ascribed to the higher hydrogen-transfer activity of E-Cat A due to the rare earths in its formulation, with E-Cat B being aimed at the maximization of olefins, RON values are essentially the same. Also, the decreasing profiles of olefin yields over both catalysts confirm the high reactivity of these compounds.

These characteristics of the catalysts are also confirmed in the analysis of the composition of gases and lighter liquid hydrocarbons. In effect, the selectivity to olefins, defined as the ratio between olefins and total hydrocarbons in the group, is higher for E-Cat B. For the example of reaction temperature 550 °C, where, as expected, because of the respective energies of activation of cracking and hydrogen-transfer reactions^{17,18} the

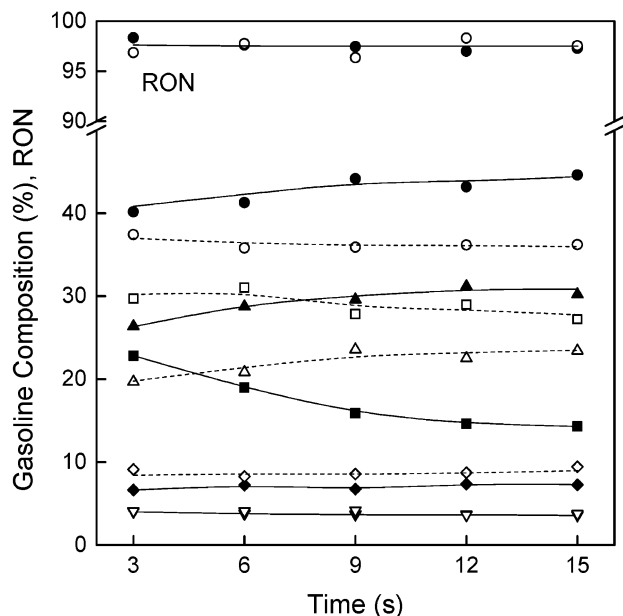


Figure 4. RON (●, ○) and product distribution in the gasoline cut obtained in the conversion of VGO at 550 °C as a function of the reaction time: E-Cat A (closed symbols, full lines); E-Cat B (open symbols, dashed lines). Symbols: aromatics (●, ○), naphthenics (◆, ◇), olefins (■, □), *n*-paraffins (▼, ▽), *i*-paraffins (▲, △).

differences are more significant, the respective selectivities to C₃, C₄, C₅, and C₆ olefins over E-Cat A are approximately 65, 40, 35, and 25%, while over E-Cat B, they are 75, 60, 55, and 45%.

Conversion of the OLEF–VGO Mixture. A 20:80 OLEF–VGO mixture was used as the feedstock to study the reconversion of the cut distilled from the bottoms stream of a depentanizer column. This ratio was selected in order to observe changes in the conversion experiments more clearly, although it is to be expected that, under actual refinery conditions, this relationship would be somewhat lower. As an example, the yields of most important hydrocarbon groups (gases, gasoline, LCO, VGO, and coke) on E-Cat A at 550 °C are shown in Figure 5 as a function of the reaction time. For comparison, results obtained using VGO are also included. As expected, the dilution effect produced by the mixture can be visualized in the lower amounts of VGO, starting at 80%, and the much higher yields of gasoline, starting at 20% (note that OLEF is included in the range of boiling points of gasoline). The yield of gases is more important as well because of the higher contribution by OLEF (under similar conditions, E-Cat A, 12 s, 550 °C, OLEF yields 25% and VGO yields 16% of gases; refer to Figures 2 and 3), while LCO yields are essentially equal. It is to be pointed out that coke yields are lower in the case of using the mixture OLEF–VGO, a fact that has to be carefully examined because of its impact on the unit's operation.

The conversion of the mixture OLEF–VGO over catalyst E-Cat B produces results with the same features. Higher yields of gasoline and gases and lower yields of VGO were observed. The lower activity of this catalyst can be noticed in the gas and coke yields, which are lower than those of E-Cat A.

These changes in the particular yields of the most important hydrocarbon groups when using the mixture OLEF–VGO cannot be explained as the simple addition of the separate contributions from the individual components of the mixture. In that sense, it is interesting

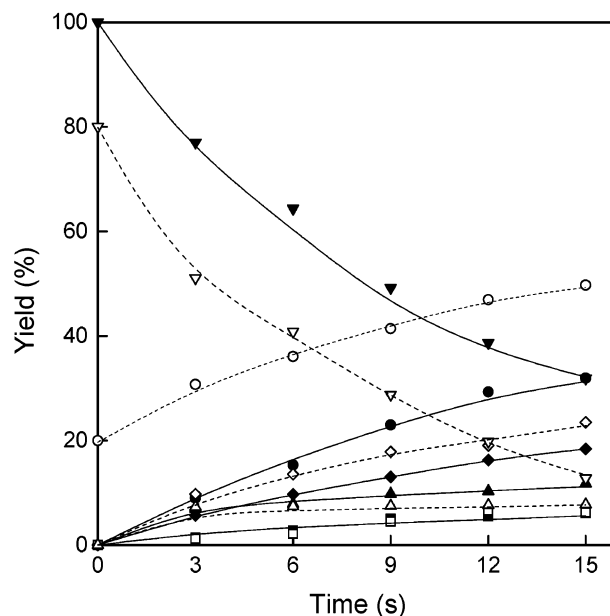


Figure 5. Yields of gases (◆, ◇), gasoline (●, ○), LCO (■, □), VGO (▼, ▽), and coke (▲, △) in the conversion of VGO (closed symbols, full lines) and the mixture OLEF–VGO (open symbols, dashed lines) over E-Cat A at 550 °C as a function of the reaction time.

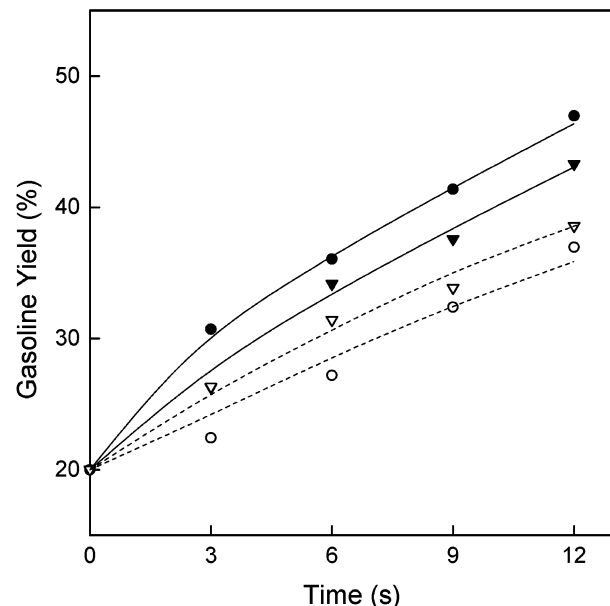


Figure 6. Yield of gasoline in the conversion of mixture OLEF–VGO (closed symbols, full lines) and values calculated from the separate conversion of each feedstock (open symbols, dashed lines): E-Cat A (●, ○); E-Cat B (▼, ▽). Temperature 550 °C.

to observe that the yield of gasoline from the mixture OLEF–VGO is higher than the one that could be calculated from the separate conversion of equivalent masses of VGO and OLEF. This is shown in Figure 6 and could be explained as the consequence of two facts: the cut OLEF cracks less than expected, according to the observed yields of C₆ and C₇ olefins (data not shown), and the VGO converts more than expected, as could be inferred from Figures 3 and 5. If this recycling approach is applied, then this fact could produce important economic benefits in the operation of the refinery, which might even overcome the loss in fuel quality. A very simple analysis of the standard (VGO cracking) and unconventional (OLEF–VGO cracking)

Table 4. Octane-Barrel Balances for VGO and OLEF-VGO Cracking Operative Strategies^a

feedstock	catalyst	gasoline yield (%)	RON	RON × yield
VGO	E-Cat A	29.3	97.0	2843
experimental	E-Cat B	28.9	98.3	2845
OLEF-VGO	E-Cat A	47.0	93.5	4391
experimental	E-Cat B	53.3	93.2	4967
OLEF-VGO	E-Cat A	37.0	95.4	3525
calculated	E-Cat B	38.6	96.5	3723

^a Experimental values are compared with those calculated from the individual conversion of each feedstock. Data are taken from experiments at 550 °C and 12 s of reaction time.

Table 5. Distribution of Hydrocarbons in the Gasoline Cut in the Cracking of VGO and OLEF-VGO at 550 °C and 15 s of Reaction Time

hydrocarbon group (%)	E-Cat A		E-Cat B	
	VGO	OLEF-VGO	VGO	OLEF-VGO
aromatics	44.6	35.3	36.2	25.7
olefins	14.3	14.0	27.2	24.9
<i>i</i> -paraffins	30.2	35.4	23.4	29.5
<i>n</i> -paraffins	3.6	4.4	3.7	4.9
naphthenics	7.3	10.9	9.4	15.0
RON	97.3	93.9	97.5	92.8

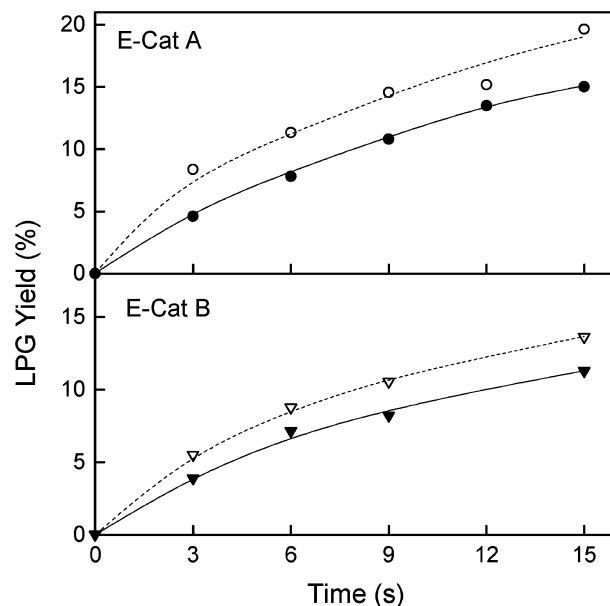
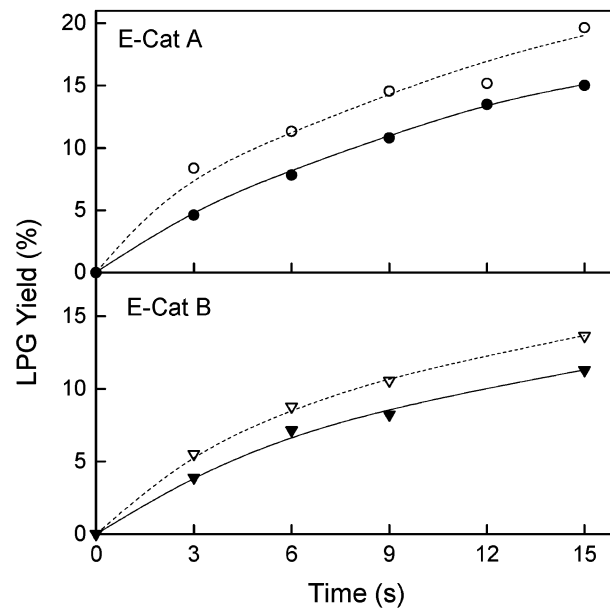
operative strategies confirms that the octane-barrel balance is improved in the case of recycling OLEF, as shown in Table 4. Benefits could also be considered in the accomplishment of fuel composition limitations like, for example, the maximum content of olefins and aromatics in gasoline.

The distribution of hydrocarbons in the gasoline cut, in comparison with the one obtained with standard feedstock VGO, is exemplified in Table 5 for both catalysts. It can be seen that the resulting RON is about 3 points lower than the one obtained with VGO. However, it is to be noted that some benefits are apparent in the distribution of components, the most marked facts being the important decrease in the amount of aromatics, which is the main reason for the decrease in RON, and the significant increase in the amount of *i*-paraffins (mainly in the C₅-C₇ group) and naphthenes. These changes are due to the substitution of part of the VGO by a feedstock with different composition, leading to more *i*-paraffins and, simultaneously, to fewer aromatics; in the case of VGO, aromatics are mainly produced from dealkylation of substituted aromatic rings present in the feedstock.

Concerning the composition of the product gas fraction, it was observed over both catalysts that the selectivities from the two feedstocks (VGO and the mixture) were very similar. However, the overall higher yields of gases when cracking OLEF-VGO include increases in the yields of light olefins and isobutane. Depending on the particular current prices of liquified petroleum gas (LPG), the increase observed in its yield (refer to Figure 7) could represent an important economical issue.

Considering some other valuable products like, for example, C₅ *i*-olefins, which are the raw materials for TAME production, their yield over both catalysts is higher when the feedstock is the mixture OLEF-VGO, as compared to VGO (refer to Figure 8). This is the consequence of starting with some olefins in the feedstock, which are easily converted to *i*-olefins and *i*-paraffins.

It should be mentioned that the implementation of this recycling option would not alter significantly the

**Figure 7.** Yield of LPG in the conversion of VGO (closed symbols, full lines) and the mixture OLEF-VGO (open symbols, dashed lines) at 550 °C: E-Cat A (●, ○); E-Cat B (▼, ▽).**Figure 8.** Yield of C₅ *i*-olefins in the conversion of VGO (closed symbols, full lines) and the mixture OLEF-VGO (open symbols, dashed lines) at 550 °C: E-Cat A (●, ○); E-Cat B (▼, ▽).

standard operation of a refinery where TAME is produced, and the capital investments are expected to be low.

The injection of the cut OLEF in a different point in the riser could be an interesting alternative. For example, the severity of cracking of the recycled stream could be increased by injecting it at locations previous to the position of the standard injection: much higher catalyst-to-oil ratios and temperatures could be achieved in that way. However, the contribution from thermal cracking could be detrimental.

Conclusions

A 60–110 °C cut obtained from the bottoms of a depentanizer column showed high crackability, with selective conversion of olefins. The conversion of a 20:

80 mixture of the cut with VGO allowed one to determine that it is feasible and profitable in comparison to the standard operation because higher yields of gasoline, LPG, and some individual products can be achieved and more octane barrels are obtained. Moreover, because of olefin conversion, it is possible to improve the composition of product gasolines. The differences in the properties of the catalysts used, particularly those associated with activity and hydrogen transfer, reflected mainly on the distribution of products. It is to be expected that the implementation of this recycle option does not require high investment costs and does not impose important modifications in the refinery configuration.

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