

FORMATION OF C₉ AND C₁₀ ALKYLAROMATIC HYDROCARBONS IN REFORMING LIGHT KEROSENE

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Earlier results [1, 2] for mono- and bimetallic reforming catalysts have indicated that naphtha feedstocks with boiling points up to 180–190 °C and having average carbon numbers higher than nine yielded a high percentage of C₉ aromatics. When reforming naphthas containing fractions boiling above 165 °C, C₁₀ aromatics are also obtained besides C₉ aromatics [3, 4]. According to the measurements of NAKAMURA et al. [5] on Pt-Al₂O₃ catalyst, maximum yields of C₁₀ aromatics can be obtained by reforming naphtha fractions boiling between 150–205 °C.

In order to study the upper limits and degree of the advantageous effects of increasing the boiling range on the yield of C₉ aromatics, experiments were carried out with kerosene boiling between 158–236 °C. These measurements aimed at studying also the distribution and amounts of C₉ and C₁₀ aromatics (among them especially pseudocumene — 1,2,4-trimethyl-benzene — which is already utilized industrially) obtainable in the parameter range applied in commercial units in the presence of Pt-Al₂O₃ catalyst.

Experimental

The experiments were carried out under pressure in a continuously operating laboratory "twin-reactor" system, including an isothermal reactor [6] filled with 100 cm³ Engelhard RD-150C catalyst. The catalyst was pretreated according to specifications of the manufacturer. Because of the high initial hydrocracking activity of the catalyst, the measured data were considered as reliable after 50 hours of operation. The experiments were carried out at a constant level of catalyst activity, checked by repeated measurements.

The light kerosene used as feedstock was of Romaskino origin and was desulphurized on 135 cm³ of Co-Mo-Al₂O₃ catalyst at 1,5 vol/vol.h liquid hourly space velocity (LHSV), 39,2 · 10⁵ Pa (40 kp/cm²) pressure and 320–340 °C. The sulphur compounds were removed from the hydrogenated product by alkaline washing. The desulphurized feedstock was subsequently dried with dehydrated Na₂SO₄ and stored on "Ersorb" molecular sieve layer.

The characteristics of the desulphurized kerosene are shown in Table 1. The hydrocarbon group composition was determined by the so-called PONA method. The average carbon number of the fraction, \bar{c} was found to be 10.9, as calculated by the following relationship (valid only in case of sulphur-free fractions):

$$\bar{c} = \frac{\bar{M}(100 - H\%)}{1201}$$

where \bar{M} is the average molar weight and H% is the hydrogen percentage which can either be determined analytically or calculated on the basis of some physical properties.

Table 1
Characteristics of the feedstock (light kerosene)

ASTM Distillation

| | |
|-----------------------|--------|
| Initial Boiling Point | 158 °C |
| 10% | 170 °C |
| 50% | 186 °C |
| 90% | 210 °C |
| End Point | 236 °C |

| | |
|-------------------------------------|--------|
| Refractive index, n_D^{20} | 1.4370 |
| Density, d_4^{20} | 0.7818 |
| Average molecular weight, \bar{M} | 152.5 |
| Average carbon number, \bar{c} | 10.9 |

Hydrocarbon Group Composition, % by weight

| | |
|------------|------|
| Paraffins | 38.5 |
| Olefins | 0.4 |
| Naphthenes | 40.5 |
| Aromatics | 20.6 |

Detailed Composition of Aromatics, % by weight

| | |
|----------------------------|------|
| C ₈ aromatics | 1.3 |
| C ₉ aromatics | 7.3 |
| C ₁₀₊ aromatics | 12.0 |

Composition of C₉ aromatics, % by weight

| | |
|--------------------------|-----|
| Pseudocumene | 2.0 |
| Other trimethyl-benzenes | 1.7 |

The C₉ and C₁₀ aromatic hydrocarbon contents of the feedstock and the aromatic hydrocarbon contents of the reformates were determined by gas-chromatography (2).

Industrial aspects ruled both setting the experimental program and choosing the values of the technological parameters. From among the parameters, pressure and the hydrogen/hydrocarbon molar ratio are essentially determined by the catalyst so only the effect of the temperature and the liquid hourly space velocity (LHSV) were studied. In order to establish the effects of the chosen independent variables (t , T_s) with a good reliability from a minimum number of runs, the program was set up on the basis of experiment planning (8).

The second-order orthogonal plan for two variables gives the possibility to describe the yields of the different aromatic hydrocarbons in the studied parameter range with regression equations which can be easily calculated from the experimental data. In case of two variables, the regression equation becomes:

$$Y = b'_0 + b_1 x_1 + b_2 x_2 + b_{12} x_1 x_2 + b_{11} x'_1 + b_{22} x'_2$$

where

Y is the objective function

b_j is the j -th coefficient of the equation

x_j is the dimensionless form of j -th independent variable

$$x_j = \frac{z_j - z_j^0}{\Delta z_j}$$

$$x'_j = x_j^2 - x_j^{\bar{}}$$

$$z_j^0 = \frac{z_j^{\max} + z_j^{\min}}{2}$$

$$\Delta z_j = \frac{z_j^{\max} - z_j^{\min}}{2}$$

In the variables z , the reaction parameters appear with their natural values, z_j^{\max} and z_j^{\min} are the extreme values of the parameter range, z_j is the actual value of the j -th parameter.

Results and discussion

The results of our measurements carried out in the parameter range 470–530 °C and 1–3 vol/vol.hour LHSV are summarized in Table 2. Regression equations were written and calculated for the following objective functions:

Y_1 — liquid yield, % by wt

Y_2 — yield of total aromatics, % by wt

Y_3 — yield of C₉ aromatics, % by wt

Y_4 — yield of C₁₀ aromatics, % by wt

Y_5 — yield of trimethyl-benzenes, % by wt

Y_6 — yield of pseudocumene, % by wt.

Table 2 Yield of aromatics

Conditions of reforming

Catalyst: RD-150C

Feed: light kerosene

Pressure: $30.3 \cdot 10^5$ Pa (31 kp/cm²)H₂/CH mole ratio: 9

| Temperature, °C | 470 | 470 | 470 | 500 |
|------------------------------------------|-------------------------------|------|------|------|
| Liquid Hourly Space Velocity, vol/volhr. | 1 | 2 | 3 | 1 |
| <i>Component</i> | <i>Yield of components in</i> | | | |
| benzene | 1.3 | 0.9 | 0.8 | 1.2 |
| toluene | 5.2 | 4.9 | 4.7 | 6.6 |
| ethyl-benzene | 1.6 | 1.5 | 1.4 | 2.0 |
| meta and para-xylene | 4.3 | 4.2 | 4.0 | 5.7 |
| iso-propyl-benzene | 0.9 | 0.9 | 0.9 | 0.4 |
| ortho-xylene | 1.3 | 1.2 | 1.1 | 2.5 |
| n-propyl-benzene | 1.7 | 1.6 | 1.5 | 1.1 |
| meta- and para-ethyl-toluene | 3.0 | 3.0 | 2.8 | 4.3 |
| mesitylene | 2.4 | 2.3 | 2.3 | 2.9 |
| ortho-ethyl-toluene | 1.5 | 1.1 | 0.9 | 2.1 |
| pseudocumene | 4.2 | 4.2 | 4.1 | 6.2 |
| n-butyl- and 1,3-diethyl-benzene | 3.2 | 2.4 | 2.4 | 3.3 |
| 1-methyl-2-n-propyl-benzene | 0.6 | 0.5 | 0.5 | 0.8 |
| hemimellitene | 2.4 | 2.2 | 2.0 | 3.0 |
| indane | 0.7 | 0.6 | 0.5 | 0.9 |
| 1,2-dimethyl-4-ethyl-benzene | 2.3 | 2.0 | 1.8 | 3.5 |
| indene | 0.4 | 0.3 | 0.2 | 0.5 |
| 1,2-dimethyl-3-ethyl-benzene | 1.5 | 1.5 | 1.4 | 0.7 |
| durene | 1.4 | 1.2 | 1.0 | 1.1 |
| iso-durene | 2.5 | 2.3 | 2.1 | 2.1 |
| 5-methyl-indane | 0.6 | 0.4 | 0.4 | 0.3 |
| prehnitene | 2.8 | 2.8 | 2.8 | 1.5 |
| benzene | 1.3 | 0.9 | 0.8 | 1.2 |
| toluene | 5.2 | 4.9 | 4.7 | 6.6 |
| C ₈ aromatics | 7.2 | 6.9 | 6.5 | 10.2 |
| C ₉ aromatics | 17.5 | 16.2 | 15.2 | 21.4 |
| C ₁₀ aromatics | 14.6 | 13.2 | 12.4 | 13.3 |
| total aromatics | 45.8 | 42.1 | 39.6 | 52.7 |
| trimethyl-benzenes | 9.0 | 8.7 | 8.4 | 12.1 |
| liquid yield | 93.1 | 94.3 | 95.2 | 82.0 |

in reforming light kerosene

| 500 2 | 500 2 | 500 2 | 500 3 | 530 1 | 530 2 | 530 3 |
|----------------------------------|----------|----------|----------|----------|----------|----------|
| percentage by weight of the feed | | | | | | |
| 1.0 | 1.0 | 1.0 | 1.1 | 1.3 | 1.1 | 1.0 |
| 5.6 | 3.9 | 5.6 | 5.0 | 6.2 | 5.5 | 5.1 |
| 1.8 | 1.8 | 1.9 | 1.6 | 2.2 | 1.8 | 1.8 |
| 4.2 | 4.1 | 4.4 | 3.7 | 7.2 | 5.8 | 4.4 |
| 1.0 | 1.0 | 1.1 | 0.7 | 0.2 | 0.2 | 0.3 |
| 2.0 | 2.0 | 1.9 | 1.9 | 3.0 | 2.2 | 2.1 |
| 1.7 | 1.7 | 1.7 | 1.8 | 1.0 | 1.2 | 0.9 |
| 4.1 | 4.2 | 4.0 | 4.0 | 5.4 | 4.4 | 4.4 |
| 2.5 | 2.6 | 2.4 | 2.4 | 2.9 | 2.2 | 2.0 |
| 1.9 | 2.0 | 1.8 | 1.9 | 2.0 | 1.9 | 1.9 |
| 5.4 | 5.8 | 5.4 | 4.5 | 7.9 | 6.9 | 6.2 |
| 4.0 | 4.2 | 3.9 | 3.1 | 2.2 | 3.1 | 3.9 |
| 0.8 | 0.7 | 0.9 | 0.6 | 1.1 | 1.1 | 1.0 |
| 2.7 | 2.8 | 2.7 | 2.4 | 1.6 | 2.1 | 2.8 |
| 0.8 | 0.8 | 0.8 | 0.8 | 0.7 | 0.7 | 0.8 |
| 2.5 | 2.6 | 2.5 | 2.2 | 2.0 | 2.4 | 3.0 |
| 0.4 | 0.4 | 0.4 | 0.4 | 0.5 | 0.4 | 0.4 |
| 1.4 | 1.5 | 1.5 | 1.6 | 0.2 | 1.2 | 1.1 |
| 1.1 | 1.1 | 1.1 | 1.4 | 1.0 | 1.3 | 1.4 |
| 2.3 | 2.4 | 2.3 | 2.7 | 2.0 | 2.2 | 2.4 |
| 0.4 | 0.4 | 0.3 | 0.4 | 0.2 | 0.3 | 0.6 |
| 2.4 | 2.3 | 2.3 | 2.0 | 1.2 | 2.0 | 1.7 |
| 1.0 | 1.0 | 1.0 | 1.1 | 1.3 | 1.1 | 1.0 |
| 5.6 | 3.9 | 5.6 | 5.0 | 6.2 | 5.5 | 5.1 |
| 8.0 | 7.9 | 7.2 | 7.2 | 12.4 | 9.8 | 8.3 |
| 20.5 | 21.3 | 21.5 | 18.7 | 22.2 | 20.0 | 16.8 |
| 14.9 | 15.2 | 14.8 | 14.2 | 9.9 | 13.6 | 17.9 |
| 50.0 | 49.9 | 50.1 | 46.2 | 52.0 | 50.0 | 49.2 |
| 10.6 | 11.2 | 10.5 | 9.3 | 12.4 | 11.2 | 11.0 |
| 89.6 | 88.0 | 87.2 | 92.8 | 65.3 | 72.8 | 78.8 |

All the yields are expressed as percentages by weight of the charge.

The experimental data were processed in a WANG 2200 computer.

The reliability of regression coefficients was checked by the *t*-test, that of the fitting of the model by the *F*-test. After carrying out the statistical tests, the following equations were obtained:

$$Y_1 = 84.13 - 10.950x_1 + 4.440x_2 + 2.850x_1x_2 - 4.439x_1'$$

$$Y_2 = 47.51 + 3.950x_1 - 2.583x_2 + 0.850x_1x_2 - 3.183x_1'$$

$$Y_3 = 18.76 + 1.683x_1 - 1.733x_2 - 2.317x_1'$$

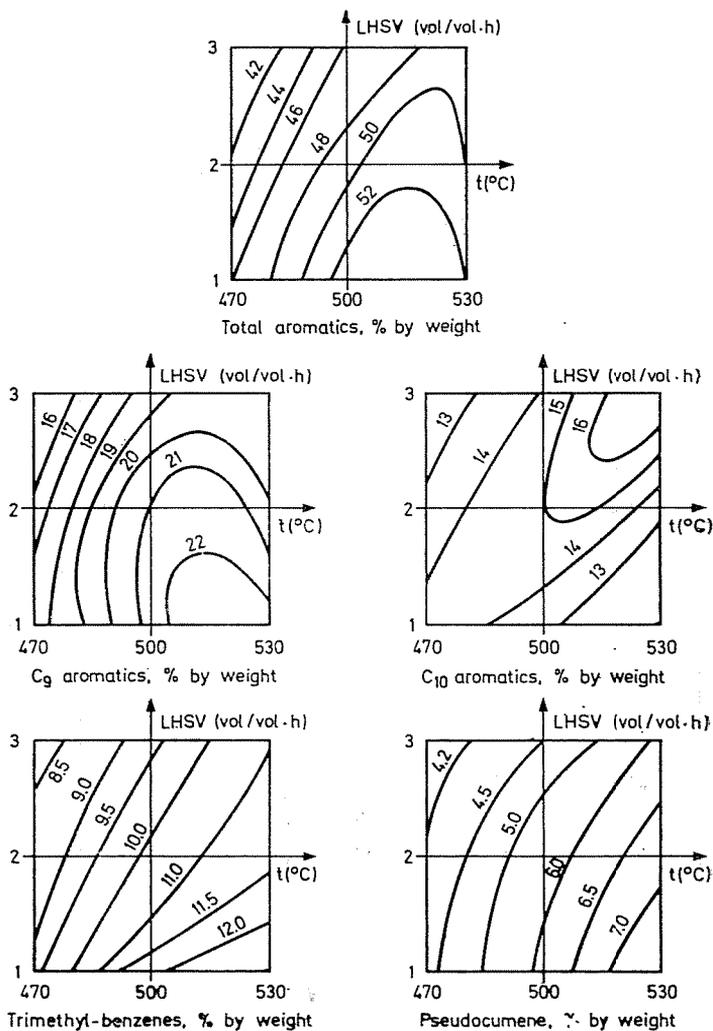


Fig. 1. Yields of aromatics from reforming light kerosene on RD-150C catalyst

$$Y_4 = 13.79 + 1.117x_2 + 2.550x_1x_2$$

$$Y_5 = 10.32 + 1.417x_1 - 0.800x_2 - 0.617x_1'$$

$$Y_6 = 5.52 + 1.417x_1 - 0.583x_2 - 0.400x_1x_2$$

In the equations, x_1 is the dimensionless form of reforming temperature, x_2 is that of LHSV.

The reliability of the regression coefficients in the above equations is 95%, the models fit the measured data with 5% error.

It is seen from the first equation that the liquid yield decreases sharply with the temperature elevation and the interaction term shows that this decrease is different for each LHSV value. The liquid yield is not very sensitive to the LHSV alteration.

Also the yield of total aromatics changes more pronouncedly with the temperature than with the LHSV. This can be clearly seen in Fig. 1 illustrating the experimental results. The comparatively low value of the coefficient of the interaction expression indicates the disadvantageous effect of the temperature elevation on the total aromatics yield caused by the decrease of the liquid yield.

Similar considerations are valid also for the yield of C₉ aromatics, though this yield depends more on the space velocity than do the other yields. This is seen in Table 3 where the corresponding b_j/b_0 values for each type of yield have been tabulated. The elevation of the temperature above 520 °C is especially disadvantageous to the yield of C₉ aromatics (partly by reducing the liquid yield), apparent from comparing the b_{11}/b_0 values. The reason for this tendency is the high sensitivity of the C₉ and C₁₀ hydrocarbon molecules to cracking, thus shifting the distribution of aromatics towards the lower carbon numbers.

The yields of trimethyl-benzenes and of pseudocumene increase monotonously with the temperature elevation and with the decrease of space velocity. The dependence on space velocity is slighter than that on the temperature.

Table 3
Relative coefficients of the regression equations

| Y \ b_j/b_0 | b_1/b_0 | b_2/b_0 | b_{11}/b_0 | b_{12}/b_0 |
|-----------------|-----------|-----------|--------------|--------------|
| Y_1 | -0.129 | 0.052 | 0.034 | -0.052 |
| Y_2 | 0.083 | -0.054 | 0.018 | -0.067 |
| Y_3 | 0.090 | -0.092 | — | -0.124 |
| Y_4 | — | +0.081 | 0.185 | — |
| Y_5 | 0.137 | -0.078 | — | -0.060 |
| Y_6 | 0.257 | -0.106 | -0.072 | — |

The interaction and second-order expressions have no marked effect on the yields.

Figure 1 shows the yield of trimethyl-benzenes and within that the yield of pseudocumene not to have extreme values in the parameter range studied. The highest yields are obtained at 530 °C and 1 vol/vol.hour LHSV, numerically 12.4 and 7.9 percent by weight, respectively. At a constant 1 vol/vol.hour space velocity the total aromatics yield and within that the yield of C₉ aromatics show a maximum as a function of temperature. On the basis of the equation

$$\frac{dY_2}{dx_1} = 3.950 + 0.850 x_2 - 6.366 x_1$$

where $x_2 = -1$, the temperature belonging to the maximum (53% by wt) of total aromatics yield is 515 °C. The maximum yield of C₉ aromatics (22.3% by wt) may be expected at 511 °C. The yield of C₁₀ aromatics has no extreme value in the parameter range studied, the elevation of temperature and increase of space velocity increase this yield up to 17.9% by wt at 530 °C and 3 vol/vol.hour LHSV.

Studying the yield data in Table 2, the following statements can be made on the individual C₉ aromatics isomers:

- The yield of iso- and normal-propyl-benzene — present in concentrations lower than 2% — decreases with decreasing space velocities and at temperatures above 500 °C.
- The yield of ortho-ethyl-toluene and the combined yield of the meta- and para-ethyl-toluene at 500 and 530 °C are 1.8–2.1 and 4.1–5.4 % by wt, resp., that is, slightly increasing with increasing reforming severity.
- Among the trimethyl-benzenes, pseudocumene has the greatest thermodynamic stability. Its yield increases from 4.1 to 7.9% by wt with increasing reforming severity (elevated temperature and reduced space velocity). The yield of mesitylene (1,3,5-trimethyl-benzene, a compound of considerable industrial interest) changes between 2.0–2.9% by wt, that of hemimellitene (1,2,3-trimethyl-benzene) between 1.6–3.0% by wt. The mesitylene to hemimellitene ratio increases with the elevation of temperature, as the rate of aromatic isomerization reactions increases.
- Indane and indene are formed in concentrations lower than 1%.
- Increasing the severity of the reforming conditions, the concentration of thermodynamically more stable alkyl-benzene isomers increases. This is seen in Table 4 where percentual distributions of the C₉ alkyl-benzenes measured at different space velocities at 530 °C are compared with the thermodynamic equilibrium data (9).

The distribution data in Table 4 indicate that the ratios of pseudocumene, mesitylene, and meta- plus para-ethyl-toluene increase with decreasing space velocities because of secondary isomerization and dealkylation.

Table 4
Distribution of C₉ alkyl-benzenes/percent by weight/at 530 °C

| Compound | LHSV, vol/vol · hr | | | Thermodynamic equilibrium |
|-----------------------------|--------------------|-------|-------|---------------------------|
| | 1 | 2 | 3 | |
| iso-propyl-benzene | 1.0 | 1.1 | 1.5 | 1.3 |
| n-propyl-benzene | 4.8 | 6.3 | 4.6 | 3.9 |
| o-ethyl-toluene | 9.5 | 10.0 | 9.7 | 8.2 |
| meta- and para-ethyltoluene | 25.7 | 23.4 | 22.6 | 31.5 |
| pseudocumene | 37.6 | 36.5 | 31.9 | 34.2 |
| mesitylene | 13.8 | 11.6 | 10.3 | 11.5 |
| hemimellitene | 7.6 | 11.1 | 14.4 | 9.4 |
| | 100.0 | 100.0 | 100.0 | 100.0 |

The amount of C₁₀ aromatics within the total aromatics depends on the conditions of reforming. Increasing the reforming severity, the distribution of aromatics is shifted towards the lower carbon numbers because of increased dealkylation. This is illustrated also by the data in Table 5, where Y_4/Y_2 values are shown.

Table 5

Fraction of C₁₀ aromatics within the total aromatics as a function of the conditions of reforming

| Temperature, °C | Y_4/Y_2 (Yield of C ₁₀ aromatics/Yield of total aromatics) | | |
|--------------------|----------------------------------------------------------------------------|-------|-------|
| | 470 | 500 | 530 |
| LHSV, vol/vol · hr | | | |
| 1 | 0.319 | 0.252 | 0.190 |
| 2 | 0.314 | 0.300 | 0.272 |
| 3 | 0.313 | 0.307 | 0.364 |

The data in Table 5 indicate that the Y_4 to Y_2 ratio is especially sensitive to the alteration of the other parameters at low space velocities and high temperatures. The bulk of C₁₀ alkyl-aromatics consists of tetra-methyl-benzenes and dimethyl-ethyl-benzenes. In the parameter range studied, iso-durene (1,2,3,5-tetramethylbenzene) and prehnithene (1,2,3,4-tetramethyl-benzene) have the highest yields, 2.0–2.5 and 1.2–2.8% by wt, respectively.

The yields of durene (1,2,4,5-tetramethyl-benzene) — the most important isomer from the industrial point of view — are between 1.0–1.4% by wt, its concentration in the reformat does not exceed 2% by wt. With increasing the severity of the reforming conditions, the concentrations of iso-durene and durene increase among the tetramethyl-benzenes.

Of the dimethyl-ethyl-benzenes, 1,2-dimethyl-4-ethyl-benzene has the highest yield reaching 3.5% by wt at 500 °C and 1 vol/vol.hour space velocity. The combined yields of n-butyl- and 1,3-diethyl-benzene aromatics may rise to 2.2–2.4% by wt while yields of the methyl-propyl-benzenes and methyl-indanes remain below 1% by wt.

In reforming kerosene, the yield of total aromatics and that of C₉ aromatics was seen to be higher than in reforming naphtha boiling between 90 and 180 °C [10]. On the other hand, our earlier results have indicated that for producing C₉ aromatics it is more advantageous to reform naphtha fractions with 35–40 °C boiling range between 130 and 190 °C than to reform kerosene (1).

This observation can be explained by the fact that kerosene contains significant amounts of C₁₁–C₁₂ hydrocarbons, which only give C₉ aromatics under severe reforming conditions. Under such conditions, however, hydrocracking is also enhanced, resulting in sharply reduced liquid yields.

Figure 2 shows aromatics yields obtained by reforming different hydrocarbon fractions on RD-150C catalyst. In reforming runs at 500 °C and 2 vol/vol.hour LHSV the yield of total aromatics as well as that of the C₉ aromatics and within that the yield of trimethyl-benzenes show a common maximum when the average carbon number of the feedstock is about 10.

The highest yield of pseudocumene can be expected already at $\bar{c} = 9.4$, attributed to the high thermodynamic stability of this compound [1]. The yield of C₁₀ aromatics reaches 15% by wt only in cases where the average carbon number of the feed is above 10.

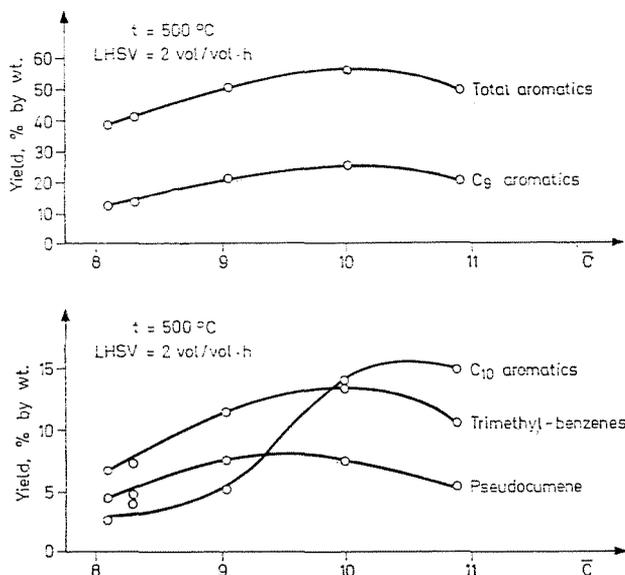


Fig. 2. Yields of aromatics from reforming different naphtha fractions with different average carbon numbers. Catalyst: RD-150C

As a conclusion it can be stated that in reforming kerosene boiling between 158–236 °C, a considerable portion of C₉ aromatics is formed from hydrocarbons having higher carbon numbers than 9. This means that reforming should be carried out at 1 vol/vol.hour space velocity and at least at 515 °C to facilitate proper dealkylation. Because of the high sensitivity of the big molecules to cracking, increased reforming severity results in sharply reduced liquid yields, causing the yields of C₉ aromatics to be lower than those obtained from heavy naphtha fractions. Thus, such an increase ($\bar{c} = 10.9$) in the average carbon number of the feedstock is not advantageous from the point of view of C₉ aromatics production.

C₁₀ aromatics are produced in significant total yields (12–16% by wt), but the amount of the individual isomers does not exceed 1–3% by wt. The concentration of the practically important durenene is not higher than 2% by wt even in reformates rich in aromatics.

Summary

Effect of the temperature and space velocity on yields of aromatics were studied in reforming light kerosene, boiling between 158 and 236 °C and with an initial aromatics percentage of 20.6% by wt. The reforming runs were carried out on RD-150C platinum-alumina catalyst at constant pressure and hydrogen/hydrocarbon mole ratio (30.3 · 10⁵ Pa, and 9, resp.). The yields of aromatics in the parameter range 470–530 °C and 1–3 vol/vol · hr were described with regression equations.

Maximum yield of C₉ aromatics (22.3% by wt) was obtained at 511 °C, 1 vol/vol · hr; that of pseudocumene amounted to 7.9% by wt at 530 °C, 1 vol/vol · hr and that of C₁₀ aromatics reached 17.9% by wt at 530 °C, 3 vol/vol · hr.

Reforming of light kerosene has shown, that a feedstock with such a high initial boiling point and average carbon number ($\bar{c} = 10.9$) isn't advantageous from the point of C₉ aromatics production: the yields of C₉ aromatics obtained earlier /1/ by reforming heavy naphtha fractions were higher than those measured in this work.

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