# *Studies on Constituents of Coal Carbonization Light Oil. II. On Saturated Hydrocarbons Contained in Low-temperature Coal Carbonization Light Oil*

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Apparently, constituents of high-temperature coal carbonization light oil are mainly aromatic hydrocarbons and those of low-temperature coal carbonization light oil, are mainly saturated hydrocarbons[1]. But, resulting data of identification of saturated hydrocarbons in low-temperature coal carbonization light oil, are scanty. In 1955, some of them have been reported by A. Jäger and G. Kattwinkel[2] and the following results have been found. 1) The saturated hydrocarbons, contained in low-temperature coal carbonization light oil, consist mainly of straight-chain paraffins. 2) The branched-chain paraffins contained in this oil are all monomethylparaffins. 3) Contents of cycloparaffins are scanty.

Ohyama[3] reported that the neutral oil contained in low-temperature coal carbonization light oil, Ube Kôsan Co., Ltd. production, amounted to about 90 per cent and also the contents of the saturated hydrocarbon in the neutral oil to about 40 per cent. However, the constituents of the individual saturated hydrocarbons in this light oil and their contents have never been ascertained. Therefore, the author has studied on the constituents of the saturated hydrocarbons contained in this oil and found five straight-chain hydrocarbons (*n*-hexane, *n*-heptane, *n*-octane, *n*-nonane and *n*-decane) and five branched-chain hydrocarbons (2-methylhexane, 2-methylheptane, 2-methyloctane, 2-methylnonane and 3-methylnonane) and four cycloparaffins (cyclohexane, methylcyclohexane, 1, 3-dimethylcyclohexane and 1, 3, 5-trimethylcyclohexane) and determined their contents approximately and compared them with the result obtained in a previous paper[4].

## Experimental

**Materials.**—Residual oil after extraction of acidic and basic oils in low-temperature coal carbonization light oil (carbonization temperature, 780°C) manufactured by Ube Kôsan Co., Ltd. in August, 1954, with dilute sulfuric acid and alkaline solution, brown colored, was used. This oil kept in a glass reagent bottle for some years and precipitation, amounting to about 1.5 per cent of this oil, was produced. The oil, removing the precipitation by filtration, had a boiling range of 92∼265°C, $d_4^{25}=0.8062$ and $n_D^{25}=1.456$. The contents of olefinic, aromatic and saturated hydrocarbons in this oil, were determined to be olefins 9.5 per cent (volume absorbed by 80% $H_2SO_4$), aromatics 49 per cent (volume absorbed by 98% $H_2SO_4$) and saturated hydrocarbons 41.5 per cent (residue) by sulfuric acid absorption method.

**Separation of the Saturated Hydrocarbon Oil.**—Olefinic and aromatic hydrocarbons contained in 320 g. of the material, were removed by extracting with concentrated sulfuric acid. The residual oil was washed by water, neutralized with 10 per cent sodium hydroxide solution, rewashed similarly and dried with calcium chloride to obtain 148 g. of crude saturated hydrocarbon oil (46.2 per cent by weight of the material). This oil was fractionated and 100 g. of 98∼220°C fraction, which had $d_4^{25}=0.7403$ and $n_D^{25}=1.417$, and 45 g. of residual oil, were obtained. Residual aromatic hydrocarbons in 100 g. of 98∼220°C fraction, were again removed completely with concentrated sulfuric acid and washed with water and neutralized with alkaline solution and rewashed similarly and dried with calcium chloride and 89 g. (27.8 per cent by weight of the material) of saturated hydrocarbon oil

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[1] M. Vahrman, *J. Appl. Chem.*, 2, 532 (1952).

[2] A. Jäger und G. Kattwinkel, *Erdöl und Köhle*, 9, 629, 706 (1955).

[3] G. Ohyama, "Studies on Ube Coal (Ube-tan no Seijô to Gôsei-yô Gasu oyobi Teion-târu no Seizô to Shori)", Ube Kôsan Co., Ltd., Japan (1951), p. 200.

[4] H. Yasui, This Bulletin, 33, 1493 (1960).

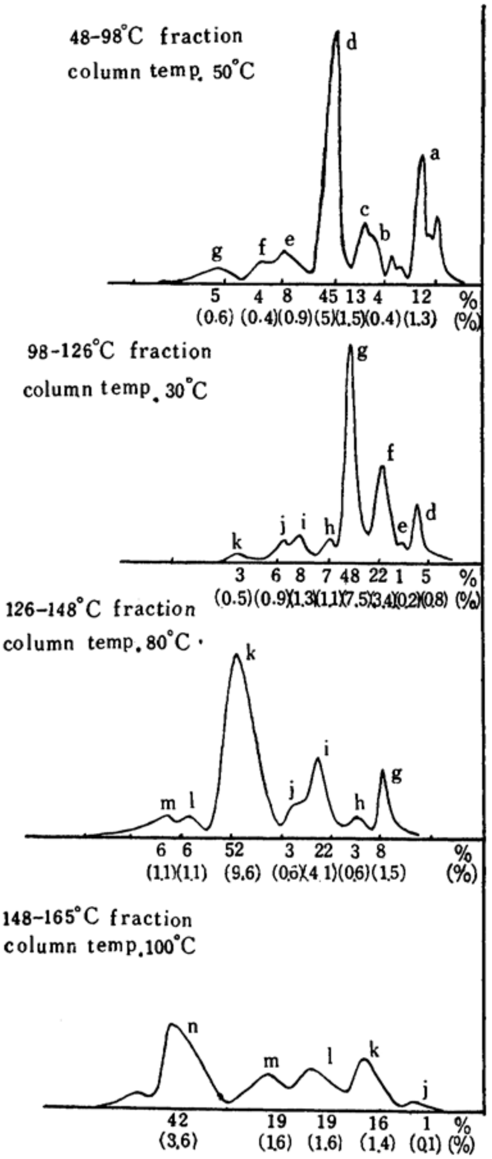

Fig. 1. Gas chromatograms of each fraction. $H_2$ flow rate 100 ml./min.

a …… *n*-Hexane
b …… Cyclohexane
c …… 2-Methylhexane
d …… *n*-Heptane
e …… Methylcyclohexane
f …… 2-Methylheptane
g …… *n*-Octane
h …… 1,3-Dimethylcyclohexane
i …… 2-Methyloctane
j …… 1,3,5-Trimethylcyclohexane
k …… *n*-Nonane
l …… 2-Methylnonane
m …… 3-Methylnonane
n …… *n*-Decane
% …… Per cent of contents to each fraction
(%) … Per cent of contents to Sample A

(Sample A), which had $d_4^{25}=0.7397$ and $n_D^{25}=1.414$, was obtained. 87 g. (110 ml.) of this oil was submitted to fractional distillation by Podbielniak equipment with metallic sodium and five fractions shown in Table I, were obtained.

TABLE I. FRACTIONS OBTAINED BY FRACTIONAL DISTILLATION OF 87 g. OF SAMPLE A

| Frac. °C | Main frac. °C | g. | Wt. % | $d_4^{25}$ | $n_D^{25}$ |
|---|---|---|---|---|---|
| 48～ 98 | 87～ 98 | 9.6 | 11.1 | 0.6877 | 1.389 |
| 98～126 | 114～121 | 13.6 | 15.6 | 0.7146 | 1.401 |
| 126～148 | 135～145 | 16.2 | 18.6 | 0.7340 | 1.409 |
| 148～165 | 150～164 | 7.5 | 8.6 | 0.7478 | 1.416 |
| 165～184 | 169～184 | 4.5 | 5.1 | 0.7481 | 1.422 |
| Residue | — | 34.4 | 39.5 | 0.7924 | 1.442 |

Analyses were carried out on 48～98°C, 98～126°C, 126～148°C and 148～165°C fractions in Table I.

**Gas Chromatographic Analysis.**—Gas chromatographic analysis of 48～98°C, 98～126°C, 126～148°C and 148°～165°C fractions was performed with a Perkin-Elmer vapor fractometer. Hydrogen was used as carrier gas and di-*n*-decylphthalate was used as liquid substrate. The identification of the peak in the chromatograms was based on synthetic hydrocarbon data. The retention time of hydrocarbons contained in these fractions was obtained from each synthetic hydrocarbon and comparing the retention times of the peak chromatogram of each synthetic hydrocarbon under identical conditions. Contents of the hydrocarbons in these fractions were calculated from the area of each measurable peak.

**Identification of *n*-Paraffn.**—*n*-Paraffins in these fractions were isolated by means of urea adducts[5]. According to the estimation *n*-paraffin was contained about 50 per cent in each fraction. It was added to urea (about three times in weight of the *n*-paraffin) moistened with methanol (about a fifth of the weight of the urea used). The suspension was stirred for 30 min. at 0°C in 98～126°C, or 20°C in 126～148°C and 148～165°C fractions, and filtered, and unreacted oils were removed. The mixture of urea adduct obtained and the residual urea was washed with anhydrous ether, removed under reduced pressure, decomposed with water, and oil (*n*-paraffin) was obtained. Unreacted oil was once more added to urea moistened with methanol and treated as mentioned above until no more oil (*n*-paraffin) was obtained by decomposition with water of urea adduct. The result was shown in Table II. As stated by the author[4] and Kuwata and co-workers[6] previously, it is difficult to isolate *n*-hexane and *n*-heptane expected to be contained mainly in 48～98°C fraction. Therefore this method was not employed.

**IR Spectra of Oils Obtained by Decomposition with Water of Urea Adducts.**—IR spectra of 48～98°C fraction and oils isolated by decomposition

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[5] J. Zimmerschied et al., *Ind. Eng. Chem.*, **42**, 1300 (1950).

[6] T. Kuwata, Y. Mizutani and E. Tamura, *J. Chem. Soc. Japan, Ind. Chem. Sec.* (*Kogyo Kagaku Zasshi*) **55**, 799 (1952).

TABLE II. ISOLATION OF *n*-PARAFFIN BY UREA ADDUCTS IN 98~126°, 126~148° AND 148~165°C FRACTION

| Fraction | | Oil isolated by urea adduct (*n*-paraffin) | | | | | Residual oil | |
|---|---|---|---|---|---|---|---|---|
| °C | g. | g. | % | b. p., °C | $d_4^{25}$ | $n_D^{25}$ | g. | % |
| 98~126 | 12 | 5.5 | 46 | 120~124 | 0.6983 | 1.396 | 5 | 42 |
| 126~148 | 14 | 6.5 | 54 | 142~151 | 0.7111 | 1.405 | 6.5 | 32 |
| 148~165 | 6.5 | 3.0 | 46 | 152~170 | 0.7248 | 1.410 | 2.2 | 34 |

with water of urea adducts of 98~126°C, 126~148°C and 148~165°C fractions, were measured with an Oh'yô Kôken infrared spectrophotometer.

**48~98°C Fraction.**—In IR spectrum bands of this fraction, those of *n*-hexane (wavelengths; 7.45, 7.73, 8.02, 8.20, 8.6, 8.79, 9.38, 9.46, 9.86, 9.95, 10.29, 11.04, 11.24, 11.31, 11.51, 12.13, 12.6, 12.94, 13.18, 13.78 $\mu$), *n*-heptane (wavelengths; 7.45, 7.68, 7.81, 8.09, 8.27, 8.78, 9.3, 9.79, 10.4, 10.74, 11.1, 11.42, 11.57, 12.01, 12.91, 13.54 13.83 $\mu$), 2-methylhexane (wavelengths; 7.3, 7.41, 7.71, 7.97, 8.18, 8.53, 8.73, 9.3, 9.53, 9.7, 9.85, 10.46, 10.63, 11.0, 11.17, 12.16, 12.81, 13.74 $\mu$), 2-methylheptane (wavelengths; 7.32, 7.46, 7.67, 8.05, 8.28, 8.54, 8.74, 9.22, 9.56, 9.98, 10.41, 10.53, 10.71, 10.88, 11.2, 11.94, 12.31, 13.04, 13.26, 13.81 $\mu$), cyclohexane (wavelengths; 7.4, 7.95, 8.7, 9.1, 9.6, 9.9, 11.1, 11.68 $\mu$) and methylcyclohexane (wavelengths; (7.28, 7.44, 7.69, 8.02, 8.62, 9.06, 9.7, 10.38, 11.02, 11.49, 11.87 $\mu$) were observed.

**Oil Isolated by Decomposition with Water of the Urea Adduct of 98~126°C Fraction.**—Physical constants (Table II) of this oil were analogous to those of *n*-octane[7] (b. p. 125.6°C, $d_4^{20}=0.703$, $n_D^{20}=1.397$). In IR spectrum bands of this oil, those of *n*-heptane, *n*-octane (wavelengths; 7.46, 7.68, 7.87, 8.16, 8.33, 8.8, 9.27, 9.65, 9.95, 10.34, 10.92, 11.1, 11.21, 11.4, 11.84, 12.6, 13.08, 13.52, 13.85 $\mu$) and *n*-nonane (wavelengths; 7.46, 7.67, 7.94, 8.22, 8.81, 9.2, 9.41, 9.58, 10.29, 10.79, 11.22, 11.4, 11.84, 12.05, 12.89, 13.29, 13.86 $\mu$) were observed.

**Oil Isolated by Decomposition with Water of the Urea Adduct of 126~148°C Fraction.**—Physical constants (Table II) of this oil were analogous to those of *n*-nonane[7] (b. p. 150.7°C, $d_4^{20}=0.7111$, $n_D^{20}=1.405$), and in IR spectrum bands of this oil, those of *n*-nonane and *n*-octane were observed.

**Oil Isolated by Decomposition with Water of the Urea Adduct of 148~165°C Fraction.**—Physical constants (Table II) of this oil denoted the values between those of *n*-nonane and *n*-decane[7] (b. p. 174°C, $d_4^{20}=0.7298$, $n_D^{20}=1.412$), and therefore the fraction was expected to be a mixture of *n*-nonane and *n*-decane. In IR spectrum bands of this oil, those of *n*-nonane and *n*-decane (wavelengths; 7.26, 7.45, 7.67, 7.82, 8.01, 8.25, 8.81, 9.16, 9.29, 9.92, 10.05, 10.86, 11.15, 11.83, 12.28, 13.0, 13.85 $\mu$) were observed.

**Gas Chromatographic Analysis.**—The gas chromatographic analysis of oils, obtained by decomposition with water of urea adducts of 98~126°C, 126~148°C and 148~165°C fraction, was performed and chromatograms of these oils were shown in Fig. 2.

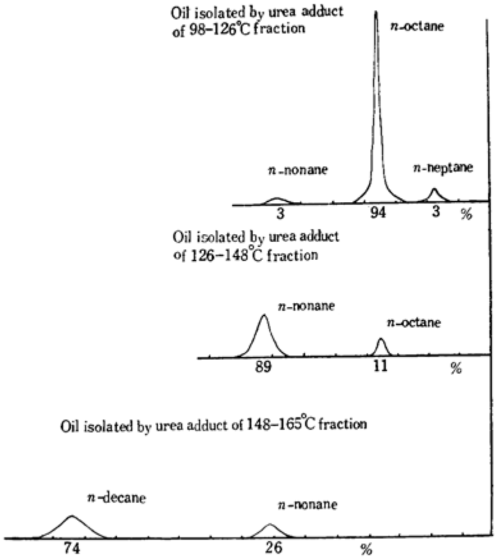

Fig. 2. Gas chromatograms of oils obtained by decomposition with water of urea adducts of each fraction.
Column temp., 100°C; $H_2$ flow rate, 98.6 ml./min.

**Identification of Cycloparaffin and Isoparaffin—IR Spectra.**—IR spectra of residual oils, in 98~126°C, 126~148°C and 148~165°C fractions, are measured with an Oh'yô Kôken spectrophotometer.

**Residual Oil in 98~126°C Fraction Shown in Table II.**—Physical constants of this oil are b. p. 110~125°C, $d_4^{25}=0.7218$, $n_D^{25}=1.404$. In IR spectrum bands of this oil, those of 2-methylheptane, 2-methyloctane (wavelengths; 7.6, 7.67, 7.86, 8.15, 8.33, 8.54, 9.14, 9.3, 9.4, 9.77, 10.18, 10.55, 10.9, 11.25, 11.42, 12.15, 12.7, 13.5, 13.8 $\mu$), 1,3-dimethylcyclohexane (wavelengths; 7.25, 7.66, 7.88, 8.36, 8.6, 8.96, 9.28, 9.45, 10.0, 10.2, 10.53, 10.71, 11.43, 11.72, 12.13 $\mu$) and 1,3,5-trimethylcyclohexane (wavelengths; 7.52, 7.92, 8.53, 8.87, 8.96, 9.35, 9.56, 9.66, 10.0, 10.63, 10.74, 11.84, 11.91, 12.78 $\mu$) were observed.

**Residual Oil in 126~148°C Fraction Shown in Table II.**—Physical constants of this oil are b. p. 139~151°C, $d_4^{25}=0.7406$ and $n_D^{25}=1.414$. In IR spectrum bands of this oil, those of 2-methyloctane, 1,3-dimethylcyclohexane, 2-methylnonane (wavelengths; 7.67, 7.93, 8.21, 8.40, 8.54, 8.76, 9.12, 9.4, 10.07, 10.4, 10.65, 10.9, 11.1, 11.48, 12.0,

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[7] G. Egloff, "Physical Constants of Pure Hydrocarbons", Vol. I, Reinhold Publ. Corp., New York (1939), p. 49.

12.32, 13.05, 13.6, 13.81 μ), 3-methylnonane (wavelengths; 7.67, 7.83, 8.16, 8.36, 8.66, 9.12, 9.43, 9.94, 11.28, 11.79, 12.2, 12.78, 12.96, 13.6, 13.8 μ) and 1, 3, 5-trimethylcyclohexane, were observed.

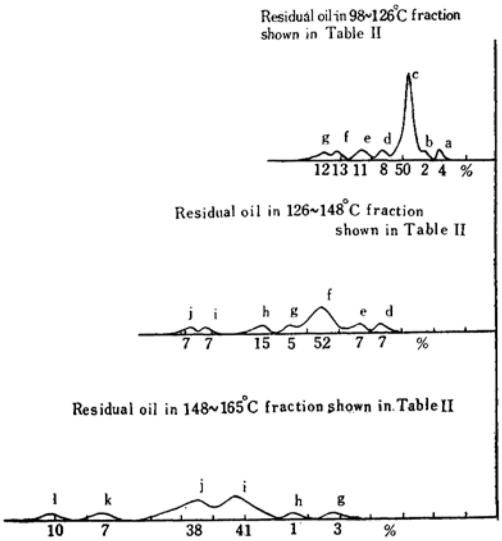

Fig. 3. Gas chromatograms of residual oils in each fraction. Column temp. 100°C; $H_2$ flow rate 98.6 ml./min.

a …… *n*-Heptane
b …… Methylcyclohexane
c …… 2-Methylheptane
d …… *n*-Octane
e …… 1, 3-Dimethylcyclohexane
f …… 2-Methyloctane
g …… 1, 3, 5-Trimethylcyclohexane
h …… *n*-Nonane
i …… 2-Methylnonane
j …… 3-Methylnonane
k …… *n*-Decane

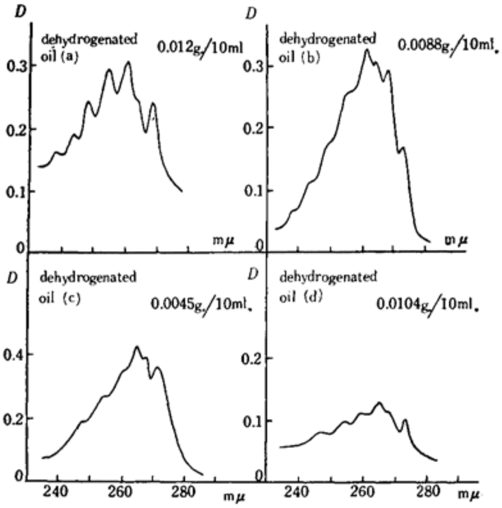

Fig. 4. Ultraviolet spectra of dehydrogenated oils.

**Residual Oil in 148∼165°C Fraction Shown in Table II.**—Physical constants of this oil are b. p. 148∼164°C, $d_4^{25}=0.7673$ and $n_D^{25}=1.425$. In IR spectrum bands of this oil, those of 2-methylnonane, 3-methylnonane and 1, 3, 5-trimethylcyclohexane were observed.

**Gas Chromatographic Analysis.**—Gas chromatographic analysis of residual oils, in 98∼126°C, 126∼148°C and 148∼165°C fractions was performed and the chromatograms were shown in Fig. 3.

**Dehydrogenation**[4,8]. — Dehydrogenation of 48∼98°C fraction (a) and residual oils (b), (c) and (d) in 98∼126°C, 126∼148°C and 148∼165°C fractions shown in Table II, was carried out with Pt-C catalyst prepared by the method described previously[4], at 290∼300°C repeatedly, until no hydrogen was producible and by this method, cycloparaffins contained in these fractions were changed to corresponding aromatic hydrocarbons. The result was shown in Table III.

TABLE III. DEHYDROGENATION
REAC. TEMP. 290∼300°C; DROPPING VELOCITY 5∼6 g./hr.

| Sample | g. | Dehydrogenated oil g. | Gas produced ml. (N. T. P.) | $H_2$* ml. (N. T. P.) |
|---|---|---|---|---|
| (a) | 4 | 2.8 | 284.8 | 263.5 |
| (b) | 4 | 3.0 | 632.9 | 623.8 |
| (c) | 2 | 1.3 | 144.1 | 136.5 |
| (d) | 2 | 1.5 | 48.1 | 40.6 |

* The gas produced by dehydrogenation was analyzed by the Hempel method, and the volume of hydrogen produced was the volume of the gas removed of that of air.

**Identification of Aromatic Hydrocarbons Produced by Dehydrogenation. — Ultraviolet Spectra of the Dehydrogenated Oils.** — Ultraviolet spectra of dehydrogenated oils, dissolved in purified ethanol, were measured with a Beckman spectrophotometer model DU, and were shown in Fig. 4.

**Nitration of Dehydrogenated Oil**[9]. — Dehydrogenated oil was added to a mixture of 1 vol. of concentrated nitric acid and 2 vol. of concentrated sulfuric acid and shaken, then boiled for five minutes and poured into cold water, and crude nitro-compounds were obtained. These compounds were separated by fractional recrystallization from ethanol solution, obtaining pure nitro-compounds. The result was shown in Table IV.

From Fig. 3 and Table IV, aromatic hydrocarcons produced by dehydrogenation were confirmed, and their original cycloparaffins and the ratio of their contents were estimated as shown in Table V.

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8) E. Berl and W. Koerber, *Ind. Eng. Chem., Anal. Ed.*, 12, 175 (1940).

9) S. P. Mulliken, "A Method for the Identification of Pure Organic Compounds", Vol. I, John Wiley & Sons Inc., New York, p. 200.

TABLE IV. NITRO-COMPOUNDS OBTAINED BY NITRATION OF DEHYDROGENATED OILS

| | Dehydrogenated oil | Nitro-compound | | | | |
|---|---|---|---|---|---|---|
| | | Obtained | | | Referred[9)] | |
| | | Crude | Recrystallized | M. p. | Compound | M. p. |
| | g. | g. | g. | °C | | °C |
| (a) | 1.0 | 0.3 | 0.04 | 87～ 90 | *m*-Dinitrobenzene | 90 |
| | | | 0.11 | 67～ 69 | 2, 4-Dinitrotoluene | 70 |
| (b) | 1.0 | 0.2 | 0.22 | 66～ 68 | 〃 | 〃 |
| | | | 0.04 | 177～179 | 2, 4, 6-Trinitro-*m*-xylene | 179 |
| | | | 0.03 | 228～231 | 2, 4, 6-Trinitro-1, 3, 5-trimethylbenzene | 232 |
| (c) | 0.75 | 0.2 | 0.03 | 177～180 | 2, 4, 6-Trinitro-*m*-xylene | 179 |
| | | | 0.04 | 228～232 | 2, 4, 6-Trinitro-1, 3, 5-trimethylbenzene | 232 |
| (d) | 1.0 | 0.04 | 0.02 | 230～232 | 〃 | 〃 |

TABLE V. CYCLOPARAFFINS CONTAINED IN (a), (b), (c) AND (d)

| | Cycloparaffin | Ratio* |
|---|---|---|
| (a) | Cyclohexane | 1 |
| | | : |
| | Methylcyclohexane | 3 |
| (b) | Methylcyclohexane | 2 |
| | | : |
| | 1, 3-Dimethylcyclohexane | 4 |
| | | : |
| | 1, 3, 5-Trimethylcyclohexane | 3 |
| (c) | 1, 3-Dimethylcyclohexane | 3 |
| | | : |
| | 1, 3, 5-Trimethylcyclohexane | 4 |
| (d) | 1, 3, 5-Trimethylcyclohexane | |

* The ratio of cycloparaffins contained in each fraction was calculated from weights of nitro-compounds obtained.

## Results and Discussion

Saturated hydrocarbons identified from the low-temperature coal carbonization light oil produced at Ube Kôsan Co. Ltd. in this research are given below.

It has become clear, that the constituents of the saturated hydrocarbons, contained in this oil, consist of five straight-chain hydrocarbon (*n*-hexane, *n*-heptane, *n*-octane, *n*-nonane and *n*-decane) and five branched-chain hydrocarbon (2-methylhexane, 2-methylheptane, 2-methyloctane, 2-methylnonane and 3-methylnonane) and four cycloparaffins (cyclohexane, methylcyclohexane, 1, 3-dimethylcyclohexane and 1, 3, 5-trimethylcyclohexane). Moreover, percentage of their contents in the saturated hydrocarbon oil (Sample A) separated from the material, is given in the gas chromatograms shown in Fig. 1, and Table VI.

The following observation is based on a study of the data presented in Table VI.

1) Saturated hydrocarbons, contained in the low-temperature coal carbonization light oil, are composed of about 91 per cent of saturated aliphatic hydrocarbon and about 9 per cent of cycloparaffinic hydrocarbon.

2) Saturated aliphatic hydrocarbons contained in this oil, are composed of two parts of *n*-paraffins and one part of isoparaffins.

3) The isoparaffins are all monomethylparaffins and these monomethylparaffins are nearly all 2-methylparaffins.

Comparing the saturated hydrocarbons, contained in high-temperature coal carbonization

TABLE VI. SATURATED HYDROCARBONS IDENTIFIED AND PERCENTAGE OF THEIR CONTENTS IN SAMPLE A

| *n*-Paraffin | | Isoparaffin | | Cycloparaffin | |
|---|---|---|---|---|---|
| Compound | % | Compound | % | Compound | % |
| *n*-Hexane | 1.3 | 2-Methylhexane | 1.5 | Cyclohexane | 0.4 |
| *n*-Heptane | 5.8 | 2-Methylheptane | 3.8 | Methylcyclohexane | 1.1 |
| *n*-Octane | 9.6 | 2-Methyloctane | 5.4 | 1, 3-Dimethylcyclohexane | 1.7 |
| *n*-Nonane | 11.5 | 2-Methylnonane | 2.7 | 1, 3, 5-Trimethylcyclohexane | 1.6 |
| *n*-Decane | 3.6 | 3-Methylnonane | 2.7 | | |
| Total | 31.8 | Total | 16.1 | Total | 4.8 |

light oil produced at Yawata Iron & Steel Co. Ltd., and identified by the author⁽⁴⁾, with those identified in this investigation, the following observations are made.

1) Saturated hydrocarbons identified from both low-temperature and high-temperature coal carbonization light oils, consist mainly of saturated aliphatic hydrocarbons of 90 per cent saturated hydrocarbons in low-temperature coal carbonization light oil, and about 85 per cent in high-temperature coal carbonization light oil.

2) In low-temperature coal carbonization light oil, the ratio of contents of *n*-paraffins and isoparaffins is about 2 : 1. In high-temperature coal carbonization light oil, about 1 : 1.5.

3) Isoparaffins, contained in low- and high-temperature coal carbonization light oils, are all monomethylparaffins, and those contained in low-temperature coal carbonization light oil are all 2-methylparaffins, except 3-methylnonane, and the monomethylparaffins contained in high-temperature coal carbonization light oil, consist of 3-methylparaffins and 4-methylparaffins except 2-methylheptane.

F. D. Rossini and B. J. Mair(10) studying the saturated hydrocarbons contained mainly in U. S. A. Mid-Continent, Ponca petroleum concluded that the saturated hydrocarbons consisted mainly of saturated aliphatic hydrocarbons, which contains a large quantity of *n*-paraffin and monomethylparaffin and a trace of branched paraffin which has many branchings. This conclusion is analogous to that of the present studies on saturated hydrocarbons contained in coal carbonization light oil.

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10) F. D. Rossini and B. J. Mair, *Ind. Eng. Chem. D. S.*, 3, 141 (1958).