Evaluation of Tobacco Quality from Pyrolytic Aspects

Part VII. Relationship between Chemical Constituents of Tobacco Leaves and Gaseous Constituents of Cigarette Smoke

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Relationship between the chemical constituents of tobacco leaves and the gaseous constituents of cigarette smoke from which \( K \) value was computed was discussed and the following presuppositions were demonstrated to be correct.

1. Fibrous substances in tobacco leaves are the main precursors of acetaldehyde, propionaldehyde, acrolein, acetone, methylethylketone, diacetyl, methanol, furan, an unknown compound, No. 6, and an unknown compound, No. 16 in cigarette smoke.

2. Sugars in tobacco leaves are the main precursors of 2-methylfuran and 2,5-dimethylfuran in cigarette smoke.

3. Resinous substances in tobacco leaves are the main precursors of isoprene and an unknown compound, No. 2 in cigarette smoke.

In previous papers,\(^1,2\) \( K \) value was proposed as a quality coefficient of tobacco leaves, and in successive papers,\(^3,4\) it was demonstrated that the aroma and/or taste of the smoke of flue-cured tobacco leaves were evaluated by \( K \) value and that \( K \) value was reasonable from the viewpoint of statistical analysis of the correlation between the gaseous constituents of cigarette smoke from which \( K \) value was computed and the aroma and/or taste of the smoke. The present paper describes the relationship between the chemical constituents of tobacco leaves and the gaseous constituents of cigarette smoke from which \( K \) value was computed.

MATERIALS AND METHODS

Materials. The cigarettes used for the analysis of the gas phase of the main stream smoke were 70×26 mm non-blended flue-cured tobaccos, conditioned at 20°C, 60% R.H. and sorted to a draw resistance. The ground tobaccos were used for analysis of chemical constituents of tobacco leaves.

Gas chromatography. The gas chromatograph used was a Perkin Elmer Model F-11 equipped with a flame ionization detector. Chromatographic separations were made on two connected columns, one was 2 m×1/8 inch o.d. stainless steel filled with 15% Carbowax 1500 on 60–80 mesh Chromosorb W, the other was 4 m×1/8 inch o.d. stainless steel filled with 15% polypropyrene glycol LB-550-X on 60–80 mesh Chromosorb W. The connected columns were operated isothermally at 30°C for 18 min and then programmed to 100°C (3°C/min) and maintained at 100°C until completion of elusion. Nitrogen was used as a carrier gas at a flow rate of 18.2 ml/min.

\(^4\) Y. Obi, M. Muramatsu, Y. Shimada and H. Sakurai, *ibid.*, No. 111, in press.
**Smoking conditions.** A smoking machine was operated to take a 35 ml of smoke during 2 seconds once per minute.

**Separation of gas phase of smoke.** Separation of the gas phase from the particulate phase was accomplished by the use of a glass fiber Cambridge filter placed immediately behind the cigarette during smoking.

**Introduction of gas phase into the gas chromatograph.** The separated gas phase at the fifth puff introduced into a 1 ml gas sampling loop connected to the by-pass of the carrier gas was directly subjected to gas chromatography for analysis by turning the 3-ways valve as carrier gas passes through the gas sampling loop.

**Measurement of peak areas of gaseous compounds.** The peak areas of each gaseous compound on gas chromatogram were measured by a planimeter.

**Analytical methods of chemical constituents of tobacco leaves.** Each procedure followed the standard analytical methods5) of Japan Monopoly Corporation.

**Gaseous compounds separated and discussed.** As described previously,1~4) gaseous compounds separated and discussed were acetaldehyde, an unknown peak, No. 2 (X-2), isoprene, propionaldehyde, an unknown peak, No. 6 (X-6), furan, acetone, acrolein, n-butyraldehyde, 2-methylfuran, methylethylketone, an unknown peak, No. 16 (X-16), methanol, diacetyl and 2,5-dimethylfuran.

**Ingredients in tobacco leaves analysed.** The ingredients in tobacco leaves analysed were crude fiber, total sugars and ether extract.

### RESULTS AND DISCUSSION

**Relationship between crude fiber content and gaseous constituents content**

Correlation between crude fiber content in tobacco leaves and corresponding gaseous constituents content is shown in Fig. 1. The gaseous constituents discussed here were acetaldehyde, propionaldehyde, n-butyraldehyde, acrolein, acetone, diacetyl, methylethylketone, methanol, diacetyl, methanol, furan, X-6 and X-16. As seen in Fig. 1, high correlation was shown giving a correlation coefficient of 0.837 (significant at 1% level).

**Gas chromatogram of the gas phase of the main stream smoke of the cigarette which was made of 96% α-cellulose paper-shreds is shown in Fig. 2.** As seen in Fig. 2, it was noted that large amounts of acetaldehyde, propionaldehyde, acrolein, acetone, methylethylketone, diacetyl, methanol, furan, X-6 and X-16 were generated by the pyrolytic degradation of α-cellulose under the same smoking conditions of tobacco cigarette. The results mentioned above supported that the fibrous substances in tobacco leaves were the main precursors of the gaseous compounds discussed above.

**Relationship between total sugars content and gaseous constituents content**

Correlation between total sugars content in

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FIG. 2. Gas Chromatogram of Gas Phase of Paper Cigarette Smoke.

Columns: Polypropylene-glycol L.B. 550-X: Chromosorb W 60--80 mesh (15:85)
4m × 1/8 inch o.d. Stainless steel (DE 208)
Carbowax 1500: Chromosorb W 60--80 mesh (15:85)
2m × 1/8 inch o.d. Stainless steel (DE 201)
N₂ flow rate: 18.2 ml/min at 30°C

FIG. 3. Correlation between Total Sugar Content of Tobacco Leaves and Gaseous Constituents Content.*

* The percentage of the sum of the peak areas on the gas chromatogram of 2-methylfuran and 2,5-dimethylfuran to the total peak area on the gas chromatogram of gaseous constituents from which K value was computed.
** Significant at 1% level.

FIG. 4. Correlation between Ether Extract Content of Tobacco Leaves and Gaseous Constituents Content.*

* The percentage of the sum of the peak areas on the gas chromatogram of isoprene and X-2 to the total peak area on the gas chromatogram of the gaseous constituents from which K value was computed.
** Significant at 1% level.

that both 2-methylfuran and 2,5-dimethylfuran

increased significantly in the main stream smoke of almost-no-sugars cigarette with added glucose, fructose and sucrose. The results mentioned above supported that sugars in tobacco leaves were the main precursors of 2-methylfuran and 2,5-dimethylfuran. As for furan, a considerable amount of it was generated by the pyrolytic degradation of \( \alpha \)-cellulose as shown in Fig. 2, and so it was suggested that furan would be derived mainly from fibrous substances in tobacco leaves.

**Relationship between ether extract content and gaseous constituents content**

Correlation between ether extract content in tobacco leaves and corresponding gaseous constituents content is shown in Fig. 4. The gaseous constituents discussed here were isoprene and X-2. As seen in Fig. 4, fairly high correlation was shown giving a correlation coefficient of 0.776 (significant at 1% level).

The gas chromatogram of the gas phase of the main stream smoke of the cigarette which was made of 96% \( \alpha \)-cellulose paper-shreds with added ether extract of tobacco leaves is shown in Fig. 5. As seen in Fig. 5, it was noted that a large amount of isoprene and X-2 were generated by the pyrolytic degradation of the ether extract of tobacco leaves. In a previous paper,\(^\text{11}\) it was reported that isoprene and X-2 were generated by the pyrolytic degradation of the non-steam-distilled neutral fraction of tobacco leaves. These facts supported that resinous substances in tobacco leaves were the main precursors of isoprene and X-2. As mentioned hitherto, it was demonstrated that there is the high correlation between the chemical constituents of tobacco leaves and the gaseous constituents of cigarette smoke from which \( K \) value was computed, and our presuppositions have been shown to be correct.

![Gas Chromatogram of Gas Phase of Smoke of Paper Cigarette Added Ether Extract of Flue Cured Tobacco Leaves](image-url)

**Fig. 5.** Gas Chromatogram of Gas Phase of Smoke of Paper Cigarette Added Ether Extract of Flue Cured Tobacco Leaves.

Same conditions as in Fig. 2.