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(54) METHOD FOR PRODUCING TOBACCO MATERIAL CONTAINING ENRICHED ESTER AROMA COMPONENTS AND COMPONENTS CONTRIBUTING TO TOBACCO FLAVOR, AND TOBACCO PRODUCT COMPRISING TOBACCO MATERIAL PRODUCED THEREBY

(57) A method for preparing a tobacco material, characterized by comprising a step of aging cut tobacco, which is to be used in manufacture of a tobacco product immediately thereafter, under a sealed condition in an atmospheric environment, and a step of adding a polyol to the cut tobacco after the aging step, as well as a tobacco product comprising the tobacco material prepared by the above method.

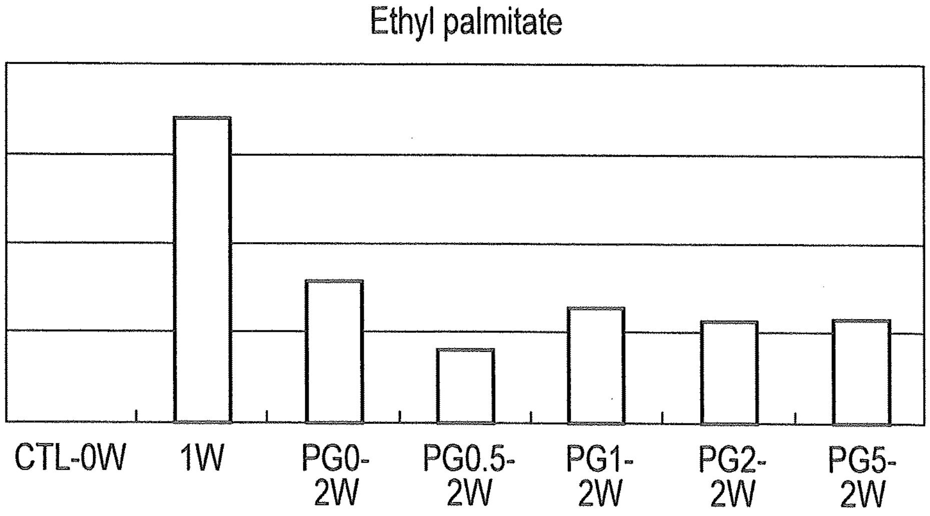

FIG. 8A

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Description

Technical Field

[0001] The present invention relates to a method for preparing a tobacco material containing enriched ester flavor components and enriched components contributing to tobacco flavor and taste, and a tobacco product comprising the tobacco material prepared by the method.

Background Art

[0002] Cultivated and harvested leaf tobacco is used to manufacture a tobacco product such as a cigarette through various processing including a drying process in a farm house, thereafter a long-term aging process for 1 to several years in a leaf processing plant, and thereafter blending and cutting processes in a manufacturing plant. The undesirable aspects of the leaf tobacco are removed through these various processing, and thus the leaf tobacco is used as a flavor-rich tobacco raw material. In order to further improve tobacco flavor and taste, flavor and taste of the leaf tobacco have been improved by adding ethyl alcohol to leaf tobacco before the aging process in the leaf processing plant, and thereafter performing a usual aging process in a barrel (Patent Document 1). Since this method utilizes an aging period in order to improve tobacco flavor and taste, the method necessitates a long period of time. In addition, since the method necessitates further processing after the aging process until the leaf tobacco is used to manufacture the tobacco product, the composition of components contributing to tobacco flavor and taste may be changed by the volatilization of flavor components or the like during the further processing.

Prior Art Document

Patent Document

[0003] Patent Document 1: Jpn. Pat. Appln. KOKAI Publication No. 56-51976

Summary of Invention

Problem to be solved by the Invention

[0004] In view of the above situations, it is an object of the present invention to provide a method for the short-time and simple preparation of a tobacco material in which composition of enriched ester flavor components and enriched components contributing to tobacco flavor and taste are less likely to change with time. It is a further object of the present invention to provide a tobacco product comprising the tobacco material prepared by the method.

Means for solving the Problem

[0005] The present inventors found that when cut tobacco which is ready for use in manufacture of a tobacco product is aged under a sealed condition in an atmospheric environment, the ester flavor components of the cut tobacco are enriched, and that when a polyol was added to the cut tobacco, further production of the ester flavor components is inhibited, which can suppress the composition change of the ester flavor components of the cut tobacco with time. Further, the present inventors found that when the conditions of the aging process are changed, the composition of the components contributing to tobacco flavor and taste including an oxidation product and a sugar decomposition product is controlled besides the ester flavor components, and thereby tobacco materials having various tobacco flavor and taste features can be prepared. Thus, the present invention has been completed.

[0006] Specifically, according to the first aspect of the present invention, there is provided a method for preparing a tobacco material, characterized by comprising a step of aging cut tobacco which is to be used in manufacture of a tobacco product immediately thereafter, under a sealed condition in an atmospheric environment; and a step of adding a polyol to the cut tobacco after the aging step.

[0007] Further, according to the second aspect of the present invention, there is provided a tobacco product comprising a tobacco material prepared by the above method.

Effects of the Invention

[0008] The present invention provides a method for the short-time and simple preparation of a tobacco material in which composition of enriched ester flavor components and enriched components contributing to tobacco flavor and taste are less likely to change with time.

Brief Description of Drawings

[0009]

FIG. 1A shows the production amount of a short-chain fatty acid methyl ester in the case where cut tobacco is aged at various temperatures (Example 2).

FIG. 1B shows the production amount of an aromatic ester in the case where cut tobacco is aged at various temperatures (Example 2).

FIG. 2A shows the production amount of ethyl crotonate in the case where cut tobacco is aged at various temperatures in the presence of ethanol.

FIG. 2B shows the production amount of ethyl palmitate in the case where cut tobacco is aged at various temperatures in the presence of ethanol.

FIG. 2C shows the production amount of menthyl formate in the case where cut tobacco is aged at various temperatures in the presence of menthol.

FIG. 2D shows the production amount of menthyl acetate in the case where cut tobacco is aged at various temperatures in the presence of menthol.

FIG. 2E shows the production amount of menthyl isovalerate in the case where cut tobacco is aged at various temperatures in the presence of menthol.

FIG. 2F shows the production amount of menthone in the case where cut tobacco is aged at various temperatures in the presence of menthol.

FIG. 2G shows the production amount of benzaldehyde in the case where cut tobacco is aged at various temperatures in the presence of benzyl alcohol.

FIG. 2H shows the production amount of furfural in the case where cut tobacco is aged at various temperatures.

FIG. 2I shows the production amount of maltol in the case where cut tobacco is aged at various temperatures.

FIG. 2J shows the production amount of methylpyrazine in the case where cut tobacco is aged at various temperatures.

FIG. 3A shows the production amount of ethyl crotonate in the case where cut tobacco is aged over various aging periods in the presence of ethanol.

FIG. 3B shows the production amount of ethyl palmitate in the case where cut tobacco is aged over various aging periods in the presence of ethanol.

FIG. 3C shows the production amount of menthyl formate in the case where cut tobacco is aged over various aging periods in the presence of menthol.

FIG. 3D shows the production amount of menthyl acetate in the case where cut tobacco is aged over various aging periods in the presence of menthol.

FIG. 3E shows the production amount of menthyl isovalerate in the case where cut tobacco is aged over various aging periods in the presence of menthol.

FIG. 4A shows the production amount of ethyl palmitate in the case where cut tobacco having various pH values is aged in the presence of ethanol.

FIG. 4B shows the production amount of ethyl linoleate in the case where cut tobacco having various pH values is aged in the presence of ethanol.

FIG. 4C shows the production amount of menthyl formate in the case where cut tobacco having various pH values is aged in the presence of menthol.

FIG. 4D shows the production amount of menthyl acetate in the case where cut tobacco having various pH values is aged in the presence of menthol.

FIG. 4E shows the production amount of menthyl isovalerate in the case where cut tobacco having various pH values is aged in the presence of menthol.

FIG. 4F shows the production amount of menthone in the case where cut tobacco having various pH values is aged in the presence of menthol.

FIG. 4G shows the production amount of benzaldehyde in the case where cut tobacco having various pH values is aged in the presence of benzyl alcohol.

FIG. 4H shows the production amount of phytol in the case where cut tobacco having various pH values is aged.

FIG. 4I shows the production amount of furfural in the case where cut tobacco having various pH values is aged.

FIG. 4J shows the production amount of maltol in the case where cut tobacco having various pH values is aged.

FIG. 4K shows the production amount of methylpyrazine in the case where cut tobacco having various pH values is aged.

FIG. 5A shows the production amount of ethyl palmitate in the case where cut tobacco to which water is added in various addition amounts is aged in the presence of ethanol.

FIG. 5B shows the production amount of ethyl linoleate in the case where cut tobacco to which water is added in various addition amounts is aged in the presence of ethanol.

FIG. 5C shows the production amount of menthyl formate in the case where cut tobacco to which water is added in various addition amounts is aged in the presence of menthol.

FIG. 5D shows the production amount of menthyl acetate in the case where cut tobacco to which water is added in various addition amounts is aged in the presence of menthol.

FIG. 5E shows the production amount of menthyl isovalerate in the case where cut tobacco to which water is added in various addition amounts is aged in the presence of menthol.

FIG. 6A shows the production amount of ethyl palmitate in the case where cut tobacco is aged in the presence of ethanol having various concentrations.

FIG. 6B shows the production amount of ethyl linoleate in the case where cut tobacco is aged in the presence of ethanol having various concentrations.

FIG. 6C shows the production amount of menthyl formate in the case where cut tobacco is aged in the presence of menthol having various concentrations.

FIG. 6D shows the production amount of menthyl acetate in the case where cut tobacco is aged in the presence of menthol having various concentrations.

FIG. 7A shows the inhibitory effect of a polyol on the production of ethyl palmitate.

FIG. 7B shows the inhibitory effect of a polyol on the production of ethyl stearate.

FIG. 7C shows the inhibitory effect of a polyol on the production of ethyl linoleate.

FIG. 7D shows the inhibitory effect of a polyol on the production of ethyl linolenate.

FIG. 7E shows the inhibitory effect of a polyol on the production of menthyl formate.

FIG. 7F shows the inhibitory effect of a polyol on the production of menthyl acetate.

FIG. 7G shows the inhibitory effect of a polyol on the production of menthyl isovalerate.

FIG. 7H shows the inhibitory effect of a polyol on the production of ethyl valerate.

FIG. 7I shows the inhibitory effect of a polyol on the production of ethyl n-hexanoate.

FIG. 7J shows the inhibitory effect of a polyol on the production of ethyl crotonate.

FIG. 7K shows the inhibitory effect of a polyol on the production of methyl acetate.

FIG. 7L shows the inhibitory effect of a polyol on the production of benzyl acetate.

FIG. 7M shows the inhibitory effect of a polyol on the production of phenylethyl acetate.

FIG. 8A shows the inhibitory effect of a polyol on the production of ethyl palmitate.

FIG. 8B shows the inhibitory effect of a polyol on the production of ethyl stearate.

FIG. 8C shows the inhibitory effect of a polyol on the production of ethyl linoleate.

FIG. 8D shows the inhibitory effect of a polyol on the production of ethyl linolenate.

FIG. 8E shows the inhibitory effect of a polyol on the production of menthyl formate.

FIG. 8F shows the inhibitory effect of a polyol on the production of menthyl acetate.

FIG. 8G shows the inhibitory effect of a polyol on the production of menthyl isovalerate.

FIG. 8H shows the inhibitory effect of a polyol on the production of ethyl valerate.

FIG. 8I shows the inhibitory effect of a polyol on the production of ethyl n-hexanoate.

FIG. 8J shows the inhibitory effect of a polyol on the production of ethyl crotonate.

FIG. 8K shows the inhibitory effect of a polyol on the production of benzyl acetate.

FIG. 8L shows the inhibitory effect of a polyol on the production of phenylethyl acetate.

FIG. 9 shows the SPME-GCMS spectra in the case where various alcohols are added to cut tobacco of flue-cured tobacco and it is aged at 40°C for 1 week.

FIG. 10 shows the SPME-GCMS spectra in the case where various acids and ethanol are added to cut tobacco of flue-cured tobacco and it is aged at 40°C for 1 week.

FIG. 11A shows the dichloromethane extraction-GC-MS spectra in the case where various essential oils or extract flavors are added to cut tobacco of flue-cured tobacco and it is aged at 40°C for 1 week.

FIG. 11B shows the SPME-GCMS spectra in the case where various essential oils or extract flavors are added to cut tobacco of flue-cured tobacco and it is aged at 40°C for 1 week.

[0010] Mode for Carrying Out the Invention Hereinafter, the present invention will be described in detail. However, the following description is not intended to limit the present invention.

[0011] A method for preparing a tobacco material of the present invention comprises a step of aging cut tobacco, which is to be used in manufacture of a tobacco product immediately thereafter, under a sealed condition in an atmospheric environment, and a step of adding a polyol to the cut tobacco after the aging step. In the aging step of this method, ester flavor components and components contributing to tobacco flavor and taste are produced from the components contained in the cut tobacco. Further production of the ester flavor components and components contributing to tobacco flavor and taste is inhibited by thereafter adding the polyol. Thereby, a tobacco material is prepared in which the composition of the enriched ester flavor components and enriched components contributing to tobacco flavor and taste are less likely to change with time.

[0012] According to a preferable aspect, a method for preparing a tobacco material of the present invention comprises a step of aging cut tobacco, which is to be used in manufacture of a tobacco product immediately thereafter, in the presence of a substrate (for example, alcohol) of an ester flavor component or component contributing to tobacco flavor and taste under a sealed condition in an atmospheric environment; and a step of adding a polyol to the cut tobacco after the aging step. In the aging step of this method, the ester flavor components are particularly produced from both the alcohol and the components contained in the cut tobacco. Further production of the ester flavor components is inhibited by thereafter adding the polyol. Thereby, a tobacco material is prepared in which the composition of the enriched ester flavor components is less likely to change with time.

[0013] In the present specification, the tobacco flavor and taste components are divided into "an ester flavor component" and "a tobacco flavor and taste component other than the ester flavor component". That is, in the present specification, when the tobacco flavor and taste components are expressed as "the ester flavor component and the component contributing to tobacco flavor and taste", the latter "component contributing to tobacco flavor and taste" means "the tobacco flavor and taste component other than the ester flavor component", specifically a tobacco flavor and taste component obtained by an oxidation reaction, a tobacco flavor and taste component obtained by an aminocarbonyl reaction, or the like.

[0014] Cut tobacco derived from any tobacco variety can be used as the cut tobacco used in the present invention. For example, stemmed leaves and midribs derived from flue-cured tobacco, Burley tobacco, and Orient tobacco, and a reconstituted sheet tobacco made from these raw tobacco materials can be used. "The cut tobacco, which is to be used in manufacture of a tobacco product immediately thereafter" means cut tobacco which is ready for use in the manufacture of a tobacco product through various processing including a drying process in a farm house, thereafter a long-term aging process for 1 to several years in a leaf processing plant, and thereafter blending and cutting processes in a manufacturing plant. When the tobacco product is a cigarette, "the cut tobacco, which is to be used in manufacture of a tobacco product immediately thereafter" is cut tobacco at a stage immediately prior to a wrapping process of the cigarette. In the case of stemmed leaves, the cut tobacco used in the present invention can generally have a cutting width of 0.6 to 1 mm in usual lamina shreds and a length of about 25 cm in maximum. In the following description, "the cut tobacco, which is to be used in manufacture of a tobacco product immediately thereafter" is merely referred to as "cut tobacco".

[0015] In the present invention, the cut tobacco is placed under a sealed condition in an atmospheric environment in order to age the cut tobacco. That is, the cut tobacco is put in an airtight vessel, and placed under a situation where the surface of the cut tobacco is in contact with air. The sealed vessel may have an arbitrary volume, and is preferably configured so that an inside temperature can be controlled. Since the surface of the cut tobacco is required to be in contact with air, it is not preferable that the cut tobacco is pressed from the upper side in the vessel and packed in the vessel. The cut tobacco can be put in the vessel, for example, so that the cut tobacco occupies about 50 to 80% of the volume of the vessel.

[0016] As the aging conditions in the method of the present invention, a temperature can be generally selected from the range of room temperature to 70°C (specifically, 20 to 70°C) (see FIGS. 1A to 1B and 2A to 2J). Since the produced amounts of a short-chain fatty acid methyl ester and aromatic ester increase with a rise in an aging temperature, as seen from the results of FIG. 1A and FIG. 1B, the aging temperature can be set to generally 20 to 70°C, preferably 30 to 70°C, and more preferably 40 to 70°C.

[0017] The ester flavor component and the component contributing to tobacco flavor and taste are produced during the aging by the following reactions:

> (i) an enzymatic reaction (for example, a reverse reaction of an ester hydrolytic enzyme) by an endogenous enzyme contained in the cut tobacco, and/or
> (ii) a chemical reaction (for example, production of an ester by a dehydration reaction, production of menthol oxide or the like by an oxidation reaction, and production of furans, reductones, and pyrazines or the like by an aminocarbonyl reaction).

[0018] Therefore, when the ester flavor component and component contributing to tobacco flavor and taste to be enriched are mainly produced by (i) the enzymatic reaction, the temperature can be set to room temperature to 50°C (specifically, 20 to 50°C). Meanwhile, when the ester flavor component and component contributing to tobacco flavor and taste to be enriched are mainly produced by (ii) the chemical reaction, the temperature can be set to 60 to 70°C. Alternatively, when the ester flavor component and component contributing to tobacco flavor and taste to be enriched are produced by both (i) the enzymatic reaction and (ii) the chemical reaction, the temperature can be set to room temperature to 70°C (specifically, 20 to 70°C).

[0019] As the aging conditions in the method of the present invention, an aging period can be generally selected from the range of 1 week to 3 months; for example, 1 to 15 weeks (see FIGS. 3A to 3E).

[0020] As the aging conditions in the method of the present invention, the pH of the cut tobacco can be generally set to 2 to 8 (see FIGS. 4A to 4K). From the results of FIGS. 4A to 4E, the production of various ester flavor components is observed when the cut tobacco having a pH value of 2 to 7 is employed, and the cut tobacco used in the method of the present invention originally has a pH value of 5 to 6. In consideration of these, the cut tobacco can be used without subjecting the cut tobacco to a pH adjusting treatment when various ester flavor components are desired to be enriched. From the results of FIGS. 4A to 4E, the production of various ester flavor components is observed also when the cut tobacco having a pH value of 2 to 3 is employed. Particularly, menthyl formate (FIG. 4C) and menthyl acetate (FIG. 4D) are dominantly produced in the vicinity of a pH of 2. Therefore, when the various ester flavor components are desired to be enriched, the pH of the cut tobacco is optionally set to 2 to 8 and then the aging of the cut tobacco is performed. As a result, cut tobacco having desired composition of ester flavor components can be obtained.

[0021] From the results of FIGS. 4F to 4K, also when the composition of the components contributing to tobacco flavor and taste (such as an oxidation reaction product (for example, menthone and benzaldehyde), an acid-decomposed component (for example, phytol), or an aminocarbonyl reaction product (for example, furans such as furfural, reductones such as maltol, and pyrazines such as methylpyrazine)) is desired to be controlled, the pH of the cut tobacco is optionally set to 2 to 8 and the aging of the cut tobacco is performed, as in the case of the control of the ester flavor components. As a result, cut tobacco having desired composition of components contributing to tobacco flavor and taste can be obtained.

[0022] The present inventors confirmed in the experiment that the pH of the cut tobacco before the aging step and the pH of the cut tobacco after the aging step do not largely fluctuate. That is, in the method of the present invention, the pH of the cut tobacco hardly fluctuates during the aging step.

[0023] The addition of water (humidification) to the cut tobacco fundamentally inhibits the production of the ester flavor components. Therefore, as the aging conditions in the method of the present invention, it is preferable that water is not added to the cut tobacco (see FIGS. 5A to 5C). The cut tobacco used in the method of the present invention originally retains moisture of 10 to 15% by weight. However, when water is added to the cut tobacco of Burley tobacco, the production of the short-chain fatty acid ester flavor components such as menthyl acetate and menthyl isovalerate increases (see FIGS. 5D and 5E). For this reason, if needed (that is, when the addition of water facilitates the production of a desired ester flavor component and component contributing to tobacco flavor and taste), water may be added to the cut tobacco. In this case, water can be generally added until the moisture in the cut tobacco reaches 80% by weight. For example, water can be added in an amount of 0.2 to 1 mL based on 1 g of the cut tobacco. Water is preferably added so that the water spreads throughout the cut tobacco by a general method used in a tobacco manufacturing process such as spraying.

[0024] As the aging conditions in the method of the present invention, the aging step is preferably performed in the presence of at least one additive selected from an alcohol, an acid, an essential oil, and an extract flavor. Specifically, the additive is added to the cut tobacco before the aging step. The alcohol, the essential oil, and the extract flavor may be added as an undiluted solution, and may be added as a diluted solution. The acid may be added as an acid-containing solution. The addition of the additive leads to increase in the amount of the substrate of the ester flavor component and component contributing to tobacco flavor and taste. Thereby, the amounts and kinds of the produced ester flavor component and component contributing to tobacco flavor and taste can be increased.

[0025] As the additive, at least one selected from alcohols such as ethanol, menthol, benzyl alcohol, isobutyl alcohol, n-butanol, isoamyl alcohol, and n-hexanol; acids such as isobutyric acid, 3-methylbutyric acid, n-hexanoic acid, and octanoic acid; and essential oils or extract flavors such as Rum, Peppermint oil, Spearmint oil, Japanese peppermint (Mentha arvensis) oil, Rosemary oil, Licorice extract, St. John's bread extract, and cocoa can be used. The essential oil or the extract flavor is a natural flavor, and contains the substrate (for example, an alcohol and/or an acid) of the ester flavor component and component contributing to tobacco flavor and taste. A different ester flavor component and different component contributing to tobacco flavor and taste can be increased depending on the kinds of the additives. For example, various kinds of ethyl esters, specifically, esters of fatty acids and ethanols such as ethyl acetate, ethyl crotonate, ethyl hexanoate, ethyl palmitate, and ethyl linoleate can be increased by adding ethanol. Similarly, the various ester flavor components can be enriched by the combination of the additive (the alcohol, acid, and natural flavor containing these) and components (an acid and alcohol) originally contained in the cut tobacco.

[0026] Further, the components contained in the additive serve as the substrate of oxidation and aminocarbonyl reactions, and produce the components contributing to tobacco flavor and taste. For example, menthol is converted to menthone by oxidation, and benzyl alcohol is converted to benzaldehyde and benzoic acid, and thus the produced components change the tobacco flavor and taste. Glucose, fructose, and maltose or the like contained in Licorice extract are converted to cycloten, furfuryl alcohol, maltol, and an aminocarbonyl reaction product by the aminocarbonyl reaction, and thus the produced components change the tobacco flavor and taste.

[0027] In the present invention, the combination of a plurality of additives, for example, the combination of ethanol and menthol and the combination of ethanol and butyric acid may be used.

[0028] The additive can be generally added in an amount of 10% by weight or less to the cut tobacco, preferably 0.4 to 10% by weight, more preferably 1 to 10% by weight, and still more preferably 1 to 5% by weight. When a plurality of additives are added, each additive can be added in an amount of 10% by weight or less to the cut tobacco, preferably 1 to 10% by weight, and more preferably 1 to 5% by weight. The additive is preferably added so that a solution of an additive or an additive-containing solution spreads throughout the cut tobacco by a general method used in a tobacco manufacturing process such as spraying. For example, ethanol can be preferably added in an amount of 1 to 10% by weight to the cut tobacco, more preferably 1 to 5% by weight, and still more preferably 2 to 4% by weight (see FIGS. 6A and 6B). Menthol can be preferably added in an amount of 1 to 10% by weight to the cut tobacco, and more preferably 4 to 10% by weight (see FIGS. 6C and 6D).

[0029] The leaf of the cut tobacco originally contains fatty acids such as formic acid, acetic acid, valeric acid, myristic acid, palmitic acid, stearic acid, oleic acid, linolic acid, and linolenic acid; and alcohols such as methanol, benzyl alcohol, and phenylethyl alcohol. Therefore, even if alcohol is not added to the cut tobacco in the method of the present invention, the ester flavor component such as methyl acetate, benzyl acetate, phenylethyl acetate, or methyl palmitate, and the component contributing to tobacco flavor and taste can be enriched from the components contained in the cut tobacco.

[0030] When the additive is added to the cut tobacco in the aging step of the method of the present invention, the additive can be used as the substrate of the ester flavor component and component contributing to tobacco flavor and taste, and thus the ester flavor component and the component contributing to tobacco flavor and taste can further be enriched.

[0031] In a preferable embodiment, a tobacco material in which a great variety of ester flavor components and components contributing to tobacco flavor and taste are enriched can be prepared by providing two or more closed vessels, aging the cut tobacco under different conditions in each of the vessels to enrich a desired ester flavor component and component contributing to tobacco flavor and taste in each of the vessels, and mixing the cut tobacco obtained from each vessel after the aging. For example, two closed vessels are provided. In one closed vessel, the method of the present invention is executed by adding 2% of ethanol to Burley type cut tobacco and aging the cut tobacco at a temperature of 20 to 50°C and in an aging period of 1 to 2 weeks, to mainly enrich the component contributing to tobacco flavor and taste with a focus on the ester flavor component. In the other closed vessel, the method of the present invention is executed by aging flue-cured type cut tobacco at a temperature of 60 to 70°C and in an aging period of 5 to 10 weeks, to mainly enrich the component contributing to tobacco flavor and taste with a focus on an oxidation product and an aminocarbonyl reaction product. Thus, the ester flavor components and the components contributing to tobacco flavor and taste may be enriched in each closed vessel in which composition of these components is different depending on each closed vessel, and the cut tobacco obtained from each closed vessels may be mixed. Thereby, a tobacco material can be prepared, in which a great variety of ester flavor components and components contributing to tobacco flavor and taste are enriched by desired composition.

[0032] In the method of the present invention, a polyol is added to the cut tobacco after the aging step. As the polyol, propylene glycol, glycerin, or a mixture thereof can be used. The polyol can be added in an amount of 0.1 to 5% by weight to the cut tobacco. The polyol is preferably added so that the polyol spreads throughout the cut tobacco by a general method used in a tobacco manufacturing process such as spraying.

[0033] Since the polyol (for example, glycerin) is a substance conventionally added to the cut tobacco in the stage of casing (addition of a casing sauce) for the purpose of securing the moisture retaining property and flavor retaining property of the cut tobacco, the application of the polyol to the cut tobacco is not problematic.

[0034] The polyol has a function of inhibiting the production of the ester flavor component, as shown in the Examples to be described below. For this reason, the further production of the ester flavor component can be inhibited by adding the polyol after the aging step. Thereby, the present invention has an advantage in that the composition of the ester flavor components produced in the aging step is less likely to change with time.

[0035] As proved in the Examples to be described below, in the method of the present invention, the composition of the produced ester flavor components and components contributing to tobacco flavor and taste can be changed by changing the aging conditions. Therefore, a person skilled in the art can appropriately design the aging conditions depending on the desired ester flavor component and component contributing to tobacco flavor and taste.

[0036] The composition of the content components (fatty acid and reducing sugar or the like) contained in the cut tobacco differs depending on the kinds of tobacco raw materials. Therefore, when the cut tobacco is aged according to the method of the present invention, the ratios of produced amounts of the ester flavor component and component contributing to tobacco flavor and taste differs depending on the kinds of tobacco raw materials. In view of the difference in these production patterns, the kinds of the tobacco raw materials are selected, and the aging conditions such as an additive (for example, alcohol), a temperature, time, a pH value, and moisture are changed. Thereby, the composition of the ester flavor components and components contributing to tobacco flavor and taste can be optionally changed.

[0037] In the method of the present invention, as described above, a plurality of aging conditions may be employed by using a plurality of closed vessels. That is, the plurality of closed vessels (for example, 2 to 3 closed vessels) are provided; the method of the present invention is executed by employing different aging conditions for each of the closed vessels to enrich the ester flavor components and components contributing to tobacco flavor and taste in each closed vessel in which composition of these compositions is different depending on each closed vessel; and the cut tobacco obtained from each closed vessel may be mixed. The composition variation of the ester flavor components and components contributing to tobacco flavor and taste can be increased by using the plurality of closed vessels.

[0038] Thus, a tobacco material having various enriched ester flavor components and various enriched components contributing to tobacco flavor and taste can be prepared by employing various aging conditions in either case where one closed vessel is used or a plurality of closed vessels are used. For example, a tobacco material having various enriched ester flavor components and various enriched components contributing to tobacco flavor and taste selected from the following can be prepared according to the method of the present invention.

(1) Esters

[0039]

- short-chain fatty acid ester, for example, ethyl crotonate, ethyl hexanoate
- long-chain fatty acid ester, for example, ethyl palmitate, ethyl linoleate
- ethyl succinate
- short-chain fatty acid menthyl, for example, menthyl formate, menthyl acetate, menthyl isovalerate
- aromatic ester, for example, benzyl acetate, phenylethyl acetate

(2) Oxidation products

[0040]

- menthone, isomenthone
- benzaldehyde, benzoic acid

(3) Aminocarbonyl reaction products

[0041]

- furans, for example, furfural, acetylfuran, furfuryl alcohol, 5-hydroxymethylfurfural
- reductones, for example, cycloten, maltol, furaneol
- pyrazines, for example, pyrazine, methylpyrazine, dimethylpyrazine

(4) Others

[0042]

- duvatrienediol decomposed product, phytol decomposed product, damascenone

[0043] The kinds of tobacco flavor and taste to which the ester flavor component and the component contributing to tobacco flavor and taste contribute are known to a person skilled in the art. For example, ethyl hexanoate, which is an ester, has a floral flower-like flavor, and contributes to the flavor of the cigarette. Ethyl palmitate has an effect of alleviating the stimulus of the cigarette. Menthone, which is an oxidation product, has a fresh mint-like flavor. Cycloten, which is an aminocarbonyl reaction product, has strong maple syrup-like sweet flavor and taste. Dimethylpyrazine contributes to roasted pleasant flavor and taste.

[0044] The tobacco material prepared according the method of the present invention can be used as a material of an arbitrary tobacco product, for example, a smoking product such as a cigarette; and a smokeless tobacco product such as a snuff or a chewing tobacco. The tobacco material prepared according to the method of the present invention may be used as the entire body of tobacco material in the tobacco product, or may be used as a part of the tobacco material in the tobacco product. The tobacco material obtained according to the present invention can be used in an arbitrary ratio when the tobacco material is used as a part of the tobacco material in the tobacco product.

[0045] In the tobacco material obtained according to the method of the present invention, the composition of the ester flavor components and components contributing to tobacco flavor and taste which are produced in the aging step are less likely to change with time. Therefore, when the tobacco material is used for the tobacco product, the enriched tobacco flavor and taste can be felt (see Examples 10 and 11 to be described below).

Examples

Example 1: Analysis of ester flavor component and component contributing to tobacco flavor and taste

[0046] A technique of simply analyzing an ester flavor component and a component contributing to tobacco flavor and taste was created. Dichloromethane extraction-gas chromatograph mass spectrometry (DCM-GCMS) was employed for the purpose of simply acquiring the profile of the entire component of cut tobacco. As a component having low polarity and low mass or a small amount of component required to be condensed was difficult to be detected in the DCM-GCMS, solid phase microextraction-gas chromatograph mass spectrometry (SPME-GCMS) was employed to analyze such components.

1-1. Dichloromethane extraction-gas chromatograph mass spectrometry (DCM-GCMS)

[0047] Cut tobacco in an amount of 0.5 g was accurately weighed in a glass screw vial (volume: 20 mL). Next, 25 μL of an acetonitrile solution of p-bromophenethyl alcohol (1 mg/mL) was added thereto as an internal standard substance. Added thereto was 10 mL of dichloromethane as an extraction solvent, and the glass screw vial was covered with a lid. The cut tobacco was subjected to shaking extraction at 200 rpm for 60 minutes at room temperature. This supernatant liquid was filtered through a membrane filter of 0.45 μm, and then moved to the GC vial, where the supernatant liquid was analyzed by GCMS equipped with an autosampler. In the obtained GC chromatogram, a ratio of peak areas of a component to be analyzed to peak areas of p-bromophenethyl alcohol was obtained. The amount of the component to be analyzed contained in each cut tobacco sample was compared with the obtained value.

<Analysis conditions>

[0048]

Apparatus: gas chromatography apparatus (6890N) manufactured by Agilent Technologies Inc.
Mass detector (5973N) manufactured by Agilent Technologies Inc.
Injected amount: 1 μL (injected in pulsed splitless mode)
Column: HP-INNOWAX (30 m x 0.25 mm (film thickness: 0.25 μm)) manufactured by Agilent Technologies Inc.
Oven: 40°C → 260°C (5°C/min)
Mass detector: TIC mode (mass number: 29 to 550)

<Components to be analyzed>

[0049]

Esters: methyl palmitate, ethyl myristate, ethyl palmitate, ethyl stearate, ethyl oleate, ethyl linoleate, ethyl linolenate, (mono, di, tri) glyceride acetate, and so forth
Oxidation products: menthone, isomenthone, benzaldehyde, benzoic acid, long-chain fatty acids such as palmitic acid, and so forth
Aminocarbonyl reaction products: acetol, acetoin, acetylpyrrole, 5-hydroxymethyl-2-furfural (5-HMF), and so forth

Others: phytol, duvatrienediol, glycerin, propylene glycol, or the like

1-2. Solid phase microextraction-gas chromatograph mass spectrometry (SPME-GCMS)

[0050] Cut tobacco in an amount of 0.5 g was accurately weighed in an SPME vial (volume: 20 mL), and a vial cap was attached to the SPME vial to seal the SPME vial. It was subjected to analysis by GCMS equipped with an SPME auto injector.

[0051] In addition, 25 μL of an acetonitrile solution of p-bromophenethyl alcohol (1 mg/mL) was added to cut tobacco in another SPME vial. It was subjected to the same analysis. A ratio of peak areas of a component to be analyzed to peak areas of p-bromophenethyl alcohol was obtained. The amount of the component to be analyzed contained in each cut tobacco sample was compared with the obtained value. For this reason, the amounts of the components to be analyzed measured in the present invention (vertical axes of FIGS. 2A to 2J, FIGS. 3A to 3E, FIGS. 4A to 4K, FIGS. 5A to 5E, FIGS. 6A to 6D, FIGS. 7A to 7M, and FIGS. 8A to 8L) represent relative amounts.

<Analysis conditions>

[0052]

Apparatus: gas chromatography apparatus (6890A) manufactured by Agilent Technologies Inc.
SPME auto injector (Combi PAL) manufactured by CTC Analytics
Mass detector (5973N) manufactured by Agilent Technologies Inc.
Injected amount: 1 μL (injected in pulsed splitless mode)
Column: INNOWAX
Oven: 40°C → 250°C (3°C/min)
Mass detector: TIC mode (mass number: 29 to 550)

<Components to be analyzed>

[0053]

Esters: (methyl acetate (C2-Me)), methyl isovalerate (isoC4-Me), methyl valerate (n-C4-Et), methyl hexanoate (n-C6-Me), methyl phenylacetate, ethyl isovalerate, ethyl valerate, ethyl crotonate, ethyl hexanoate, ethyl heptanoate, ethyl octanoate, ethyl nonanoate, ethyl decanoate, menthyl formate, menthyl acetate, menthyl isovalerate, benzyl formate, phenethyl formate, benzyl acetate, phenethyl acetate, and so forth

Oxidation products: short-chain fatty acids such as acetic acid, benzaldehyde, benzoic acid, phenylacetic acid, and so forth

Aminocarbonyl reaction products: furfural, acetylfuran, 5-methylfurfural, 2-furanmethanol, cycloten, maltol, furaneol, pyrazine, methylpyrazine, 2,5-dimethylpyrazine, 2,6-dimethylpyrazine, and so forth

Example 2: Effect of aging temperature in absence of alcohol

2-1. Aging

[0054] One g of cut tobacco at a stage immediately prior to a wrapping process of a cigarette was placed at various temperatures in a temperature-controllable closed vessel (glass screw vial: 5 mL) for 1 week. As the cut tobacco, Brazilian Burley tobacco and Brazilian flue-cured tobacco were used. As the aging temperature, 5°C, 22°C, 30°C, 40°C, 50°C, 60°C, and 70°C were employed. Cut tobacco subjected to no aging treatment was used as control.

2-2. Analysis

[0055] An ester flavor component and a component contributing to tobacco flavor and taste were analyzed according to the method described in Example 1.

2-3. Results

[0056] The analysis results of a short-chain fatty acid methyl ester and aromatic ester are shown in FIGS. 1A and 1B, respectively. In FIGS. 1A and 1B, the analysis result of the Burley tobacco is shown on the left side, and the analysis result of the flue-cured tobacco is shown on the right side.

[0057] In FIG. 1A, C2-Me represents methyl acetate; isoC4-Me represents methyl isovalerate; n-C4-Me represents methyl valerate; and n-C6-Me represents methyl hexanoate.

[0058] From the results of FIG. 1A, the produced amount of the short-chain fatty acid methyl ester showed a tendency to increase with increase in temperature. Under the conditions of 50°C and 60°C, the produced amount of the short-chain fatty acid methyl ester was particularly high. In both the Burley tobacco and the flue-cured tobacco, the produced amount of C2-Me (methyl acetate) was the highest. Although methyl isovalerate (isoC4-Me) was detected in the Burley tobacco, methyl isovalerate was not detected in the flue-cured tobacco.

[0059] From the results of FIG. 1B, the produced amount of methyl phenylacetate particularly increased with increase in temperature. In the Burley tobacco, the produced amounts of benzyl acetate and phenethyl acetate in addition to methyl phenylacetate increased with increase in temperature. In the flue-cured tobacco, the produced amounts of benzyl acetate, phenethyl formate, and phenethyl acetate in addition to methyl phenylacetate increased with increase in temperature.

Example 3: Effect of aging temperature under presence of alcohol

3-1. Aging step

[0060] One g of cut tobacco at a stage immediately prior to a wrapping process of a cigarette was placed at various temperatures in a temperature-controllable closed vessel (glass screw vial: 5 mL) for 1 week. As the cut tobacco, Brazilian Burley tobacco and Brazilian flue-cured tobacco were used. As the aging temperature, -40°C, 5°C, 22°C, 30°C, 35°C, 40°C, 45°C, 50°C, 60°C, or 70°C were employed.

[0061] Before aging, a menthol/ethanol solution (corresponding to ethanol of 2% by weight and menthol of 1% by weight based on cut tobacco) was added to the cut tobacco by syringe injection.

3-2. Analysis

[0062] An ester flavor component and a component contributing to tobacco flavor and taste were analyzed according to the method described in Example 1.

3-3. Results

[0063] The analysis results are shown in FIGS. 2A to 2J. In FIGS. 2A to 2J, black circles represent Burley tobacco, and white circles represent flue-cured tobacco.

[0064] From the results of FIGS. 2A to 2B, the high produced amount of ethyl crotonate was shown at a temperature of 50°C or more, preferably 60°C or more, and more preferably 60 to 70°C; and the high produced amount of ethyl palmitate was shown at a temperature of 20°C or more, and preferably 20 to 50°C.

[0065] From the results of FIGS. 2C to 2E, the high produced amount of menthyl formate was shown at a temperature of 60°C or more, and preferably 60 to 70°C; the high produced amount of menthyl acetate was shown at a temperature of 20°C or more, and preferably 20 to 50°C; and the high produced amount of menthyl isovalerate was showed at a temperature of 20 to 60°C, and preferably 30 to 50°C.

[0066] From the results of FIGS. 2F to 2G, the high produced amount of menthone was shown at a temperature of 60°C or more, preferably 60 to 70°C; and the high produced amount of benzaldehyde was shown at a temperature of 20°C or more, preferably 60°C or more, and more preferably 60 to 70°C.

[0067] From the results of FIGS. 2H to 2J, the high produced amounts of furfural, maltol, and methylpyrazine were shown at a temperature of 60°C or more, and preferably 60 to 70°C. In the flue-cured tobacco, the production of furans and reductones was superior. Meanwhile, in the Burley tobacco, the production of pyrazines was superior.

Example 4: Effect of aging period under presence of alcohol

4-1. Aging step

[0068] One g of cut tobacco at a stage immediately prior to a wrapping process of a cigarette was placed at a temperature of 40°C in a temperature-controllable closed vessel (glass screw vial: 5 mL) for various aging periods. As the cut tobacco, Brazilian Burley tobacco and Brazilian flue-cured tobacco were used. As the aging period, 0 week, 1 week, 2 weeks, 4 weeks, 8 weeks, or 12 weeks was employed.

[0069] Before aging, ethanol of 2% by weight based on cut tobacco was weighed by a syringe and added to cut tobacco, and menthol of 1% by weight based on cut tobacco was weighed in powder form and added to cut tobacco. After the addition, the vessel was sealed, and the cut tobacco in the vessel was sufficiently stirred to uniformize the cut tobacco.

4-2. Analysis

[0070] An ester flavor component and a component contributing to tobacco flavor and taste were analyzed according to the method described in Example 1.

4-3. Results

[0071] The analysis results are shown in FIGS. 3A to 3E. In FIGS. 3A to 3E, black circles represent Burley tobacco, and white circles represent flue-cured tobacco.

[0072] From the results of FIGS. 3A to 3B, the high produced amount of ethyl crotonate was shown in the case of an aging period of 5 weeks or more, and preferably 5 to 12 weeks; and the high produced amount of ethyl palmitate was shown in the case of an aging period of 1 to 5 weeks, and preferably 1 to 2 weeks.

[0073] From the results of FIGS. 3C to 3E, the high produced amounts of menthyl formate, menthyl acetate, and menthyl isovalerate were shown in the case of an aging period of 1 week or more, and preferably 1 to 5 weeks.

Example 5: Effect of pH of cut tobacco under presence of alcohol

5-1. Aging step

[0074] In the present Example, cut tobacco at a stage immediately prior to a wrapping process was used as a material to prepare cut tobacco having various pH values. Specifically, the pH was adjusted by spraying a predetermined amount of 1N sulfuric acid aqueous solution or a 1N sodium hydroxide aqueous solution to the cut tobacco and uniformly mixing the cut tobacco.

[0075] One g of pH-adjusted cut tobacco was placed and aged at a temperature of 40°C in a temperature-controllable closed vessel (glass screw vial: 5 mL) for 1 week. As the cut tobacco, Brazilian Burley tobacco and Brazilian flue-cured tobacco were used. The pH of the cut tobacco was set to 2 to 10.

[0076] Before aging, ethanol of 2% by weight based on cut tobacco was weighed by a syringe and added to pH-adjusted cut tobacco, and menthol of 1% by weight based on cut tobacco was weighed in powder form and added to pH-adjusted cut tobacco. After the addition, the vessel was sealed, and the cut tobacco in the vessel was sufficiently stirred to uniformize the cut tobacco.

5-2. Analysis

[0077] An ester flavor component and a component contributing to tobacco flavor and taste were analyzed according to the method described in Example 1.

[0078] The pH of the cut tobacco was obtained by adding 10 mL of distilled water to 0.5 g of the cut tobacco, performing shaking extraction of 200 rpm for 30 minutes, and measuring pH of the obtained supernatant liquid by a pH meter (Mettler Toledo).

5-3. Results

[0079] The analysis results are shown in FIGS. 4A to 4K. In FIGS. 4A to 4K, black circles represent Burley tobacco, and white circles represent flue-cured tobacco.

[0080] From the results of FIGS. 4A to 4B, the high produced amount of ethyl esters (ethyl palmitate, ethyl linoleate) was shown when cut tobacco having a pH of 3 or less (preferably, a pH of 2 to 3) or a pH of 4 to 7 was used.

[0081] From the results of FIGS. 4C to 4E, the high produced amount of menthyl formate was shown when cut tobacco having a pH of 3 or less (preferably, a pH of 2 to 3) was used; the high produced amounts of menthyl acetate and menthyl isovalerate were shown when cut tobacco having a pH of 3 or less (preferably, a pH of 2 to 3) or a pH of 4 to 7 was used.

[0082] From the results of FIGS. 4F to 4G, the high produced amounts of menthone and benzaldehyde were shown when cut tobacco having a pH of 4 or less (preferably, a pH of 2 to 3) was used.

[0083] From the results of FIG. 4H, phytol showed a tendency to be decomposed when cut tobacco having a pH of 4 or less (preferably, a pH of 2 to 3) was used.

[0084] From the results of FIGS. 4I to 4K, the high produced amount of furfural was shown when cut tobacco having a pH of 3 or less (preferably, a pH of 2 to 3) was used; the high produced amount of maltol was shown when cut tobacco having a pH of 5 or less (preferably, a pH of 2 to 5) was used; the high produced amount of methylpyrazine was shown when cut tobacco having a pH of 7 or more (preferably, a pH of 7 to 9) was used. In the flue-cured tobacco, the production of furans and reductones was superior. Meanwhile, in the Burley tobacco, the production of pyrazines was superior.

Example 6: Effect of humidity conditioning of cut tobacco under presence of alcohol

6-1. Aging step

[0085] In the present Example, various predetermined amounts of water were added to cut tobacco at a stage immediately prior to a wrapping process of a cigarette by spraying and uniformly mixing.

[0086] One g of water-added cut tobacco was placed and aged at a temperature of 40°C in a temperature-controllable closed vessel (glass screw vial: 5 mL) for 1 week. As the cut tobacco, Brazilian Burley tobacco and Brazilian flue-cured tobacco were used. As the amount of water to be added, 0 μL, 50 μL, 100 μL, and 200 μL were employed based on 0.5 g of the cut tobacco.

[0087] Before aging, ethanol of 2% by weight based on cut tobacco was weighed by a syringe and added to water-added cut tobacco, and menthol of 1% by weight based on cut tobacco was weighed in powder form and added to water-added cut tobacco. After the addition, the vessel was sealed, and the cut tobacco in the vessel was sufficiently stirred to uniformize the cut tobacco.

6-2. Analysis

[0088] An ester flavor component was analyzed according to the method described in Example 1.

6-3. Results

[0089] The analysis results are shown in FIGS. 5A to 5E. In FIGS. 5A to 5E, black circles represent Burley tobacco, and white circles represent flue-cured tobacco.

[0090] From the results of FIGS. 5A to 5E, when water was added to the flue-cured tobacco, the production of ethyl palmitate and ethyl linoleate was inhibited, and the changes in the production of the menthyl formate, menthyl acetate, and menthyl isovalerate were not observed. Meanwhile, when water was added to the Burley tobacco, the production of menthyl acetate and menthyl isovalerate increased.

Example 7: Effect of alcohol concentration

7-1. Aging step

[0091] One g of cut tobacco at a stage immediately prior to a wrapping process of a cigarette was placed and aged at a temperature of 40°C in a temperature-controllable closed vessel (glass screw vial: 5 mL) for 1 week. As the cut tobacco, Brazilian Burley tobacco and Brazilian flue-cured tobacco were used.

[0092] Before aging, ethanol having various concentrations was weighed in a predetermined amount by a syringe and added to cut tobacco, or alternatively menthol having various concentrations was weighed in a predetermined amount in powder form and added to cut tobacco. After the addition, the vessel was sealed, and the cut tobacco in the vessel was sufficiently stirred to uniformize the cut tobacco. As the ethanol concentration, 1% by weight, 2% by weight, 4% by weight, 10% by weight, and 40% by weight were employed based on the weight of the cut tobacco. As the menthol concentration, 0.4% by weight, 1% by weight, 4% by weight, and 10% by weight were employed based on the weight of the cut tobacco.

7-2. Analysis

[0093] An ester flavor component was analyzed according to the method described in Example 1.

7-3. Results

[0094] The analysis results are shown in FIGS. 6A to 6D. In FIGS. 6A to 6D, black circles represent Burley tobacco, and white circles represent flue-cured tobacco.

[0095] From the results of FIGS. 6A to 6D, when ethanol was added as a substrate in an amount of 1 to 5% by weight, and more preferably 2 to 4% by weight to the cut tobacco, the produced amounts of ethyl palmitate and ethyl linoleate significantly increased. When menthol was added as a substrate in an amount of 4 to 10% by weight to the cut tobacco, the produced amounts of menthyl formate and menthyl acetate significantly increased.

Example 8: Effect of addition of polyol

Example 8-A: Effect of addition of polyol prior to aging

8-A1. Polyol addition and subsequent aging

[0096] An aqueous solution containing glycerin and propylene glycol at a ratio of 4:1 was added to cut tobacco at a stage immediately prior to a wrapping process of a cigarette by spraying and uniformly mixing, its humidity was conditioned in a conditioning chamber (22°C, 4 days) to prepare cut tobacco. The addition amounts of glycerin and propylene glycol to the cut tobacco were set to 2% by weight and 0.5% by weight, respectively. Further, a menthol/ethanol solution (corresponding to ethanol of 2% by weight and menthol of 1% by weight based on cut tobacco) was added to the cut tobacco by syringe injection.

[0097] One g of the cut tobacco treated as described above was placed and aged at a temperature of 40°C in a temperature-controllable closed vessel (glass screw vial: 5 mL) for 1 week. As the cut tobacco, Brazilian Burley tobacco and Brazilian flue-cured tobacco were used.

8-A2. Analysis

[0098] An ester flavor component was analyzed according to the method described in Example 1.

8-A3. Results

[0099] The analysis results are shown in FIGS. 7A to 7M. FIGS. 7A to 7M show, in order from the left, the following cases:

a case where a polyol (glycerin and propylene glycol) was not added to the flue-cured tobacco (-);
a case where a polyol (glycerin and propylene glycol) was added to the flue-cured tobacco (+);
a case where a polyol (glycerin and propylene glycol) was not added to the Burley tobacco (-); and
a case where a polyol (glycerin and propylene glycol) was added to the Burley tobacco (+).

[0100] In both the flue-cured tobacco and Burley tobacco, it was shown that the production of various esters was more or less inhibited by adding the polyol.

Example 8-B: Effect of addition of polyol after aging for 1 week

8-B1. Aging and subsequent additional aging under polyol addition conditions

[0101] One g of cut tobacco at a stage immediately prior to a wrapping process of a cigarette was weighed in a screw vial and an SPME vial. Ethanol, menthol, or benzyl alcohol of 1% by weight was added thereto. Ethanol and benzyl alcohol were weighed in a predetermined amount by a syringe, and menthol was weighed in a predetermined amount in powder form. After the addition, the vial was sealed, and the cut tobacco in the vial was sufficiently stirred to uniformize the cut tobacco.

[0102] One g of the cut tobacco treated as described above was placed and aged at a temperature of 40°C for 1 week. After that, propylene glycol was added to a screw vial and SPME vial containing the aged cut tobacco by syringe injection. Further, the cut tobacco was placed and aged at a temperature of 40°C for 1 week. The addition concentration of propylene glycol was set to 0, 0.5, 1, 2, and 5% by weight. As the cut tobacco, Brazilian flue-cured tobacco was used.

8-B2. Analysis

[0103] An ester flavor component was analyzed according to the method described in Example 1.

8-B3. Results

[0104] Graphs of FIGS. 8A to 8L show, in order from the left, the analysis results of the following cut tobacco:

cut tobacco which was not aged (0-week control (CTL-OW));
1-week aged cut tobacco which was aged only for the first 1 week (1W);

2-week aged cut tobacco which was further aged for 1 week without adding propylene glycol after aging for 1 week (PG0-2W);
2-week aged cut tobacco which was further aged for 1 week with 0.5% by weight of propylene glycol added after aging for 1 week (PG0.5-2W);
2-week aged cut tobacco which was further aged for 1 week with 1% by weight of propylene glycol added after aging for 1 week (PG1-2W);
2-week aged cut tobacco which was further aged for 1 week with 2% by weight of propylene glycol added after aging for 1 week (PG2-2W); and
2-week aged cut tobacco which was further aged for 1 week with 5% by weight of propylene glycol added after aging for 1 week (PG5-2W).

[0105] FIGS. 8A to 8D show the production of ethyl esters of various long-chain fatty acids in the aging treatment of the cut tobacco to which 1% by weight of ethanol was added as a substrate. In FIGS. 8A to 8D, the produced amounts of the various ethyl esters decreased in the 2-week aged cut tobacco (PG0-2W) as compared with the 1-week aged cut tobacco (1W). This corresponds to the tendency of the change with time shown in FIG. 3B. In the 2-week aged cut tobacco (PG0.5-2W, PG1-2W, PG2-2W, and PG5-2W) to which propylene glycol was added, the produced amount of the esters was less than that of the 2-week aged cut tobacco (PG0-2W) to which propylene glycol was not add, irrespective of the concentration of propylene glycol. The ester production reaction showed a tendency to be inhibited by adding propylene glycol.

[0106] FIGS. 8E to 8G show the production of menthyl esters of various short-chain fatty acids in the aging treatment of the cut tobacco to which 1% by weight of menthol was added as a substrate. In FIGS. 8E to 8G, the produced amounts of the various menthyl esters increased in the 2-week aged cut tobacco (PG0-2W) as compared with the 1-week aged cut tobacco (1W). This corresponds to the tendency of the change with time shown in FIGS. 3C to 3E. In the 2-week aged cut tobacco (PG0.5-2W, PG1-2W, PG2-2W, and PG5-2W) to which propylene glycol was added, the produced amount of the esters was less than that of the 2-week aged cut tobacco (PG0-2W) to which propylene glycol was not added, irrespective of the concentration of propylene glycol. The ester production reaction showed a tendency to be inhibited by adding propylene glycol.

[0107] FIGS. 8H to 8J show the production of ethyl esters of various short-chain fatty acids in the aging treatment of the cut tobacco to which 1% by weight of ethanol was added as a substrate. In FIGS. 8H to 8J, the produced amounts of the various ethyl esters increased in the 2-week aged cut tobacco (PG0-2W) as compared with the 1-week aged cut tobacco (1W). This corresponds to the tendency of the change with time shown in FIG. 3A. In the 2-week aged cut tobacco (PG0.5-2W, PG1-2W, PG2-2W, and PG5-2W) to which propylene glycol was added, the produced amount of the esters was less than that of the 2-week aged cut tobacco (PG0-2W) to which propylene glycol was not added, irrespective of the concentration of propylene glycol. The ester production reaction showed a tendency to be inhibited by adding propylene glycol.

[0108] FIG. 8K shows the production of benzyl acetate in the aging treatment of the cut tobacco to which 1% by weight of benzyl alcohol was added as a substrate. In FIG. 8K, the produced amount of benzyl acetate in the 1-week aged cut tobacco (1W) was almost equal to that of the 2-week aged cut tobacco (PG0-2W). Further, there was not much different between the above produced amount and those of the 2-week aged cut tobacco (PG0.5-2W, PG1-2W, PG2-2W, and PG5-2W) to which propylene glycol was added. However, as the addition concentration of propylene glycol increased, the ester production reaction showed a tendency to be slightly inhibited.

[0109] FIG. 8L shows the production of phenethyl acetate in the aging treatment of the cut tobacco to which an alcohol was not added. The produced amount of phenethyl acetate in the 1-week aged cut tobacco (1W) was almost equal to that of the 2-week aged cut tobacco (PG0-2W). Further, there was not much different between the above produced amount and those of the 2-week aged cut tobacco (PG0.5-2W, PG1-2W, PG2-2W, and PG5-2W) to which propylene glycol was added. However, as the addition concentration of propylene glycol increased, the ester production reaction showed a tendency to be slightly inhibited.

[0110] From the above results, the production of various esters showed a tendency to be much or less inhibited by adding the polyol.

Example 9: Production of flavor component in the case where various alcohols were added and then aging is performed

9-1. Aging step

[0111] One g of cut tobacco to which various alcohols were added before aging was placed and aged at 40°C in a temperature-controllable closed vessel (glass screw vial: 5 mL) for 1 week. As the cut tobacco, Brazilian flue-cured tobacco was used. Cut tobacco to which alcohol was not added was used as the control.

[0112] The kinds and addition concentrations of the added alcohols are shown below.

2% by weight of isobutyl alcohol
2% by weight of n-butanol
2% by weight of isoamyl alcohol
2% by weight of n-hexanol
2% by weight of propylene glycol
2% by weight of benzyl alcohol

9-2. Analysis

[0113] An ester flavor component was analyzed according to the method described in Example 1.

9-3. Results

[0114] Dichloromethane extraction-GCMS and SPME-GCMS chromatograms of the aged cut tobacco samples to which various alcohols were added were shown in FIG. 9. The production of the ester corresponding to the kinds of the added alcohols was confirmed.

[0115] In FIG. 9, "C1" represents formate ester; "C2" represents acetate ester; "C5" represents valerate ester; and "C6" represents hexanoate ester.

Example 10: Production of flavor component in the case where various acids and ethanol are added and then aging is performed

10-1. Aging step

[0116] Various acids and ethanol were added before aging, and the cut tobacco was aged as in Example 9. The kinds and addition concentrations of the added acids, and the addition concentrations of ethanol are shown below. The following addition concentrations are represented by % by weight based on the cut tobacco.

1% by weight of isovaleric acid + 1% by weight of ethanol
1% by weight of 3-methylvaleric acid + 1% by weight of ethanol
1% by weight of n-hexanoic acid + 1% by weight of ethanol
1% by weight of n-octanoic acid + 1% by weight of ethanol

10-2. Analysis

[0117] An ester flavor component was analyzed according to the method described in Example 1.

10-3. Results

[0118] The SPME-GCMS chromatograms of the aged cut tobacco samples to which various acids and ethanol were added were shown in FIG. 10. The production of an ethyl ester corresponding to the added acid was confirmed.

[0119] In FIG. 10, when isovaleric acid and ethanol were added, the production of ethyl isovalerate was confirmed; when 3-methylvaleric acid and ethanol were added, the production of ethyl 3-methylvalerate was confirmed; when n-hexanoic acid and ethanol were added, the production of ethyl n-hexanoate was confirmed; and when n-octanoic acid and ethanol were added, the production of ethyl n-octanoate was confirmed.

Example 11: Production of flavor component in the case where essential oil or extract flavor is added and then aging is performed

11-1. Aging step

[0120] Various essential oils or extract flavors were added before aging, and cut tobacco was aged as in Example 9. The kinds and addition concentrations of the added essential oils or extract flavors are shown below. The following addition concentrations are represented by % by weight based on the cut tobacco.

1% by weight of Cinnamon leaf oil
1% by weight of Rosemary oil
1% by weight of Peppermint oil
1% by weight of Spearmint oil
1% by weight of St. John's bread extract
1% by weight of Licorice extract

11-2. Analysis

[0121] An ester flavor component was analyzed according to the method described in Example 1.

11-3. Results

[0122] Dichloromethane extraction-GCMS and SPME-GCMS chromatograms of the aged cut tobacco samples to which various essential oils or extract flavors were added were shown in FIGS. 11A and 11B. The production of various esters was confirmed based on the composition of alcohols and acids contained in the added essential oil or extract flavor. This result showed that cut tobacco having the composition of various esters was obtained by adding various essential oils and extract flavors.

[0123] In FIG. 11A, "C14" represents ethyl myristate; "C16" represents ethyl palmitate; and "C18" represents ethyl stearate. In FIG. 11B, "C2" represents ethyl acetate; "C5" represents ethyl valerate; and "C6" represents ethyl hexanoate.

Example 12: Organoleptic evaluation of snuff

12-1. Preparation of organoleptic evaluation sample and evaluation method

[0124] Before aging, a menthol/ethanol solution (corresponding to ethanol of 2% by weight and menthol of 1% by weight based on cut tobacco) was added to cut tobacco by syringe injection. One g of the obtained cut tobacco was placed and aged at 40°C in a temperature-controllable closed vessel (glass screw vial: 5 mL) for 1 week. As the cut tobacco, Malawian flue-cured tobacco, Japanese Burley tobacco, and Greek Oriental tobacco were used.

[0125] A propylene tube (φ 8 mm i.d.) was filled with

0.2 g of the aged cut tobacco. Both ends were covered with acetate filters having a width of 4 mm. A rubber heater was wound around the outside of the propylene tube, and it was heated to 40°C to be used as a smoking evaluation sample.

[0126] An expert panel of five judges test-smoked the smoking evaluation sample for the organoleptic evaluation, and evaluated the differences in flavors, tastes, and somatic sensations according to the difference among tobacco raw materials.

12-2. Results

[0127] The results of the organoleptic evaluation are described below. The enriched flavor and taste experienced by the panels in the smoking evaluation sample as compared with control (a smoking evaluation sample using cut tobacco subjected to no aging treatment) are described below.

(1) Malawian flue-cured tobacco

[0128] Strong fruit and herb-like flavors, strong sweet flavor, and cigarette-like smoking sensation.

[0129] Clear and cold menthol feeling having less stimulus.

(2) Japanese Burley tobacco

[0130] Strong dried fruit and wine-like flavors, and strong sweet flavor.

[0131] Raw flavor unique to Japanese Burley leaf remains, but it is weak.

(3) Greek Oriental tobacco

[0132] Strong floral sweet, strong smoking sensation, and strong cold sense of menthol.

[0133] Stimulus is weak. Green herbal flavor of Oriental tobacco remains, but it is weak.

Example 13: Organoleptic evaluation of cigarette

13-1. Preparation of organoleptic evaluation sample and evaluation method

[0134] Cut tobacco was aged as in Example 10 to prepare cut tobacco. Cut tobacco of Seven Stars Lights Menthol which was not flavored after cutting was extracted from a rolled cigarette paper, and it was filled with 800 mg of the above aged cut tobacco, to be used as a smoking evaluation cigarette.

[0135] An expert panel of five judges test-smoked the cigarette sample, and evaluated the difference in exterior aroma and smoking taste according to the difference among tobacco raw materials.

13-2. Results

[0136] The results of organoleptic evaluation are described below. The enriched flavor and taste experienced by the panels in the smoking evaluation sample as compared with control (a smoking evaluation sample using cut tobacco subjected to no aging treatment) are described below.

1) Flue-cured tobacco

[0137]

Exterior aroma: strong wine and Rum-like aroma.
Smoking taste: extremely fine and soft smoke quality, fruity sweet flavor.
Strong and lasting cold sense of menthol, and salivation effect.
Weak stimulus.

2) Burley tobacco

[0138]

Exterior aroma: aged Raisin-like, brandy tone fruity aroma.
Smoking taste: dried fruit-like sweet flavor, Burley-like flavor, weak menthol sense.

(3) Oriental tobacco

[0139]

Exterior aroma: strong Rum and raisin-like aroma, and Oriental tobacco-like green herbal aroma.
Weak smell of acid involving fermentation feeling unique to Oriental tobacco.
Smoking taste: Rum and raisin-like flavor, weak smell of fermentation, salivation effect, and slightly strong stimulus.

[0140] As described above, the present invention has been described according to various embodiments and Examples. However, the present invention is not limited to these embodiments and Examples. The present inventors have first found the technique of controlling the ester flavor component and the component contributing to tobacco flavor and taste by the aging treatment of the cut tobacco, and the technique creates cut tobacco having the intended tobacco flavor and taste feature by changing the content components contained in the cut tobacco, the additive such as alcohol or acid, and the aging treatment conditions. It goes without saying that the tobacco variety or the natural tobacco material containing the tobacco variety used in the Examples, and the additive such as ethanol and menthol used in the Examples can be applied to the present invention. General natural tobacco materials and general flavor additives including alcohol and acid may be also widely applied to the present invention.

Claims

1. A method for preparing a tobacco material, **characterized by** comprising:

    a step of aging cut tobacco, which is to be used in manufacture of a tobacco product immediately thereafter, under a sealed condition in an atmospheric environment; and
    a step of adding a polyol to the cut tobacco after the aging step.

2. The method according to claim 1, **characterized in that** the aging step is performed in the presence of at least one additive selected from an alcohol, an acid, an essential oil, and an extract flavor.

3. The method according to claim 1 or 2, **characterized in that** the cut tobacco is aged at a temperature ranging from room temperature to 70°C in the aging step.

4. The method according to claim 1 or 2, **characterized in that** the cut tobacco is aged over the period ranging from 1 week to 3 months in the aging step.

5. The method according to claim 1 or 2, **characterized in that** the cut tobacco has a pH of 2 to 8 in the aging step.

6. The method according to claim 2, **characterized in that** the additive is an alcohol, and the alcohol is at least one selected from ethanol, menthol, benzyl alcohol, isobutyl alcohol, n-butanol, isoamyl alcohol, and n-hexanol.

7. The method according to claim 2, **characterized in that** the additive is an acid, and the acid is at least one selected from isobutyric acid, 3-methylbutyric acid, n-hexanoic acid, and octanoic acid.

8. The method according to claim 2, **characterized in that** the additive is an essential oil or an extract flavor, and the essential oil or the extract flavor is at least one selected from Rum, Peppermint oil, Spearmint oil, Japanese peppermint oil, Rosemary oil, Licorice extract, St. John's bread extract, and cocoa.

9. The method according to claim 2, **characterized in that** the additive is an alcohol, and the alcohol is added in an amount of 0.4 to 10% by weight based on the cut tobacco.

10. A tobacco product comprising a tobacco material prepared by the method according to any one of claims 1 to 9.

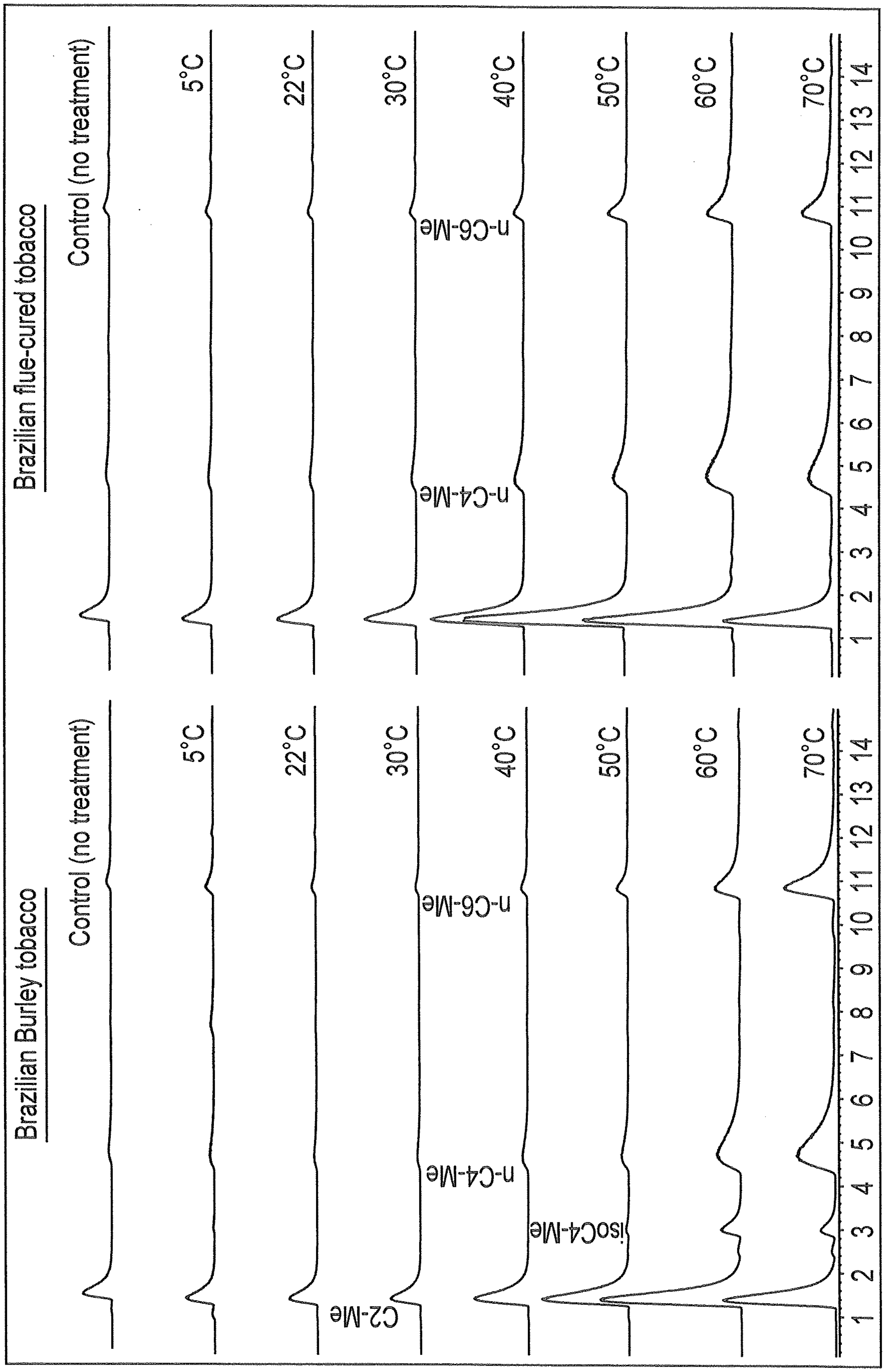
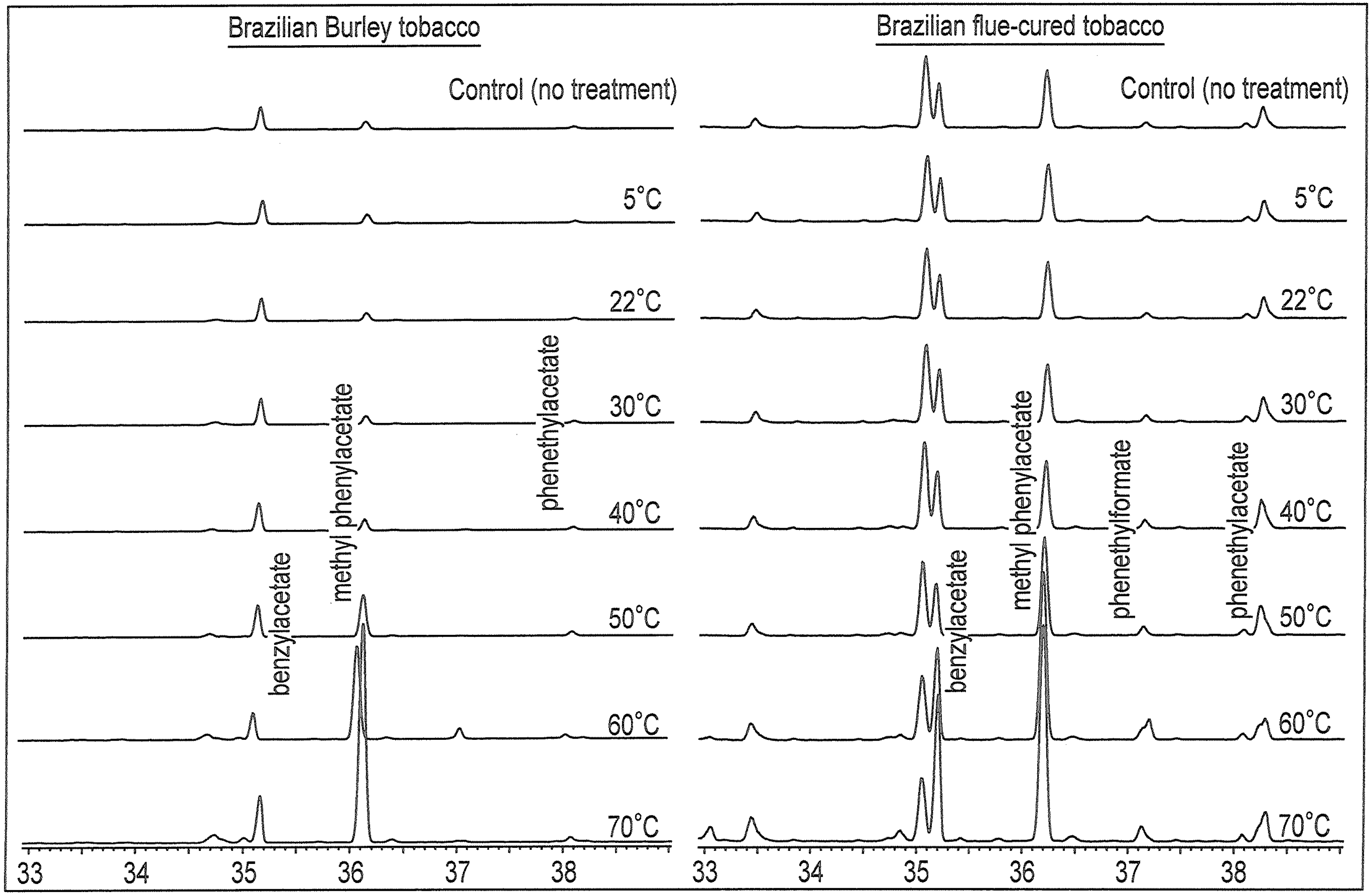
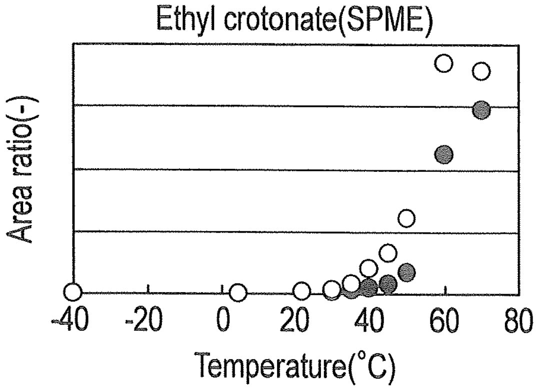

Brazilian Burley tobacco
Control (no treatment)
5°C
22°C
30°C
40°C
50°C
60°C
70°C
C2-Me
isoC4-Me
n-C4-Me
n-C6-Me
1 2 3 4 5 6 7 8 9 10 11 12 13 14
Brazilian flue-cured tobacco
Control (no treatment)
5°C
22°C
30°C
40°C
50°C
60°C
70°C
n-C4-Me
n-C6-Me
1 2 3 4 5 6 7 8 9 10 11 12 13 14

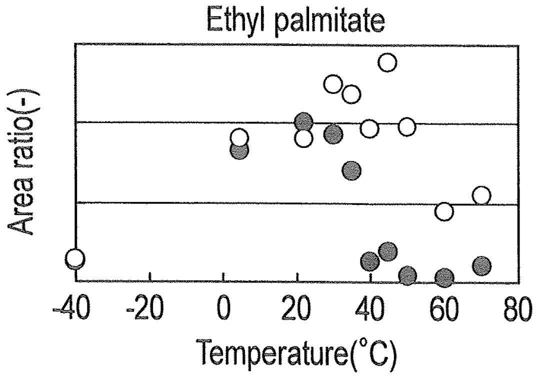
FIG. 1A


Brazilian Burley tobacco
Control (no treatment)
5°C
22°C
30°C
40°C
50°C
60°C
70°C
benzylacetate
methyl phenylacetate
phenethylacetate
33
34
35
36
37
38
Brazilian flue-cured tobacco
Control (no treatment)
5°C
22°C
30°C
40°C
50°C
60°C
70°C
benzylacetate
methyl phenylacetate
phenethylformate
phenethylacetate
33
34
35
36
37
38

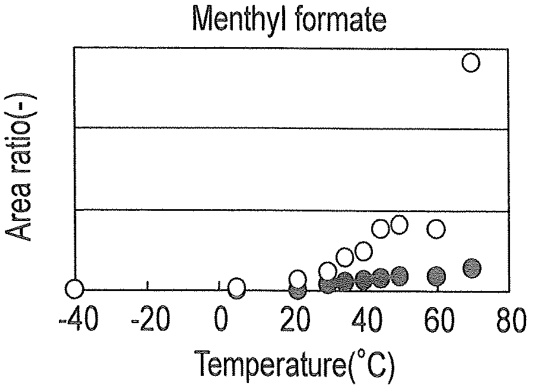
F I G. 1B



F I G. 2A



F I G. 2B



F I G. 2C

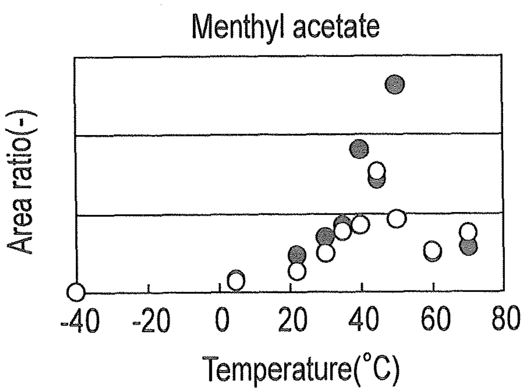


FIG. 2D

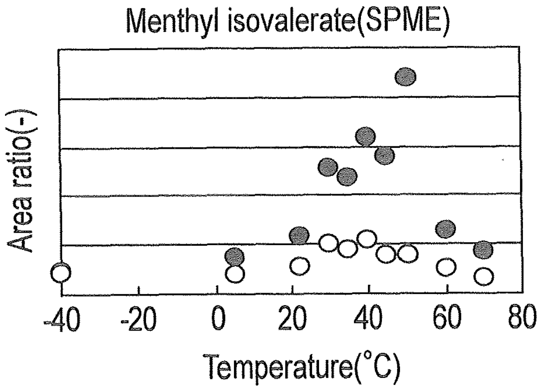


FIG. 2E

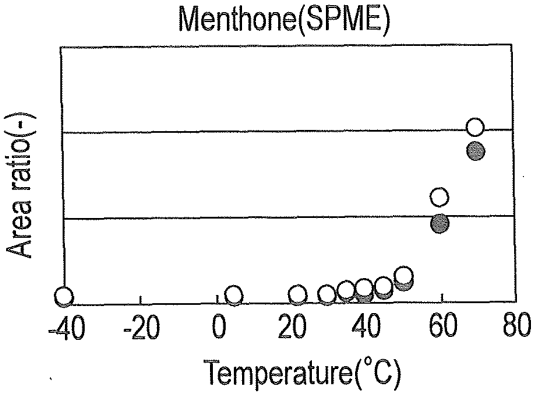


FIG. 2F

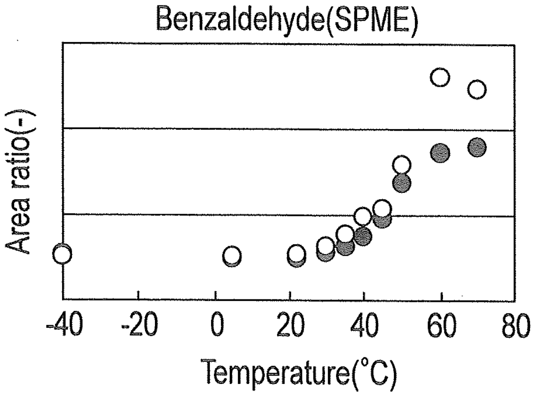


FIG. 2G

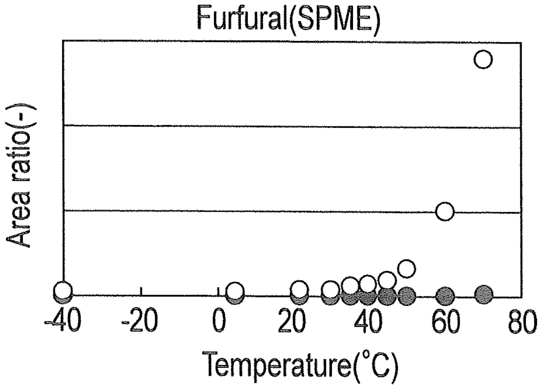


FIG. 2H

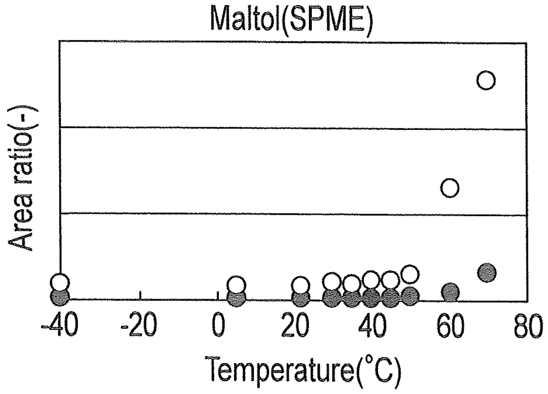


FIG. 2I

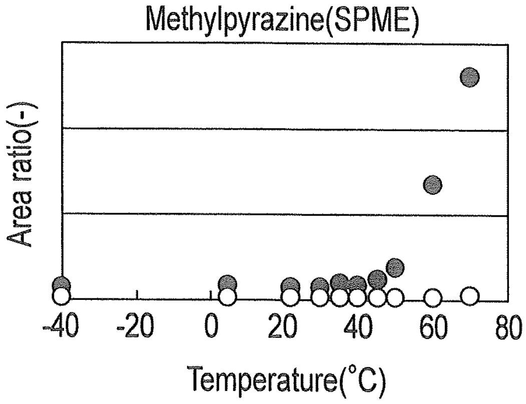


FIG. 2J

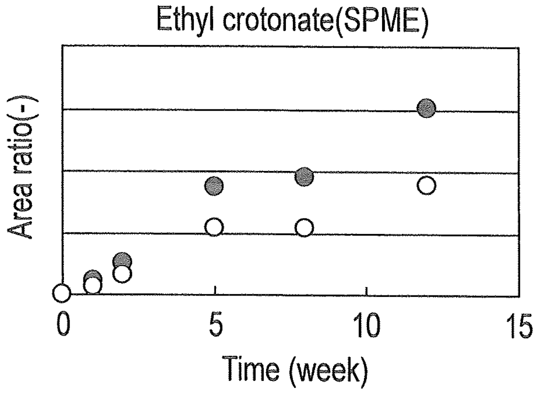


FIG. 3A

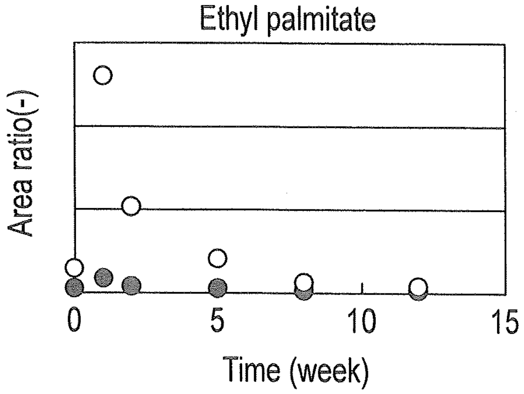
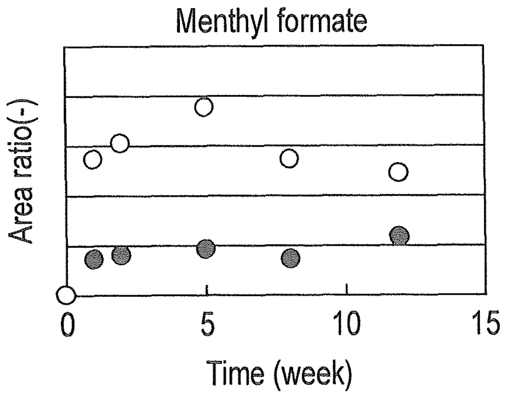
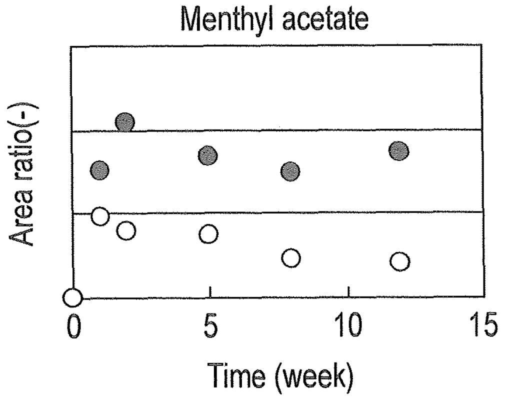


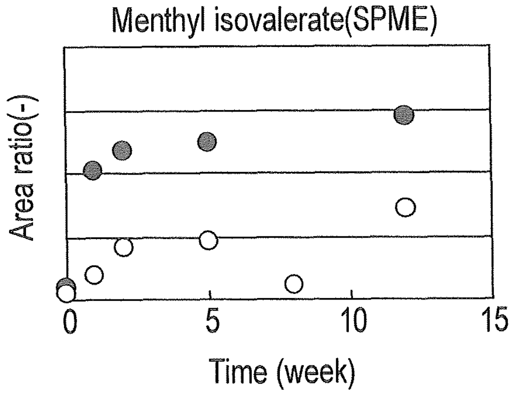
FIG. 3B



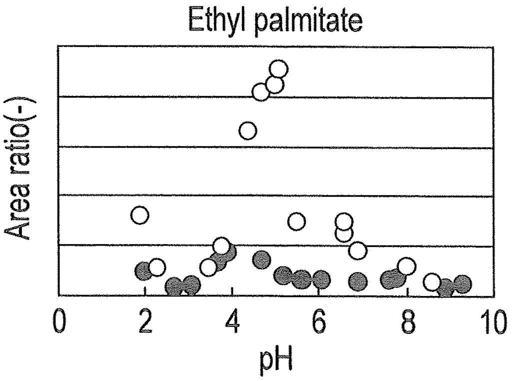
F I G. 3C



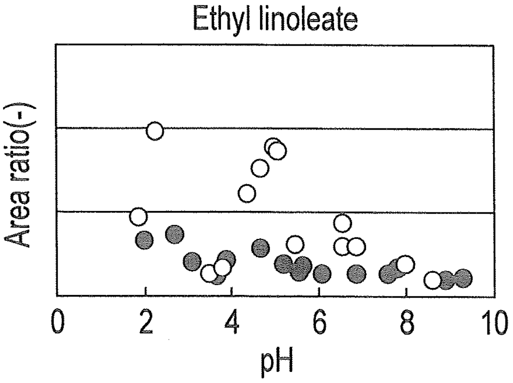
F I G. 3D



F I G. 3E


Ethyl palmitate
Area ratio(-)
0
2
4
6
8
10
pH

F I G. 4A


Ethyl linoleate
Area ratio(-)
0
2
4
6
8
10
pH

F I G. 4B

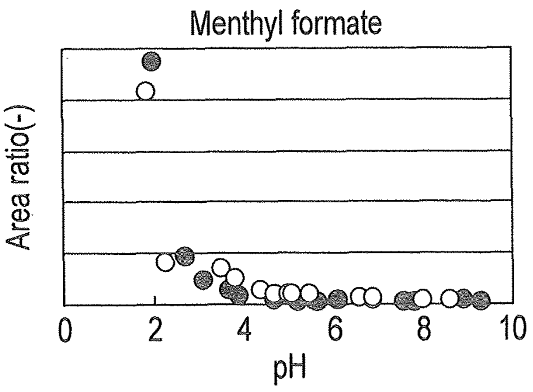


FIG. 4C

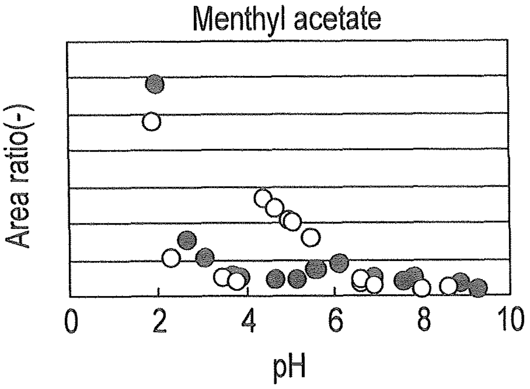


FIG. 4D

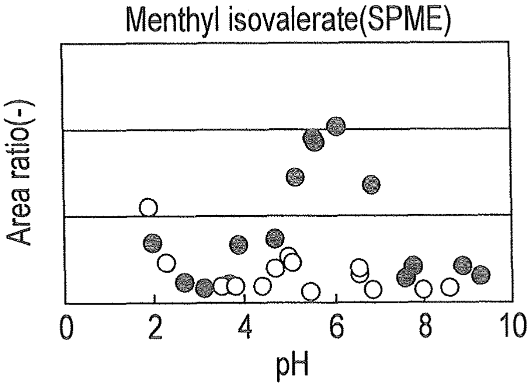


FIG. 4E

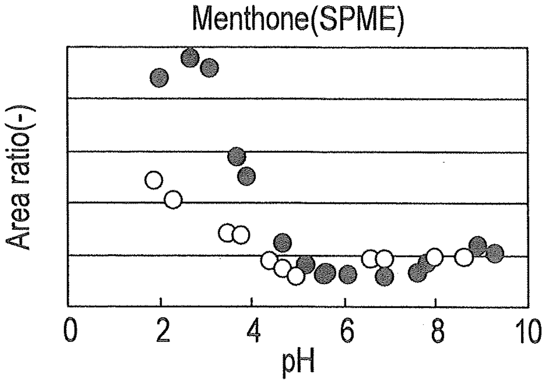


FIG. 4F

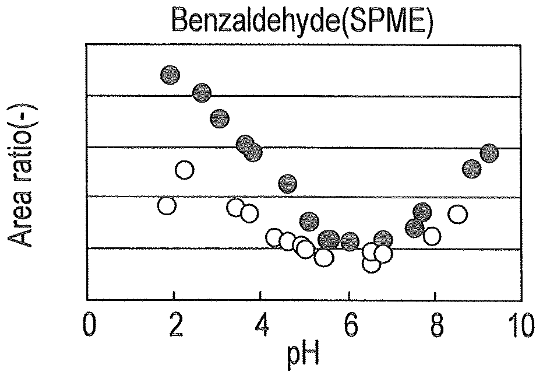


FIG. 4G

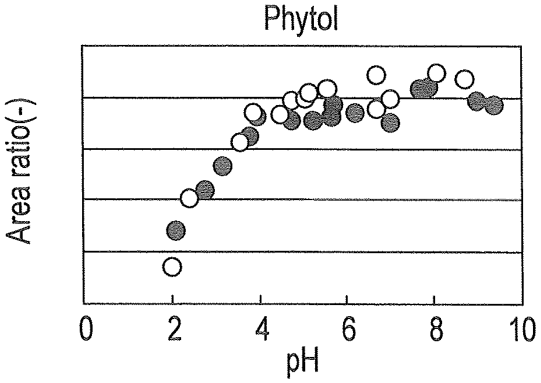


FIG. 4H

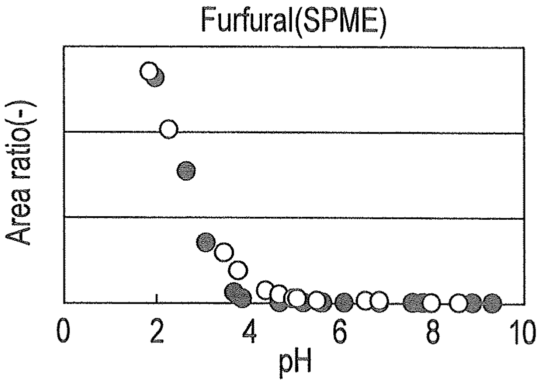


FIG. 4I

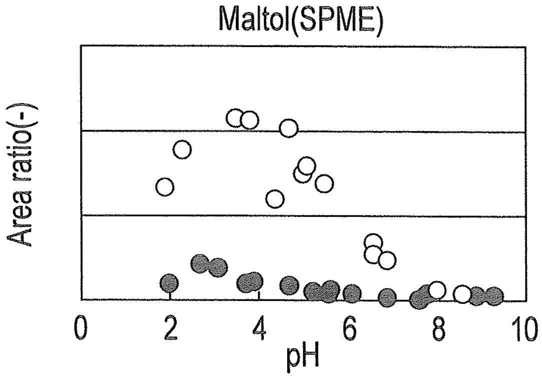


FIG. 4J

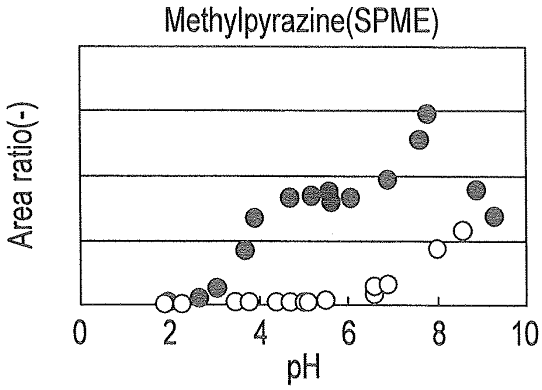


FIG. 4K

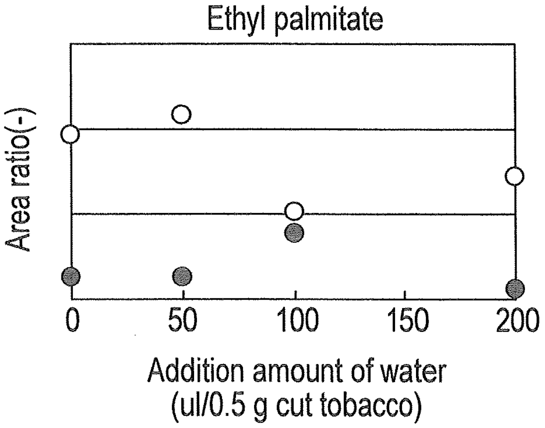

Ethyl palmitate
Area ratio(-)
0
50
100
150
200
Addition amount of water
(ul/0.5 g cut tobacco)

FIG. 5A

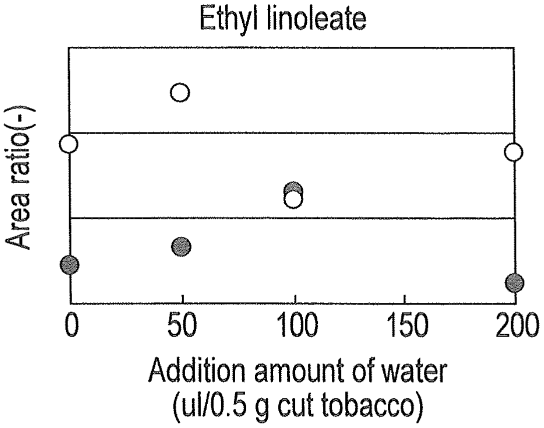

Ethyl linoleate
Area ratio(-)
0
50
100
150
200
Addition amount of water
(ul/0.5 g cut tobacco)

FIG. 5B

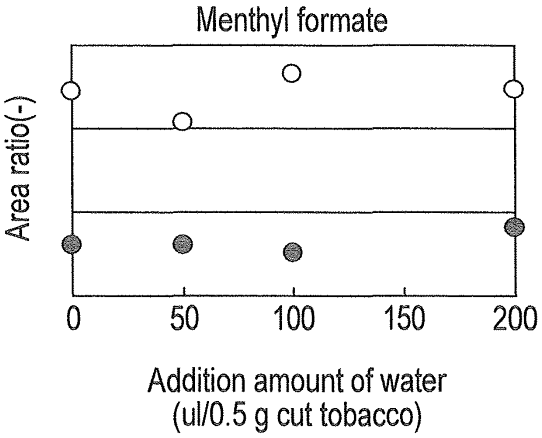


FIG. 5C

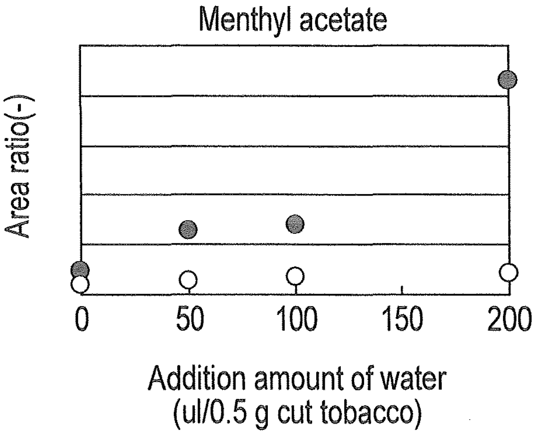


FIG. 5D

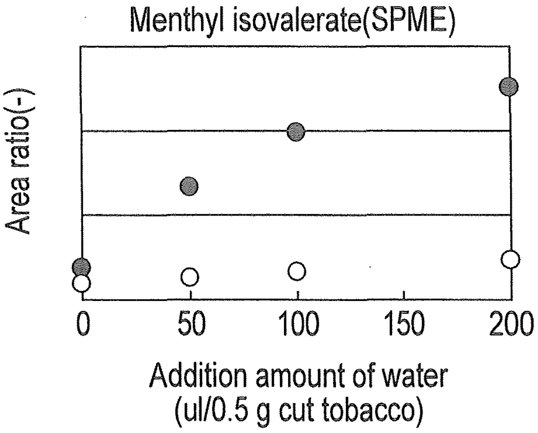


FIG. 5E

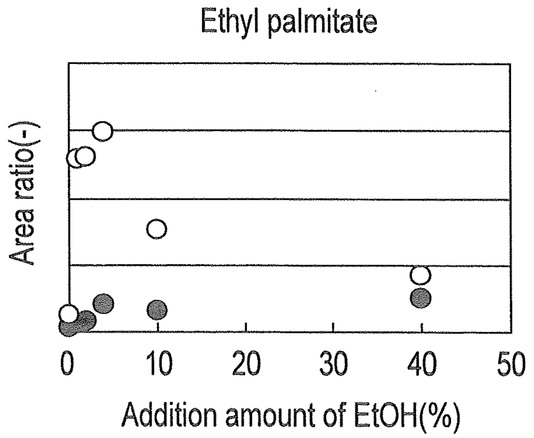

Ethyl palmitate
Area ratio(-)
0
10
20
30
40
50
Addition amount of EtOH(%)

FIG. 6A

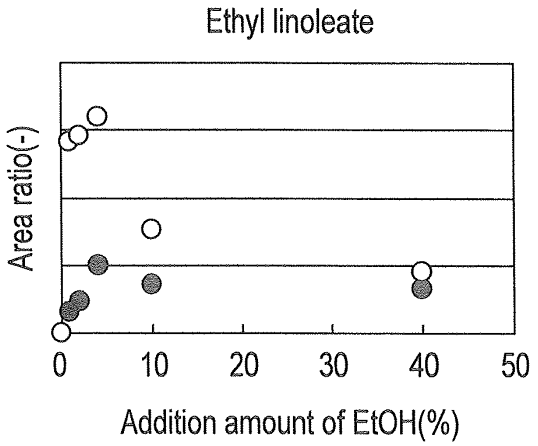

Ethyl linoleate
Area ratio(-)
0
10
20
30
40
50
Addition amount of EtOH(%)

FIG. 6B

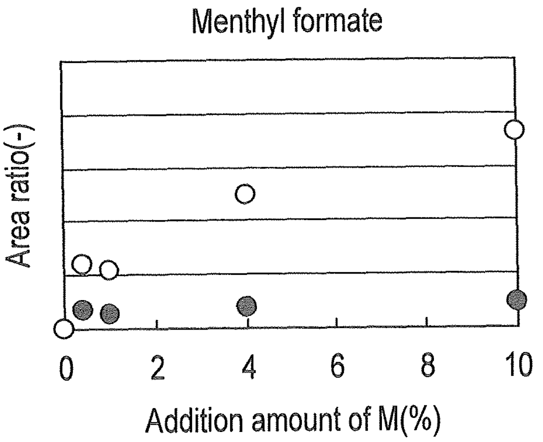

Menthyl formate
Area ratio(-)
0
2
4
6
8
10
Addition amount of M(%)

FIG. 6C

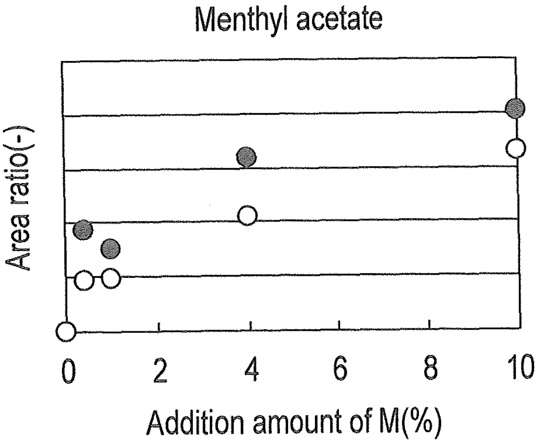

Menthyl acetate
Area ratio(-)
0
2
4
6
8
10
Addition amount of M(%)

FIG. 6D

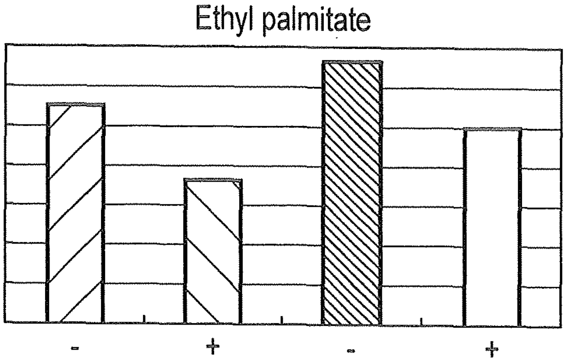


FIG. 7A

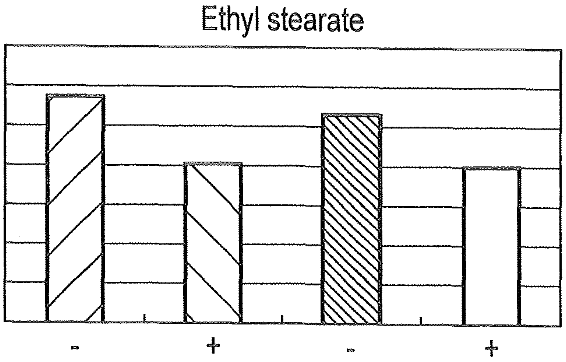


FIG. 7B

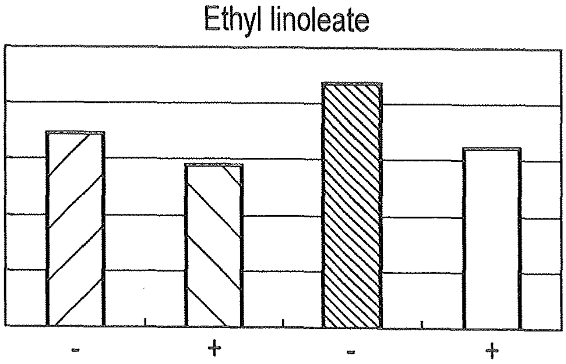


FIG. 7C

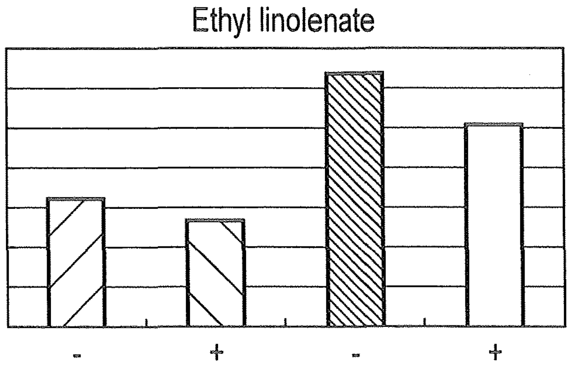


FIG. 7D

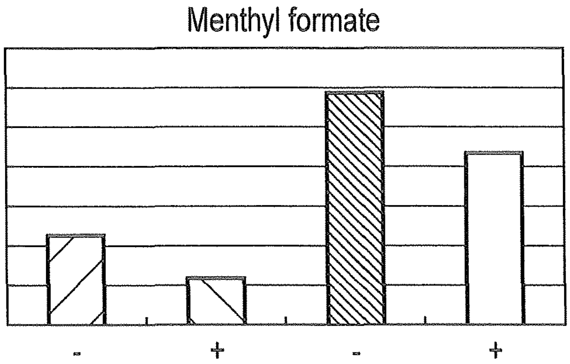


FIG. 7E

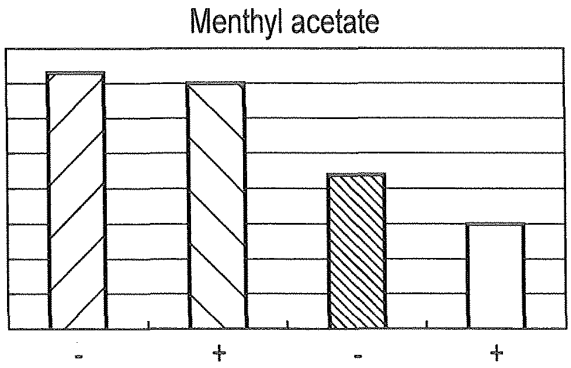
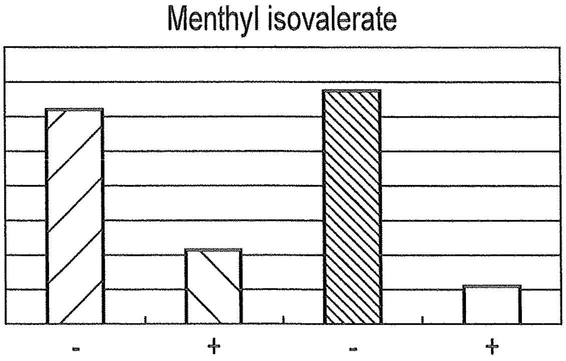
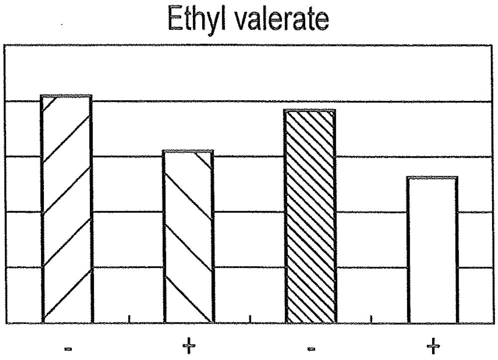


FIG. 7F



F I G. 7G



F I G. 7H

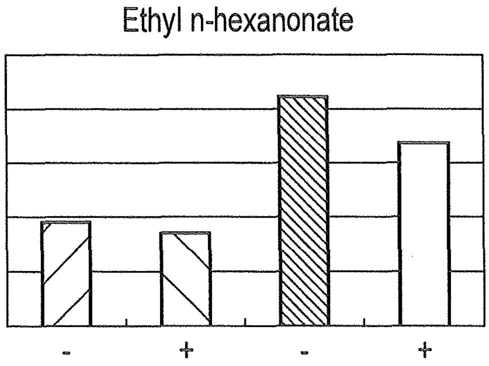


FIG. 7I

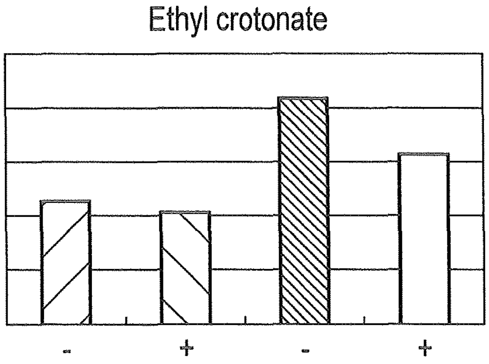


FIG. 7J

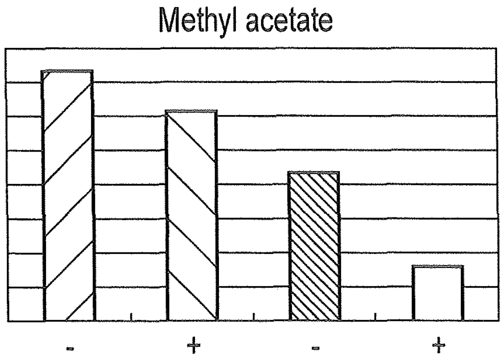


FIG. 7K

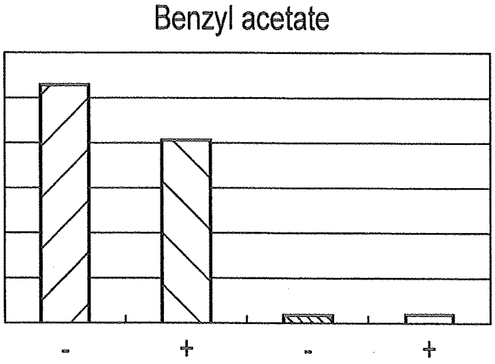


FIG. 7L

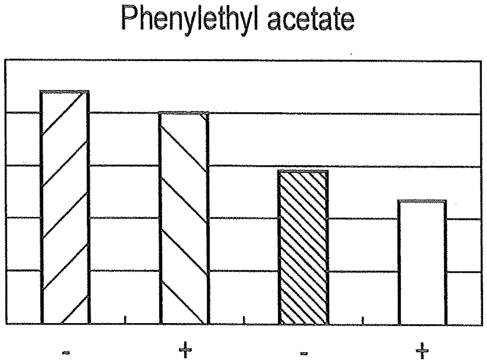


FIG. 7M

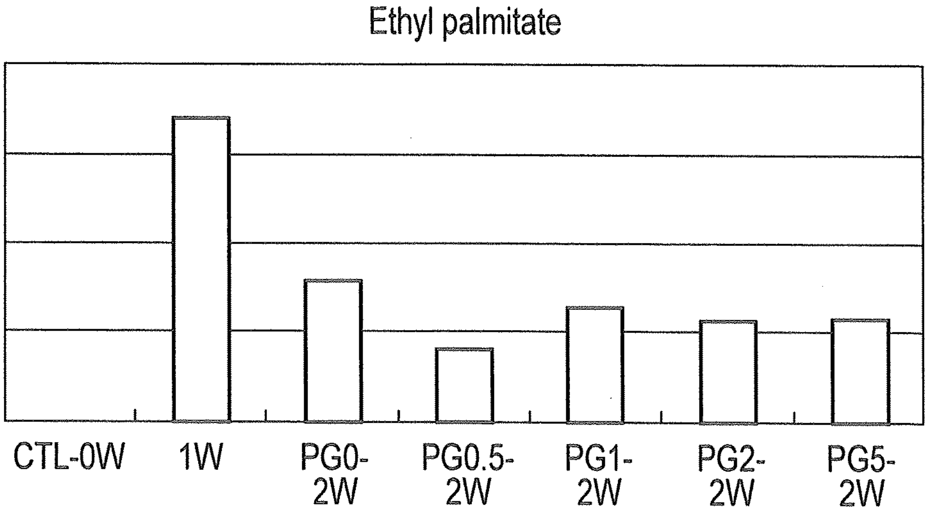

Ethyl palmitate
CTL-0W
1W
PG0-2W
PG0.5-2W
PG1-2W
PG2-2W
PG5-2W

FIG. 8A

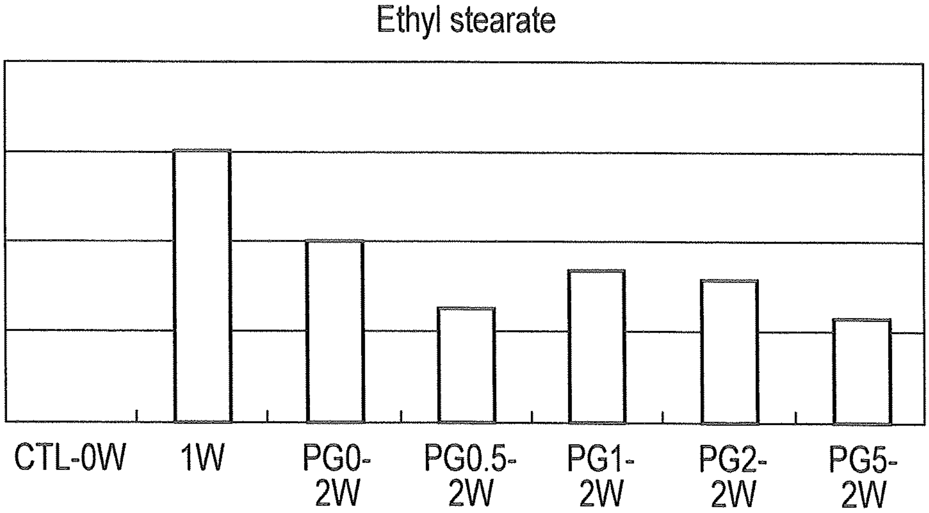

Ethyl stearate
CTL-0W
1W
PG0-2W
PG0.5-2W
PG1-2W
PG2-2W
PG5-2W

FIG. 8B

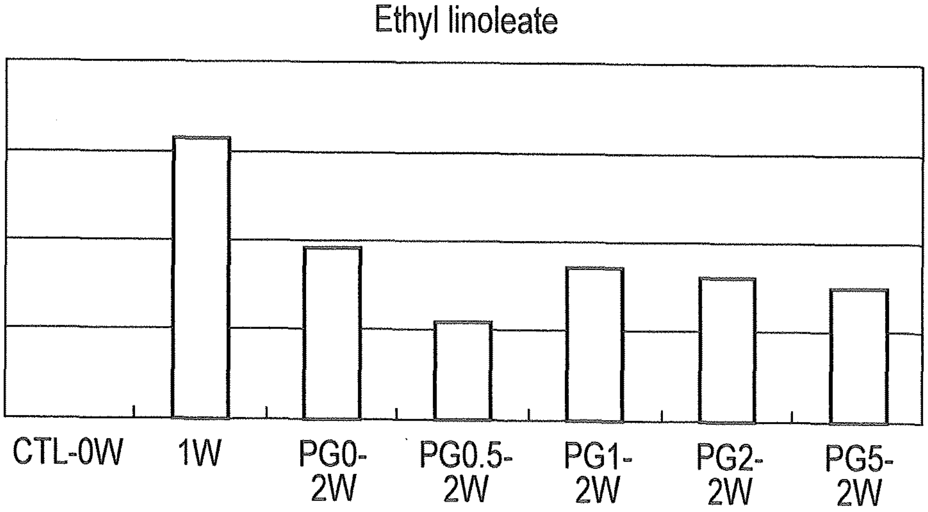

Ethyl linoleate
CTL-0W
1W
PG0-2W
PG0.5-2W
PG1-2W
PG2-2W
PG5-2W

FIG. 8C

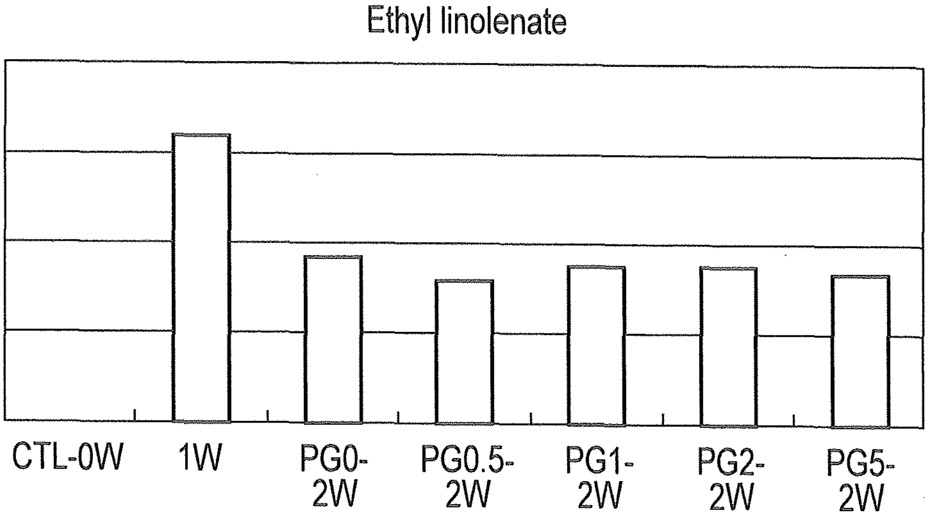
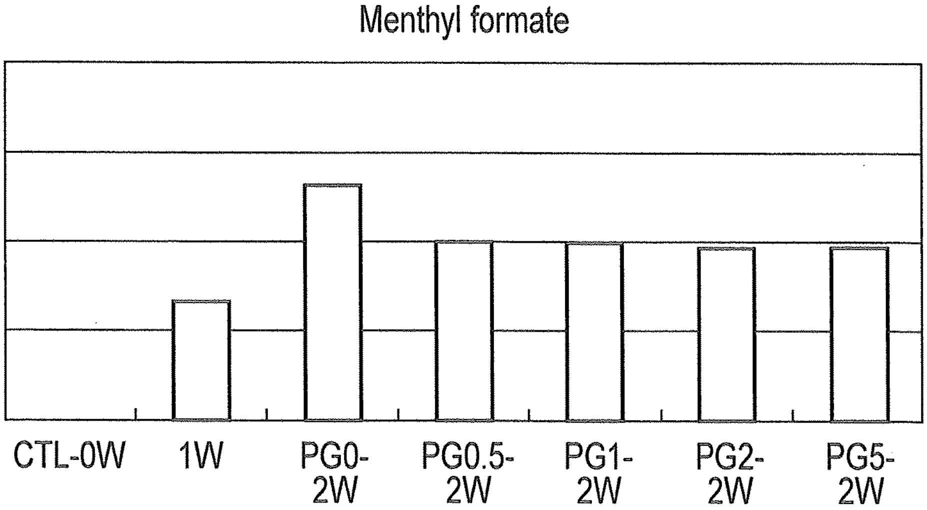
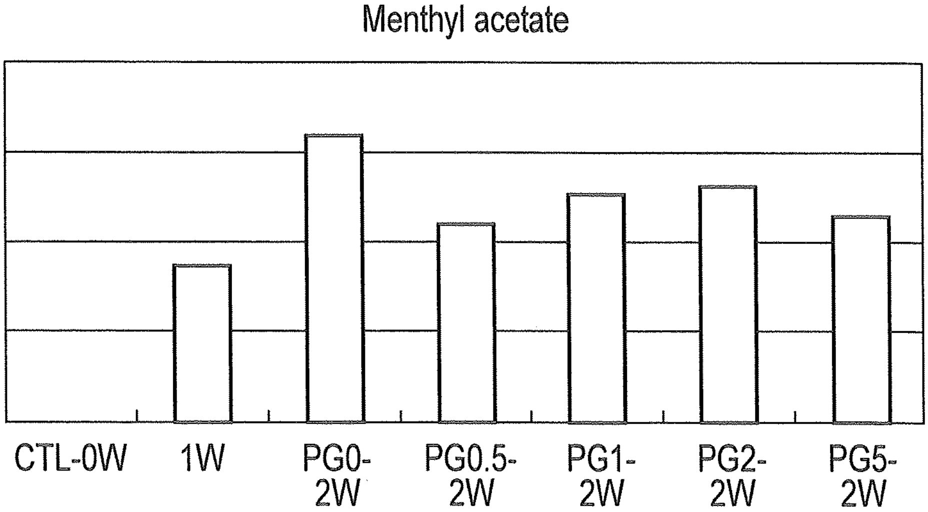

Ethyl linolenate
CTL-0W
1W
PG0-2W
PG0.5-2W
PG1-2W
PG2-2W
PG5-2W

FIG. 8D


Menthyl formate
CTL-0W
1W
PG0-2W
PG0.5-2W
PG1-2W
PG2-2W
PG5-2W

F I G. 8E


Menthyl acetate
CTL-0W
1W
PG0-2W
PG0.5-2W
PG1-2W
PG2-2W
PG5-2W

F I G. 8F

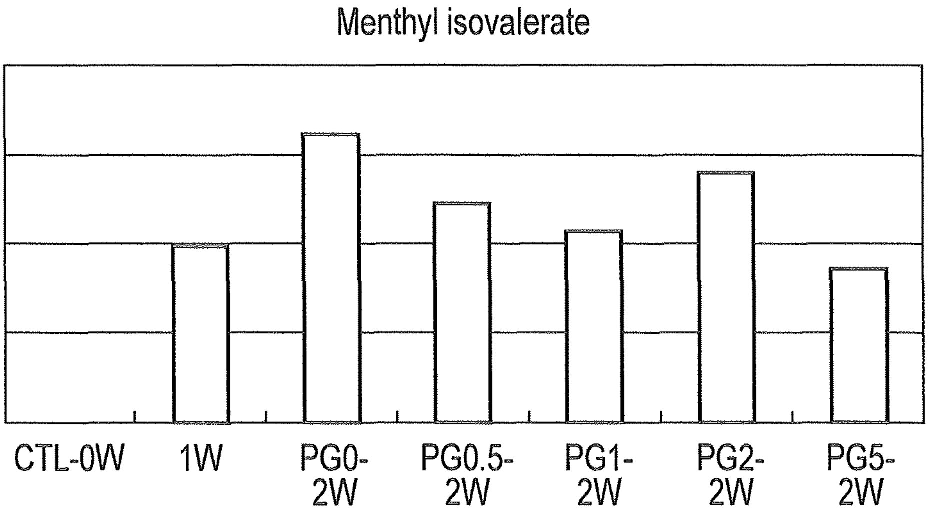

Menthyl isovalerate
CTL-0W
1W
PG0-2W
PG0.5-2W
PG1-2W
PG2-2W
PG5-2W

FIG. 8G

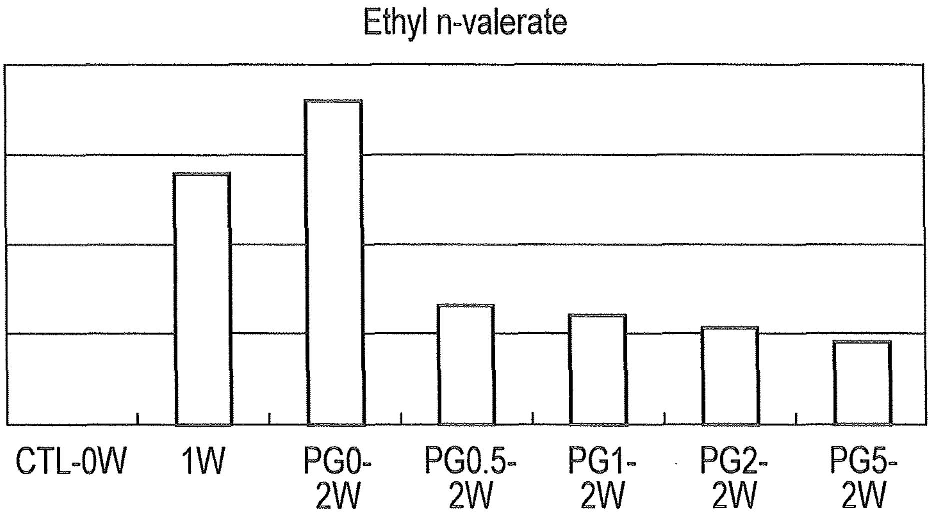
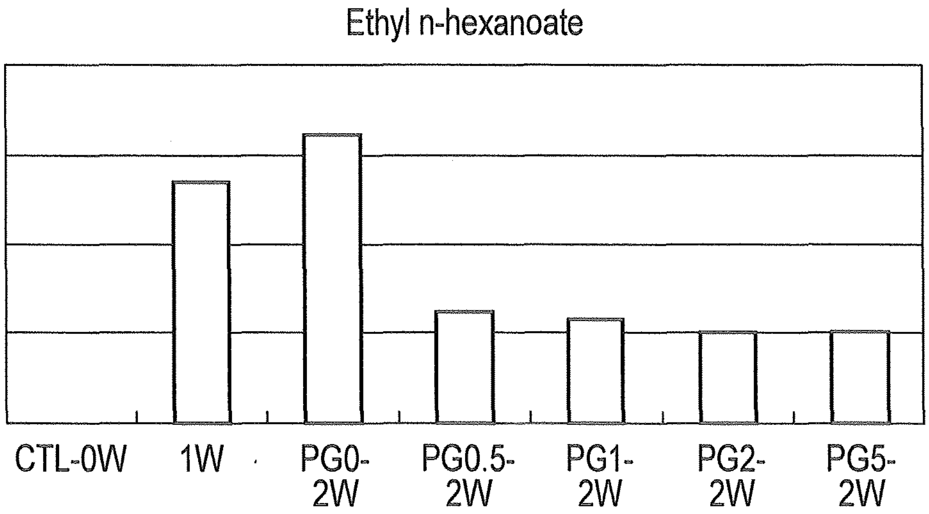
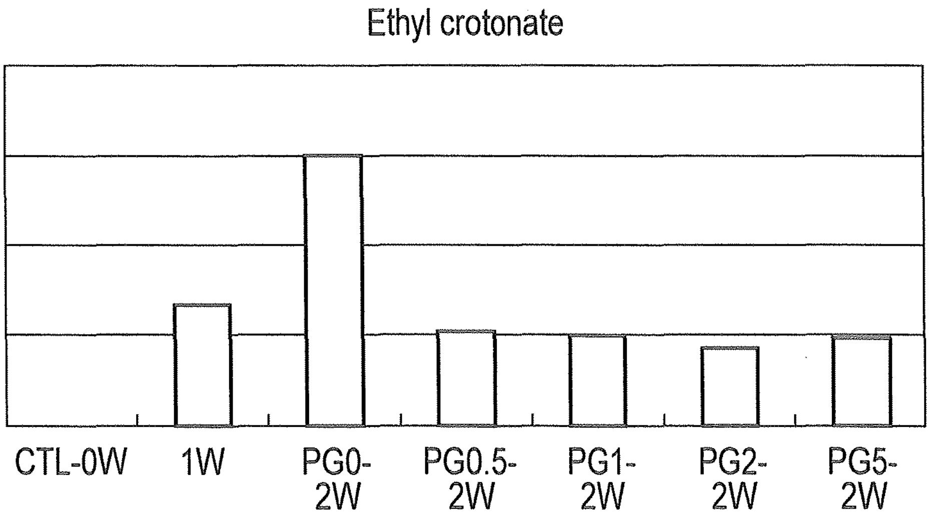

Ethyl n-valerate
CTL-0W
1W
PG0-2W
PG0.5-2W
PG1-2W
PG2-2W
PG5-2W

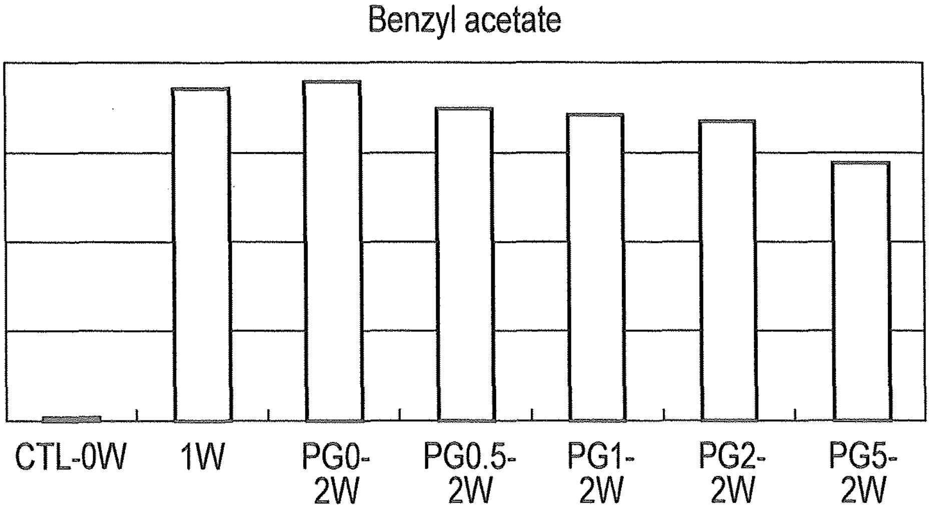
FIG. 8H


Ethyl n-hexanoate
CTL-0W
1W
PG0-2W
PG0.5-2W
PG1-2W
PG2-2W
PG5-2W

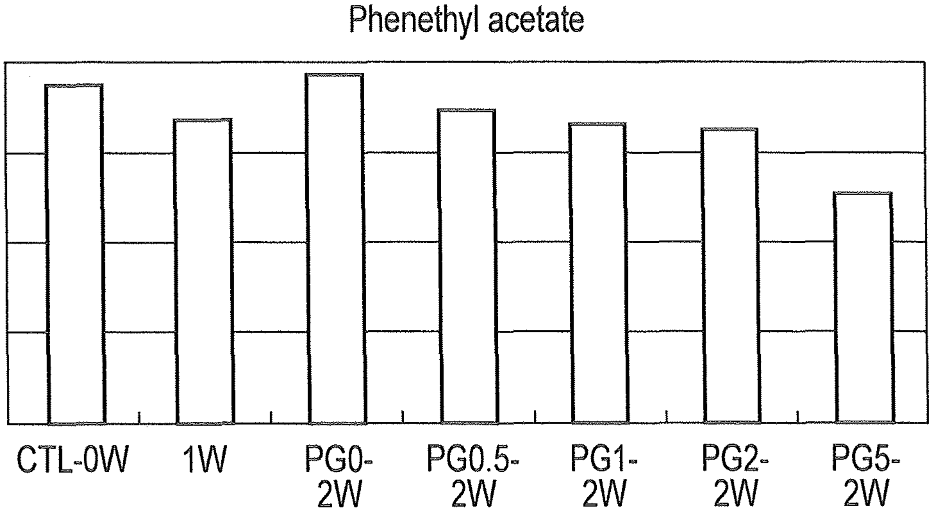
F I G. 8I


Ethyl crotonate
CTL-0W
1W
PG0-2W
PG0.5-2W
PG1-2W
PG2-2W
PG5-2W

F I G. 8J


Benzyl acetate
CTL-0W
1W
PG0-2W
PG0.5-2W
PG1-2W
PG2-2W
PG5-2W

F I G. 8K


Phenethyl acetate
CTL-0W
1W
PG0-2W
PG0.5-2W
PG1-2W
PG2-2W
PG5-2W

F I G. 8L

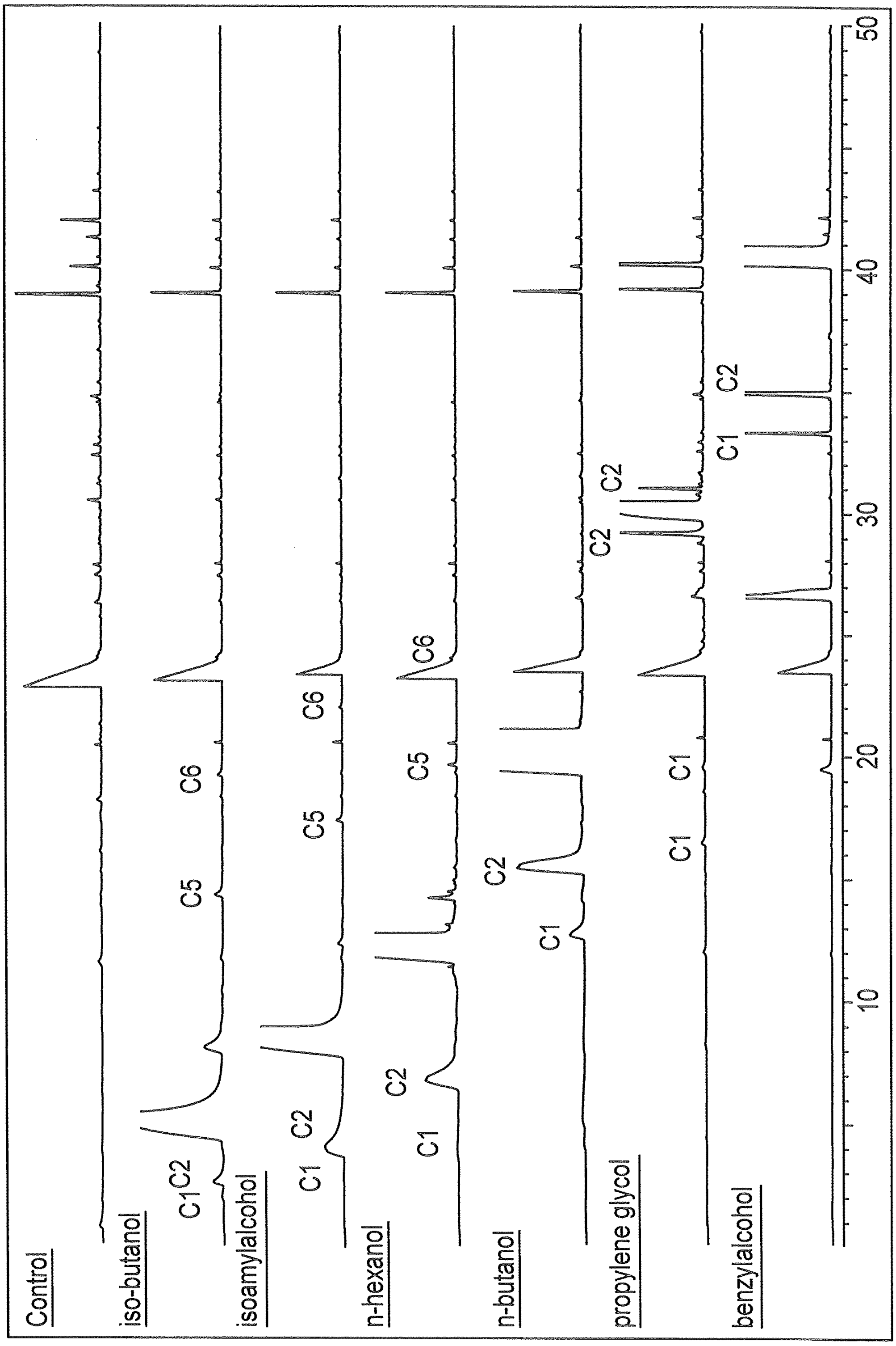
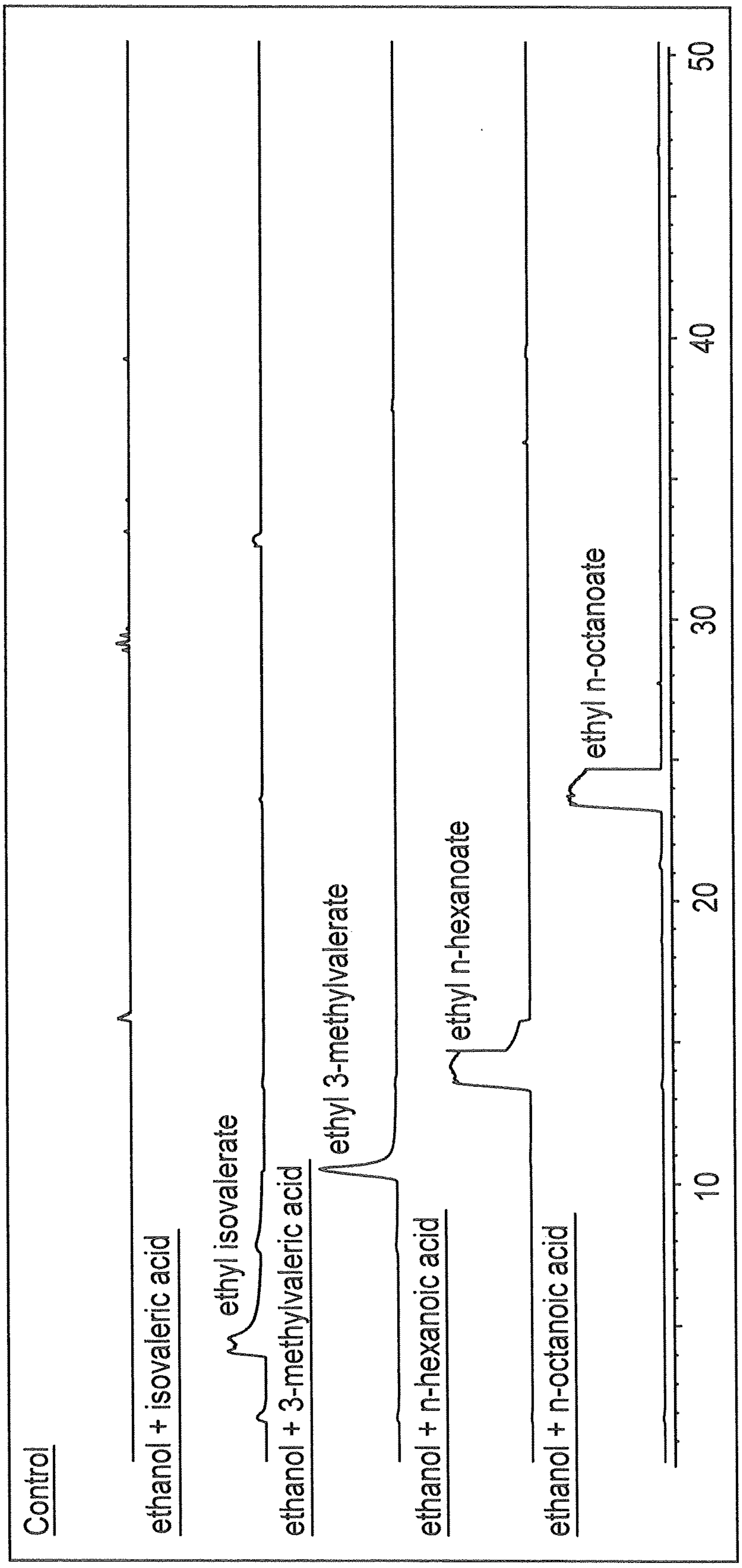

Control
iso-butanol
C1
C2
C5
C6
isoamylalcohol
C1
C2
C5
C6
n-hexanol
C1
C2
C5
C6
n-butanol
C1
C2
propylene glycol
C1
C1
C2
C2
benzylalcohol
C1
C2
10
20
30
40
50

FIG. 9


Control
ethanol + isovaleric acid
ethyl isovalerate
ethanol + 3-methylvaleric acid
ethyl 3-methylvalerate
ethanol + n-hexanoic acid
ethyl n-hexanoate
ethanol + n-octanoic acid
ethyl n-octanoate
10
20
30
40
50

F I G. 10

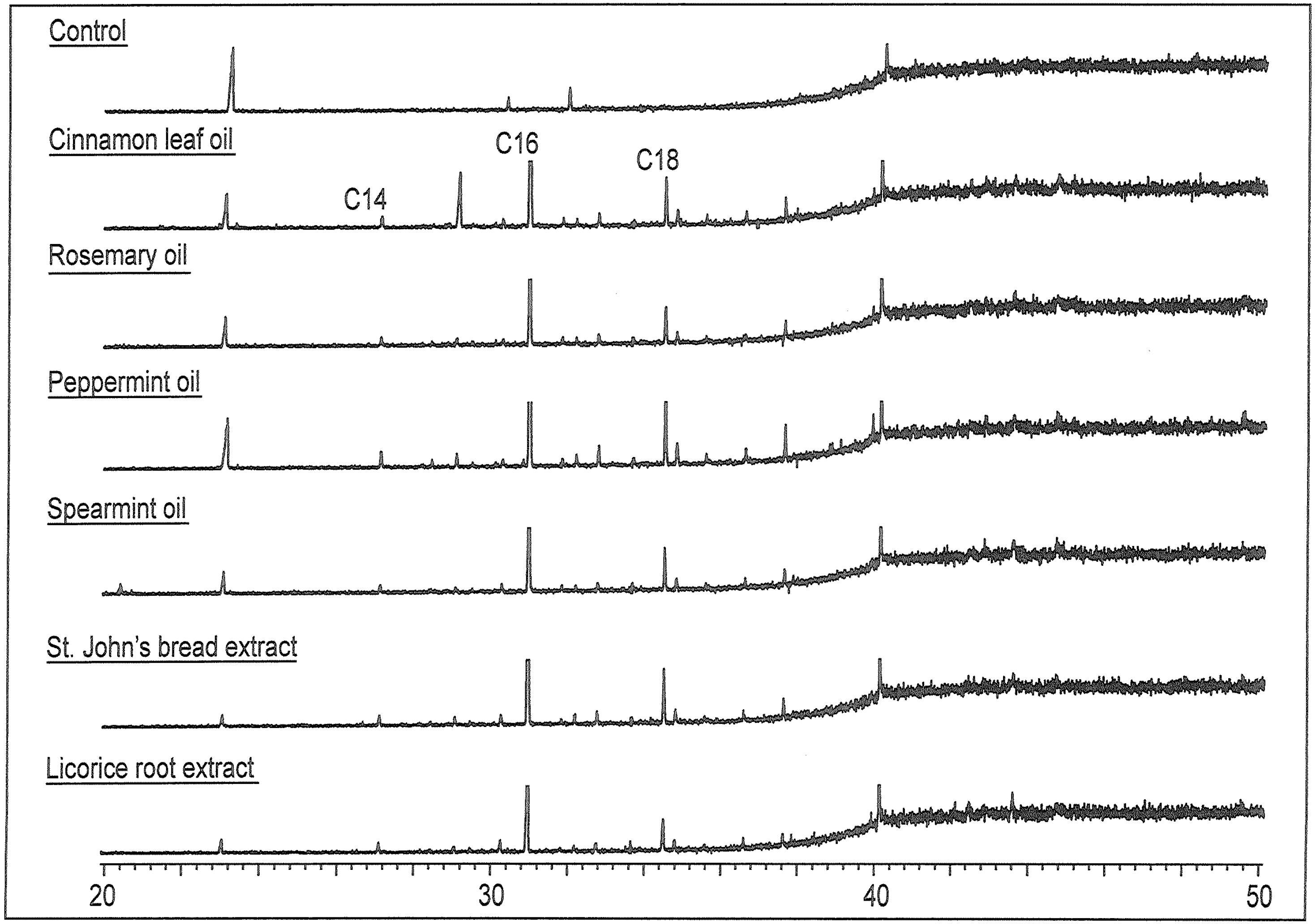
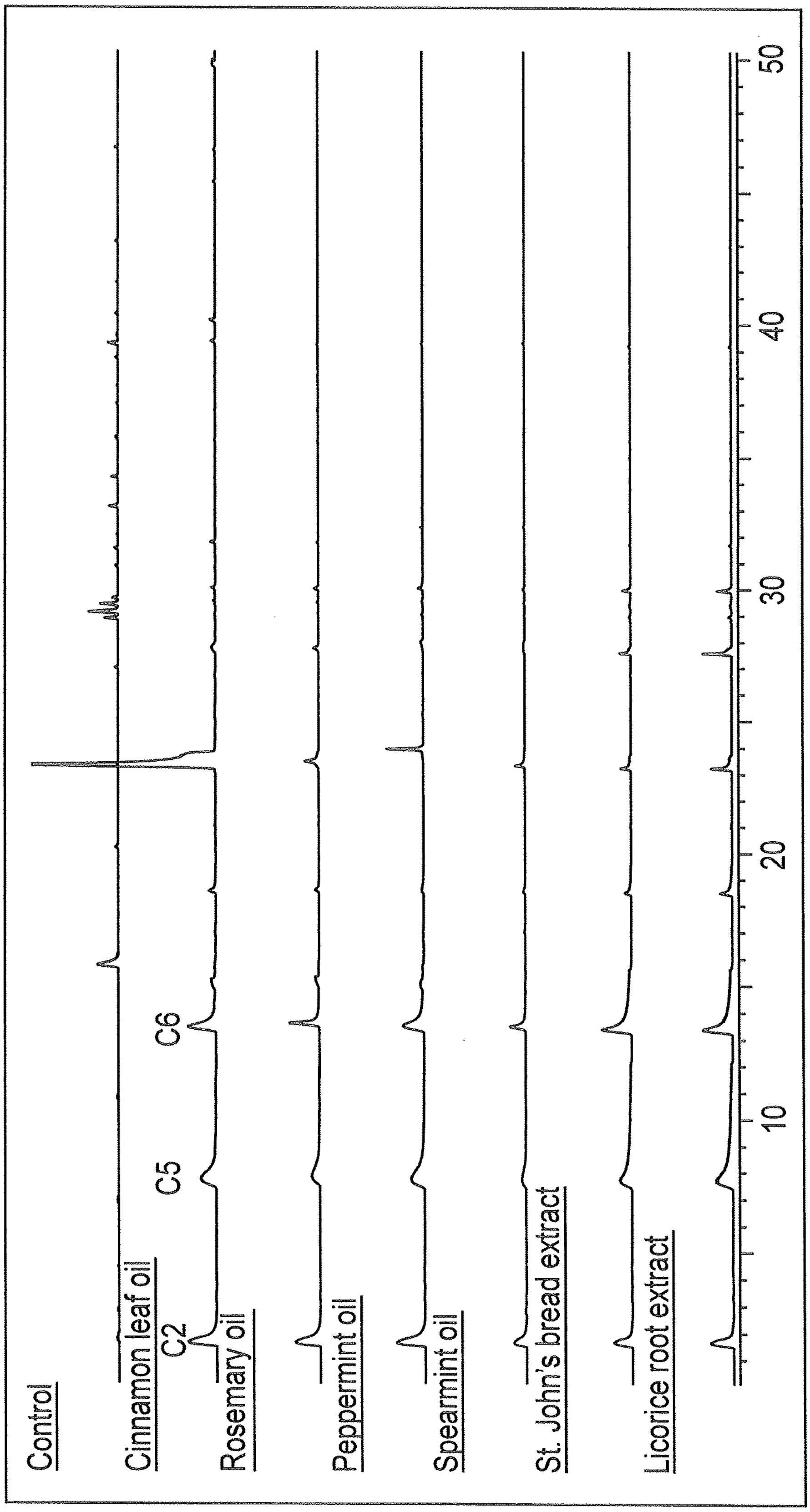

Control
Cinnamon leaf oil
C14
C16
C18
Rosemary oil
Peppermint oil
Spearmint oil
St. John's bread extract
Licorice root extract
20
30
40
50

FIG. 11A


Control
Cinnamon leaf oil
Rosemary oil
Peppermint oil
Spearmint oil
St. John's bread extract
Licorice root extract
C2
C5
C6
10
20
30
40
50

F I G. 11B

INTERNATIONAL SEARCH REPORT	International application No. PCT/JP2011/080082

A. CLASSIFICATION OF SUBJECT MATTER
A24B3/12(2006.01)i

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
A24B3/04, A24B3/12

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Jitsuyo Shinan Koho	1922-1996	Jitsuyo Shinan Toroku Koho	1996-2012
Kokai Jitsuyo Shinan Koho	1971-2012	Toroku Jitsuyo Shinan Koho	1994-2012

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	JP 57-022545 B2 (Nippon Senbai Kosha), 13 May 1982 (13.05.1982), entire text; all drawings & JP 56-51976 A	1-10
Y	JP 124767 C2 (Chosen Sotokufu Senbai Kyokucho), 18 May 1938 (18.05.1938), entire text & JP 13-000479 B1	1-10

☒ Further documents are listed in the continuation of Box C. ☐ See patent family annex.

* Special categories of cited documents:

“A” document defining the general state of the art which is not considered to be of particular relevance

“E” earlier application or patent but published on or after the international filing date

“L” document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

“O” document referring to an oral disclosure, use, exhibition or other means

“P” document published prior to the international filing date but later than the priority date claimed

“T” later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

“X” document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

“Y” document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

“&” document member of the same patent family

Date of the actual completion of the international search 21 June, 2012 (21.06.12)	Date of mailing of the international search report 03 July, 2012 (03.07.12)
Name and mailing address of the ISA/ Japanese Patent Office Facsimile No.	Authorized officer Telephone No.

Form PCT/ISA/210 (second sheet) (July 2009)

INTERNATIONAL SEARCH REPORT

International application No.
PCT/JP2011/080082

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	JP 59-033345 B2 (R.J. Reynolds Tobacco Co.), 15 August 1984 (15.08.1984), entire text; all drawings & JP 57-208977 A & US 4449541 A & EP 67601 A1 & DE 3272008 D & AU 8283982 A & BR 8202891 A & CA 1179568 A & AT 20697 T & AU 549327 B & AT 20697 E	1-10
Y	JP 2003-000219 A (Japan Tobacco Inc.), 07 January 2003 (07.01.2003), paragraphs [0015] to [0016]; fig. 1 (Family: none)	1-10
Y	JP 2008-538911 A (Philip Morris Products S.A.), 13 November 2008 (13.11.2008), paragraph [0009] & US 2007/0012328 A1 & US 2007/0095356 A1 & EP 1909603 A2 & WO 2006/120570 A2 & DE 602006009944 D & CA 2606527 A & KR 10-2008-0005588 A & NO 20076163 A & CN 101222861 A & EA 200702372 A & ZA 200708896 A & AT 446023 T & HK 1113645 A & BRA PI0611153	7-8

REFERENCES CITED IN THE DESCRIPTION

Patent documents cited in the description

- JP 56051976 A **[0003]**