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(54) **TOBACCO RAW MATERIAL, METHOD FOR MANUFACTURING SAME, AND TOBACCO PRODUCT**

(57) The present invention provides a tobacco raw material having a good smoking flavor and a method for producing the same. Provided is a tobacco raw material having a ratio, which is expressed as "water-extracted material weight/hexane-extracted material weight," be-

tween the weight of an extracted material obtained by extraction using n-hexane as a solvent and the weight of an extracted material obtained following the extraction by further extraction using water as a solvent, of 0 or more and 1.5 or less.

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DescriptionTECHNICAL FIELD

5 **[0001]** The present invention relates to tobacco raw material and a method of production thereof.

BACKGROUND ART

10 **[0002]** Known methods for adjusting the smoking flavor of tobacco include art in which specific components are extracted and removed from a leaf tobacco material, and art that entails carrying out an extraction operation on a leaf tobacco material so as to obtain an extract and an extraction residue, carrying out a given treatment, etc. on the extract, then re-applying the treated extract to the extraction residue.

15 **[0003]** A known technique for extracting and removing specific components from a leaf tobacco material involves removing lipid components from tobacco using a mixed solvent of an aliphatic hydrocarbon and a lower alcohol (Patent Document 1).

[0004] A known technique for carrying out an extraction operation on a leaf tobacco material and re-applying the extract to the extraction residue involves the method of carrying out extraction on tobacco raw material with a surfactant-containing solution, removing the surfactant and polypeptides within the extract, and then adding the resulting solution to the tobacco raw material (Patent Document 2).

20 **[0005]** A method for reducing nitrosamines from tobacco raw material that entails using a supercritical fluid to treat the tobacco raw material and removing nitrosamines from the extract obtained by such treatment is also known (Patent Document 3).

25 **[0006]** Yet another technique that is known involves condensing the smoking flavor components present in a leaf tobacco material on the surface of the leaf tobacco via the operations of immersing the leaf tobacco material under applied pressure in carbon dioxide that has been rendered into a supercritical liquid, and then lowering the pressure to atmospheric pressure (Patent Document 4).

[0007]

Patent Document 1: Japanese Examined Patent Publication No. S58-4544

30 Patent Document 2: Japanese Translation of PCT Application No. H7-505521

Patent Document 3: Japanese Patent No. 3931084

Patent Document 4: Japanese Patent Application Laid-open No. H8-23952

DISCLOSURE OF THE INVENTION

35 **[0008]** Leaf tobacco materials contain many components of differing polarities. Such components are broadly divided into low-polarity, high-polarity and bipolar components. When carrying out the removal of these components by extraction, such as when carrying out extraction with a low-polarity solvent, the low-polarity and bipolar component groups within the leaf tobacco are removed.

40 **[0009]** By contrast, when carrying out extraction with a high-polarity solvent, the high-polarity and bipolar components within the leaf tobacco material are removed.

[0010] Hence, with a single extraction using a low-polarity or high-polarity solvent, in addition to the low-polarity or high-polarity components, the group of bipolar components is also removed. This sometimes has a large influence on the smoking flavor of the tobacco raw material.

45 **[0011]** Also, in the case of art which, after carrying out extraction, adds the extract to tobacco raw material, the group of components obtained by a single extraction includes numerous components. When the extract has been treated (such as by contact with an antioxidant), in addition to the components to be removed, the components which affect the smoking flavor are also thought to change. Hence, there seems to be a possibility of a large influence on the smoking flavor.

50 **[0012]** It is therefore an object of this invention to provide a tobacco raw material having a good smoking flavor. Another object is to provide a method of producing such a tobacco raw material.

[0013] The inventors have conducted extensive investigations, as a result of which they have discovered that a tobacco raw material, having a ratio, which is expressed as "water-extracted material weight/hexane-extracted material weight," between the weight of an extracted material obtained by extraction using n-hexane as a solvent and the weight of an extracted material obtained following the extraction by further extraction using water as a solvent, of 0 or more and 1.5 or less, has an excellent smoking flavor.

[0014] In addition, the inventors have also discovered that the above problems can be resolved by a method of production that includes: a first extraction step of extracting a leaf tobacco material using a solvent having a dielectric constant of 1 or more and less than 25 to obtain an extract and an extraction residue; a second extraction step of,

following the first extraction step, extracting the extraction residue obtained in the first extraction step with a solvent having a dielectric constant of 25 or more to obtain an extraction residue; and the step of re-applying the extract obtained in the first extraction step to the extraction residue obtained in the second extraction step so as to form a leaf tobacco raw material. These discoveries ultimately led to the present invention.

[0015] The invention is recited below.

[1] A tobacco raw material having a ratio, which is expressed as "water-extracted material weight/hexane-extracted material weight," between the weight of an extracted material obtained by extraction using n-hexane as a solvent and the weight of an extracted material obtained following the extraction by further extraction using water as a solvent, of 0 or more and 1.5 or less.

[2] The tobacco raw material according to [1], wherein the sum of the weight of an extracted material obtained by extraction using n-hexane as the solvent and the weight of an extracted material obtained following the extraction by further extraction using water as the solvent is from 5 to 17 wt % of the weight of the tobacco raw material before the extractions.

[3] A method of producing a tobacco raw material, which method includes: a first extraction step of extracting a leaf tobacco material using a solvent having a dielectric constant of 1 or more and less than 25 to obtain an extract and an extraction residue; a second extraction step of, after the first extraction step, extracting the extraction residue obtained in the first extraction step using a solvent having a dielectric constant of 25 or more to obtain an extraction residue; and the step of re-applying the extract obtained in the first extraction step to the extraction residue obtained in the second extraction step so as to form a leaf tobacco raw material.

[4] The production method according to [3], wherein the solvent used in the first extraction step is a solvent having a dielectric constant of from 1 to 20 and the solvent used in the second extraction step is water.

[5] The production method according to [3] or [4], wherein the solvent used in the first extraction step is one or more selected from among n-hexane, tetrahydrofuran and ethanol.

[6] A tobacco raw material obtained by the production method according to any one of [3] to [5].

[7] A tobacco product which uses the tobacco raw material according to [1], [2] or [6].

[0016] In this specification, "leaf tobacco material" refers to a material used to produce "tobacco raw material", and means leaf tobacco prior to passing through treatment such as extraction steps. Also, "tobacco raw material" refers to the product obtained by applying treatment such as extraction steps to the "leaf tobacco material".

[0017] The invention provides a tobacco raw material having a good smoking flavor, and also provides a method of producing the same.

BRIEF DESCRIPTION OF THE DIAGRAMS

[0018]

FIG. 1 is a graph showing sensory evaluation results for Example 3 and Comparative Example 2.

FIG. 2 is a graph showing sensory evaluation results for Example 4 and Comparative Example 3.

FIG. 3 is a graph showing sensory evaluation results for Example 5 and Comparative Example 4.

FIG. 4 is a graph showing sensory evaluation results for Example 6 and Comparative Example 1.

FIG. 5 is a graph showing sensory evaluation results for Comparative Examples 5 and 6.

FIG. 6 is a graph showing sensory evaluation results for Comparative Example 7.

MODE FOR CARRYING OUT THE INVENTION

[0019] The invention is described in detail below by way of embodiments, examples and the like. However, the invention is not limited to the following embodiments and examples, and may be practiced using any modifications thereto insofar as they do not depart from the spirit and scope of the invention.

<Production Method of the Invention>

[0020] The inventive method of producing a tobacco raw material includes: a first extraction step of extracting a leaf tobacco material using a solvent having a dielectric constant of 1 or more and less than 25 to obtain an extract and an extraction residue; a second extraction step of, after the first extraction step, extracting the extraction residue obtained in the first extraction step using a solvent having a dielectric constant of 25 or more; and the step of re-applying the extract obtained in the first extraction step to the extraction residue obtained in the second extraction step so as to form a leaf tobacco raw material.

[0021] The inventive method of producing a tobacco raw material includes, as a first extraction step, the step of extracting a leaf tobacco material using a solvent having a dielectric constant of 1 or more and less than 25 so as to obtain an extract and an extraction residue.

[0022] The leaf tobacco material that may be used in this invention is not particularly limited as to type. Illustrative examples include genus *Nicotiana* varieties such as flue-cured and burley varieties of *nicotiana tabacum*, and the brasilia variety of *nicotiana rustica*.

[0023] These leaf tobacco materials are cut to a suitable size (having a particle diameter of about 0.425 to 1.18 mm) using suitable means such as a grinding mill. Use may be made of such materials that have been dried by a known method.

[0024] When the leaf tobacco material obtained by the foregoing treatment is extracted using a solvent having a dielectric constant of 1 or more and less than 25, the desired smoking flavor components contained in the leaf tobacco material can be extracted.

[0025] The solvent used in the first extraction step is more preferably one having a dielectric constant of 1 or more and 20 or less.

[0026] Illustrative examples of solvents having a dielectric constant of 1 or more and less than 25 include ethanol, isopropyl alcohol, ethyl acetate, linear or branched alkanes of 5 to 10 carbons, n-propyl alcohol, butanol, benzene, toluene, xylene, benzyl alcohol, acetone, tetrahydrofuran, diethyl ether, 1,4-dioxane, chloroform and dichloromethane.

[0027] Examples of the alkanes include pentane, n-hexane, cyclohexane, heptane, octane, nonane and decane.

[0028] Of the above, embodiments that do not use a low-molecular-weight organic ester such as ethyl acetate can be exemplified.

[0029] Solvents that may be preferably used include ethanol, n-hexane, heptane, diethyl ether and tetrahydrofuran.

[0030] The solvent having a dielectric constant of 1 or more and less than 25 may be of a single type used alone or a plurality of such solvents may be used in admixture. In cases where a mixed solvent is used, the dielectric constant of the mixed solvent is adjusted to 1 or more and less than 25.

[0031] Extraction may be carried out at normal temperature and pressure, or may be carried out with warming to, for example, about 40 to 80°C and under an applied pressure of from 1,000 to 2,000 psi. When indicating the pressure in the SI unit system, calculation is carried out at a conversion factor of 1 psi $\cong$ 6,894.757 Pa.

[0032] The extraction time is not particularly limited and may be, for example, from 5 minutes to 1 hours.

[0033] Extraction may be carried out once or may be carried out a plurality of times. Extraction may be carried out using the subsequently described accelerated solvent extraction system.

[0034] In the first extraction step, an operation for separating the extract and the extraction residue may be suitably provided. For example, the method of applying the extracted material to filter paper or a sieve having a suitable pore size, and thereby separating it into an extract and an extraction residue may be used.

[0035] The resulting extraction residue is dried by a suitable method. The drying method is exemplified by freeze-drying.

[0036] By additionally carrying out, on the extraction residue obtained via the first extraction step, an extraction operation using a solvent having a dielectric constant of 25 or more, components within the leaf tobacco material that were not extracted in the first extraction step can be extracted.

[0037] The solvent having a dielectric constant of 25 or more that is used in the second extraction step is not particularly limited. Illustrative examples include water, methanol, and mixed solvents containing these.

[0038] The second extraction step, as with the first extraction step, may be carried out at normal temperature and pressure as the extraction conditions. Extraction may also be carried out with warming to about 40 to 80°C and under an applied pressure of 1,000 to 2000 psi.

[0039] The extraction time is not particularly limited, and may be set to from 5 minutes to 1 hour.

[0040] Extraction may be carried out once or may be carried out a plurality of times. Also, extraction may be carried out using the subsequently described accelerated solvent extraction system.

[0041] In the first extraction step and the second extraction step, because the solvent used in the second extraction step has a higher dielectric constant than the solvent used in the first extraction step, components within the leaf tobacco material that were not extracted in the first extraction step can be extracted in the second extraction step. The extract obtained in the second extraction step is not re-applied to the extraction residue.

[0042] In the second extraction step, unwanted components contained in the leaf tobacco material can be removed from the extraction residue obtained in the first extraction step.

[0043] Examples of unwanted components include high-polarity components, such as low-molecular-weight organic acids and ammonia.

[0044] In the production method of the invention, the order of the first extraction step and the second extraction is important. That is, it is important to use a relatively low-polarity solvent as the solvent in the initial extraction step, and to use a relatively high-polarity solvent having a higher dielectric constant than the solvent used in the first extraction step as the solvent in the next extraction step. If this order is reversed, a tobacco raw material having a good smoking flavor cannot be obtained.

[0045] It is possible to carry out the first extraction step a plurality of times and then carry out the second extraction

step a plurality of times, or to carry out the first extraction step and the second extraction step in succession and then repeat this by again carrying out the first extraction step and again carrying out the second extraction step.

[0046] From the standpoint of simplifying the production steps, it is desirable for the total number of steps represented by the first extraction and second extraction steps to be small.

[0047] The production method of the invention includes the step of, after passing through the second extraction step, re-applying the extract obtained in the first extraction step to the extraction residue obtained by passing through the second extraction step so as to form a leaf tobacco raw material.

[0048] In this step, by re-applying the extract obtained in the first extraction step to the extraction residue obtained in the second extraction step, components that were extracted in the first extraction step and are desirable for the smoking flavor of the tobacco raw material are returned to the tobacco raw material.

[0049] When re-applying the extract to the extraction residue, having the extract spread uniformly throughout the extraction residue is desirable from the standpoint of maintaining uniformity of quality as a tobacco raw material.

[0050] The extract obtained in the first extraction step, the extract obtained in the first extraction step may be re-applied directly as is, or may be re-applied after concentrating the solvent only. Alternatively, the extract may be subjected to salting-out treatment.

[0051] After the extract has been re-applied to the extraction residue, the solvent is removed by a suitable method.

[0052] It is possible in this way to obtain the tobacco raw material of the invention.

[0053] The mode of use of the tobacco raw material obtained by the production method of the invention is exemplified by use at normal temperature and use in a warmed state, and the smoking flavor when used in either of these ways is excellent.

[0054] Modes of use at normal temperature or under heated conditions are exemplified by use in which leaf tobacco obtained by the above treatment is filled into, for example, a tube having a bore of 0.7 mm and used.

[0055] Modes of use in a warmed state are exemplified by use in which the leaf tobacco is warmed to, for example, about 40 to 70°C.

<Tobacco Raw Material of the Invention>

[0056] The tobacco raw material of the invention has a ratio, which is expressed as "water-extracted material weight/hexane-extracted material weight" (or "W/H ratio"), between the weight of an extracted material obtained by extraction using n-hexane as a solvent (also referred to below as "hexane-extracted material") and the weight of an extracted material obtained by extraction using water as a solvent (also referred to below as "water-extracted material"), of 0 or more and 1.5 or less.

[0057] This tobacco raw material, by having such a W/H ratio, is a tobacco raw material with a good smoking flavor. At a W/H ratio greater than 1.5, i.e., at a content of water-extracted material that is too large, the tobacco raw material has a smoking flavor that is adulterated.

[0058] In another embodiment, the W/H ratio may have a value of 0.3 or more and 0.6 or less.

[0059] The sum of the weight of the hexane-extracted material and the water-extracted material in the tobacco raw material of the invention may be from 5 to 17 wt %, based on the weight of the tobacco raw material prior to extraction. In another embodiment, this may be from 5 to 14 wt %.

[0060] Production of tobacco raw material having such a W/H ratio may involve the use of, for example, the above-described tobacco raw material production method of the invention.

[0061] The following method may be used as the method of measuring, within the tobacco raw material, the amount of extracted material that can be obtained by extraction using n-hexane as the solvent and the amount of extracted material that can be obtained by subsequent extraction using water as the solvent. Specifically, an accelerated solvent extraction system (ASE 200, from Dionex) is used and a sample-filled cell is prepared by placing 1.0 g/cell of tobacco raw material in a pressure-resistant extraction cell made of metal and having a volume of 22 mL. The cell is mounted in the accelerated solvent extraction system, and extraction is carried out under the conditions shown in the table below.

Table 1

Solvent	PREHAT (min)	STATIC (min)	FLUSH% /CYCLES	PURGE (sec)	PRESSURE (psi)	Temperature (°C)	Number of vials
n-Hexane	0	10	150 % / 3cycles	90	2000	70	1
Ultrapure water	0	10	150 %/3cycles	90	2000	100	3

[0062] In the above extraction, the extract is transferred to a specialized glass vial for the solvent extraction system, this extract being collected following extraction and used to measure the amount of extraction (see below for the method). A single cycle in solvent extraction consists of pressurized filling of the solvent, warming, standing at rest, purging (transfer of solvent to the vial) and, once again, pressurized filling of the solvent. In the case of n-hexane, three cycles are repeated and the total amount of liquid obtained from one cell is collected in one vial. When water extraction is successively carried out, a total of nine cycles are repeated and the total amount of liquid obtained from three cycles is collected in one vial. Hence, a total of three vials can be obtained from one cell.

[0063] Measurement of the amount of n-hexane extraction is carried out as follows.

[0064] A 1 vial/1 cell n-hexane extract is transferred to a pre-weighed 100 mL round-bottomed flask using n-hexane, ethyl acetate and ethanol, in addition to which the vial is rinsed out, thereby collecting all of the extracted material in the flask. The round-bottomed flask is placed on a rotary evaporator and the contents are vacuum-concentrated, then dried at normal temperature and reduced pressure for 3 hours with a vacuum oil pump. Concentration is carried out using a rotary evaporator equipped with trapping balls, and the liquid temperature of the water tank is set to 25°C or less. The weight of the flask after drying is measured, and the difference is treated as the amount of n-hexane extracted material.

[0065] Measurement of the water-extracted material is carried out as follows.

[0066] A 3 vial/1 cell water extract is transferred, using ultrapure water, to a pre-weighed 500 mL interchangeable ground joint round-bottomed flask for freezing, in addition to which the vial is rinsed out so that all of the extracted material is recovered in the flask. The round-bottomed flask and its contents are frozen in the pre-freezing tank of a freeze-drying system and then connected to the freeze-drying system and dried. The weight of the flask after drying is measured, and the difference is treated as the yield of water-extracted material.

EXAMPLES

[0067] The invention is described more fully by way of examples. However, the invention, insofar as it does not depart from the spirit and scope thereof, is not limited to the following examples.

[0068] The invention is described more fully below by way of examples. However, the invention, insofar as it does not depart from the spirit and scope thereof, is not limited to the following examples.

<Example 1>

[0069] Leaf tobacco material (a domestically produced burley variety) was ground using a grinding mill.

[0070] The grinding mill used was an SK-M10 Sample Mill from Kyoritsu Riko K.K. The ground material was then immediately applied to a sieve, giving a ground leaf tobacco material having a particle size of from 0.425 mm to 1.18 mm. Next, 2 g of this ground leaf tobacco material was precisely weighed and filled into a metal cell.

[0071] The voids that formed during filling were filled using stainless steel beads. The cell used was a 22 mL extraction cell made by Dionex, and the beads were 2.2 mm diameter beads from Central Scientific Commerce, Inc. Solvent was delivered to this cell, and an accelerated solvent extraction system was used to carry out extraction on the ground leaf tobacco material.

[0072] The extractor used was an ASE-200 accelerated solvent extraction system from Dionex.

[0073] Solvents having a dielectric constant of 1 or more and less than 25, such as n-hexane (dielectric constant, 2) and ethanol (dielectric constant, 24), were used for extraction.

[0074] In extraction with the above extractor using these solvents, the sequence consisting of first filling the cell with solvent, warming the cell, letting it stand at rest, then forcing out the solvent within the cell was repeated three times. The temperature during warming and the pressure conditions during standing at rest were as follows for the respective solvents.

n-Hexane: 70°C, 2,000 psi

Ethanol: 80°C, 2,000 psi

[0075] The volume of extract obtained with these solvents was about 40 mL. The extract was concentrated using an evaporator and, using the same solvent as that used during extraction, was brought to a constant volume of 6.25 mL in a suspended state. Extraction with water (dielectric constant, 80) as the solvent was then carried out. At the time of this extraction, the same sequence was followed as for extraction with a solvent having a dielectric constant of 1 or more and less than 25, but the temperature during warming and the pressure during standing at rest were set to 100°C and 2,000 psi. Also, the three cycles carried out during extraction with a solvent having a dielectric constant of 1 or more and less than 25 was changed to nine cycles during extraction with water. The total volume of the extract was about 120 mL.

[0076] The extraction residue remaining in the cell following extraction was completely recovered and pre-frozen, then furnished to freeze-drying. An FD-81 Freeze Dryer from EYELA was used for pre-freezing and freeze-drying.

[0077] The entire amount of extract prepared as described above with low-polarity solvent was uniformly re-applied to the dried extraction residue, thereby giving a tobacco raw material according to the invention.

[0078] The solvents used in the respective examples were as follows.

Example in which n-hexane was used: Example 1

Example in which ethanol was used: Example 2

[0079] An example in which a tobacco raw material obtained using methanol (dielectric constant, 33) as the solvent in the first extraction step was treated as Comparative Example 1.

[0080] The ammonia contents were measured for the tobacco raw materials in Examples 1 and 2 and Comparative Example 1. Measurement of the ammonia content in the tobacco raw material was carried out by the following procedure.

<Ammonia Analysis>

[0081] Untreated raw material (180 mg) or treated raw material (having the weight obtained when 180 mg of untreated raw material has been treated) was immersed and extracted for 30 minutes in 5 mL of pure water. The extract was then filtered with a 10 KDa ultrafiltration membrane. The filtrate was analyzed with a capillary electrophoresis chromatograph (Agilent Technologies). The absorbance was measured at a wavelength of 280 nm and the ammonia was quantitatively determined as ammonium ions using an aqueous solution of ammonium chloride diluted to a suitable concentration as the standard solution.

[0082] The quantitatively determined values converted to amounts based on the raw material weight are shown in Table 2. For the treated raw material, conversion was carried out using the raw material weight when untreated (in order to eliminate the influence arising from the weight loss disparity due to treatment).

Table 2

	Ctrl	n-Hexane	Ethanol	Methanol
Domestic burley tobacco	3066.9	0.0	483.8	1564.6
Brazilian flue-cured tobacco	365.3	0.0	-	-

Note: In the table, "-" means not measured. Numerical values other than the controls in the table are values obtained by converting quantitatively measured values to amounts based on the weight of the raw material (ppm (W.B.)).

[0083] It is apparent from the results in Table 2 above that, in cases where a solvent having a dielectric constant of 1 or more and less than 25 was used in the first extraction step and, after passing through the second extraction step, the extract obtained in the first extraction step was re-applied to the extraction residue, compared with cases in which a solvent having a dielectric constant of 25 or more was used in the first extraction step, a large amount of ammonia was selectively removed.

<Sensory Evaluation>

[0084] With regard to tobacco raw material obtained by the production method of the invention, sensory evaluations of the sort described below were carried out in Example 3, where the tobacco raw material was produced under the same conditions as in Example 1; in Comparative Example 2, where it was obtained by carrying out only the operation of re-applying the extract obtained from the first extraction step (no second extraction step); in Example 4 where it was obtained by, aside from using a Brazilian flue-cured variety as the leaf tobacco material, carrying out the same operations as in Example 1; in Comparative Example 3, where it was obtained by carrying out only the operation of re-applying the extract obtained from the first extraction step in Example 4 (no second extraction step); in Example 5, where it was obtained from the same leaf tobacco material and operations as in Example 3; and in Comparative Example 4, where it was obtained from the same leaf tobacco material and operations as in Comparative Example 2. The following operations were carried out during sensory evaluation.

(Examples 3 and 4, and Comparative Examples 2 and 3)

[0085] Untreated material that had been ground and sieved (particle size, from 0.425 mm to 1.18 mm) and the respective tobacco raw materials (Examples 3 and 4, and Comparative Examples 2 and 3) were filled into quartz tubes (9 mm (o.d.) x 7 mm (i.d.) x 55 mm (L)).

[0086] The fill weight was set to 180 mg for untreated raw material and in cases where no extracted material was removed. In the case of a treated raw material that included an extracted material removing step, taking into account the amount of weight loss due to not re-applying the water-extracted portion, the fill weight was set to 109 mg (the weight of the raw material obtained when 180 mg of raw material was treated). The sample was kept from scattering upon inhalation during evaluation by stopping both ends of the quartz tube with stainless steel mesh. These quartz tubes were heated to 60°C, inhalation was carried out from one end of the tube, and the results compared with untreated raw material were examined.

<Sensory Evaluation Method>

[0087] The sensory evaluation (change in smoking satisfaction) criteria are shown below. The degree of change relative to untreated raw material was rated using the 7-step scale expressed in terms of the language shown below. The number of subjects for each evaluation was set at ten people. The least significant difference (LSD) was used to determine the significance of differences in the sensory evaluation results due to the treatment method. The results are shown in FIGS. 1 and 2.

- (1) very strong: +3
- (2) strong: +2
- (3) somewhat strong: +1
- (4) no change: ± 0
- (5) somewhat weak: -1
- (6) weak: -2
- (7) very weak: -3

[0088] From the results in FIGS. 1 and 2, in tests by the least significant difference method, there was a 95% significant difference between the examples of the invention and the comparative examples, and so effects due to the production method of the invention were observed.

[0089] From this, regardless of differences in the variety of the leaf tobacco material, such as domestic burley tobacco and Brazilian flue-cured tobacco, the tobacco raw material obtained by the production method of the invention was found to achieve an excellent smoking flavor.

(Example 5 and Comparative Example 4)

[0090] Untreated material that had been ground and sieved (particle size, from 0.425 mm to 1.18 mm) and the respective tobacco raw materials (Example 5 and Comparative Example 4) were filled into plastic tubes (9 mm (o.d.) x 7 mm (i.d.) x 110 mm (L)).

[0091] The fill weight was set to 360 mg for untreated raw material and in cases where no extracted material was removed. In the case of treated raw material that included an extracted material removing step, taking into account the amount of weight loss due to not re-applying the water-extracted portion, the fill weight was set to 218 mg (the weight of the raw material obtained when 360 mg of raw material was treated). The sample was kept from scattering upon inhalation during evaluation by stopping both ends of the plastic tube with nonwoven fabric mesh. Using these plastic tubes, inhalation was carried out from one end of the tube at room temperature, and the results in compared with untreated raw material were examined. The results are shown in FIG. 3.

[0092] Evaluations on Example 5 and Comparative Example 4 were carried out in the same way as for Examples 3 and 4 and Comparative Examples 2 and 3. As a result, in tests by the least significant difference method, there was a 95% significant difference between Example 5 and Comparative Example 4. Hence, effects due to the production method of the invention were observed.

<Example 6>

[0093] Aside from using tetrahydrofuran (dielectric constant: 8) as the solvent for extraction in the first extraction step, a tobacco raw material was produced by the same procedure as in Example 1.

[0094] The tobacco raw material of Example 6 (104.94 mg) and the tobacco raw material produced in Comparative Example 1 (using methanol as the solvent for extraction in the first extraction step; 134.55 mg) were subjected to sensory evaluations (change in smoking satisfaction) by the same method as in Examples 3 and 4 and Comparative Examples 2 and 3. The results are shown in FIG. 4.

<Comparative Examples 5 and 6>

[0095] Leaf tobacco material of a domestic burley variety was subjected to grinding treatment in the same way as in Example 1, and the first extraction step was not carried out; only the second extraction step was carried out (only extraction with water was carried out). The extract obtained by this second extraction step was re-applied to the extraction residue, thereby producing a tobacco raw material for Comparative Example 5.

[0096] Also, leaf tobacco material of a domestic burley variety was subjected to grinding treatment in the same way as in Example 1 and the second extraction step (extraction with water) was initially carried out, following which the first extraction step (extraction with n-hexane) was carried out. Next, only the extract obtained by carrying out extraction with water was re-applied to the extraction residue, thereby producing a tobacco raw material for Comparative Example 6.

[0097] Sensory evaluations (change in smoking satisfaction) on the tobacco raw materials obtained in Comparative Examples 5 and 6 were carried out by the same method as described above. The weight of the tobacco raw material from Comparative Example 5 used in the sensory evaluation was 180 mg, and the weight of the tobacco raw material from Comparative Example 6 used in sensory evaluation was 172.9 mg. The results are shown in FIG. 5.

<Comparative Example 7>

[0098] Leaf tobacco material of a domestic burley variety was subjected to grinding treatment in the same way as in Example 1, the second extraction step was carried out first (i.e., extraction with water was carried out first), following which the first extraction step was carried out (i.e., extraction with n-hexane was subsequently carried out), and the extract thus obtained (n-hexane extract only) was re-applied to the extraction residue, thereby producing a tobacco raw material for Comparative Example 7.

[0099] Sensory evaluation (change in smoking satisfaction) was carried out for Comparative Example 7 by the same method as described above for the tobacco raw materials obtained in Examples 3 and 4 and Comparative Examples 2 and 3. The weight of the tobacco raw material from Comparative Example 7 used in the sensory evaluation was 83.86 g. The results are shown in FIG. 6.

<W/H Ratios of Tobacco Raw Materials>

[0100] The W/H ratios of the respective tobacco raw materials obtained in Example 1 (in which n-hexane was used in the first extraction step), Example 2 (in which ethanol was used in the first extraction step), Example 6 (in which tetrahydrofuran was used in the first extraction step), and Comparative Example 1 (in which methanol was used in the first extraction step) were measured. As a reference example, the W/H ratio for untreated tobacco raw material (burley variety) was also measured. The measurement method is as described above.

[0101] Filling of the samples used in extraction was carried out as follows.

[0102] Additive-free filter paper was set in a pressure-resistant metal extraction cell, and the cell was filled with 10 g of prewashed and weighed stainless steel balls. Next, the tobacco raw materials obtained in Examples 1, 2 and 6, Comparative Example 1 and the reference example were weighed out, with an even balance, in respective amounts of 1.0 g/cell onto powder paper and, using a suitable aid such as a powder funnel, the weighed samples were placed in the already prepared 22 mL cells in which the filter paper and stainless steel balls had been set; sample remaining on the powder paper was wiped several times with the stainless steel balls and added to the cell. In addition, voids were eliminated by packing the cell with stainless steel balls until it was filled to the top. Once the cell was completely filled, the top side was closed and the cell was tapped on a desk to bury any gaps and even out the contents, after which the empty space was filled with stainless steel balls.

[0103] Next, under the conditions shown in Table 1, extraction with n-hexane and water (MilliQ water) was carried out for 3 cycles and 9 cycles, respectively, and the weight of extracted material obtained by extraction with each of these solvents was determined. Measurement was carried out twice for each of the tobacco raw materials, and the weight of the extracted material in each case was indicated as an average of the resulting measurements.

[0104] The W/H ratio and the ratio of the extracted material weight relative to the tobacco raw material before the extraction treatment were determined from the resulting extracted material weights. The results are shown in Table 3.

Table 3

	Example 1	Example 2	Example 6	Comparative Example 1	Reference Example
n-Hexane extracted material weight ratio (%)	7.23	10.38	8.60	7.80	6.75

(continued)

	Example 1	Example 2	Example 6	Comparative Example 1	Reference Example
Water-extracted material weight ratio (%)	2.55	5.43	2.70	16.45	39.55
W/H ratio	0.4	0.5	0.3	2.1	5.9
Extracted material weight/Tobacco raw material weight (%)	9.8	15.8	11.3	24.2	46.3

[0105] The apparatuses, solvents and products used were as follows.

<Apparatuses Used>

- [0106]** Accelerated solvent extraction system (ASE 200, from Dionex)
[0107] Even balance (PM 2500, from Mettler)
[0108] Freeze dryer with pre-freezer (FD-81, from Tokyo Rikakikai Co., Ltd.)
[0109] Vacuum oil pump (ULVAC GCD-051X, from ULVAC KIKO Inc.)

<Solvents Used>

- [0110]** n-Hexane (high-performance liquid chromatography (HPLC) grade, from Wako Pure Chemical Industries, Ltd.)
[0111] Water (MilliQ water from Nihon Millipore K.K., obtained with MQ Gradient water purification system)
[0112] Ethyl acetate (high-performance liquid chromatography (HPLC) grade, from Wako Pure Chemical Industries, Ltd.)
[0113] Ethanol (high-performance liquid chromatography (HPLC) grade, from Wako Pure Chemical Industries, Ltd.)
Note: The ethyl acetate and ethanol were used when transferring n-hexane extracts to round-bottomed flasks.

<Products Used>

- [0114]** Powder paper: 10 cm square sheets
[0115] Interchangeable ground joint round-bottom flasks: 100 mL (Sibata Scientific Technology, Ltd.)
[0116] Interchangeable ground joint round-bottom flasks for freeze-drying: 500 mL (EYELA K.K.)
[0117] Pressure-resistant metal extraction cells, 22 mL (Dionex)
[0118] Additive-free filter paper for extraction cells (Dionex)
[0119] Stainless steel balls, particle size, 2.3 mm (Central Scientific Commerce, Inc.)

Claims

1. A tobacco raw material having a ratio, which is expressed as "water-extracted material weight/hexane-extracted material weight," between the weight of an extracted material obtained by extraction using n-hexane as a solvent and the weight of an extracted material obtained following the extraction by further extraction using water as a solvent, of 0 or more and 1.5 or less.
2. The tobacco raw material according to claim 1, wherein the sum of the weight of an extracted material obtained by extraction using n-hexane as the solvent and the weight of an extracted material obtained following the extraction by further extraction using water as the solvent is from 5 to 17 wt % of the weight of the tobacco raw material before the extractions.
3. A method of producing a tobacco raw material, comprising: a first extraction step of extracting a leaf tobacco material using a solvent having a dielectric constant of 1 or more and less than 25 to obtain an extract and an extraction residue; a second extraction step of, after the first extraction step, extracting the extraction residue obtained in the first extraction step using a solvent having a dielectric constant of 25 or more to obtain an extraction residue; and the step of re-applying the extract obtained in the first extraction step to the extraction residue obtained in the second extraction step so as to form a leaf tobacco raw material.

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4. The production method according to claim 3, wherein the solvent used in the first extraction step is a solvent having a dielectric constant of from 1 to 20 and the solvent used in the second extraction step is water.
5. The production method according to claim 3 or 4, wherein the solvent used in the first extraction step is one or more selected from among n-hexane, tetrahydrofuran and ethanol.
6. A tobacco raw material obtained by the production method according to any one of claims 3 to 5.
7. A tobacco product which uses the tobacco raw material according to claim 1, 2 or 6.

FIG. 1

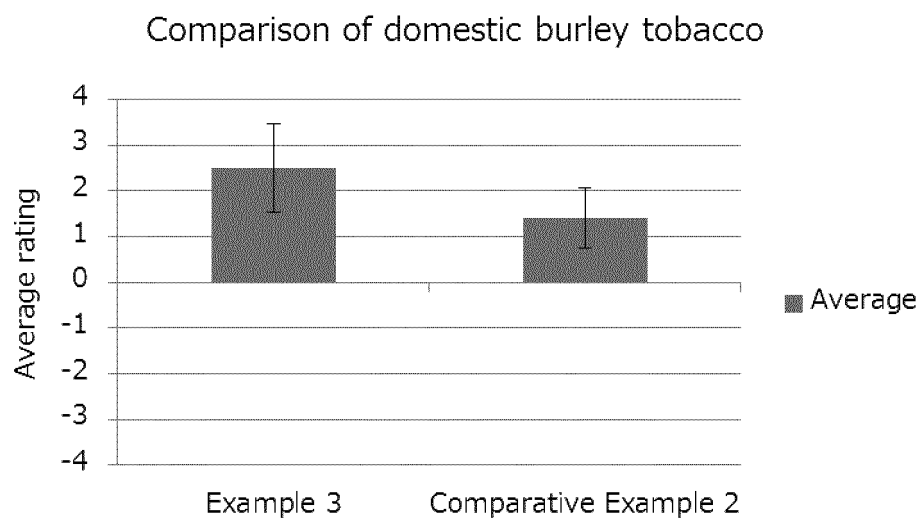
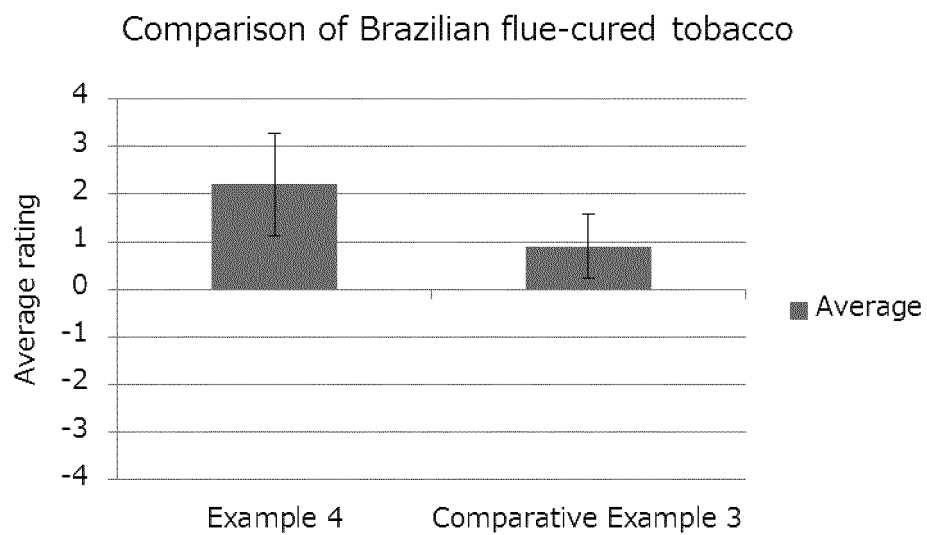


FIG. 2



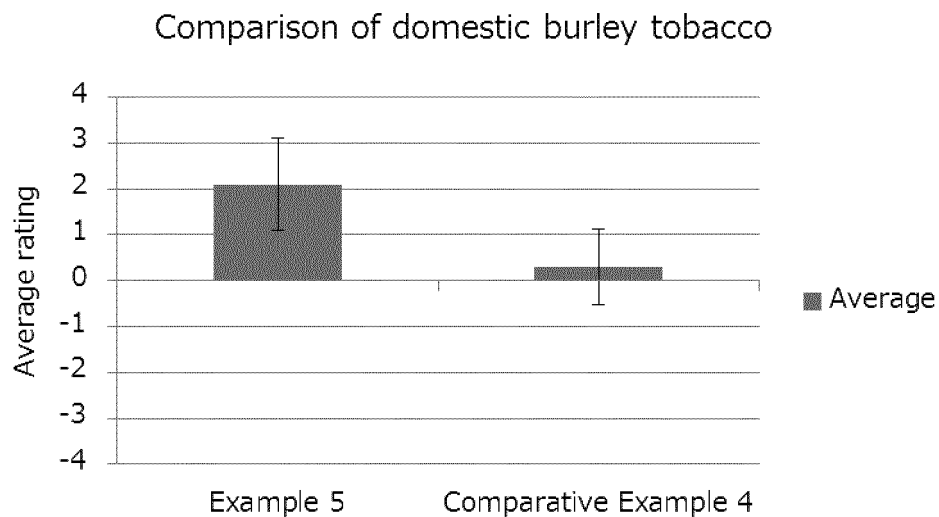
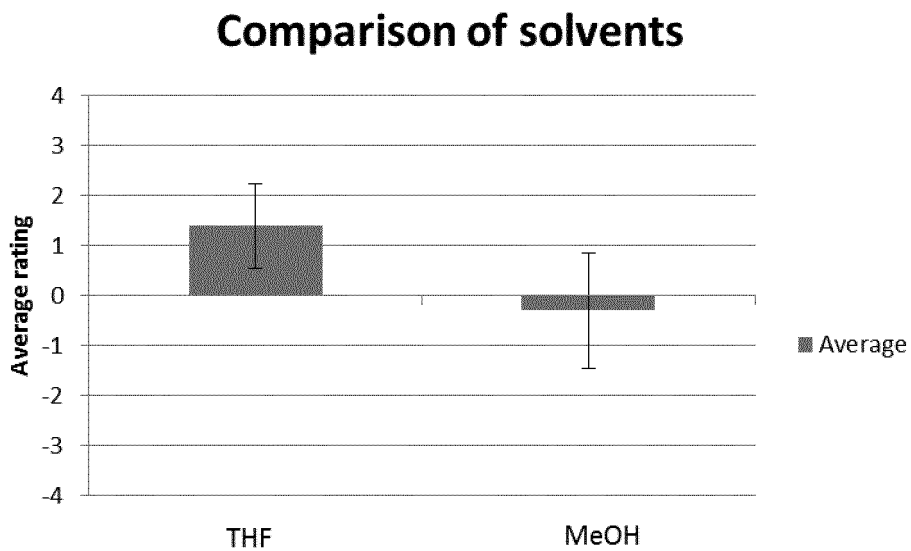


FIG. 4



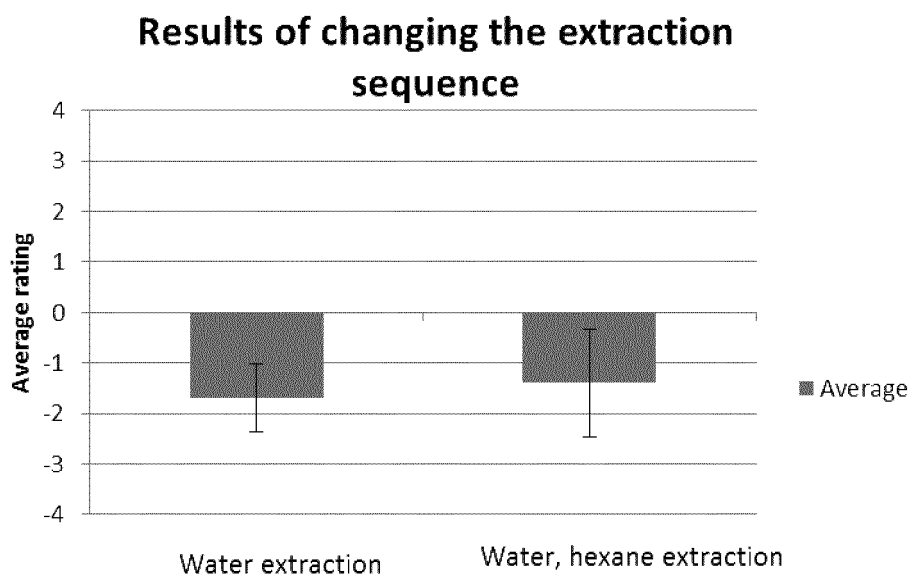
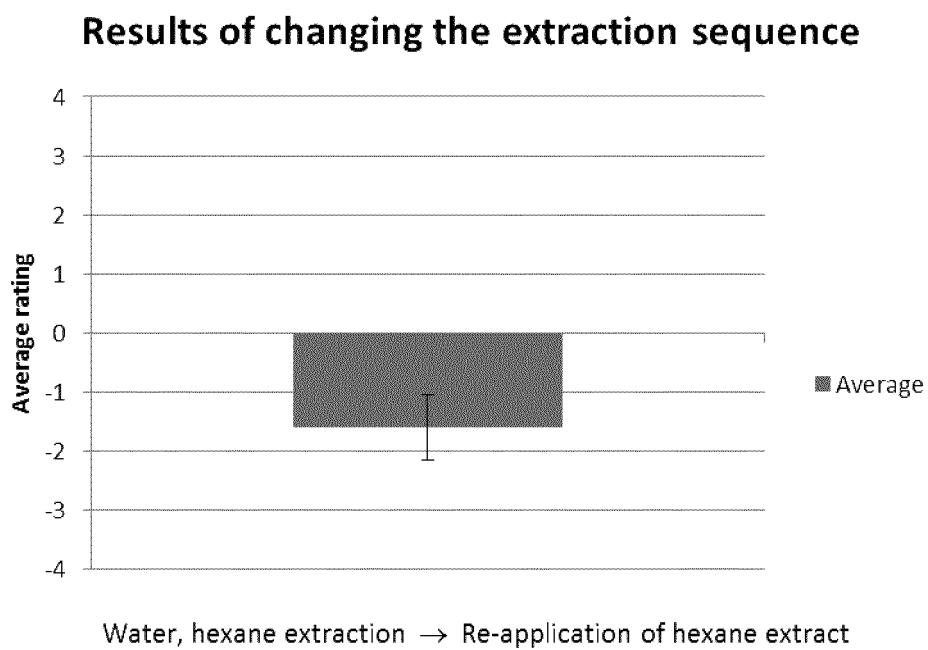


FIG. 6



INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP2014/072244

A. CLASSIFICATION OF SUBJECT MATTER

A24B15/26(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)

A24B15/26

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 1996-2014

Kokai Jitsuyo Shinan Koho 1971-2014 Toroku Jitsuyo Shinan Koho 1994-2014

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.
X A	JP 2011-10656 A (Novozymes A/S), 20 January 2011 (20.01.2011), paragraphs [0015] to [0032], [0098] to [0101] & US 6298859 B1 & WO 2000/002464 A1	1-2, 7 3-6
A	JP 52-1098 A (AMF Inc.), 06 January 1977 (06.01.1977), & US 4200113 A & GB 1532083 A & DE 2613569 A & FR 2314677 A & CH 603082 A	1-7



Further documents are listed in the continuation of Box C.



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Date of the actual completion of the international search

21 November, 2014 (21.11.14)

Date of mailing of the international search report

02 December, 2014 (02.12.14)

Name and mailing address of the ISA/
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