

[TITLE OF THE INVENTION]

Tobacco product containing purified natural (L)-menthol

[TECHNICAL FIELD]

[0001]

The present invention relates to a tobacco product containing purified natural (L)-menthol, and more particularly, to a tobacco product containing purified natural (L)-menthol for smoking with unpleasant taste eliminated and a refreshing feeling added by addition of activated carbon to crude menthol.

[BACKGROUND ART]

[0002]

Menthol is an organic compound, a kind of a cyclic monoterpene alcohol, and is a mint-smelling volatile colorless crystal. Some menthol exist in diastereomer and enantiomer, and natural (L)-menthol is widely used not only for toothpastes, confectionery, mouth refrigerants, etc. but also for pharmaceuticals, due to its partial vasodilator action and skin irritant action etc.

[0003]

Menthol has been conventionally used as flavor of tobacco. When raw leaf tobacco is burned, it has a peculiar stimulus, off smell such as green smelling or fishy smelling, unpleasant taste such as astringency and bitterness, addition to its peculiar smoking flavor and taste. Thus, licorice, sugar, honey, rum, fruit juice, vanilla, etc. have been empirically used to soften off smell and unpleasant taste.

[0004]

People's preference in tobacco taste has recently become diverse and products with refreshing and rich smoking flavor and taste have been increasingly popular. Among ingredients contained in a so called modifier for tobacco flavor and taste, particularly natural (L)-menthol plays a large role.

[0005]

Most of commercially-available tobacco products are

cigarettes (a paper-wrapping tobacco), but other products are known, such as a cigar wrapped by not a paper but a tobacco leaf, a snuff product for people to enjoy its aroma without lighting it, and a chewing tobacco product for people to enjoy its taste by directly chewing a mixture of tobacco leaves. For example, a snuff product, so-called SNUS, which people enjoy by putting a small packet containing tobacco powder between the gum and the upper lip, has been widely used in Northern Europe and chewing tobacco products are very popular among baseball players. These tobacco products also contain natural (L)-menthol as a modifier for smoking flavor and taste.

[0006]

As a menthol, natural (L)-menthol and synthetic (L)-menthol are used. Synthetic (L)-menthol is obtained in higher purity than natural (L)-menthol, but is inferior to natural (L)-menthol in terms of flavor, since it does not contain flavor ingredient impurities derived from raw mint oil. Natural (L)-menthol is extracted from a plant, such as a mint leaf, and thus contains impurities. The content of this impurities affects quality and flavor of the natural (L)-menthol. For that reason, methods of purifying natural (L)-menthol have been considered from various aspects. One of them includes a method for contacting crude menthol with supercritical carbon dioxide, and for fractionating, extracting, and removing impurities (for example, see patent document 1). This method can remove ingredients such as unpleasant smell (taste), bitter components, weed-like smell, substances with strongly unfavorable taste and resinoid substances. However, this method utilizes supercritical carbon dioxide and thus requires expensive devices. Accordingly, the method is hard to industrialize.

[0007]

One of other methods includes a method of dissolving crude menthol in acetonitrile and then cooling the solution to crystallize the menthol (for example, see patent document 2). This method yields natural (L)-menthol with more than 99% of

chemical purity and more than 99% of optical purity, but working environment to implement the method is limited since it requires that menthol should be dissolved in an organic solvent (acetonitrile).

[0008]

As described above, even if using the art indicated by the patent document 1 or 2, it would be difficult to easily and inexpensively provide a purified natural (L)-menthol with a refreshing feeling and mint-like flavor and a tobacco product containing the purified natural (L)-menthol .

[PRIOR ART DOCUMENTS]

[PATENT LITERATURE]

[0009]

[Patent document 1] Japanese Patent No. 3935987

[Patent document 2] Japanese Patent No. 5014846

[SUMMARY OF THE INVENTION]

[PROBLEMS TO BE SOLVED BY THE INVENTION]

[0010]

The present invention aims to solve the above-described problems of the prior art and relates to a tobacco product containing purified natural (L)-menthol with unpleasant taste eliminated and having a refreshing feeling and mint-like flavor derived from natural products.

[MEANS TO SOLVE THE PROBLEMS]

[0011]

The invention according to claim 1 relates to a tobacco product containing a purified natural (L)-menthol with a refreshing feeling and mint-like flavor which chroma measured by following methods (1) to (3) is equal to or less than 1.
(1) a step of subjecting the purified natural (L)-menthol in heated and melted state to distillation under reduced pressure to obtain a residue distilled under reduced pressure.
(2) a step of adding ethanol to the residue obtained at the step

(1) to prepare an ethanol solution.

(3) a step of calculating chroma with a color difference meter pursuant to ISO 7724 (JIS Z8722) using the ethanol solution obtained at the step (2) as a color difference measurement sample.

[0012]

The invention according to claim 2 relates to the tobacco product of claim 1, wherein the above-mentioned chroma is within a range of 0.4 to 0.6.

[0013]

The invention according to claim 3 relates to the tobacco product of claim 1 or 2, wherein the invention is a cigarette comprising a wrapping part consisting of shredded tobacco and a tobacco wrapping paper to wrap the shredded tobacco, a filter part consisting of a filter and a filter wrapping paper to wrap the filter, a tip paper for connecting the above-mentioned wrapping part and the above-mentioned filter part by wrapping at least a part of the above-mentioned wrapping part and the above-mentioned filter part.

[0014]

The invention according to claim 4 relates to the tobacco product of claim 3 wherein the above-mentioned purified natural (L)-menthol is contained in the above-mentioned shredded tobacco.

[0015]

The invention according to claim 5 relates to the tobacco product of claim 1 wherein the residue distilled under reduced pressure of the above-mentioned purified natural (L)-menthol is less than 0.1 % by weight.

[EFFECT OF THE INVENTION]

[0016]

According to the invention of claim 1, measuring an ethanol solution prepared by adding ethanol to the residue after the distillation under reduced pressure of purified natural (L)-menthol using a color difference meter pursuant to ISO 7724 enables confirmation of the existence of impurities which has

big influence on preference. In such a case, it is possible to prepare the purified natural (L)-menthol having a refreshing feeling and mint-like flavor from the purified natural (L)-menthol which chroma is equal to or less than 1. The above-mentioned purified natural (L)-menthol is included in a tobacco product so as to have a refreshing feeling and mint-like flavor.

[0017]

According to the invention of claim 2, when the chroma of the purified natural (L)-menthol evaluated by a color difference meter in a similar method as described in claim 1 is within a range of 0.4 to 0.6, a tobacco product containing the purified natural (L)-menthol having a refreshing feeling and mint-like flavor stronger than those of the tobacco product containing the purified natural (L)-menthol of claim 1 could be prepared (see Table 1).

[0018]

According to the invention of claim 3, the purified natural (L)-menthol of claim 1 is contained in a part of the cigarette so that it can have a refreshing feeling and mint-like flavor.

[0019]

According to the invention of claim 4, purified natural (L)-menthol of claim 1 is contained in the above-mentioned shredded tobacco, and thus it is possible to easily transfer flavor of the purified natural (L)-menthol to smoke during smoking, thereby obtaining a cigarette having a refreshing feeling and mint-like flavor.

[0020]

According to the invention of claim 5, since the residue distilled under reduced pressure of the above-mentioned purified natural (L)-menthol is less than 0.1 % by weight, and thus, it is possible to obtain a purified natural (L)-menthol having a refreshing feeling and mint-like flavor with little impurities by a relatively easy step of contacting the purified natural (L)-menthol with activated carbon, and accordingly to use the obtained purified natural (L)-menthol to provide a

tobacco product having a refreshing feeling and mint-like flavor.

[BRIEF DESCRIPTION OF THE DRAWINGS]

[0021]

[Fig. 1] Fig.1 is a schematic view of a cigarette having one kind of filter.

[Fig. 2] Fig.2 is a schematic view of a cigarette having two kinds of filters.

[Fig. 3] Fig.3 (A) is a cross sectional view of a cigarette having a string containing purified natural (L)-menthol in the center of the filter and (B) is a sectional view of A-A.

[Fig.4] Fig.4 (A) is a cross sectional view of a cigarette having a breakable capsule containing purified natural (L)-menthol in the cavity inside the filter and (B) is a cross sectional view of the cigarette when the capsule in the cavity is broken.

[Fig.5] Fig.5 (A) is a schematic view of a cigar and (B) is a cross sectional view of the cigar.

[Fig.6] Fig.6 (A) is a schematic view of a pipe for sucking flavor and taste and (B) is a cross sectional view of the pipe.

[Fig. 7] Fig.7 is a schematic view of a wet smokeless tobacco product, (B) is a cross sectional view of A-A, and (C) shows a method of using the wet smokeless tobacco product.

[Fig. 8] Fig.8 shows a method of using a chewing tobacco product.

[DETAILED DESCRIPTION OF THE INVENTION]

[0022]

The tobacco product containing the purified natural (L)-menthol according to the present invention and methods of manufacturing and evaluating the same will be set forth below.

[0023]

First, the tobacco product containing the purified natural (L)-menthol of the present invention is explained with reference to the figures as below.

[0024]

1. Tobacco product containing purified natural (L)-menthol

The tobacco product of the present invention contains the purified natural (L)-menthol having a refreshing feeling and mint-like flavor, wherein chroma measured by following evaluation methods is equal to or less than 1, preferably in a range of 0.4 to 0.6. A suitable cigarette to contain the purified natural (L)-menthol of the present invention is explained with reference to Fig.1 to Fig.4.

[0025]

Fig. 1 is a schematic view of a cigarette having one kind of filter.

A cigarette comprises a wrapping part (2) consisting of shredded tobacco (6) and a tobacco wrapping paper (5) to wrap the shredded tobacco (6), a filter part (1) consisting of a filter (7) and a filter wrapping paper (3) to wrap the filter (7), and a tip paper (4) for connecting the above-mentioned wrapping part (2) and the above-mentioned filter part (1) by wrapping at least a part of the above-mentioned wrapping part (2) and the above-mentioned filter part (1).

The shredded tobacco (6) is made by blending shredded tobacco leaves (flue-cured tobacco, Burley or the like), shredded stem, shredded sheet tobacco, and expanded tobacco. The tobacco wrapping paper (5) includes flax and/or wood pulp as a raw material, may include calcium carbonate to reduce the smell of burning paper and to adjust the burning rate of the paper to that of the shredded tobacco, and may contain citrate as a combustion improver. The tobacco wrapping paper (5) with fine pores provided to adjust the amount of air inflows may be used. The filter (7) is generally a cellulose acetate filter made from cellulose acetate fiber, but neo sheet, rayon, paper, etc. may be used. Generally, but not limited to, a paper is used as a material of the filter wrapping paper (3). Generally, wood pulp, calcium carbonate, and titanium oxide are used as a material of the tip paper (4), but other materials with fine pores provided to adjust the amount of air inflows may be used.

In such a case, purified natural (L)-menthol can be

contained in the shredded tobacco (6) and/or the filter (7).
[0026]

Fig. 2 is a schematic view of a cigarette having two kinds of filters.

The cigarette has the substantially same structure as the cigarette having one kind of filter of Fig. 1, but a filter part (1) consists of two kinds of filters; a cellulose acetate filter (9) and a charcoal filter (10). Filter part (1) further includes an inner filter wrapping paper (8) to wrap the cellulose acetate filter (9) and the charcoal filter (10), and a filter wrapping paper (3) to join the cellulose acetate filter (9) and the charcoal filter (10).

The charcoal filter (10) has activated carbon incorporated into a double structure or a triple structure made by combining cellulose acetate fiber, neo sheet, rayon, paper, etc. The inner filter wrapping paper (8) is generally made from, but not limited to, a paper, as well as the filter wrapping paper (3).

In such a case, the purified natural (L)-menthol can be contained in the shredded tobacco (6) and/or, the cellulose acetate filter (9), and/or the charcoal filter (10), as well as the cigarette having one kind of filter.

[0027]

Fig. 3(A) is a cross sectional view of a cigarette having a string containing purified natural (L)-menthol in the center of the filter and (B) is a sectional view of A-A. The cigarette has the substantially same structure as the cigarette having one kind of filter of Fig. 1, but has a string (11) located substantially in the central part of the filter.

The string (11) is generally made of, but not limited to, cotton and used with the purified natural (L)-menthol applied thereon.

In such a case, the purified natural (L)-menthol can be contained in the filter (7) and/or shredded tobacco (6) in addition to the string (11).

[0028]

Fig. 4(A) is a cross sectional view of a cigarette having

a breakable capsule containing purified natural (L)-menthol in the cavity inside the filter and (B) is a cross sectional view of the cigarette when the capsule in the cavity is broken. The cigarette has the substantially same structure as the cigarette having one kind of filter of Fig. 1, but has a breakable capsule (12) in the cavity inside the filter.

The breakable capsule (12) is filled up with the purified natural (L)-menthol and is broken by pressure applied by fingers before or during smoking to release the purified natural (L)-menthol in the breakable capsule (12) to the filter (7). Shell of the breakable capsule (12) is made of gelatin etc. and the capsule contains 1-10 mg of purified natural (L)-menthol.

In such a case, the purified natural (L)-menthol can be contained in the filter (7) and/or shredded tobacco (6) in addition to the breakable capsule (12). Although not illustrated in Fig. 4, the cigarette could include a string in the center of the filter and the purified natural (L)-menthol can be contained in that string, as shown in Fig. 3.

[0029]

As for a cigarette, purified natural (L)-menthol may be applied inside a cigarette packaging container. The method of application may include, but not limited to, inserting aluminum-laminated paper coated with the purified natural (L)-menthol into the cigarette packaging container, and directly applying the purified natural (L)-menthol inside the packaging container.

Applying the purified natural (L)-menthol inside the cigarette packaging container enables flavor of the purified natural (L)-menthol to be transferred to the shredded tobacco (6) and/or the filter (7) of the cigarette during storage of the cigarette.

[0030]

Cigar tobaccos suitable for containing the purified natural (L)-menthol of the present invention will be explained with reference to Fig. 5.

[0031]

Fig. 5 is (A) a schematic view and (B) a cross sectional view of a cigar tobacco. The cigar tobacco includes a filler (13) containing tobacco leaves or shredded tobacco, a binder (14) optionally wrapping the filler with tobacco leaves, and a wrapper (15) wrapping the filler or the binder with tobacco leaves.

In such a case, the purified natural (L)-menthol can be contained in the tobacco leaves or shredded tobacco.

[0032]

A snuff product suitable for containing the purified natural (L)-menthol of the present invention will be explained with reference to Figs. 6 and 7.

[0033]

The snuff product includes a snuff, a pipe for sucking flavor and taste, a wet smokeless tobacco product, or the like. Fig. 6 shows a structure and a method of using the pipe for sucking flavor and taste, and fig. 7 shows a structure and a using method of the wet smokeless tobacco product.

Snuff refers to dried or wet tobacco powder. A pinch of the dried powder is put on the hollow in the base of the thumb and forefinger on the back of the hand and sucked through the nose. The wet tobacco powder is applied on the gum, which is a common use for enjoying the snuff.

In such a case, the purified natural (L)-menthol can be contained in the tobacco powder.

[0034]

Fig. 6 is (A) a schematic view and (B) a cross sectional view of a pipe for sucking flavor and taste, which is a snuff product. The pipe for sucking flavor and taste comprises a tube-shaped pipe body (16), a mouthpiece (18) to be removably inserted into one end of the pipe body (16), a cap (17) for covering the mouthpiece (18), an exchangeable flavor-generating cartridge (19) accommodated in the pipe body (16) for emitting flavor and taste, and shredded tobacco (6) filled up in the cartridge (19).

In such a case, the purified natural (L)-menthol can be

contained in the shredded tobacco (6) in the cartridge (19).
[0035]

Fig. 7 is (A) a schematic view, (B) a cross sectional view along A-A of a wet smokeless tobacco product, and shows (C) a method of using a wet smokeless tobacco product (22). The wet smokeless tobacco product (22) comprises shredded tobacco (6) containing moisture content, a water permeable packaging material (21) for wrapping the shredded tobacco (6), a sealing part (20) for sealing the water permeable packaging material (21).

The moisture content of the shredded tobacco (6) may be between 20-70%. The packaging material is generally, but not limited to, a non-woven fleece and should be a water permeable material. The material of the seal is not specifically limited. The wet smokeless tobacco product (22) is put between the upper lip and the gum for use, as shown in Fig. 7 (C).

In such a case, the purified natural (L)-menthol can be contained in the shredded tobacco (6) containing moisture content.

[0036]

A chewing tobacco product suitable for containing the purified natural (L)-menthol of the present invention will be explained with reference to Fig. 8.

[0037]

Fig. 8 shows a method of using a chewing tobacco product (23).

The chewing tobacco product (23) includes a plug, a twist, a scrap, a chewing gum, etc. The chewing tobacco product (23) is directly put into the mouth and chewed for use, as shown in Fig. 8.

The tobacco leaf or shredded tobacco is compressed into a sheet shape, then wrapped with an upper wrapping leaf and compressed again to provide a plug. A sheet of the above-mentioned shredded tobacco is generally square, but the shape is not specifically limited. In such a case, the purified natural (L)-menthol can be contained in the above-mentioned

tobacco leaf, shredded tobacco, and/or the upper wrapping leaf.

The tobacco leaf is twisted-and-wound, wrapped in the upper wrapping leaf, then twisted again, and pressurized to provide a twist. In such a case, the purified natural (L)-menthol can be contained in the above-mentioned tobacco leaf and/or the upper wrapping leaf.

A scrap is a shredded tobacco itself to be directly put into the mouth and chewed. In such a case, the purified natural (L)-menthol can be contained in the above-mentioned shredded tobacco.

A chewing gum is obtained by processing of a tobacco component without the use of tobacco leaf or shredded tobacco. In such a case, the purified natural (L)-menthol can be contained in the chewing gum with the tobacco component.

[0038]

2. Manufacturing Method of Purified natural (L)-menthol

Next, a method of manufacturing purified natural (L)-menthol will be explained.

Crude menthol according to the present invention is obtained by extraction, such as steam distillation, from *Mentha aquatica*, *Mentha arvensis*, *Mentha longifolia*, *Mentha pulegium*, *Mentha spicata*, *Mentha piperita*, etc. which belong to the genus of *Mentha* of the Lamiaceae family. A place of production and a kind of these materials are not specifically limited. The crude menthol obtained from the essential oil extracted through steam distillation etc. from dried leaves and other shoots of these genres of *Mentha* is used for the present invention.

[0039]

The crude menthol may better have as much (L)-menthol as possible, and preferably 90% or more of (L)-menthol. The extraction method is not specifically limited. For example, the crude menthol may be a natural (L)-menthol obtained by crystallization and centrifugation of essential oil, and be further refined by distillation, column chromatography, etc.

[0040]

[Heating and Melting Process]

The extracted crude menthol is heated and melted into a liquid crude menthol before addition of the activated carbon. Since the melting point of menthol is around 42 °C, the heating temperature may be, but not specifically limited to, 45-100 °C, preferably 55-60 °C.

[0041]

[Adding Process of Activated Carbon]

The activated carbon is added to this liquid crude menthol, where the amount of activated carbon added may be 0.1 to 10 wt%, preferably 1 to 4 wt%, of the crude menthol. It is not preferable that the amount is less than 0.1 wt%, since the impurities in the crude menthol cannot be sufficiently removed and consequently the unfavorable taste cannot be removed. Also, it is not preferable that the amount exceeds 10 wt%, since the excessive impurities of the crude menthol are removed and the mint-like flavor is lost. The mint-like flavor refers to a flavor of natural mint.

[0042]

[Stirring Process]

Then, the crude menthol is stirred with being heated in order to prevent from solidifying, so that the impurities are absorbed to the activated carbon. At this time, the temperature for heating the crude menthol may be in the range of 50 °C to 80 °C, preferably 55 °C to 65 °C, and the stirring time may be in the range of 30 minutes to 24 hours, preferably 2 to 5 hours. A heat retention means and a stirring means are not specifically limited to this.

[0043]

[Removing Process of Activated Carbon]

Next, the stirred liquid menthol is filtered to remove the activated carbon. A filtering method and an apparatus to be used are not specifically limited in this process, and a pressure filtration or filtration under reduced pressure can be used. In the Examples below, the pressure filtration was used.

[0044]

[Recrystallization Process]

The obtained filtrate is cooled down to be recrystallized. A cooling rate is, not specifically limited, but preferably slow, such that the temperature of the filtrate may be decreased, for example, at about 1-5 °C/min. For instance, a melting container is left under normal temperature to decrease a liquid temperature of a menthol melting solution, etc.

[0045]

3. Evaluating Method of Purified Natural (L)-Menthol

Next, an evaluating method of the purified natural (L)-menthol will be explained as follows.

In order to evaluate the smoking taste of the recrystallized purified natural (L)-menthol, it is added to a tobacco. For a comparison between samples, a certain amount of the purified menthol was added to each sample. The evaluation of the smoking taste consists of two evaluation criteria, refreshing feeling and mint-like flavor. Refreshing feeling refers to refreshing feeling which remains in the mouth after smoking, and the mint-like flavor refers to a flavor of natural mint. This purified natural (L)-menthol having refreshing feeling and mint-like flavor is blended into the tobacco to obtain a tasty menthol tobacco.

[0046]

In order to measure an amount of the impurities contained in the above-mentioned purified natural (L)-menthol, the purified menthol is heated and melted, and distilled under reduced pressure. The purified menthol is distilled at 2 kPa or less with the heating temperature of 80 °C. The heating temperature may be in the range of 75-95 °C. 10-30 ml of ethanol, more preferably 20 ml of ethanol, is added to the obtained residue. This ethanol solution is used as a color difference measurement sample.

[0047]

$L^*a^*b^*$ value of the color difference measurement sample is measured using a color difference meter according to ISO 7724 (JIS Z8722) to calculate chroma, thereby evaluating a degree of purification. The smoking taste correlates with the chroma,

i.e., the higher the evaluation of the smoking taste is, the lower the chroma is. A purified natural (L)-menthol with the chroma being 1 or less is used for tobacco product of the present invention.

When the purified menthol was used as a color difference measurement sample, there was little difference in color, and thus it was difficult to find out any correlation between the smoking taste and chroma. After the keen examination by the inventors with respect to the above-mentioned matter, the purified menthol was distilled under reduced pressure in a heating and melting state to obtain a residue. Then, the inventors added ethanol to the residue and used the ethanol solution as a color difference measurement sample to find out a correlation between the smoking taste and chroma is found out.

[0048]

4. Purified Natural (L)-Menthol

Finally, the purified natural (L)-menthol of the present invention is explained as follows.

The purified natural (L)-menthol of the present invention contained in the tobacco product is characterized in that chroma measured by the above-mentioned evaluating method is 1 or less. The purified natural (L)-menthol with the chroma being 1 or less can have refreshing feeling and mint-like flavor, and this purified natural (L)-menthol is blended with the tobacco product to obtain a tasty menthol tobacco product.

[0049]

The purified natural (L)-menthol of the present invention contained in the tobacco product does not depend on the (L)-menthol content, but it can be efficiently obtained by contacting the activated carbon with preferably 90% or more of crude menthol and filtering the menthol. Removing an appropriate amount of impurities by the activated carbon allows for easy purification of the purified natural (L)-menthol having refreshing feeling and mint-like flavor. After the removal of the activated carbon by filtration, the filtrate is recrystallized to finally obtain a purified natural (L)-menthol.

The (L)-menthol content is preferably 97% or more, and more preferably 99% or more.

[0050]

Also, the residue obtained by distillation under reduced pressure of the above-mentioned purified natural (L)-menthol is preferably less than 0.1 wt%. This allows for a production of a purified natural (L)-menthol having refreshing feeling and mint-like flavor with little impurities by the relatively easy process of contacting the purified natural (L)-menthol with the activated carbon.

[EXAMPLES]

[0051]

Hereinafter, the effects of the present invention will be made clearer by illustrating Examples relating to the tobacco product containing the purified natural (L)-menthol according to the present invention. However, the present invention is not limited to the following Examples.

[0052]

A naturally-derived crude menthol was heated, melted, and stirred after addition of the activated carbon. The activated carbon was then removed and the menthol was recrystallized from the filtrate to give a purified natural (L)-menthol. At this time, the obtained purified menthol went through sensory evaluation. The purified menthol was also distilled and ethanol was added to the residue. $L^*a^*b^*$ value was measured by setting this purified menthol as a color difference measurement sample to calculate chroma.

[0053]

[Preparation Example 1]

500 g of crude menthol was heated and melted and activated carbon (0 wt%, 1 wt%, 2 wt%, 4 wt%) was added to the menthol. It was then stirred for 0, 2.5, 5, or 15 hours under hot bath at 60 °C to make the activated carbon absorb the impurities. It was then vacuum filtered by a 0.5 micrometer membrane filter to remove the activated carbon. The resulting filtrate was cooled down by being left under normal temperature and

recrystallized. The purified Shirasagi (for brewing) made by Japan EnviroChemicals, Limited was used as activated carbon in this Example.

Next, production procedures of a filter cigarette using a purified natural (L)-menthol will be explained as follows. First, the obtained purified natural (L)-menthol was dissolved into a solvent solution to prepare a loading solution. Ethanol was used as a solvent solution. The loading solution may contain other perfumes as well as the natural (L)-menthol and the solvent solution is not limited to ethanol. Also, a blending ratio of a natural (L)-menthol and solvent solution may be, but not specifically limited to, 9:1-5:5. Then, the loading solution was added to shredded tobacco (American Blend, no perfume added after shredding) by a method such as spraying and use of a nozzle so that the (L)-menthol to shredded tobacco ratio is 10,000 ppm, and the shredded tobacco was processed to a filter cigarette. The menthol may be added not only to the shredded tobacco but also to a filter in the cigarette, but not specifically limited to them. The similar procedures were used for preparing a cigarette in Preparation Examples 2 and 3.

Next, sensory evaluation of smoking taste will be explained as follows. The evaluation was performed by 14 trained expert assessors smoking a cigarette. The evaluation of the smoking taste consists of two evaluation criteria, intensity of refreshing feeling and quality and intensity of mint-like flavor. When the purified natural (L)-menthol having refreshing feeling and mint-like flavor is blended into a tobacco product, a tasty tobacco product can be obtained.

250g of the purified natural (L)-menthol was distilled with 2 kPa or less at 80 °C. 20 ml of 95% ethanol was added to the resulting residue. 95% ethanol was used as ethanol, but it is not limited to this. $L^*a^*b^*$ value was measured by using this menthol as a color difference measurement sample using a color difference meter ZE-2000 (made by Nippon Denshoku Industries Co., Ltd) pursuant to ISO 7724 (JIS Z8722) to calculate chroma. A transmission method was used as a measuring method. Results

are shown in Table 1. The purified natural (L)-menthol with the chroma being more than 1 was described as Comparative Examples 1 and 2 and that with the chroma being 1 or less was described as Examples 1, 2, 3, and 4, respectively.

The Chroma can be obtained by calculating $L^*a^*b^*$, which was measured by a measurement color difference meter, using the following formula. L^* , a^* , and b^* (a combination of these three values refer to $L^*a^*b^*$ value) correspond to lightness of a color ($L=0$ is black diffuse color and $L=100$ is a white diffuse color), a position between red and green (the negative value of "a" is close to green and the positive value is close to red), and a position between yellow and blue (the negative value of "b" is close to blue and the positive value is close to yellow), respectively.

[0054]

$$\text{Chroma } (C^*) = \sqrt{(a^*)^2 + (b^*)^2}$$

[0055]

[Preparation Example 2]

6,000 g of crude menthol was distilled with 2 kPa or less at 80 °C. The menthol concentrated to the weight of 3,000 g was used as a concentrated product having twice amounts of the impurities. Some distillates are used in Preparation Example 3.

500 g of this concentrated product having twice amounts of the impurities was heated and melted and activated carbon (0 wt%, 1 wt%, 2 wt%, 4 wt%) was added to the product. It was then stirred for 0, 2.5, 5, or 15 hours under hot bath at 60 °C to make the activated carbon absorb the impurities. It was then vacuum filtered by a 0.5 micrometer membrane filter to remove the activated carbon. The resulting filtrate was cooled down by being left under normal temperature and recrystallized.

The loading solution containing the obtained purified natural (L)-menthol was added to shredded tobacco so that the (L)-menthol to shredded tobacco ratio is 10,000 ppm, as shown

in Preparation Example 1, and the shredded tobacco was processed to a filter cigarette (American Blend, no perfume added after shredding). Then, it went through sensory evaluation of smoking taste. The evaluation of the smoking taste consists of two evaluation criteria, intensity of refreshing feeling and quality and intensity of mint-like flavor. When the natural (L)-menthol having refreshing feeling and mint-like flavor is blended into a tobacco product, a tasty tobacco product can be obtained.

250 g of crude menthol was also distilled with 2 kPa or less at 80 °C. 20 ml of 95% ethanol was added to the resulting residue. $L^*a^*b^*$ value was measured by using this menthol as a color difference measurement sample using a color difference meter ZE-2000 (made by Nippon Denshoku Industries Co., Ltd) pursuant to ISO 7724 (JIS Z8722) to calculate chroma. Results are shown in Table 1. The purified natural (L)-menthol with the chroma being more than 1 was described as Comparative Examples 3, 4, 5, 6, and 7 and that with the chroma being 1 or less was described as Example 5, respectively.

[0056]

[Preparation Example 3]

1,500 g of some distillates obtained in Preparation Example 2 and 1,500 g of crude menthol were blended to give a concentrated product having 1/2 times amounts of the impurities.

500 g of this concentrated product having 1/2 times amounts of the impurities was heated and melted and activated carbon (0 wt%, 1 wt%, 2 wt%, 4 wt%) was added to the product. It was then stirred for 0, 2.5, 5, or 15 hours under hot bath at 60 °C to make the activated carbon absorb the impurities. It was then vacuum filtered by a 0.5 micrometer membrane filter to remove the activated carbon. The resulting filtrate was cooled down by being left under normal temperature and recrystallized.

The loading solution containing the obtained purified natural (L)-menthol was added to shredded tobacco so that the (L)-menthol to shredded tobacco ratio is 10,000 ppm, as shown in Preparation Examples 1 and 2, and the shredded tobacco was

processed to a filter cigarette (American Blend, no perfume added after shredding). Then, it went through Sensory evaluation of smoking taste. The evaluation of the smoking taste consists of two evaluation criteria, intensity of refreshing feeling, and quality and intensity of mint-like flavor. When the natural (L)-menthol having refreshing feeling and mint-like flavor is blended into a tobacco product, a tasty tobacco product can be obtained.

250 g of crude menthol was also distilled with 2 kPa or less at 80 °C. 20 ml of 95% ethanol was added to the resulting residue. L*a*b* value was measured by using this menthol as a color difference measurement sample using a color difference meter ZE-2000 (made by Nippon Denshoku Industries Co., Ltd) pursuant to ISO 7724 (JIS Z8722) to calculate chroma. Results are shown in Table 1. The purified natural (L)-menthol with the chroma being 1 or less was described as Example 6, 7, 8, 9, 10, and 11.

[0057]

[Table 1]

	Sample Name	Amount of Activated Carbon Added (%)	Stirring Time (h)	Color Measurement (Transmission)				Evaluation of Smoking Taste	
				L*	a*	b*	Chroma (C*)	Refreshing Feeling	Mint-like flavor
Preparation Example 1	Comparative Example 1	0	0	97.65	-0.25	2.01	2.02	C	D
	Comparative Example 2	1	5	99.03	-0.10	1.13	1.14	C	C

	Examp e 1	2	2.5	99 .5 0	-0 .0 8	0. 80	0.8 0	B	B
	Examp e 2	2	5	99 .3 9	-0 .0 5	0. 84	0.8 5	B	B
	Examp e 3	2	15	99 .6 3	-0 .1 6	0. 62	0.6 4	A	B
	Examp e 4	4	5	99 .8 1	-0 .1 4	0. 51	0.5 3	A	A
Prepar ation Examp e 2	Compar ative Examp e 3	0	0	89 .7 2	0. 02	5. 05	5.0 5	D	D
(2 x Impuri ties)	Compar ative Examp e 4	1	5	98 .6 4	-0 .1 9	1. 62	1.6 3	C	D
	Compar ative Examp e 5	2	2.5	98 .3 4	-0 .1 4	1. 44	1.4 4	C	D
	Compar ative Examp e 6	2	5	98 .2 5	-0 .0 9	1. 47	1.4 7	C	D
	Compar ative Examp e 7	2	15	98 .4 8	-0 .0 6	1. 44	1.4 4	C	D
	Examp e 5	4	5	99 .4 0	-0 .1 6	0. 93	0.9 4	B	C

Preparation Example 3	Example 6	0	0	99 .8 5	-0 .1 7	0. 78	0.8 0	B	B
(1/2 x Impurities)	Example 7	1	5	99 .5 2	-0 .0 3	0. 52	0.5 2	A	A
	Example 8	2	2.5	99 .4 5	0. 00	0. 44	0.4 4	A	A
	Example 9	2	5	99 .5 5	-0 .1 1	0. 43	0.4 5	A	A
	Example 10	2	15	99 .6 3	-0 .1 3	0. 42	0.4 4	A	A
	Example 11	4	5	99 .7 6	-0 .0 8	0. 30	0.3 1	A	B

[0058]

[Evaluation]

Trained 14 panelists performed sensory tests for refreshing feeling and mint-like flavor of a cigarette in smoking. They conducted the evaluations on a 10-point scale and further evaluated their averages as follows:

- 8.0 or more point: Grade A (very strong refreshing feeling, or very good and strong mint-like flavor),
- 7.0 or more - less than 8.0 point: Grade B (strong refreshing feeling, or good and strong mint-like flavor),
- 6.0 or more - less than 7.0 point: Grade C (slightly weak refreshing feeling or mint-like flavor),
- Less than 6.0 point: Grade D (weak refreshing feeling or mint-like flavor).

[0059]

"Stirring time" in Table 1 represents the time during which

the heated and melted purified natural (L)-menthol is stirred under hot bath at 60 °C after addition of the activated carbon.
[0060]

From Table 1, it was found out that, generally, the more the activated carbon was added to the sample, the higher grades the refreshing feeling and mint-like flavor acquired in the evaluation of the smoking taste. However, when too much amount of activated carbon was added to the sample with respect to the amount of impurities, the evaluation of mint-like flavor fell (see Example 11).
[0061]

Also, the longer the stirring time became, the stronger the refreshing feeling became. However, in Preparation Examples 2 and 3, no difference was confirmed among the evaluations by the smoking tastes depending on the stirring time.
[0062]

In all Examples, there was a tendency that the lower the chroma was, the higher the evaluation of the smoking taste was. However, if the chroma becomes 0.31 or less (see Example 11), the mint-like flavor will be lost. Also, if the chroma was 1 or less, the evaluations of refreshing feeling and mint-like flavor were relatively high. And if the chroma was within the range of 0.4 to 0.6, the grades of both refreshing feeling and mint-like flavor were very high, Grade A.
[0063]

In Preparation Examples 1 and 2, the more the activated carbon was added to the sample, the higher the L* value was, but there were little difference among the L* values between Examples 1 and 2. No correlation between an amount of activated carbon added and L* values was seen in Preparation Example 3.
[0064]

In all Preparation Examples, the lower the b value was, the higher the evaluation of the smoking taste was, which had similar tendencies as in the evaluation with chroma, i.e., . That is, it was found out that, when there were many impurities in the purified natural (L)-menthol, yellow color was shown in

the evaluation by the color difference meter.

[0065]

From above, since the chroma is correlated with the evaluation of the smoking taste, if we use the chroma as an indicator using this evaluation method, the smoking taste can be evaluated not only with sensory evaluation but also with numerical values. Also, generally, the more the activated carbon was added, the higher evaluation of the smoking taste the purified natural (L)-menthol can obtain. That is, it was found out that, if the purified natural (L)-menthol has fewer impurities, it can be highly tasty, but on the contrary, if the menthol has too few impurities, the evaluation of the smoking taste falls, especially mint-like flavor is lost. Accordingly, very small amounts of impurities are believed to be required for the natural (L)-menthol having refreshing feeling and mint-like flavor. And the tobacco product according to the present invention containing this purified natural (L)-menthol enables a user to enjoy both refreshing feeling and mint-like flavor.

[0066]

[Analysis and Measurement of Metallic Element Included in Residue]

Next, in order to confirm that the impurities are reduced or removed by activated carbon treatment, the metallic elements included in the crude menthol before the activated carbon treatment and the residues of the purified menthol after the activated carbon treatment were analyzed and measured by the following method.

[0067]

5000g of the crude menthol before the activated carbon treatment and 5000g of the purified menthol after the activated carbon treatment prepared on the same condition as Example 4 of Preparation Example 1 were provided, respectively, and heated and melted at 60 °C. They were distilled with 2 kPa or less at 80 °C to give 0.396 g and 0.829g of residues, respectively. With respect to these samples, elementary

analysis was conducted by ICP MS using the following reagents and measuring instruments.

[0068]

[Table 2]

Reagent Name	Grade
Nitric acid	Suprapur(Made by Merck & Co., Inc.)
Hydrogen peroxide	Ultrapur(Made by Merck & Co., Inc.)
Multi-element standard solution XVI	Certipur(Made by Merck & Co., Inc.)

[0069]

[Table 3]

Measuring Instrument	Note
ICP MS	Thermo X series 2(Made by Thermo Scientific, Inc.)
Microwave sample decomposing device	Made by Milestone Co., Ltd.
Detector	Mass detector
Data collection	Electronic data collection system
Carrier gas	Argon gas(super high purity)
Balance	Made by Sartorius AG
Micropipets	1-10 microliter capacity and 10-1000 microliter capacity

[0070]

[Preparation Method of Analytical Sample]

0.1 g-0.2 g of the obtained residues were transferred to a container made by Teflon (Registered Trademark) and 5 ml of nitric acid solution was added to the residues.

Further, 1 ml of H₂O₂ was added to the container and the

container was sealed. This container was installed in the microwave sample decomposing device to conducted a decomposition treatment for 20 minutes at 150 °C.

The container was then cooled and taken out from the microwave sample decomposing device. The residues in the container were transferred to a 100 ml measuring flask. The flask was filled up to 100 ml with ultrapure water and well shaken.

This sample solution was used for analysis and measurement of the metallic elements.

[0071]

A blank without the residues added was also prepared by the above-mentioned method.

Also, standard solution for creating a calibration curve was prepared using multi-element standard solution XVI and nitric acid solution.

Specifically, 0.2% nitric acid solution was first added to 1 ml of the multi-element standard solution so that the flask was filled up to 100 ml with the solution to prepare 1000 ppb of standard stock solution.

1 ppb, 3 ppb, 5 ppb, 10 ppb, 15 ppb, and 20 ppb of standard solution were prepared using this standard stock solution and 1.0% nitric acid solution.

[0072]

Measured results were converted into concentration (ppm) using the following formula.

$$\frac{\{(\text{sample read} - \text{blank read}) \times \text{volume of sample solution}\}}{(\text{weight of residue (g)} \times 1000)}$$

Results are shown in Table 4.

[0073]

[Table 4]

	Element Name	Detection Limit	Before Activated Carbon Treatment	After Activated Carbon Treatment
1	Iron	0.05	3670.92	38.74
2	Zinc	0.05	5773.62	154.12
3	Sodium	0.05	2640.91	91.08

4	Magnesium	0.05	840.48	13.28
5	Aluminum	0.05	1893.18	16.45

Unit:ppm

[0074]

Table 4 shows that, if activated carbon treatments are conducted, the amounts of the metallic elements decrease to 1 / 29 - 1/118 of the amounts without activated carbon treatments.

That is, the amounts of impurities are found to substantially decrease due to the activated carbon treatments.

Thus, the large amounts of impurities containing metallic elements, such as the above-mentioned iron, zinc, sodium, magnesium, and aluminum, are removable from the menthol by the addition and purification of the activated carbon.

Accordingly, it is possible to easily and inexpensively provide a tobacco product containing a purified natural (L)-menthol with fewer impurities by adding the activated carbon to the tobacco product containing the purified natural (L)-menthol of the present invention.

[INDUSTRIAL APPLICABILITY]

[0075]

The tobacco product containing the purified natural (L)-menthol of the present invention is conveniently used for a cigarette, cigar, snuff product, and chewing tobacco product.

[EXPLANATION OF NUMERALS]

[0076]

- 1 Filter Part
- 2 Wrapping Part
- 3 Filter Wrapping Paper
- 4 Tip Paper
- 5 Tobacco Wrapping Paper
- 6 Shredded Tobacco
- 7 Filter
- 8 Inner Filter Wrapping Paper
- 9 Cellulose Acetate Filter
- 10 Charcoal Filter

- 11 String
- 12 Breakable Capsule
- 13 Filler
- 14 Binder
- 15 Wrapper
- 16 Pipe Body
- 17 Cap
- 18 Mouthpiece
- 19 Flavor-generating Cartridge
- 20 Sealing Part
- 21 Water Permeable Packaging Material
- 22 Wet Tobacco Product
- 23 Chewing Tobacco Product

[CLAIMS]

1. A tobacco product containing a purified natural (L)-menthol with a refreshing feeling and mint-like flavor
5 which chroma measured by following methods (1) to (3) is equal to or less than 1;

(1) a step of subjecting the purified natural (L)-menthol in heated and melted state to distillation under reduced pressure to obtain a residue distilled under
10 reduced pressure,

(2) a step of adding ethanol to the residue obtained at the step (1) to prepare an ethanol solution, and

(3) a step of calculating chroma with a color difference meter pursuant to ISO 7724 (JIS Z8722) using
15 the ethanol solution obtained at the step (2) as a color difference measurement sample.

2. The tobacco product according to claim 1, wherein said chroma is within a range of 0.4 to 0.6.

20

3. The tobacco product according to claim 1 or 2, wherein the tobacco product is a cigarette comprising a wrapping part consisting of shredded tobacco and a tobacco wrapping paper to wrap the shredded tobacco, a filter part
25 consisting of a filter and a filter wrapping paper to wrap the filter, a tip paper for connecting said wrapping part and said filter part by wrapping at least a part of said wrapping part and said filter part.

4. The tobacco product according to claim 3, wherein said purified natural (L)-menthol is contained in said shredded tobacco.
- 5 5. The tobacco product according to claim 1, wherein the residue distilled under reduced pressure of said purified natural (L)-menthol is less than 0.1 % by weight.