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(54) Fragrant patchouli oil and a perfume composition comprising said patchouli oil.

(57) The present invention is directed to a patchouli oil which contains a certain amount of patchouli alcohol having an improved fragrance, and a perfume composition comprising said patchouli alcohol.

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FRAGRANT PATCHOULI OIL AND A PERFUME COMPOSITION COMPRISING SAID PATCHOULI OIL

The present invention is directed to a patchouli oil which contains a certain amount of patchouli alcohol having an improved fragrance, and a perfume composition comprising said patchouli alcohol.

Patchouli oil is utilized as a fragrance in a wide variety of fields including its use as a perfume for soap. However, because patchouli oil itself has a herbal, spicy, balsamic and woody odor, it cannot be combined into a combination perfume in a large amount, and thus the amount that can be used is limited.

On the other hand, it is known that the characteristic fragrant component in patchouli oil is patchouli alcohol, which is contained in patchouli oil in amount of about 30% by weight. Patchouli oil has a very interesting odor, namely, it has a camphoraceous, woody, earthy and amber odor. Isolation of patchouli alcohol can be done by distilling patchouli oil to enhance the content of patchouli alcohol and crystallizing it. However, this crystallization is not necessarily efficient and, as a result, the patchouli alcohol obtained is very expensive. The value of patchouli oil as a perfume and the cost for manufacturing it are off-balanced.

It has been reported that an important fragrant component of patchouli oil is norpatchoulenol and patchouli alcohol is almost odorless (P. Teissire, P. Manpetit and B. Corbier, Recherches (RBD), 19 , 8 (1974)). On the contrary, it has also been reported that patchouli alcohol is still an important fragrant component for perfumes (B.D. Mookheriee, K.K. Liahat and I.D. Hill, "Essential Oils" Allured Publishing Corp. (1981) p. 274).

However, it was not known at all until the present invention that when the concentration of patchouli alcohol is increased, the existence of norpatchoulenol is essential and it makes the whole odor both rich and strong.

The present invention was accomplished based on the above findings.

Accordingly, the present invention is directed to a fragrant patchouli oil which comprises 70 to 99.6% by weight of patchouli alcohol, and 0.4 to 5.0% by weight of norpatchoulenol.

The present inventors have found that an essential oil which has a rich, powerful and fresh odor can be obtained by distilling a patchouli oil and by precisely controlling the content of both norpatchoulenol and patchouli alcohol while removing the hydrocarbons which have a nasty top note odor and which are associated with patchouli oil which comprises 70 to 99.6% by weight of patchouli alcohol, and 0.4 to 5.0% by weight of norpatchoulenol.

The present invention is also directed to a combination perfume which comprises a perfume base; and a fragrance emitting effective amount of patchouli oil which comprises 70 to 99.6% by weight of patchouli alcohol, and 0.4 to 5.0% by weight of norpatchoulenol.

Fig. 1 is the capillary gas chromatography chart of patchouli oil in Example 2.

Fig. 2 is the capillary gas chromatography chart of patchouli oil in Comparative Example 2.

Horizontal axes in Fig. 1 and Fig. 2 represent retention time (unit:minute) and vertical axes represent strength respectively. A. represents the peak for norpatchoulenol and B. represents the peak for patchouli alcohol.

The present inventors have conducted research in order to find a method for increasing the content of patchouli alcohol by means of distillation without recrystallization. Namely, they have studied a method which consists of distilling off the low boiling portion of a patchouli oil by rectification and then distilling the main distillate which includes the patchouli alcohol. By means of this method, the content of patchouli alcohol can be increased up to around 85% by weight.

Based upon this original assumption, it was assumed that a higher content of patchouli alcohol resulted in a decrease in odor of the other components which impeded the odor of the patchouli alcohol itself and, as a result, enhanced the value patchouli oil has as a perfume. However, in fact, patchouli oil having a high content of patchouli alcohol, for example, 85% by weight, does not have a powerful and characteristic odor, and as a result is less attractive for use as a perfume material.

The present inventors have made intensive studies in order to improve this less attractive effect and, as a result, found that when a patchouli oil contains a larger proportion of a low boiling point material and the content of patchouli alcohol is lowered a little, the patchouli oil has an excellent fragrance. Namely, the present inventors have found that norpatchoulenol, which has a lower boiling point than that of patchouli alcohol, surprisingly stresses or modifies the odor of patchouli alcohol, the high boiling point material.

A sample which contains about 80% by weight of patchouli alcohol and which is well evaluated with respect to odor was fractionated by using preparative liquid chromatography to prepare a sample in which only norpatchoulenol was removed.

The odor of the above described sample where norpatchoulenol was removed was compared with (a) the odor of a sample prior to fractionation by preparative liquid chromatography and (b) with the odor of a

sample which was a combination of all of the fractions. As a result, it was found that the odor of the sample with norpatchoulenol removed lost a volumerous and sweet tone similar to another sample where the low boiling point portion was sufficiently removed by rectification to increase the content of patchouli alcohol up to 85% by weight.

In the case of practicing the present invention, it is preferred to use rectification in order to efficiently remove the nasty top note smell. The theoretical plates of the rectification are five or more, preferably ten or more. In the case of practicing the present invention, the distillation can be conducted under ordinary pressure or reduced pressure. It is unfavorable for product quality to expose patchouli alcohol to heat for a long period of time. Accordingly, distillation under reduced pressure is preferable, generally in the range of from 0.01 to 50 mm Hg; however the pressure is not necessarily limited to this range.

Although the temperature at the top of a distillation column depends upon the number of rectification plates and reduced pressure, it is generally in the range of from 50 to 300 ° C.

One purpose of the present invention is to obtain a patchouli oil comprising 70.0 to 99.6% by weight of patchouli alcohol and 0.40 to 5.0% by weight of norpatchoulenol by means of distillation.

In the present invention, when a patchouli oil containing patchouli alcohol of less than 70% by weight is distilled, (a) hydrocarbons with low boiling points distill off, then (b) patchouli alcohol containing norpatchoulenol (hereinafter, referred to as the first main distillate) distills off, and (c) finally patchouli alcohol containing scarce amounts of norpatchoulenol distills off (hereinafter referred to as the second main distillate).

A variety of methods for distillation can be used. For example, one method is to fractionate the first main distillate, combining some of the fractions of the first main distillate which the second main distillate. Another method is to distill the patchouli oil to collect a distillate having the same component as the combination of fractions described above in the form of one distillate by controlling the number of plates of the distillation column, the reduced pressure and the distillation temperature.

The patchouli oil of the present invention which comprises an increased amount of patchouli alcohol and a certain amount of norpatchoulenol does not have a green or earthy top note, but rather a mild sweet, woody, balsamic and amber-like odor. The patchouli oil of the present invention is better balanced in odor than that of patchouli alcohol itself because it comprises an appropriate amount or norpatchoulenol.

Therefore, a perfume composition of the present invention is very useful as a high-grade fragrance compound material for soaps, cosmetics, perfumes, etc.

The present invention is described in detail by way of the following examples. The present invention, however, is not limited to these examples.

Examples 1-6 and Comparative Examples 1-2

Respective amounts of patchouli oil (manufactured by T. Hasegawa Co., Ltd., Tokyo, Japan) was distilled off to remove the respective amounts of initial distillates as shown in Table 1 using a distillation column having the number of theoretical plates described in Table 1 under initial distillation conditions (reflux ratio, temperature, pressure).

Then, the residue was distilled without a distillation column under the main distillate conditions (temperature, pressure) as shown in Table 1. Patchouli oil of the present invention (example 1-6) and patchouli oil of the comparative examples (comparative example 1-2) were obtained.

Each patchouli oil obtained was analyzed by capillary gas chromatography to measure the content of patchouli alcohol and norpatchoulenol. Each patchouli oil obtained was also evaluated with respect to its aromaticity. The results are shown in Table 1. Furthermore, as to the patchouli oil of Example 2, its capillary gas chromatography chart is shown in Figure 1.

Conditions of the capillary gas chromatography were column = methyl silicon (ø = 0.2 nm x 25 n). Carrier gas = Helium (15 ml/min) and heating conditions were 80-200 ° C/20 min.

TABLE 1

EXAMPLE	THEORETICAL STEPS	REFLUX RATIO (close/open)	CHARGE AMOUNT (Kg)	INITIAL DISTILLATE		MAIN DISTILLATE		CONTENT (area ratio %)		AROMATICITY	
				BOILING POINT (°C/mmHg)	YIELD (Kg)	BOILING POINT (°C/mmHg)	YIELD (Kg)	PATCHOULI ALCOHOL	NORPATC-HOULENO-L	ODOR	EVALUATION
1	10	5:1	1.95	<120/5	1.33	<111/0.18	0.62	78.7	0.49	volumed, sweet soft woody balsamic	good
2	10	2:1	1.40	<112/3	0.87	<110/0.18	0.53	81.8	0.54	volumed, sweet soft woody balsamic	good
3	10	2:1	0.58	<112/3	0.32	<110/0.18	0.26	78.9	0.59	volumed, sweet soft woody balsamic	good
4	50	1:2	38.0	<112/1.8	24.7	<157/4	13.0	83.9	0.65	green volumeness sweet	good
5	10	2:1	0.27	<112/3	0.15	<147/3	0.12	77.1	0.92	volumed, sweet soft woody balsamic	good
6	50	1:2	38.0	<107/1.8	23.7	<157/4	13.8	78.3	1.53	green volumeness sweet	good
COMPARATIVE EXAMPLE											
1	50	1:2	38.0	<114/1.8	25.5	<158/4	12.0	85.4	0.36	weak top note non-volume	bad
2	20	2:1	1.92	<132/5	1.21	<115/0.2	0.66	85.8	0.00	hard non-sweet	bad
sensual test by 10 panelists											

Comparative Example 3

Two thousand four hundred and ten (2410) mg of patchouli oil obtained in Example 2 was separated into a 2240 mg portion containing no norpatchoulenol and a 170 mg portion containing norpatchoulenol by using high pressure liquid chromatography (HPLC) packed with Inertsil Prep-SIL (manufactured by Gasukuro Kogyo, Inc.), normal phase column, and 3% ethyl acetate/hexane as eluent.

One hundred and seventy (170) mg of the portion containing norpatchoulenol was further separated by using HPLC packed with Inertsil Prep-ODS (manufactured by Gasukuro, Kogyo, Inc.), reversed phase column and acetonitrile-water (6/4) as eluent to obtain 30 mg of norpatchoulenol and a 140 mg portion having no norpatchoulenol.

One hundred and forty (140) mg of the remaining portion was combined with the above described 2240 mg of the portion, containing no norpatchoulenol, to obtain 2380 mg of the patchouli oil which has same composition as "patchouli oil" which is obtained by removing only norpatchoulenol from the patchouli oil of Example 2.

The patchouli oil obtained was analyzed by capillary gas chromatography to measure the content of patchouli alcohol and norpatchoulenol. The patchouli oil obtained does not have a volumerous and sweet odor.

A capillary gas chromatography chart of the patchouli oil is shown in Figure 2.

Considering the results of the above-mentioned Examples and Comparative Examples, it is realized that the aromatic character of patchouli oil can be improved remarkably by containing more than 0.4% by weight of norpatchoulenol in the patchouli oil.

The amount of patchouli oil of the present invention in fragrance compositions is not limited to any range. However, a preferred range of the patchouli alcohol in fragrance compositions is from 1% to 50% by weight based on the total weight of said composition.

Example 7 (Example of floral bouquet combination perfume)	
	Parts by Weight
Sagetone V [1]	100
Sandal mysore core [2]	20
γ - Methyl ionon	100
Pearllide DEP [3]	100
Methyl dihydroxy jasmonate	200
Rose base	100
Galbanum oil	10
Pollenal V [4]	10
Lemon oil	50
Bergamot oil	100
Fruitate [5]	10
	800

(1) Sagetone V: Product name of Kao Corp. Spiro (bornane-3,1-cyclopenta-2-one)
(2) Sandal mysore core: Product name of Kao Corp. Spiro 2-methyl-4-(2.2.3-trimethyl-3-cyclopenten-1-yl)-2-buten-1-ol
(3) Pearllide DEP: Product name of Kao Corp. 1,3,4,6,7,8-hexahydro-4,6,6,7,8,8-hexamethylcyclopenta-γ-2-benzopyran diethyl phthalate solution
(4) Pollenal II: Product name of Kao Corp. 2-cyclohexyl propanal
(5) Fruitate: Product name of Kao Corp. Ethyl [5,2,1,$0^{2,6}$] decan 2-carboxylate

Floral bouquet perfume compositions having a fresh and rich odor can be obtained by adding 200 parts by weight of the invention composition in Example 1 into the above-mentioned combination perfume.

Claims

1. Fragrant patchouli oil which comprises 70 to 99.6% by weight of patchouli alcohol, and 0.4 to 5.0% by weight of norpatchoulenol.
2. A combination perfume composition which comprises a perfume base; and a fragrance emitting effective amount of patchouli oil of Claim 1.
3. The composition according to Claim 2 wherein said fragrance emitting effective amount of said patchouli oil is present in an amount of from 1% to 50% by weight based on the total weight of said composition.
4. Use of a patchouli oil of Claim 1 as a fragrance.

FIG. 1

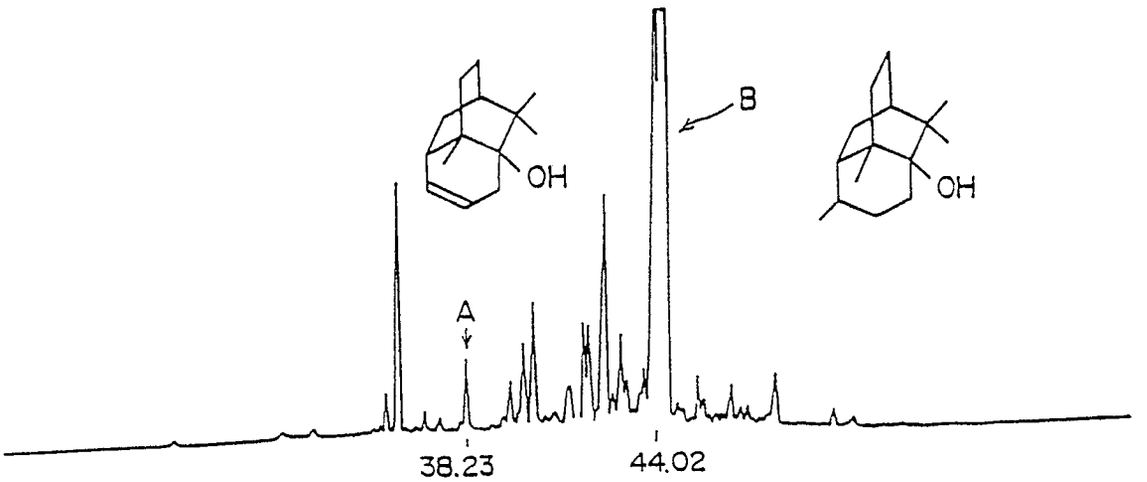

FIG. 2