(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization

International Bureau





(10) International Publication Number WO 2014/005614 A1

- (43) International Publication Date 9 January 2014 (09.01.2014)
- (51) International Patent Classification: A24B 15/28 (2006.01)
- (21) International Application Number:

PCT/EP2012/062821

(22) International Filing Date:

2 July 2012 (02.07.2012)

(25) Filing Language:

English

(26) Publication Language:

English

- (71) Applicant (for all designated States except US): SYM-RISE AG [DE/DE]; Mühlenfeldstraße 1, 37603 Holzminden (DE).
- (72) Inventors; and
- (75) Inventors/Applicants (for US only): SCHEIDELER, Gerhard [DE/DE]; Weintalstraße 5, 37671 Hoexter (DE). SIMON, Heike [DE/DE]; Corveyblick 5, 37603 Holzminden (DE). TEIWES, Holger [DE/DE]; Braunschweigerstraße 42, 37603 Holzminden (DE). MUNAJAT, Rokiah [SG/SG]; Block 285D, #02-64 Toh Guan Road, Singapore 604285 (SG). KWIE, Cher Weh [SG/SG]; Block 115, Rivervale Walk #02-29, Singapore 540115 (SG). YIP, Lorraine [SG/SG]; 53 West Coast Way, #02-05 Carabelle, Singapore 127016 (SG).

- (74) Agent: FABRY, Bernd; IP2 Patentanwalts GmbH, An der Eickesmühle 33, 41238 Mönchengladbach (DE).
- (81) Designated States (unless otherwise indicated, for every kind of national protection available): AE, AG, AL, AM, AO, AT, AU, AZ, BA, BB, BG, BH, BR, BW, BY, BZ, CA, CH, CL, CN, CO, CR, CU, CZ, DE, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IS, JP, KE, KG, KM, KN, KP, KR, KZ, LA, LC, LK, LR, LS, LT, LU, LY, MA, MD, ME, MG, MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PE, PG, PH, PL, PT, QA, RO, RS, RU, RW, SC, SD, SE, SG, SK, SL, SM, ST, SV, SY, TH, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, ZA, ZM, ZW.
- (84) Designated States (unless otherwise indicated, for every kind of regional protection available): ARIPO (BW, GH, GM, KE, LR, LS, MW, MZ, NA, RW, SD, SL, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, RU, TJ, TM), European (AL, AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT, LU, LV, MC, MK, MT, NL, NO, PL, PT, RO, RS, SE, SI, SK, SM, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).

Published:

with international search report (Art. 21(3))



A method of flavouring a smoking product

Field of invention

5

10

15

20

30

35

The present invention belongs to the area of smoking products and refers to a method for improved flavouring of tobacco and tobacco products by using pellitorine.

State of the art

The flavour of smoking products consists of two parts: the aroma and the taste. In general what is perceived through the olfactory epithelium in the nasal cavity is referred to as `aroma`, whereas the term `taste` is generally used to describe the sensory impact that is perceived via the mouth, especially the tongue. The flavour sensation experienced upon consumption, especially the taste, provides the final analysis of tobacco prior to consumption thereof. Visual and olfactory (smell) signals already give a first indication but only after intake of the smoke into the mouth the final decision is made either to ingest or to reject the product. Sweet taste is usually a signal that the product is safe (appetising) leading to ingestion of the tobacco. Bitter and sour are usually experienced as repulsive taste sensations that can lead to rejection. Temperature is another measure by which the smoking product is judged just as well as aching sensations like capsaicin (hot pepper) and certain chemicals (like carbon dioxide, menthol), also summarized as trigeminal effects.

It is well known that tobacco used for the preparation of cigarettes, for example, is composed essentially of a mixture of different types of tobacco which gives the characteristic flavour and aroma that is desired in the tobacco smoke produced. Thus, cigarettes currently manufactured usually contain mixtures of Virginia, Maryland or Kentucky tobacco in combination with oriental or Turkish tobacco.

It is also common practice to employ flavouring substances and humectants as additives to these tobacco mixtures to further enhance the organoleptic properties thereof. From the state of the art a huge number of very different chemical compounds have been proposed as flavourings for improving the organoleptic performance of a smoking product, like a cigarette or a cigar. For example, various derivatives of 6 to 12 membered aliphatic ring structures are proposed in **US 4,076,854**, **US 4,107,209** and **US 4,75,251** (all International Flavours and Fragrances Inc.). **US 4,963,193** (Givaudan) suggests oxoionyl esters, while diterpenols are the preferred flavouring agents of **US 5,296,461** (Unilever). Very common is the use of ascorbic acid, for example in combination with chlorophyll compounds and inorganic salts as disclosed in **US 6,200,391** (Ohshiro).

40 At first glance it seems surprising that there is still a need for new and improved compounds taking into account the huge prior art that deals with the generic problem of flavouring a

tobacco or smoking product. Nevertheless, common flavouring agents suffer from not sufficiently boosting aroma and trigeminal effects, especially with respect to the typical so-called "Kretek" (i.e. clove or clove oil) aroma of cigarettes which are very common in the South East Asian region. Another problem that is not solved in a satisfying way is the boosting and enhancing of mouth-watering and sweet impression on the tongue.

In the context of the invention reference is made to European patent EP 1517880 B1 (Symrise) describing a process for obtaining the isomer cis-pellitorine by transamidation of decadienoic acid esters with isobutylamine in the presence of a catalyst and the use of the product for flavouring for example oral care products and chewing gums. EP 1562893 B1 (Symrise) covers the use of trans-pellitorine as a flavouring agent. Compositions of cis- or transpellitorine and hesperitin are also known for improved sweetness (EP 1977655 B1, Symrise). Combinations of cis- or trans-pellitorine and other cooling agents are subject to WO 2009 021558 A1 (Symrise), the use of trans-pellitorine for masking unpleasant flavours is described in EP 2058297 A2 (Symrise).

Therefore, it has been the object of the present invention to solve the disadvantages of the prior art described above and in particular to provide a new flavouring agent for smoking products, that improves the aromas in general and the *Kretek* aroma in particular and that simultaneously enhances mouth-watering and sweet impression on the tongue.

20 Description of the invention

5

10

25

30

35

Object of the present invention is a method of flavouring a smoking product by treating it with a working amount of pellitorine. More particularly, said smoking product is selected from the group consisting of cigarettes, cigarillos, cigars, snuff tobacco, chewing tobacco, water pipe tobacco, shisha tobacco, hookah tobacco, roll-your-own tobacco and electric cigarettes.

Surprisingly, it has been observed that pellitorine provides a long-lasting, acid-free mouth-watering effect and increases salivation compared to a control level by up to 150 %. Also over longer storage times the flavour profile stays constant. A major advantage linked to the present invention is that the treatment for example of tipping paper, especially Kretek tipping paper leads in deed to a more natural and sweet sugar-like perception.

Pellitorine

Pellitorine (CAS 28836-52-7, FEMA 4148) stands for the isomeric forms of decadienoic acid-N-isobutylamide, which may be present as the *trans*-form (2E, 4E) or the *cis*-form (2E, 4Z) or in the form of the (2Z, 4Z) or (2Z, 4E) isomers:

The structure of pellitorine was first reported by *Jacobson* in **JACS 71(1)**, p 366-367 (1949). It was originally extracted from the roots of *Piper nigrum* and was recently suggested by *Cheng*

et al. for its anti-cancer properties [Molecules 15, p 2398-2404 (2010)]. The product is available in the market for example under the trademark Optaflow® from Symrise.

Pellitorine can be administered directly to the tobacco rod, but also to the filters or the tipping papers. Preferably, the active is brought on the tobacco products, the filters or the paper in form of a solution in an organic solvent, such as for example C_1 - C_4 aliphatic alcohols, preferably ethanol or (iso)propylalcohol, alkylene glycols, like e.g. ethylene glycol, propylene glycol, diethylene glycol or dipropylene glycol, triacetin and their mixtures. The solutions may contain the Pellitorine in amounts of about 1 to about 25, preferably about 2 to about 20 and more preferably about 5 to about 15 % b.w.

5

10

15

20

25

30

35

40

45

It is also possible to treat the smoking products by pellitorine capsules. In case encapsulated pellitorine is used the active is incorporated into the tobacco or into the filters. The compositions are typically encapsulated by means of a solid covering material, which is preferably selected from starches, degraded or chemically or physically modified starches (in particular dextrins and maltodextrins), gelatins, gum arabic, agar-agar, ghatti gum, gellan gum, modified and non-modified celluloses, pullulan, curdlan, carrageenans, alginic acid, alginates, pectin, inulin, xanthan gum and mixtures of two or more of said substances.

The solid covering material is preferably selected from gelatin (preferred are pork, beef, chicken and/or fish gelatins and mixtures thereof, preferably comprising at least one gelatin with a bloom value of greater than or equal to 200, preferably with a bloom value of greater than or equal to 240), maltodextrin (preferably obtained from maize (corn), wheat, tapioca or potato, preferred maltodextrins have a DE value of 10 - 20), modified cellulose (for example cellulose ether), alginates (for example Na-alginate), carrageenan (beta-, iota-, lambda- and/or kappa carrageenan), gum arabic, curdlan and/or agar-agar. Gelatin is preferably used, in particular, because of its good availability in different bloom values. Particularly preferred, especially for oral use are seamless gelatin or alginate capsules, the covering of which dissolves very rapidly in the mouth or bursts when chewing. Production may take place, for example, as described in **EP 0389700 A1, US 4,251,195, US 6,214,376, WO 2003 055587** or **WO 2004 050069 A1.**

The capsules, however, may also represent micro-capsules. "Microcapsules" are understood to be spherical aggregates with a diameter of about 0.1 to about 5 mm which contain at least one solid or liquid core surrounded by at least one continuous membrane. More precisely, they are finely dispersed liquid or solid phases coated with film-forming polymers, in the production of which the polymers are deposited onto the material to be encapsulated after emulsification and coacervation or interfacial polymerization. In another process, liguid active principles are absorbed in a matrix ("microsponge") and, as microparticles, may be additionally coated with film-forming polymers. The microscopically small capsules, also known as nanocapsules, can be dried in the same way as powders. Besides single-core microcapsules, there are also multiple-core aggregates, also known as microspheres, which contain two or more cores distributed in the continuous membrane material. In addition, single-core or multiple-core microcapsules may be surrounded by an additional second, third etc. membrane. The membrane may consist of natural, semisynthetic or synthetic materials. Natural membrane materials are, for example, gum arabic, agar agar, agarose, maltodextrins, alginic acid and salts thereof, for example sodium or calcium alginate, fats and fatty acids, cetyl alcohol, collagen, chitosan, lecithins, gelatin, albumin, shellac, polysaccharides, such as starch or dextran, polypeptides, protein hydrolyzates, sucrose and waxes.

Semisynthetic membrane materials are inter alia chemically modified celluloses, more particularly cellulose esters and ethers, for example cellulose acetate, ethyl cellulose, hydroxypropyl cellulose, hydroxypropyl cellulose, hydroxypropyl methyl cellulose and carboxymethyl cellulose, and starch derivatives, more particularly starch ethers and esters. Synthetic membrane materials are, for example, polymers, such as polyacrylates, polyamides, polyvinyl alcohol or polyvinyl pyrrolidone.

Examples of known microcapsules are the following commercial products (the membrane material is shown in brackets) Hallcrest Microcapsules (gelatin, gum arabic), Coletica Thalaspheres (maritime collagen), Lipotec Millicapseln (alginic acid, agar agar), Induchem Unispheres (lactose, microcrystalline cellulose, hydroxypropylmethyl cellulose), Unicetin C30 (lactose, microcrystalline cellulose, hydroxypropylmethyl cellulose), Kobo Glycospheres (modified starch, fatty acid esters, phospholipids), Softspheres (modified agar agar) and Kuhs Probiol Nanospheres (phospholipids).

The active principles are released from the microcapsules by mechanical, thermal, chemical or enzymatic destruction of the membrane, normally during the use of the preparations containing the microcapsules. Despite the fact that the state of the art a huge range of possibilities for the encapsulation of actives, methods according to which a shell is obtained by coazervation, precipitation or polycondensation of anionic and cationic polymers has been quite suitable for the formation of stable capsules. Particularly, a preferred process for the encapsulation of active principles according to the present invention is characterised in that it comprises the steps of

- (a) preparing a matrix from gel formers, cationic polymers and active principles;
- (b) optionally dispersing said matrix in an oil phase; and

5

10

15

20

25

30

35

(c) treating said dispersed matrix with aqueous solutions of anionic polymers and optionally removing the in phase in the process.

Of course, anionic and cationic polymers in steps (a) and (c) can be exchanged.

- (i) **Gel formers.** In the context of the invention, preferred gel formers are substances which are capable of forming gels in aqueous solution at temperatures above 40° C. Typical examples of such gel formers are heteropolysaccharides and proteins. Preferred thermogelling heteropolysaccharides are agaroses which may be present in the form of the agar agar obtainable from red algae, even together with up to 30% by weight of non-gel-forming agaropectins. The principal constituent of agaroses are linear polysaccharides of Galactose and 3,6-anhydro-L-galactose with alternate 1,3- and 1,4-glycosidic bonds. The heteropolysaccharides preferably have a molecular weight of 110,000 to 160,000 and are both odourless and tasteless. Suitable alternatives are pectins, xanthans (including xanthan gum) and mixtures thereof. Other preferred types are those which in 1% by weight aqueous solution still form gels that do not melt below 80° C. and solidify again above 40° C. Examples from the group of thermogelling proteins are the various gelatines.
- 40 (ii) **Anionic polymers.** Salts of alginic acid are preferred for this purpose. The alginic acid is a mixture of carboxyl-containing polysaccharides with the following idealized monomer unit:

5

10

15

20

25

30

35

The average molecular weight of the alginic acid or the alginates is in the range from 150,000 to 250,000. Salts of alginic acid and complete and partial neutralization products thereof are understood In particular to be the alkali metal salts, preferably sodium alginate ("algin") and the ammonium and alkaline earth metal salts. Mixed alginates, for example sodium/magnesium or sodium/calcium alginates, are particularly preferred. In an alternative embodiment of the invention, however, carboxymethyl celluloses and anionic chitosan derivatives, for example the carboxylation and above all succinylation products are also suitable for this purpose.

(iii) Cationic polymers. Chitosans are biopolymers which belong to the group of hydrocolloids. Chemically, they are partly de-acetylated chitins differing in their molecular weights which contain the following – idealized - monomer unit:

$$[\begin{array}{c|cccc} CH_2OH & OH & NHR \\ O & O & O \\ OH & NH_2 & CH_2OH \\ \end{array} \\$$

In contrast to most hydrocolloids, which are negatively charged at biological pH values, chitosans are cationic biopolymers under these conditions. The positively charged chitosans are capable of interacting with oppositely charged surfaces and are therefore used in cosmetic hair-care and body-care products and pharmaceutical preparations.

In a preferred embodiment of the invention a 1 to 10 and preferably 2 to 5% by weight aqueous solution of the gel former, preferably agar agar, is normally prepared and heated under reflux. A second aqueous solution containing the cationic polymer, preferably chitosan, in quantities of 0.1 to 2 and preferably 0.25 to 0.5% by weight and the active principle in quantities of 0.1 to 25 and preferably 0.25 to 10% by weight is added in the boiling heat, preferably at 80 to 100 ° C.; this mixture is called the matrix. Accordingly, the charging of the microcapsules with active principles may also comprise 0.1 to 25% by weight, based on the weight of the capsules. If desired, water-insoluble constituents, for example inorganic pigments, may also be added at this stage to adjust viscosity, generally in the form of aqueous or aqueous/alcoholic dispersions. In addition, to emulsify or disperse the active principles, it can be useful to add emulsifiers and/or solubilisers to the matrix. After its preparation from gel former, cationic polymer and active principle, the matrix optionally is very finely dispersed in an oil phase with intensive shearing in order to produce small particles in the subsequent encapsulation process. It has proved to be particularly advantageous in this regard to heat the matrix to temperatures in the range from 40 to 60° C while the oil phase is cooled to 10 to 20° C. The actual encapsulation, i.e. formation of the membrane by contacting the cationic polymer in the matrix with the anionic polymers, takes place in the

third step. To this end, it is advisable to wash the matrix - dispersed in the oil phase - with an aqueous ca. 0.1 to 3 and preferably 0.25 to 0.5% by weight aqueous solution of the anionic polymer, preferably the alginate, at a temperature in the range from 40 to 100 and preferably 50 to 60° C. and, at the same time, to remove the oil phase if present. The resulting aqueous preparations generally have a microcapsule content of 1 to 10% by weight. In some cases, it can be of advantage for the solution of the polymers to contain other ingredients, for example emulsifiers or preservatives. After filtration, microcapsules with a mean diameter of preferably 1 to 3 mm are obtained. It is advisable to sieve the capsules to ensure a uniform size distribution. The microcapsules thus obtained may have any shape within production-related limits, but are preferably substantially spherical.

Pellitorine is administered at a working amount which means a concentration sufficient to achieve the desired performance. Typically, pellitorine is added to the tobacco products in an amount of

- (a) about 1 to about 50, preferably about 5 to about 40 g calculated on 100 kg of tobacco, or
- (b) about 1 to about 10, preferably about 2 to about 8 g calculated on tipping paper having a size of 900 x 5,000 m, or
- (c) about 1 to about 30, preferably about 5 to about 20 g calculated on cigarette filters having a size of 100 x 3,000 m.

Additives

5

10

15

20

25

30

35

40

In a preferred embodiment of the present invention the tobacco products are treated with mixtures of pellitorine and auxiliary agents, said auxiliary agents being selected from the group consisting of flavours, cooling agents, sweeteners, plant extracts and solvents.

A. Flavours

Suitable flavours or flavourings include, but are not limited to, mint, such as peppermint and spearmint, chocolate, licorice, citrus and other fruit flavours, gamma octalactone, vanillin, ethyl vanillin, breath freshener flavours, spice flavours such as cinnamon, methyl salicylate, linalool, bergamot oil, geranium oil, lemon oil, and ginger oil. Other suitable flavours may include flavour compounds selected from the group consisting of an acid, an alcohol, an ester, an aldehyde, a ketone, a pyrazine, combinations or blends thereof and the like.

B. Cooling agents

The compositions may also contain one or more flavours with a physiological cooling effect (cooling agents), which are preferably selected here from the following list: menthol and menthol derivatives (for example L-menthol, D-menthol, racemic menthol, isomenthol, neoisomenthol, neomenthol) menthylethers (for example (I-menthoxy)-1,2-propandiol, (I-menthoxy)-2-methyl-1,2-propandiol, I-menthyl-methylether), menthylesters (for example menthylformiate, menthylacetate, menthylisobutyrate, menthyllactates, L-menthyl-Llactate, L-menthyl-D-lactate, menthyl-(2-methoxy)acetate, menthyl-(2-methoxyethoxy-

)acetate, menthylpyroglutamate), menthylcarbonates (for example menthylpropyleneglycolcarbonate, menthylethyleneglycolcarbonate, menthylglycerolcarbonate or mixtures thereof), the semi-esters of menthols with a dicarboxylic acid or derivatives thereof (for example mono-menthylsuccinate, mono-menthylglutarate, mono-menthylmalonate, Omenthyl succinic acid ester-N,N-(dimethyl)amide, O-menthyl succinic acid ester amide), menthanecarboxylic acid amides (in this case preferably menthanecarboxylic acid-Nethylamide [WS3] or N^{α} -(menthanecarbonyl)glycinethylester [WS5], as described in **US** 4,150,052, menthanecarboxylic acid-N-(4-cyanophenyl)amide or menthanecarboxylic acid-N-(4-cyanomethylphenyl)amide as described in WO 2005 049553 A1, methanecarboxylic acid-N-(alkoxyalkyl)amides), menthone and menthone derivatives (for example L-menthone glycerol ketal), 2,3-dimethyl-2-(2-propyl)-butyric acid derivatives (for example 2,3-dimethyl-2-(2-propyl)-butyric acid-N-methylamide [WS23]), isopulegol or its esters (I-(-)-isopulegol, I-(-)-isopulegolacetate), menthane derivatives (for example p-menthane-3,8-diol), cubebol or synthetic or natural mixtures, containing cubebol, pyrrolidone derivatives of cycloalkyldione derivatives (for example 3-methyl-2(1-pyrrolidinyl)-2-cyclopentene-1-one) or tetrahydropyrimidine-2-one (for example iciline or related compounds, as described in WO 2004/026840), further carboxamides (for example N-(2-(pyridin-2-yl)ethyl)-3-pmenthanecarboxamide or related compounds), (1R,2S,5R)-N-(4-Methoxyphenyl)-5-methyl-2-(1-isopropyl)cyclohexane-carboxamide [WS12], oxamates (preferably those described in EP 2033688 A2) and their mixtures. Both, the flavours and the cooling agents can be present in amounts of about 0 to about 0.5% by weight, and preferably, from about 0.05 to about 0.2% by weight.

C. Sweeteners

5

10

15

20

25

30

35

40

Examples of sweeteners which may be employed together with pelitorine include sugars, for example, monosaccharides of 5 or 6 carbon atoms, such as arabinose, xylose, ribose, glucose, mannose, galactose, fructose, dextrose, or sorbose or mixtures of two or more of the foregoing monosaccharides; disaccharides, for example, sucrose, such as cane or beet sugar, lactose, maltose or cellobiose; polysaccharides, such as partially hydrolyzed starch or dextrin, as well as polyols, such as sorbitol, mannitol, xylitol, mixtures thereof and mixtures with one or more of the above sugars. The mixtures may also contain an artificial sweetener, such as, for example, aspartame, cyclamate, or a saccharin or other sweetener as set out hereinafter, the artificial sweetener being present in an amount of from about 0 to about 0.5% by weight, and preferably, from about 0.05 to about 0.2% by weight. Examples of artificial sweeteners which may be employed herein include sodium, calcium or ammonium saccharin salts, dihydrochalcones, rebaudiosides, mogrosides,glycyrrhizin, dipotassium glycyrrhizin, glycyrrhizic acid ammonium salt, L-aspartyl-L-phenylalamine methyl ester, (aspartame), the sodium or potassium salt of 3,4-dihydro-6-methyl-1,2,3-oxathiazine-4-one-2,2-dioxide (Ace-sulfame-K), as well as extracts of Stevia rebaudiana (Stevioside), Richardella dulcifica (Miracle Berry), Dioscoreophyllum cumminsii (Serendipity Berry) or Luo Han Guo, cyclamate salts, and the like, or mixtures of any two or more of the above. Where longlasting sweetness is desired, the sweetener may be employed in particulate form so as to have an average particle size of less than 150 microns, and preferably less than 100 microns.

Industrial application

5

Additional objects of the present invention are directed to a smoking product selected from the group consisting of cigarettes, cigarillos, cigars, snuff tobacco, chewing tobacco, water pipe tobacco, shisha tobacco, hookah tobacco, roll-your-own tobacco and electric cigarettes, flavoured with a working amount of pellitorine, and a tobacco rod, a filter or a tipping paper flavoured with a working amount of pellitorine.

Examples

Example 1 and Comparative Examples C1 and C2

5 Cigarette filters were treated with a composition comprising

10.0 % b.w. pellitorine (Optaflow® A, consists >95% of trans-isomer)

30.0 % b.w. ethanol

30.0 % b.w. diethylmalonate (food grade)

29.8 % b.w. 1,2-propylene glycol

10 0.2 % b.w. peppermint oil

at a concentration of about 50 g in 100,000 sticks. The filters were used to make cigarettes and the flavoured cigarettes were compared to conventional unflavoured cigarettes (C1) and cigarettes flavoured by a composition where the pellitorine was replaced by the same amount of benzyl benzoate (C2) by a group consisting of 20 experienced consumers. The cigarettes were determined with regard to four different performance features and judged on a scale from 1 (= very week) to 10 (very strong). The results are presented in the following table 1 and represent the average of three test runs.

Table 1Evaluation of flavoured and unflavoured Kretek cigarettes

Parameter	C1 (Control)	C2	1
Sweetness	2	6	9
Clean mouth feeling/sensation during and after smoking	4	6	8
Intensity of the overall impact and smoke taste	4	7	8
Increase of fineness and elegance of to- bacco aroma	4	5	8

The examples clearly illustrate the superior properties of pellitorine compared to a standard flavouring agent for Kretek cigarettes.

15

Claims

5

10

20

35

40

 A method of flavouring a smoking product by treating it with a working amount of pellitorine.

- 2. The method of Claim 1, wherein the smoking product is selected from the group consisting of cigarettes, cigarillos, cigars, snuff tobacco, chewing tobacco, water pipe tobacco, shisha tobacco, hookah tobacco, roll-your-own tobacco and electric cigarettes.
- 3. The method of Claim 1, wherein pellitorine encompasses the (2E, 4E), (2E, 4Z), (2Z, 4Z) and (2Z, 4E) stereoisomers and their mixtures.
 - 4. The method of Claim 1, wherein pellitorine is administered to the tobacco rod, the filters or the tipping papers.
 - 5. The method of Claim 1, wherein pellitorine is administered as a solution in an organic solvent.
- 15 6. The method of Claim 5, wherein the organic solvents is selected from the group consisting of C_1 - C_4 aliphatic alcohols, alkylene glycols, triacetin and their mixtures.
 - 7. The method of Claim 1, wherein pellitorine is administered as a capsule.
 - 8. The method of Claim 7, wherein pellitorine is encapsulated by solid covering materials, which is preferably selected from starches, degraded or chemically or physically modified starches, gelatins, gum arabic, agar-agar, ghatti gum, gellan gum, modified and non-modified celluloses, pullulan, curdlan, carrageenans, alginic acid, alginates, pectin, inulin, xanthan gum and mixtures of two or more of said substances or by coazervation.
- 9. The method of Claim 1, wherein pellitorine is administered to the tobacco product in an amount of
 - (a) about 1 to about 50 g calculated on 100 kg of tobacco, or
 - (b) about 1 to about 10 g calculated on tipping paper having a size of 900 x 5,000 m, or
 - (c) about 1 to about 30 g calculated on cigarette filters having a size of 100 x 3,000 m.
- 30 10. The method of Claim 1, wherein the tobacco products are treated with mixtures of pellitorine and auxiliary agents, said auxiliary agents being selected from the group consisting of flavours, cooling agents, sweeteners, plant extracts and solvents.
 - 11. The method of Claim 10, wherein the flavours are selected from the group consisting of peppermint and spearmint, chocolate, licorice, citrus and other fruit flavours, gamma octalactone, vanillin, ethyl vanillin, breath freshener flavours, spice flavours such as cinnamon, methyl salicylate, linalool, bergamot oil, geranium oil, lemon oil, and ginger oil.
 - 12. The method of Claim 10, wherein the cooling agents are selected from the group consisting of menthol and menthol derivatives, menthylethers, menthylesters, menthyl-carbonates, semi-esters of menthols with a dicarboxylic acid or derivatives thereof,

menthanecarboxylic acid amides, menthone and menthone derivatives, menthane derivatives, cubebol or synthetic or natural mixtures, containing cubebol, pyrrolidone derivatives of cycloalkyldione derivatives or tetrahydropyrimidine-2-one, carboxamides, oxamates and their mixtures.

- The method of Claim 10, wherein the sweeteners are selected from the group consisting of sugars, aspartame, cyclamate, saccharin, sodium, calcium or ammonium saccharin salts, dihydrochalcones, rebaudiosides, mogrosides, glycyrrhizin, dipotassium glycyrrhizin, glycyrrhizic acid ammonium salt, L-aspartyl-L-phenylalamine methyl ester, (aspartame), sodium or potassium salt of 3,4-dihydro-6-methyl-1,2,3-oxathiazine-4-one-2,2-dioxide, and extracts of Stevia rebaudiana, Richardella dulcifica, Dioscore-ophyllum cumminsii and Luo Han Guo.
 - 14. A smoking product selected from the group consisting of cigarettes, cigarillos, cigars, snuff tobacco, chewing tobacco, water pipe tobacco, shisha tobacco, hookah tobacco, roll-your-own tobacco and electric cigarettes, flavoured with a working amount of pellitorine.

15

15. A tobacco rod, a filter or a tipping paper flavoured with a working amount of pellitorine.

INTERNATIONAL SEARCH REPORT

International application No PCT/EP2012/062821

a. classification of subject matter INV. A24B15/28

ADD.

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)

A24B

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

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

EPO-Internal, WPI Data, BIOSIS, FSTA, INSPEC

2010/226864 A1 (OERTLING HEIKO [DE] ET 9 September 2010 (2010-09-09) agraph [0003] agraph [0076] agraph [0082] - paragraph [0086] 439 878 B2 (SHIGEYOSHI HATASA, ET AL) August 1973 (1973-08-29)	1-8,10, 11,14,15
August 1973`(1973-08-29)	10,12,
e 2 - page 4	1,,10
March 2011 (2011-03-29)	1-4,7, 14,15
	7 914 825 B2 (SIEGEL SVEN [DE] ET AL) March 2011 (2011-03-29) umn 12, line 48 - column 13, line 11

Further documents are listed in the continuation of Box C.	See patent ramily annex.	
Special categories of cited documents : "A" document defining the general state of the art which is not considered to be of particular relevance	"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art "&" document member of the same patent family	
 "E" earlier application or patent but published on or after the international filing date "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) "O" document referring to an oral disclosure, use, exhibition or other means "P" document published prior to the international filing date but later than the priority date claimed 		
Date of the actual completion of the international search	Date of mailing of the international search report	
28 February 2013	06/03/2013	
Name and mailing address of the ISA/	Authorized officer	
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016	Dimoula, Kerasina	

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2012/062821

ategory* Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT tegory* Citation of document, with indication, where appropriate, of the relevant passages US 2007/221236 A1 (KIEFER JESSE [US] ET AL) 27 September 2007 (2007-09-27) page 2, paragraph 22 page 7, paragraph 63 - paragraph 64 page 14, paragraph 133	Relevant to claim No. 1,2,14, 15

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No
PCT/EP2012/062821

US 2010226864 A1 09-09-2010 AT 545639 T 15-03-2012	Patent document cited in search report	Publication date	Patent family member(s)	Publication date
AU 4682072 A 13-09-1973 US 7914825 B2 29-03-2011 EP 2022503 A1 11-02-2009 US 2009041872 A1 12-02-2009 US 2007221236 A1 27-09-2007 EP 2129234 A1 09-12-2009	US 2010226864	09-09-2010	EP 2168957 A2 ES 2379751 T3	31-03-2010 03-05-2012
US 2009041872 A1 12-02-2009 US 2007221236 A1 27-09-2007 EP 2129234 A1 09-12-2009	AU 439878	B2 29-08-1973		
	US 7914825	B2 29-03-2011		
WO 2008124667 A1 16-10-2008	US 2007221236	A1 27-09-2007	US 2007221236 A1	27-09-2007