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(54) **FUNCTIONAL CIGARETTE FILTER CONTAINING NATURAL PLANT MATERIAL, CIGARETTE, AND METHOD FOR PRODUCING SAME**

(57) Disclosed is a cigarette filter including: natural plant based granules composed of a natural plant material and a binder; and a filter portion to accommodate therein the natural plant based granules, in which the binder is hydroxy propyl methyl cellulose (HPMC), and a weight ratio of the natural plant material and the binder is in a range of 95 to 99% : 5 to 1%. Further, disclosed are a preparation method of the cigarette filter, and a cigarette including the cigarette filter.

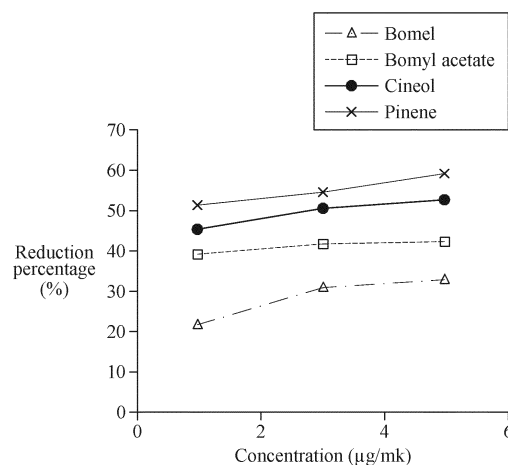


FIG. 8

**Description**

## TECHNICAL FIELD

5 **[0001]** The following description relates to a cigarette filter containing natural plant material that reduces cigarette-derived odor ingredient and intraoral halitosis inducing substance, and a cigarette including the filter and a preparation method thereof.

## BACKGROUND ART

10 **[0002]** In general, in order to prepare cigarettes, various kinds of leaf tobacco are mixed and processed to give the desired aroma and taste. Next, the processed leaf tobacco mixture is cut to prepare a cigarette filler, and the cigarette filler is rolled with a cigarette paper to prepare a filter-free cigarette. Then, if necessary, the filter is attached to the filter-free cigarette.

15 **[0003]** The cigarette is one of the most popular preferences, and consumers get a sense of satisfaction using smoking the cigarette. In particular, research on adding natural plant material to cigarettes is being actively conducted so that consumers may feel the scent of natural plant material that suits their preferences simultaneously with smoking. Moreover, after the smoking, cigarette odors that are left in the smoker's mouth or hands are perceived as negative by neighbors. Thus, research on cigarettes with less cigarette odor and preparation methods thereof is being actively conducted.

20 **[0004]** For example, the effect of reducing the intraoral halitosis ingredient may be identified as the volatile ingredient is delivered into the smoke under the chemical, antibacterial and masking actions of natural plant material-based volatile substances. In this connection, gas molecules fly at a speed of 100 m/s to 1,000 m/s, and they collide with other molecules about at least 20,000 times during travel thereof by 1 mm, thereby to realize the above effect. Moreover, the effect of reducing halitosis inducing ingredients derived from cigarettes may be expected using the chemistry and adsorption actions of non-volatile substances based on the natural plant material. Therefore, there is a need for a study on an optimized method to reduce cigarette-derived odor ingredient and intraoral halitosis inducing substance by applying natural plant material to a cigarette filter.

## DISCLOSURE OF INVENTION

30 Technical Subject

**[0005]** A purpose of the present disclosure is to provide a cigarette filter including natural plant based granules composed of a natural plant material and a binder; and a filter portion to accommodate therein the natural plant based granules, in which the binder is hydroxy propyl methyl cellulose (HPMC), and a weight ratio of the natural plant material and the binder is in a range of 95% to 99% : 5% to 1%.

**[0006]** Another purpose of the present disclosure is to provide a cigarette including the cigarette filter.

40 **[0007]** Another purpose of the present disclosure is to provide a preparation method of a cigarette filter, the method including inserting a binder into a natural plant material to prepare natural plant based granules; drying and classifying the natural plant based granules; and adding the classified natural plant based granules to a cigarette filter, in which the binder is hydroxy propyl methyl cellulose (HPMC) of a viscosity of 50 cps, in which a weight ratio of the natural plant material and a binder is in a range of 95% to 99% : 5% to 1%.

45 **[0008]** However, the purpose to be solved by the present disclosure is not limited to the purposes mentioned above, and other purposes that are not mentioned may be clearly understood by those with ordinary knowledge in the relevant technical field from the following description.

Technical Solutions

50 **[0009]** According to an aspect, there is provided a cigarette filter including natural plant based granules composed of a natural plant material and a binder; and a filter portion to accommodate therein the natural plant based granules, in which the binder is hydroxy propyl methyl cellulose (HPMC), and a weight ratio of the natural plant material and the binder is in a range of 95% to 99% : 5% to 1%.

55 **[0010]** According to one embodiment, the natural plant material may include at least one selected from rosemary, pine needle, peppermint, spearmint, coffee, pineapple, chamomile, orange, eucalyptus, thyme, geranium, jasmine, rosemary, lavender, lemongrass, pine needle, clover, sage, taxol, bergamot, basil, thyme, valerian, hyssop, tea tree, myrrh, and juniper.

**[0011]** According to one embodiment, a hardness of the natural plant based granule may be in a range of 90.0% to 99.0%.

[0012] According to one embodiment, a size of the natural plant based granule may be in a range of 0.25 mm to 2.0 mm.

[0013] According to one embodiment, a moisture content of the natural plant based granule may be in a range of 5% to 12% of a total weight of the natural plant based granules.

[0014] According to one embodiment, the cigarette filter may be a single filter or a multiple filter.

5 [0015] According to one embodiment, the cigarette filter may further include a herb oil capsule containing an oil ingredient extracted from the natural plant material.

[0016] According to another aspect, there is provided a cigarette including the cigarette filter.

[0017] According to one embodiment, the cigarette may reduce methylmercaptan as a halitosis inducing ingredient.

10 [0018] According to one embodiment, the cigarette may reduce a cigarette odor inducing ingredient including ammonia, acrolein, crotonaldehyde, methyl ethyl ketone (MEK), and 1,3-butadiene, acrylonitrile, benzene, and pyridine by 9% to 55%.

[0019] According to still another aspect, there is provided a preparation method of a cigarette filter, the method including inserting a binder into a natural plant material to prepare natural plant based granules; drying and classifying the natural plant based granules; and adding the classified natural plant based granules to a cigarette filter, in which in the preparing of the natural plant based granules, the binder is hydroxy propyl methyl cellulose (HPMC) of a viscosity of 50 cps, in which a weight ratio of the natural plant material and a binder is in a range of 95% to 99% : 5% to 1%.

15 [0020] According to one embodiment, the preparing of the natural plant based granules may include adding 2% of hydroxy propyl methyl cellulose (HPMC) of a viscosity of 50 cps as a binder to 50% ethanol to produce a binder solution, and mixing pulverized natural plant powders and the binder solution with each other to form a mixture and stirring the mixture, and shaping and classifying the mixture.

[0021] According to one embodiment, the drying of the natural plant based granules may include one of freeze-drying, hot air drying, warm air drying, and fluidized bed drying methods.

[0022] According to one embodiment, a size of the classified natural plant based granule may be in a range of 0.25 mm to 2.0 mm.

25 [0023] According to one embodiment, the adding of the classified natural plant based granules to the cigarette filter may include adding a herb extract to acetate tow using a spraying method or a nozzle based spraying method (TJNS).

[0024] According to one embodiment, the method of preparing the cigarette filter may further include injecting a herb oil capsule containing an oil ingredient extracted from the natural plant material into the cigarette filter using a capsule injection facility.

30 [0025] According to one embodiment, in the adding of the classified natural plant based granules to the cigarette filter, an added amount of the granules may be in a range of 0.5 to 4 mg/mm Tip.

## EFFECTS

35 [0026] The cigarette filter including the natural plant based granules according to the present disclosure absorbs, binds, and neutralizes the cigarette-derived odor ingredient or halitosis inducing ingredient, thus reducing the unique cigarette-derived odor and halitosis derived from a mouth of a cigarette smoker. In addition, in addition to the natural plant based granules, the cigarette filter may further include the herb extract or the herb oil capsule, thereby to enhance the halitosis reduction effect, thus providing a cigarette that minimizes discomfort to non-smokers as well as smokers.

40 [0027] Moreover, according to the present disclosure, the natural plant based granules may have a hardness of 90.0% to 99.0% and a moisture content of 5% to 12%. Due to the natural plant based granules having the above characteristics, activated carbon as a second additive applied to a cigarette filter may not be used. When applying the natural plant based granules to the cigarette filter, the filter productivity may be improved.

45 [0028] The effect of the present disclosure is not limited to the above effect, and may include all effects that may be deduced from the detailed description of the present disclosure or a configuration of the invention described in the claims.

## BRIEF DESCRIPTION OF DRAWINGS

### [0029]

50 FIG. 1 shows an example of a triple complex filter according to the present disclosure.  
 FIG. 2 shows an example of a double complex filter according to the present disclosure.  
 FIG. 3 shows another example of a double complex filter according to the present disclosure.  
 FIG. 4 shows another example of a double complex filter according to the present disclosure.  
 55 FIG. 5 is a graph showing a result of identifying a standardized quantification of each of an index ingredient and an index fragrance ingredient of pine needle and rosemary raw material through LC/MS/MS analysis.  
 FIG. 6 is a graph showing the result of analyzing the fragrance ingredient in smoke of pine needle and rosemary through SPME analysis.

FIG. 7 is a graph showing the analysis results of fragrance carrying ingredients in smoke of the products (NRP5-2, NRP5-4) 90 days after the preparation of the cigarette product (NRP5-2) having pine needle granules added thereto and the cigarette product (NRP5-4) having rosemary granules added thereto. In this connection, common fragrance carrying ingredient between the two products are borneol, bonyl acetate and 1,8-cineol, whose contents are indicated in ug/pad in the graph.

FIG. 8 is a graph showing the results of identifying reducing ability for methyl mercaptan as halitosis inducing ingredients based on the concentrations of borneol, bonyl acetate, cineol, and pinene as fragrance carrying ingredients in smoke of pine needle and rosemary.

FIG. 9 is a graph showing the results of identifying reducing ability for the halitosis inducing ingredients by borneol, bonyl acetate, cineol, and pinene as fragrance carrying ingredients in smoke of pine needle and rosemary, based on measurement of the sedimentation of the halitosis ingredients through absorption analysis.

FIG. 10 is a graph showing the results of the Ames test of the mainstream smoke solid phase fraction (TPM) and the cell function effect evaluation results of the mainstream smoke solid phase fraction (TPM).

## BEST MODE FOR CARRYING OUT THE INVENTION

**[0030]** Hereinafter, embodiments are described in detail with reference to the accompanying drawings. However, since various changes may be made to the embodiments, the scope of the rights of the present application is not particularly limited to these embodiments. It should be understood that all changes, equivalents and substitutes to the embodiment are included in the scope of rights of the disclosure.

**[0031]** The terminology used herein is for the purpose of describing particular embodiments only and is not intended to limit the present disclosure. As used herein, the singular forms "a" and "an" are intended to include the plural forms as well, unless the context clearly indicates otherwise. It will be further understood that the terms "comprises", "comprising", "includes", and "including" when used in this specification, specify the presence of the stated features, numbers, steps, operations, elements, components or combinations thereof, but do not preclude the presence or addition of one or more other features, numbers, steps, operations, elements, components, and/or combinations thereof.

**[0032]** Unless otherwise defined, all terms including technical and scientific terms used herein have the same meaning as commonly understood by one of ordinary skill in the art to which this inventive concept belongs. It will be further understood that terms, such as those defined in commonly used dictionaries, should be interpreted as having a meaning that is consistent with their meaning in the context of the relevant art and will not be interpreted in an idealized or overly formal sense unless expressly so defined herein.

**[0033]** The same reference numbers in different figures represent the same or similar elements, and as such perform similar functionality. Further, descriptions and details of well-known steps and elements are omitted for simplicity of the description. Furthermore, in the following detailed description of the present disclosure, numerous specific details are set forth in order to provide a thorough understanding of the present disclosure. However, it will be understood that the present disclosure may be practiced without these specific details. In other instances, well-known methods, procedures, components, and circuits have not been described in detail so as not to unnecessarily obscure aspects of the present disclosure.

**[0034]** The present disclosure relates to a cigarette filter to which natural plant based granules are applied in order to reduce the cigarette-derived odor ingredient and intraoral halitosis inducing substance. According to an aspect, there is provided a cigarette filter including natural plant based granules composed of a natural plant material and a binder; and a filter portion to accommodate therein the natural plant based granules, in which the binder is hydroxy propyl methyl cellulose (HPMC), and a weight ratio of the natural plant material and the binder is in a range of 95 to 99% : 5 to 1%.

**[0035]** The present disclosure aims to provide a cigarette filter having granularized natural plant material added thereto to deliver a fresh and clean flavor evenly from the first sip to the last sip during smoking, and to neutralize and remove bad odors associated with smoking.

**[0036]** According to one embodiment, the natural plant material may include at least one selected from rosemary, pine needle, peppermint, spearmint, coffee, pineapple, chamomile, orange, eucalyptus, thyme, geranium, jasmine, rosemary, lavender, lemongrass, pine needle, clover, sage, taxol, bergamot, basil, thyme, valerian, hyssop, tea tree, myrrh, and juniper. However, the present disclosure is not limited thereto. The natural plant materials processed in accordance with the present disclosure may be applied alone or in combination with each other as long as the plants are allowed under the Food Additives Act. Plant parts containing the largest amount of natural scent ingredients (for example, leaf, stem, root, fruit, etc.) may be used in a natural state or may be processed in a suitable manner for application to the cigarettes.

**[0037]** In the following examples, pine needle and rosemary were selected as natural plant materials, and the ingredients and efficacy thereof were analyzed.

**[0038]** When the pine needle and rosemary based natural plant materials of the present disclosure are applied to the cigarette filter, the effect of reducing cigarette-derived halitosis ingredients and increasing the expression of natural plant flavor may be identified. Cigarette-derived halitosis ingredient reduction effect may be realized by chemical neutralization

of volatile fragrance ingredients such as terpenes, chemical reactions of fragrance ingredients such as polyphenol, and physical adsorption by granule porous mass and dietary fiber. The effect of increasing the expression of natural plant flavor may be implemented via the volatilization of the volatile fragrance ingredients. However, the disclosure is not limited thereto.

**[0039]** According to the present disclosure, the binder may hydroxy propyl methyl cellulose (HPMC) which may be easily coupled to the natural plant material and allow the size and the hardness of natural plant based granules to be proper. However, the disclosure is not limited thereto. The binder of the present disclosure may be one of hydrophilic polymer, hydrophobic polymer, monosaccharide, disaccharide, sugar alcohol, and hydroxy propyl methyl cellulose (HPMC). The hydrophilic polymer may be one selected from a group consisting of polyethylene glycol, polyethylene oxide, gelatin, starch, dextran sulfate, sugar, carboxymethyl cellulose (CMC), chitosan, hydroxy ethyl cellulose, hydroxy propyl methyl cellulose, methyl cellulose, arabinogalactan, polyvinyl alcohol, polyacrylic acid, polyvinylpyrrolidone, gum arabic and sodium alginate. In one example, the hydrophobic polymer may be one selected from the group consisting of ethyl cellulose, methacrylic acid polymer and esters thereof, polyethylene, polyamide, hydroxy propyl methyl cellulose phthalate, polyethylenevinyl acetate, cellulose nitrate, silicone and poly(lactide-co-glycolic acid). In one example, the monosaccharide may be one selected from the group consisting of glucose, fructose, tricellulose and galactose. The disaccharide may be one selected from the group consisting of xylose, D-mannose, sorbose, lactose, and maltose. Moreover, the sugar alcohol may be one selected from the group consisting of mannitol, sorbitol, xylitol, glycerin, glycerol and arabitol. The HPMC based binder may be tested under conditions of viscosity of 2, 15, 20, and 50 cps. Preferably, a binder with a viscosity of 50 cps is used.

**[0040]** In this connection, the content of the binder combined with the natural plant material is 1 to 5%, preferably 2%, so that the weight ratio of the natural plant material and the binder is 98% : 2%. In one example, when preparing the binder, extracts of various natural plants may be added thereto to improve the adsorption capacity. The alcohol concentration for extracting the extract may be 0 to 100%, preferably 50%. In one example, to increase the formability of natural plant based granules powder, inorganic binders selected from silica, silicate and bentonite may be further added thereto in a weight ratio of 1 to 30% based on the natural plant material. In one example, the natural plant based granules are prepared as wet or dry granules which are added to the filter so that the natural plant scent may be uniformly carried from the beginning to the end during smoking.

**[0041]** The natural plant based granules are preferably prepared as wet and dry granules, but may include extract, capsule, and mixture types.

**[0042]** The size of each of the natural plant based granules may be 0.25 to 2.0 mm, preferably, 0.5 to 1.0 mm. In this connection, when the size of the prepared natural plant based granules is smaller than 0.25 mm, in the cigarette filter preparation, a scattering (flying) amount of granules increases, thus causing filter contamination. Conversely, when the size of natural plant based granules is greater than 2.0 mm, it is difficult to ensure uniformity of the quality of the cigarette filter because a uniform amount of granules may not be added to the cigarette filter.

**[0043]** The moisture content of the natural plant based granules made under optimal granularization conditions is 5 to 12% of the total weight of the natural plant based granules, preferably 7 to 10%. If the granule has moisture content outside the above range, the granule may be broken, and the broken fine powder may cause filter contamination problems. Therefore, both productivity and quality of the cigarette filter to which the granule is applied may be deteriorated.

**[0044]** The hardness of the natural plant based granules is preferably 90.0 to 99.0%. When the weight ratio of the natural plant material and the binder is 98% : 2% and the hydroxy propyl methyl cellulose (HPMC) binder having the viscosity of 50 cps is used, the hardness of the natural plant based granules may be in a range of 90.0 to 99.0%. In this case, the granule applied to the cigarette filter is not broken or deformed, and further, a target amount of the granule to be injected into the cigarette filter may be injected. In one example, when the amount of the granule added to the cigarette filter is 3 mg/mm Tip, the maximum scent may be expressed, and the workability efficiency in applying the granule to the filter may be further improved.

**[0045]** The present disclosure relates to a cigarette filter including a filter portion that accommodates the natural plant based granules therein. The natural plant based granules are not added to the cigarette filler portion, but are added to the cigarette filter portion. In the conventional scheme where the natural plant based granules are added to the cigarette filler portion, when the cigarette is burned, the natural plant scent is deformed via thermal decomposition or synthesis, thereby to induce bad odors (irritating odor, fish odor, burnt odor, spicy odor, sulfur odor, egg odor). In accordance with the present disclosure, the natural plant based granules are added only to the cigarette filter portion, and are not added to the cigarette filler portion, such that the original natural plant flavor is delivered to smokers while the natural plant scent is not deformed via thermal decomposition or synthesis.

**[0046]** In one example, the cigarette filter according to the present disclosure may be a cigarette filter further including a herb oil capsule containing an oil ingredient extracted from a natural plant material. As a result, a cigarette filter with increased natural plant fragrance and enhanced fragrance retention may be provided. The herb oil capsule may include an extract extracted from pine needle and rosemary based natural plant material.

**[0047]** Pine needle extract includes pine, eucalyptus, rosemary, menthol, and MCTG as a pine needle natural oil, but

is not limited thereto.

**[0048]** Moreover, two types of rosemary extracts, that is, citrus based rosemary (Herb-401) and spicy based rosemary may be used as the rosemary extract. The citrus based rosemary may include orange, mandarin, cognac, ylang ylang, ling, genranium, menthol, benzyl bentoate, triethyl citrate, PG, and ethanol. The spicy based rosemary (Herb-402) may include mandarin, ylang ylang, rosemary, lavandin, vitiver, cassia, Jasmine, menthol, benzoate, triethyl citrate, PG, and ethanol. However, the present disclosure is not limited thereto.

**[0049]** The cigarette filter according to the present disclosure may be a single filter or the multiple filters.

**[0050]** The cigarette filter according to the present disclosure may be a 27 mm single filter. The single filter may cause the problem in which granules are exposed and released to the outside.

**[0051]** Moreover, the cigarette filter according to the present disclosure may be a multiple filter. The multiple filter system may include a triple complex filter system having the cavity filter portion, a 15 + 12 mm double complex filter system, and a 9 + 18 mm double complex filter system, but is not limited thereto. The preferred cigarette filter system according to the present disclosure may be selected based on the added amount of the natural plant based granules and workability at which the granules are applied to the cigarette filler side portion (TE portion) and the mouth side portion (ME portion). The preferable examples of the cigarette filter system of the present disclosure may include a cigarette filter of a 9 + 18 mm double complex filter system and a cigarette filter of a 15 + 12 mm double complex filter system, but may not be limited thereto.

**[0052]** Hereinafter, more detailed description of the present disclosure will be made with reference to the drawings. The drawings exemplified in the present disclosure are only examples, and the disclosure is not limited thereto, and it is obvious that they may be easily changed.

**[0053]** A cigarette filter according to an embodiment of the present disclosure will be described in detail with reference to FIG. 1 to FIG. 4.

**[0054]** FIG. 1 to FIG. 4 are cross-sectional views schematically showing a cigarette according to embodiments of the present disclosure, respectively.

**[0055]** The cigarette according to the present disclosure includes a cigarette filler portion 10 that is burned by fire and a cigarette filter portion 20 that filters cigarette smoke. The cigarette filler portion 10 and the cigarette filter portion 20 may be connected to each other by a tipping paper.

**[0056]** In the multiple filter, two or more filters may be included therein. For example, the description thereof is as follows. The cigarette filter portion 20 may include a mouth side portion (ME portion) 22, the filler side portion (TE portion) 21, and optionally a cavity portion 23. Further, each of the portions 21, 22 and 23 may include fibrous or filamentary acetate tow.

**[0057]** FIG. 1 is a schematic cross-sectional view of a cigarette including a cigarette filter of a 9 + 18 mm double complex filter system.

**[0058]** As shown in FIG. 1, the cigarette filter of the 9 + 18 mm double complex filter system of the present disclosure may include a filler side portion (TE portion) 21 made by adding natural plant based granules 31 to a fibrous or filamentary acetate tow, and a mouth side portion (ME portion) 22 composed of an acetate tow or a carbon fiber. In one example, the natural plant based granules 31 may be added to the mouth side portion (ME portion) 22.

**[0059]** Moreover, the cigarette filter of the 9 + 18 mm double complex filter system of the present disclosure may include a mouth side portion (ME portion, 9 mm) 22 made by adding a herb extract (flavoring liquid) 32 to fibrous or filamentary acetate tow, and a filler side portion (TE portion, 18 mm) 21 made by adding natural plant based granules 31 or herb extract (flavoring liquid) 32 (10  $\mu$ l) to the fibrous or filamentary acetate tow.

**[0060]** In one example, the natural plant based granules 31 may be injected on an unfolded acetate tow band in a free falling manner, and then the acetate tow band may be rolled into a rod form.

**[0061]** In one example, the herb extract may be different from the natural plant based granules and may be in a liquid or powder form. The herb extract may be a natural extract from a herb plant or a synthetic flavoring liquid.

**[0062]** In one example, the herb extract of the present disclosure may be added using a spraying method or a nozzle based spraying method (TJNS). The spraying method may be an acid bath based spraying method. Moreover, the herb oil capsules may be put into the filter using a capsule injection facility. The capsule injection facility may be a device disclosed in Korean Patent Application No. 10-2009-0101822, but is not limited thereto. Various types of devices to inject the capsules into the filter may be used.

**[0063]** The scheme of adding the herb extract or the herb oil capsule to the cigarette filter portion may be equally applied to a process of preparing another cigarette filter according to the present disclosure as described below.

**[0064]** FIG. 2 is a schematic view showing a cigarette including a cigarette filter of a triple complex filter system including a cavity filter portion.

**[0065]** As shown in FIG. 2, a filler side portion (TE portion) 21 may be made of a fibrous or filamentary acetate tow or carbon fiber.

**[0066]** In one example, a mouth side portion (ME portion) may contain natural plant based granules 31, and may further contain the herb extract (flavoring liquid) 32 in the fibrous or filamentary acetate tow.

**[0067]** In one example, the natural plant based granules 31, each having a size of approximately 0.5 to 1.0 mm may be included in a certain area of the cigarette filter portion 20 in a filled form. In this case, the acetate tow may not be included therein. The filter filled with only the natural plant based granules 31 and free of the acetate tow may be referred to as a cavity filter. FIG. 2 includes a cavity portion 23 filled with the natural plant based granules 31.

**[0068]** FIG. 3 is a cross-sectional view schematically showing a cigarette including a cigarette filter of a 15 + 12 mm double complex filter system (108 × 24.2 mm, PD 460 mm H<sub>2</sub>O).

**[0069]** As shown in FIG. 3, the cigarette filter of the 15 + 12 mm double complex filter system of the present disclosure may include a mouth side portion (ME portion, 15 mm) 22 having the herb extract (flavoring liquid) 32 added thereto, and a filler side portion (TE portion, 12 mm) 21 having the natural plant based granules 31 and the herb extract (flavoring liquid) 32 added thereto. In another example, the natural plant based granules 31 may be added to the mouth side portion (ME portion) 22.

**[0070]** FIG. 4 is a schematic cross-sectional view of a cigarette including a cigarette filter of another 15 + 12 mm double complex filter system (108 × 24.2 mm, PD 460 mm H<sub>2</sub>O).

**[0071]** As shown in FIG. 4, a herb oil capsule 33 may be added to the filler side portion (TE portion, 12 mm) of the cigarette filter of the 15 + 12 mm double complex filter system according to the present disclosure. Moreover, the cigarette filter of the 15 + 12 mm double complex filter system according to the present disclosure may include the filler side portion (TE portion, 12 mm) 21 having the herb oil capsule 33 added thereto, and a mouth side portion (ME portion, 15 mm) 22 having the herb extract (flavoring liquid) 32 having added thereto.

**[0072]** Moreover, the natural plant based granules 31 may be further added to the mouth side portion (ME portion, 15 mm) 22.

**[0073]** The present disclosure provides a cigarette including the cigarette filter as described above.

**[0074]** The cigarette according to the present disclosure may reduce methyl mercaptan as a halitosis inducing ingredient.

**[0075]** Moreover, the cigarette according to the present disclosure may reduce a cigarette odor inducing ingredient including ammonia, acrolein, crotonaldehyde, methyl ethyl ketone (MEK), 1,3-butadiene, acrylonitrile, benzene, and pyridine by 9 to 55%.

**[0076]** In following Examples 24-1 and 24-2, the physical properties, cigarette smoke ingredient, and halitosis inducing substance reducing ability of the cigarette according to the present disclosure were identified. Moreover, we identified fragrance ingredients related to halitosis reduction in mainstream smoke of the cigarette, and identified reducing ability for stinking and toxic ingredients in the mainstream smoke thereof.

**[0077]** It was identified that the cigarette including a cigarette filter containing rosemary extract and rosemary oil capsule as prepared in a preferred example, that is, Example 24-2 has the highest reducing effect of the halitosis inducing ingredient, that is, methyl mercaptan. Example is not limited thereto. Other Examples and characteristics tests of the cigarettes according to the present disclosure may refer to in detail Examples 24-1 and 24-2 below.

**[0078]** The cigarette filter according to the present disclosure may be prepared by a following preparation method.

**[0079]** There is provided a preparation method of a cigarette filter, the method including inserting a binder into a natural plant material to prepare natural plant based granules; drying and classifying the natural plant based granules; and adding the classified natural plant based granules to a cigarette filter, in which the binder is hydroxy propyl methyl cellulose (HPMC) of a viscosity of 50 cps, in which a weight ratio of the natural plant material and a binder is in a range of 95 to 99% : 5 to 1%.

**[0080]** The preparing of the natural plant based granules may include adding and dissolving 2% of hydroxy propyl methyl cellulose (HPMC) of a viscosity of 50 cps as a binder to 50% ethanol to produce a binder solution, and mixing pulverized natural plant powders and the produced binder solution with each other to form a mixture and stirring the mixture, and shaping and classifying the mixture.

**[0081]** The drying of the natural plant based granules may include one of freeze-drying, hot air drying, warm air drying, and fluidized bed drying. When applying the hot air drying, the natural plant based granules may be dried for 90 minutes to 300 minutes at a drying temperature of 40 degrees C to 70 degrees C, preferably, at a drying temperature of 45 degrees C for 90 minutes. In this case, the natural plant based granules with optimal granule physical properties and improved natural fragrance intensity may be obtained.

**[0082]** In the classifying of the natural plant based granules, a size of the classified natural plant based granule may be in a range of 0.25 to 2.0 mm and more preferably, 0.5 to 1.0 mm. When a size of the classified natural plant based granule is smaller than 0.25 mm, the scattering (flying) amount of the granules increases during the cigarette filter preparation, thereby causing filter contamination. Conversely, when the granule size exceeds 2.0 mm, a uniform amount of the granules may not be injected into the cigarette filter, thereby to make it difficult to ensure uniformity of the cigarette filter quality.

**[0083]** In the adding of the classified natural plant based granules to the cigarette filter, the natural plant based granules in the cigarette filter may be injected on an unfolded acetate tow band in a free falling manner. Then, the remaining granule is reused and added again thereto. In the meantime, the adding of the classified natural plant based granules

to the cigarette filter may include further adding a herb extract (flavoring liquid) to acetate tow using an acid bath based spraying method or a nozzle based spraying method (TJNS).

[0084] Moreover, the method of preparing the cigarette filter may further include injecting a herb oil capsule containing an oil ingredient extracted from the natural plant material into the cigarette filter using a capsule injection facility.

[0085] In the adding of the classified natural plant based granules to the cigarette filter, an added amount of the granules may be in a range of 0.5 to 4 mg/mm Tip. When the added amount of the granules is smaller than 0.5 mg/mm Tip, the natural scent expression is low. On the contrary, when the added amount thereof exceeds 4 mg/mm Tip, there may be a problem that the granules may not be added to the cigarette filter.

[0086] Hereinafter, the present disclosure will be described in more detail based on Experimental Examples and Examples. However, the Experimental Examples and Examples below are set forth only to help to understand the present disclosure, and do not limit the scope of the present disclosure.

### **Experimental Example 1: Analysis experiment of ingredients of natural plant material**

[0087] An experiment was conducted to quantify the index ingredients of pine needle and rosemary as natural plant materials.

#### **Index ingredient analysis experiment of pine needle**

[0088] An experiment was conducted to quantify the index ingredients of pine needle as a natural plant based raw material.

[0089] 50 ml of 80% methanol aqueous solution containing internal standard substance (Nicarbazine 40  $\mu$ g/ml) was added to 0.5 g of the natural plant material pine needle sample. The mixture was shaken at 200 rpm for 30 minutes, and was filtered using a 0.2  $\mu$ m syringe filter. Then, the filtered product was analyzed via LC/MS/MS. LC/MS/MS analysis conditions were as follows: a column was Phenomenex Luna C18[2] 5  $\mu$ m, 100A, 150  $\times$  2.0 mm, a column temperature was 20 degrees C, and a sample injection amount was 10  $\mu$ l. The filtering was performed using a 0.2  $\mu$ m syringe filter, and then LC/MS/MS analysis was performed. In one example, the LC/MS/MS analysis experiment was based on a scheme in which the retention time of the sample may be significantly changed while slightly and continuously changing the concentration of the organic solvent in a mobile phase. In particular, the mass was analyzed based on a gradient method which is a useful method for a short separation time of a wide range of a hydrophobic molecular sample having a larger elution time. Moreover, the organic solvent applied to the water-soluble filter (PVDF) in this experiment was 0.1% formic acid dissolved in water (H<sub>2</sub>O). The organic solvent applied to the oil-soluble filter (PTFE) was 0.1% formic acid dissolved in methanol (MeOH).

[0090] Based on a result of the experiment, kaempferol as the index ingredient of the pine needle was identified. In one example, recommended plants from which the index ingredient of the pine needle is obtained may be sea pine and Siberian pine but may not be limited thereto. The result of identifying the standardized quantification of the index ingredient of the pine needle via the LC/MS/MS analysis is shown in FIG. 5 (see FIG. 5).

#### **Analysis experiment of index ingredient of rosemary**

[0091] An experiment was conducted to quantify the index ingredients of rosemary as a natural plant based raw material.

[0092] 100  $\mu$ l of methanol solution was added to 0.1 g of the natural plant material rosemary sample. The mixture was shaken at 200 rpm for 60 minutes, and was filtered using a 0.2  $\mu$ m syringe filter. Then, 0.2 ml of the filtered solution was put into a 25 ml flask. 1 ml of the internal standard substance (Nicarbazine 1  $\mu$ g/ml) was added thereto. The product was analyzed via LC/MS/MS. The LC/MS/MS analysis conditions were applied in the same manner as in the pine needle ingredient analysis experiment.

[0093] As a result of the experiment, rosmarinic acid as the index ingredient of rosemary was identified. In one example, recommended plants from which the index ingredient of rosemary is obtained may be Tunisia and Moroccan rosemary, but are not limited thereto. FIG. 5 shows the standardized quantification of the rosemary's index ingredient via LC/MS/MS analysis (see FIG. 5).

### **Examples 1 to 11: Pine needle granule preparation**

#### **Pine needle granule preparation**

[0094] We prepared the pine needle granules of Examples 1 to 11.

[0095] The pine needle granules of Examples 1 to 11 were prepared by adding a binder to the raw material pine needle powers (leaf, stem, and root) and drying the mixture for 10 to 300 hours at 45 degrees C or 65 degrees C temperature



condition. The weight ratio of the pine needle powder and the binder was 98% : 2%.

**[0096]** The raw material pine needle (leaves, stems, and roots) was dried at room temperature to about 50 degrees C for 24 to 120 hours, and was pulverized using a grinder (pin crusher), and then classified with a 150  $\mu$ m sieve. Powders passing through the 150  $\mu$ m sieve were used. The binder employed a mixture of 2% hydroxy propyl methyl cellulose (HPMC) having one of viscosity 2, 15, and 50 cps, and 50% ethanol.

**[0097]** Among the pine needle granules of Examples 1 to 11, the pine needle granule of Example 6 had the most desirable physical properties and natural fragrance expression effect, and thus was prepared. The crushed pine needle powders were put into a flow shear mixer (Germany, Lodige company). A binder solution containing a mixture of 2% of hydroxy propyl methyl cellulose (HPMC) of the viscosity of 50 cps and 50% alcohol was added thereto and the mixture was stirred. The mixture of the pine needle powder and the binder was shaped and was classified using a sieve of 0.25 to 2.0 mm in diameter, preferably 0.5 to 1.0 mm. The classified mixture was subjected to hot air drying at 45 degrees C for 90 minutes. Thus, the pine needle granules of Example 6 having a size of 0.5 to 1.0 mm was prepared.

**[0098]** Moreover, in Examples 1 to 5 and Examples 7 to 11 in which the viscosity (cps) of hydroxy propyl methyl cellulose (HPMC) and the drying scheme of the granule, the drying temperature (degrees C), and the drying time duration (min) thereof varied, various pine needle granules were prepared (see Table 1).

#### **Experiment of properties of pine needle granules**

**[0099]** An experiment was conducted to identify the physical properties and natural fragrance expression effects of the pine needle granules prepared in Examples 1 to 11 (see Table 1).

**[0100]** It was identified that the pine needle granule prepared in Example 6 as a preferred example had physical properties of 98.1% hardness, 98.4% granularity, 0.431 g/cc filling density, and 7.9% moisture. The hardness of the pine needle granule prepared in Example 6 under optimal granularization conditions increased by 13.6% compared to that of the granule prepared in Example 1, and the total fragrance ingredient of the pine needle in Example 6 increased by about 60% compared to that of the granule in Example 11.

**[0101]** It was identified that the pine needle granule prepared in Example 6 as a preferred example had minimized moisture change in the cigarette filler and exhibited the greatest natural fragrance expression. However, the Example is not limited thereto. The physical properties and natural fragrance expression effect of the pine needle granules of Examples 1 to 5 and Examples 7 to 11 may also refer to Table 1 shown below.

[Table 1]

Identification experiment of physical properties and natural fragrance expression  
effect of pine needle granules

Granule preparation condition	Example	1	2	3	4	5	6	7	8	9	10	11
	Viscosity (cps) (%)	2	15	15	50	50	50	50	50	50	50	50
	Drying scheme	Hot air	Hot air	Hot air	Hot air	Hot air	Hot air	Hot air	Hot air	Hot air	Fluidized bed	Fluidized bed
	Drying Temp. (°C)	65	45	65	45	65	45	45	45	45	45	45
	Drying duration (min)	180	300	180	300	180	90	150	210	300	10	30
Pine needle	Hardness (%)	84.5	97.3	96.7	96.3	97.2	98.1	98.8	97.7		93.6	97.2
	Granularity (%)						98.4	99.3	98.0		99.6	99.6
	Filling density (g/cc)	0.434	0.465	0.447	0.489	0.496	0.431	0.408	0.393		0.404	0.417
	Moisture (%)	3.4	3.1	1.9	4.0	3.6	7.9	3.0	2.4		5.5	3.5
	Total fragrance ingredient	184	231	183	271	218	289	249	212	221	210	171

#### Examples 12 to 22: Rosemary granule preparation

##### Rosemary granule preparation

[0102] The rosemary granules of Examples 12 to 22 were prepared.

[0103] The conditions and methods of preparing the pine needle granule were applied in the same way to the rosemary granule preparation of Examples 12 to 22.

[0104] Among the rosemary granules of Examples 12 to 22, the rosemary granule of Example 17 had the most desirable physical properties and natural fragrance expression effect, and thus was prepared.

[0105] Moreover, in Examples 12 to 16 and Examples 18 to 22 in which the viscosity (cps) of hydroxy propyl methyl cellulose (HPMC) and the drying scheme of the granule, the drying temperature (degrees C), and the drying time duration (min) thereof varied, various rosemary granules were prepared (see Table 2).

##### Experiment of properties of rosemary granules

[0106] An experiment was conducted to identify the physical properties and natural fragrance expression effects of the rosemary granules prepared in Examples 12 to 22 (see Table 2).

[0107] It was identified that the rosemary granule prepared in Example 17 as a preferred example had physical

properties of 92.0% hardness, 99.0% granularity, 0.436 g/cc filling density, and 9.9% moisture. The hardness of the rosemary granule prepared in Example 17 under optimal granularization conditions increased by 20% compared to that of the granule prepared in Example 12, and the total fragrance ingredient of the rosemary in Example 17 increased by about 100% compared to that of the granule in Example 14.

**[0108]** It was identified that the rosemary granule prepared in Example 17 as a preferred example had minimized moisture change in the cigarette filler and exhibited the greatest natural fragrance expression. However, the Example is not limited thereto. The physical properties and natural fragrance expression effect of the rosemary granules of Examples 12 to 16 and Examples 18 to 22 may also refer to Table 2 shown below.

[Table 2]

Identification experiment of physical properties and natural fragrance expression effect of rosemary granules

Granule preparation condition	Example	12	13	14	15	16	17	18	19	20	21	22
	Viscosity (CPS) (%)	2	15	15	50	50	50	50	50	50	50	50
	Drying scheme	Hot air	Hot air	Hot air	Hot air	Hot air	Hot air	Hot air	Hot air	Hot air	Fluidized bed	Fluidized bed
	Drying Temp. (°C)	65	45	65	45	65	45	45	45	45	45	45
	Drying duration (min)	180	300	180	300	180	90	150	210	300	10	30
Rosemary	Hardness (%)	72.0	87.8	89.8	83.8	89.5	92.0			83.8		
	Granularity (%)						99.0					
	Filling density (g/cc)	0.310	0.369	0.355	0.333	0.373	0.436			0.333		
	Moisture (%)	2.8	3.9	3.4	4.1	3.6	9.9			4.1		
	Total fragrance ingredient		2028	1384	2105	1519	2970			1750		

#### Example 23: Cigarette filter preparation

**[0109]** The natural plant materials applied to the cigarette filters of Examples 23-1 and 23-2 were herb oil capsules of two types based on rosemary and pine needle, herb extract (flavoring liquid) of two types based on rosemary and pine needle, and natural plant based granules of two types based on rosemary and pine needle.

**[0110]** The natural plant based granules employed the pine needle granule of Example 6 and the rosemary granule

of Example 17 which have the highest physical properties and natural scent expression effect of the granule, among the natural plant based granules prepared in Examples 1 to 22, and the cigarette filter containing them was prepared.

#### **Example 23-1: 9 + 18 mm double complex filter preparation**

[0111] A cigarette filter of a double complex filter system was prepared which included a filler side portion (TE portion) 21 made by adding the natural plant based granules 31 to fibrous or filamentary acetate tow, and a mouth side portion (ME portion) 22 made of acetate tow or carbon fiber.

[0112] Moreover, a cigarette filter having the natural plant based granules 31 added to the mouth side portion (ME portion) 22 was prepared.

[0113] Moreover, a cigarette filter of a double complex filter system composed of the mouth side portion (ME portion, 9 mm) 22 made by adding the herb extract (flavoring liquid) 32 to the fibrous or filamentary acetate tow, and a filler side portion (TE portion, 18 mm) 21 made by adding the natural plant based granules 31 and the herb extract (flavoring liquid) 32 (10  $\mu$ l) to the fibrous or filamentary acetate tow was prepared (see FIG. 1).

#### **Example 23-2: Preparation of triple complex filter including cavity filter portion**

[0114] A cigarette filter of a triple complex filter system composed of the mouth side portion (ME portion) 22 made by adding the herb extract (flavoring liquid) 32 to the fibrous or filamentary acetate tow, a cavity portion 23 having the natural plant based granules 31 added thereto, and a filler side portion (TE portion) 21 made of the acetate tow or carbon fiber was prepared.

[0115] Moreover, a cigarette filter of a triple complex filter system in which the natural plant based granules 31 were added to the mouth side portion (ME portion) 22 was prepared (see FIG. 2).

#### **Example 23-3: 15 + 12 mm double complex filter preparation**

[0116] The natural plant materials applied to the cigarette filter of Example 23-3 were herb oil capsules of 2 types based on rosemary and pine needle (oil was Hanbit fragrance, and the capsule was prepared by Futuretech), the herb extracts (flavoring liquid) of 2 types based on rosemary and pine needle (which were prepared by Natural Solution), and one type of natural plant based granules (mixture of rosemary : pine needle = 2 : 8, which was prepared by Natural Way).

[0117] The natural plant based granules employed a mixture at 2 : 8 ratio of the pine needle granule of Example 6 and the rosemary granule of Example 17 which have the highest physical properties and natural scent expression effect, among the natural plant based granules prepared in Examples 1 to 22, and the cigarette filter containing them was prepared as follows.

[0118] A cigarette filter of a double complex filter system (108  $\times$  24.2 mm, PD 460 mm H<sub>2</sub>O) composed of a mouth side portion (ME portion, 15 mm) 22 having the herb extract (flavoring liquid) 32 added thereto, and a filler side portion (TE portion, 12 mm) 21 having the natural plant based granules 31 and the herb extract (flavoring liquid) 32 added thereto.

[0119] Moreover, the cigarette filter in which the natural plant based granules 31 were added to the mouth side portion (ME portion) 22 was prepared (see FIG. 3).

[0120] Further, a cigarette filter of a double complex filter system (108  $\times$  24.2 mm, PD 460 mm H<sub>2</sub>O) composed of a filler side portion (TE portion, 12 mm) having the herb oil capsule 33 added thereto, and a mouth side portion (ME portion, 15 mm) filled with fibrous or filamentary acetate tow was prepared.

[0121] Moreover, a cigarette filter in which the natural plant based granules 31 were further added to the mouth side portion (ME portion, 15 mm) was prepared.

[0122] A cigarette filter in which the herb extract (flavoring liquid) 32 was further added to each of the filler side portion (TE portion, 12 mm) and the mouth side portion (ME portion, 15 mm) (see FIG. 4).

[0123] The herb extract (flavoring liquid) 32 may be added to fibrous or filamentary acetate tow using an acid bath based spraying method or a nozzle based spraying method (TJNS).

[0124] Detailed descriptions of the preparation of the cigarette filter of the 15 + 12 mm double complex filter system may refer to a following table 3.

[Table 3]

Preparation of 15 + 12 mm double complex filter									
Examples		Raw materials							
		Flavoring liquid				TE portion (12mm)			
		ME portion (15mm)		TE portion(12mm)		Types	Added amount (mg)		
		Types	Added amount (mg)	Types	Added amount (mg)				
1	ACE-Du al ( Control)	TEC	8±2	TEC	8±2	-	-		
2	NG	TEC	8±2	TEC	8±2	Natural plant based granules	24±2		
3	RE+NG	TEC	8±2	TEC	8±2	Natural plant based granules	24±2		
		Rosemary extract	8±2	Rosemary extract	8±2				
4	PE+NG	TEC	8±2	TEC	8±2	Natural plant based granules	24±2		
		Pine needle extract	8±2	Pine needle extract	8±2				
5	CR+RE	TEC	8±2	TEC	8±2	Rosemary based oil capsule	14.6		
		Rosemary extract	8±2						
6	CP+PE	TEC	8±2					Pine needle based oil capsule	18.9
		Pine needle extract	8±2						
NG: Natural Plant Granule, RE: Rosemary Extract, PE: Pine Extract, RC: Rosemary capsule, PC: Pine capsule, ME: Mouth side portion, TE: Filler side portion									

#### Identification physical properties of cigarette filter of 15 + 12 mm double complex filter system

**[0125]** The physical properties of cigarette filter of 15 + 12 mm double complex filter as prepared in Example 23-3 were identified (see Table 4).

[Table 4]

complex identification result of physical properties of cigarette filter (15 + 12 mm double filter) as prepared in Example 23-3

Examples			Preparation configuration		Physical properties			
			portion	Raw materials	Weight	Circumference	EPD	Out of roundness
					mg	mm	mm WG	%
1	ACE-Dual (Control )	Control	ME portion	-	818	24.24	454	96.9
			TE portion	-				
2	NG	Test group	ME portion	-	880	24.12	460	97.5
			TE portion	Natural plant based granules				
3	RE+NG		ME portion	Rosemary extract	900	24.28	424	97.2
			TE portion	Rosemary extract + Natural plant based granules				
4	PE+NG		ME portion	Pine needle extract	895	24.26	413	96.9
			TE portion	Pine needle extract Natural plant+ based granules				
5	RE+RC		ME portion	Rosemary extract	870	24.26	435	96.3
			TE portion	Rosemary based oil capsule				
6	PE+PC		ME portion	Pine needle extract	879	24.28	428	96.3
			TE portion	Pine needle based oil capsule				

NG: Natural Plant Granule, RE: Rosemary Extract, PE: Pine Extract, RC: Rosemary capsule, PC: Pine capsule, ME: Mouth side portion, TE: Filler side portion EPD (Encapsulated Pressure Drop): Resistance of suction in smoking as measured after a perforation formed in an outer face of the filter was blocked

**Example 24: Cigarette preparation****Example 24-1: Preparation of cigarette including cigarette filter prepared in Example 23-1**

**[0126]** A cigarette was prepared by connecting a cigarette filter portion 20 prepared in Example 23-1 (9 + 18 mm double complex filter) and a secondary non-flavored filler portion 10 with each other (see FIG. 1).

**Experiment of cigarette characteristics**

**[0127]** The cigarette prepared in Example 24-1 was identified as having the following characteristics.

**Analysis of fragrance ingredient carrying related to halitosis reduction in cigarette smoke**

**[0128]** Using the cigarette prepared in Example 24-1, an experiment was conducted to analyze the carrying of the fragrance ingredient in the smoke based on the pine needle and rosemary.

**[0129]** Using a solid phase microextraction (SPME) method, the fragrance ingredients in each of pine needle and rosemary based smokes were tested.

**[0130]** To extract fragrance ingredients from smokes based on pine needle and rosemary, SPME fibers (PDMS/DVB) to be applied to 0.5 g of each sample of pine needle and rosemary were heated at 80 degrees C and were subjected to extraction for 30 mins. The process of pretreatment of the sample taking 5 minutes for fiber adsorption and 3 minutes for fiber removal proceeded. In one example, the fiber includes PA (polyacrylate), PDMS (polydimethylsiloxane), CAR (Carboxen)/PDMS, or PDMS/DVB (divinylbenzene), but is not limited thereto. PDMS/DVB (divinylbenzene) was used in this experiment. The fiber employed MPS (Multi Purpose Sampler) from GERSTEL and was injected to GC-MS (GC-MS in which 5973 MSD system and 5975C inert XL mass spectrometer from Agilent are connected to each other).

**[0131]** The analysis of the fragrance ingredient carrying in the smoke based on pine needle and rosemary was conducted via quantitative analysis after pretreatment of samples of pine needle and rosemary. During the sample pretreatment, the smoking employed 20 cigarettes and a 44 mm filter pad. We collected the cigarette smoke on the Cambridge filter pad using dry ice. An impinger was connected thereto and the smoke was subjected to extraction alternately for 1 hour with 20 ml methanol (MeOH), and then was subjected to filtration through a 0.45  $\mu$ m pore diameter PVDF filter.

**[0132]** Next, the pretreated sample was quantitatively analyzed via GC/MS, SIM mode analysis method.

**[0133]** It was identified based on a result of the experiment that the fragrance carrying ingredient in the smoke of pine needle included borneol, bonyl acetate, 1,8-cineol, delta-cadinene, alpha-cadinene, limonene, and benzaldehyde. It was identified based on a result of the experiment that the fragrance carrying ingredient in the smoke of rosemary included borneol, bonyl acetate, 1,8-cineol, beta-caryophyllene, alpha-humulene, delta-terpineol, and alpha-terpineol (see FIG. 6).

**[0134]** DB-Wax column (length 30 m, I.D. 0.25 mm, film thickness 0.25  $\mu$ m) from Agilent was used for efficient separation of fragrance ingredients from smokes of pine needle and rosemary in the quantitative analysis of fragrance ingredients in the smokes. Further, helium gas as inert gas was used as a carrier gas. A flow rate thereof was 1 ml/min, and the sample was injected into the instrument at an injection temperature of 250 degrees C in a splitless mode.

**[0135]** In order to detect fragrance ingredients in smokes of pine needle and rosemary, a temperature program of GC-MS was set such that the temperature was maintained at 35 degrees C for 10 minutes, then was raised to 100 degrees C at a rate of 2 degrees C/min, and then was raised to 200 degrees C at a rate of 1 degree C/min, and then was raised at a rate of 3 degrees C/min to 230 degrees C, and then was maintained for 10 minutes. The separated ingredient enters MS along with the helium gas. To detect this ingredient, the ion source was set to 250 degrees C, the transfer line was set to 240 degrees C, and the quadrupole was set to 150 degrees C. The ionization energy was 70 eV, which was commonly used in electron impact (EI).

**[0136]** On 90 days after preparing a cigarette including a cigarette filter to which natural plant material pine needle and rosemary based granules were applied, the cigarette was used to analyze the fragrance ingredients of pine needle and rosemary (see FIG. 7). Based on the experimental result, it was identified that some of the ingredients of the pine needle and of rosemary were carried in the smoke as shown in Table 5 below. Therefore, it was identified that the cigarette including the cigarette filter to which the natural plant material pine needle and rosemary based granules were applied had the possibility of expressing natural scent and imparting functionality.

[Table 5]

Analysis result of fragrance ingredient carrying in common smoke of pine needle and rosemary			
Examples	Content ( $\mu\text{g}/\text{pad}$ )		
	1,8-Cineol	Bornyl acetate	Borneol
NRP5-2(Pine needle granule)	4.425	0.005	0.067
NRP5-4 (Rosemary granule)	1.385	0.004	0.005

#### Identification of reduction of halitosis inducing substance

**[0137]** Using the cigarette prepared in Example 24-1, an experiment was conducted to identify reducing ability for the methyl mercaptan as a representative halitosis ingredient in the cigarette smoke.

**[0138]** Pine needle and rosemary based powders, granules, oils, and flavoring liquid, as well as pine needle based fragrance carrying ingredients, that is, alpha-pinene, bornyl acetate, and rosemary based fragrance carrying ingredients, that is, 1,8-cineol, and borneol were added to the cigarette filter. Thus, a cigarette including the cigarette filter was prepared to identify methyl mercaptan reducing ability.

**[0139]** A content of methyl mercaptan as used was 1 ppm [ $1 \mu\text{g}/\text{ml}$ ] 2 ml. When the pine needle and rosemary based powders and granules were used, 10 mg of the sample was extracted with 2 ml of water and was used. In one example, when using the oil and flavoring liquid based on pine needle and rosemary, 2 ml thereof was used. As for a single ingredient, 3 ppm ( $3 \mu\text{g}/\text{ml}$ ) of the sample was extracted with 2 ml of water and was used.

**[0140]** The analysis experiment of reducing ability for methyl mercaptan included a pretreatment process of the sample, and the analysis process under the headspace sampler condition and the GC/MS-SIM condition. First, 10 mg of pine needle and rosemary based powders and granules sample were put into a 20 ml vial, which was immersed in 2 ml of water for 1 hour. Next, 2 ml of 0.2 M potassium phosphate was added thereto and the pH was adjusted to 7.5. Thereafter, 2 ml of methyl mercaptan ( $1 \mu\text{g}/\text{ml}$ ) was added thereto and the vial was sealed, followed by stirring with a Vortex mixer for 5 seconds. In the headspace sampler, reaction occurred at 42 degrees C for 6 minutes, and methyl mercaptan isolated into headspace was directly analyzed via GC/MS. An area of methyl mercaptan obtained as a result of the GC/MS analysis was compared with that of the control to determine the reduction percentage.

$$\text{Deodorizing Activity (DA, \%)} = [\text{C-S}]/\text{C} \times 100$$

- C: methyl mercaptan area of control
- S: Methyl mercaptan area of test group
- See Tokita, F. et al. Nippon Nogeikagaku Kaishi 58(6), p585 to 589 [1984].

**[0141]** The GC/MS instrument based analysis conditions were set as follows. Headspace Sampler conditions were set as follows: culture temperature: 42 degrees C, culture duration: 6 minutes, and transfer liner temperature: 100 degrees C.

**[0142]** The GC/MS-SIM conditions were set as follows: DB-624(60 m, 0.25 mm, 0.25  $\mu\text{m}$ ) column was used; Oven Temp: 35 degrees C for 10 minutes; post run: 250 degrees C for 30 minutes. Injector/Interface Temp was 150 degrees C or 220 degrees C. For injection, Split 10:1, flow rate 1 ml/min, methyl mercaptan ion (m/z): quantitative ion 47; and qualitative ion 48 and 45.

**[0143]** Pine needle and rosemary based powders, granules, oil, and filter flavoring liquid, and fragrance carrying ingredients in smoke, that is, cineol, bornyl acetate, and borneol were added to the cigarette filter to identify methyl mercaptan reducing abilities, which are shown in Table 6 below (see FIG. 8). In one example, based on a result of the experiment, it was identified that the cigarette with the filter having the pine needle and rosemary based granules added thereto had the methyl mercaptan reducing ability. However, the direct analysis of cigarette was limited in detection due to the difference in smoking amounts.



[Table 6]

Identification result of reducing ability for methyl mercaptan as halitosis ingredient by cigarette having filter having pine needle and rosemary based granules added thereto			
Pine needle	Methyl mercaptan Reducing ability (%)	Rosemary	Methyl mercaptan Reducing ability (%)
Powder	99.7	Powder	99.6
Granule	99.7	Granule	99.6
Oil	72.0	Oil	57.0
Flavoring liquid	60.0	Flavoring liquid	-
$\alpha$ -Pinene	54.7	1,8-Cineol	51.9
Bonyl acetate	41.7	Borneol	30.7

#### Measurement of sedimentation of halitosis ingredient using absorbance analysis

[0144] Using the cigarette prepared in Example 24-1, the sedimentation of halitosis ingredient thereof was measured via absorbance analysis. To this end, an experiment was conducted to identify the reducing ability for the halitosis ingredient derived from oral microbes when smoking the cigarette with the filter having the pine needle and rosemary based substances added thereto.

[0145] The experiment for measuring the sedimentation of halitosis ingredients was as follows. Hydrogen sulfide in a volatile sulfur compound as mouth odor ingredient generated when cultivating oral bacteria in saliva was combined with iron in a liquid state such that iron sulfide (FeS) in a form of a black precipitate occurred. Thus, the sedimentation of halitosis ingredient was analyzed. First, whole saliva was incubated for 48 hours at 37 degrees C under anaerobic conditions. Then, 50 ml of oil based test substance or 20  $\mu$ l of a single ingredient was added to 0.5 ml cultured saliva. 0.05 g surfactant was added thereto. The saliva was mixed with 0.05 g ferrous sulfate II (FeSO<sub>4</sub>). The mixture was incubated for 24 hours at 37 degrees C under anaerobic conditions. Thus, absorbance of the culture was measured at 700 nm to determine sedimentation of halitosis ingredients.

[0146] Pine needle and rosemary based oil, flavoring liquid, fragrance carrying ingredient in smoke, that is, alpha-pinene, 1,8-cineol, bonyl acetate, and borneol were added to a cigarette filter to identify halitosis ingredient reducing ability thereof, which are shown in Table 7 below (see FIG. 9). In one example, based on a result of the experiment, we identified that reducing ability for the halitosis ingredient derived from oral microorganisms was exhibited by the cigarette with the filter having the pine needle and rosemary based substances. However, there was a limit in the detection in the direct analysis of cigarette due to the difference in smoking amount.

[Table 7]

Identification of halitosis ingredient reducing ability by cigarette including filter having pine needle and rosemary based oil, flavoring liquid, fragrance carrying ingredient in smoke added thereto			
Pine needle	Halitosis ingredient (Hydrogen sulfide) Reducing ability(%)	Rosemary	Halitosis ingredient (Hydrogen sulfide) Reducing ability (%)
Oil	19.2	Oil	48.5
$\alpha$ -Pinene	50.0	1,8-Cineol	37.0
Bonyl acetate	36.0	Borneol	45.0

#### Identification of reducing ability for stinking and toxic ingredient in cigarette smoke

[0147] Using the cigarette prepared in Example 24-1, an experiment was conducted to identify reducing ability for stinking and toxic ingredients in cigarette smoke.

[0148] 3 mg/mm Tip of each of rosemary and pine needle based granules was added to a cigarette filter for test. Based on a result of measuring the reduction percentage (%) of the stinking and toxic ingredients per tar in smoke, it was identified that ammonia, acrolein, crotonaldehyde, methyl ethyl ketone (MEK), 1,3-butadiene, acrylonitrile, benzene, and pyridine were reduced by 9 to 55%. Carbonyl ingredient as a cigarette odor inducing ingredient included in tar was acrolein, crotonaldehyde and methyl ethylene ketone (MEK). The volatile organic compounds include 1,3-butadiene,

acrylonitrile, and benzene. The semi-volatile organic compound is pyridine.

**[0149]** The results of analyzing reducing ability for the stinking and toxic ingredients in smoke by the cigarette including the filter to which pine needle and rosemary based granules were applied are shown in Table 8 below.

[Table 8]

Identification of reducing ability for stinking and toxic ingredients in smoke by cigarette including filter to which pine needle and rosemary based granules are applied								
Natural plant based granules (mg/mm Tip)		Carbonyl			VOCs			Semi-VOCs
	Ammonia (%)	Acrolein (%)	Crotonaldehyde (%)	MEK (%)	1,3-butadiene (%)	Acrylonitrile (%)	Benzene (%)	Pyridine (%)
NRP5-4 (Rosemary granule)	12.9	9.9	52.1	16.4	15.2	12.9	12.1	31.9
NRP5-2 (Pine needle granule)	19.2	1.3	54.1	16.9	9.9	9.4	10.8	20.4

#### Evaluation of characteristics and sensory properties sense of cigarette

**[0150]** Using the cigarette prepared in Example 24-1, identification of smoke carrying ingredient among fragrance ingredients, identification of hazardous ingredient in smoke, sense or taste evaluation, function evaluation, safety evaluation, and storage evaluation were performed.

**[0151]** Cigarette in which natural plant based granules and flavoring liquid were not added to the cigarette filter (control NRP5-1), cigarette in which pine needle based granule was added to the filter (NRP5-2), cigarette in which pine needle granule and flavoring liquid were added to the filter (NRP5-3), cigarette in which rosemary based granule was added to the filter (NRP5-4), and cigarette in which rosemary granule and flavoring liquid were added to the filter (NRP5-5) were tested in terms of the above items and the results are shown in Table 9 below.

[Table 9]

Comparison of characteristics and sensory properties of control group (NRP5-1 free of granule and flavoring liquid) and experimental groups (NRP5-2 to NRP5-5)

Examples			Smoke carrying ingredient among fragrance ingredients	Hazardous ingredient in smoke	Function evaluation	Taste or sense evaluation	Safety evaluation	Storage evaluation
Control	NRP5-1	No addition	⊙	⊙	⊙	⊙	⊙	⊙
Pine needle	NRP5-2	Granule	⊙	⊙	⊙	⊙	⊙	⊙
	NRP5-3	Granule + Flavoring liquid		⊙	⊙	⊙		⊙
Rosemary	NRP5-4	Granule	⊙	⊙	⊙	⊙	⊙	⊙
	NRP5-5	Granule + Flavoring liquid		⊙	⊙	⊙		⊙

**Taste or sense evaluation**

**[0152]** An experiment was conducted using the cigarette prepared in Example 24-1 to evaluate the taste or sense improvement of the cigarette and the effect of reducing the halitosis by the natural plant based granules.

**[0153]** A blind test was conducted on 48 experienced sensory evaluation panels, and scores were scored according to a 7-point scale. The average value was indicated in following Tables 10 to 13 based on the paired t test method. In one example, 48 people as the sensory evaluation panels were divided into four smoker panel groups of 12 people each. As a result of the test, it was identified that compared to the control, the rosemary granule-applied samples exhibited increase in preference of external appearance before smoking, reduced odor and taste during smoking, and increase in harmony and smoothness of throat, increase in freshness in taste after smoking and decrease in unpleasant remaining odor. Further, as a result of the test, it was identified that compared to the control, the pine needle granule-applied samples exhibited reduced odor and taste during smoking, and increase in smoothness of throat, increase in freshness in taste after smoking and decrease in unpleasant remaining odor.

[Table 10]

Comparison of control (NRP5-1, no added granule and flavoring liquid) and test group (NRP5-2, pine needle granule as added), and evaluation of cigarette taste or sense properties and halitosis reduction effect								
NRP5-1 vs NRP5-2_ Pine needle granule	NRP5-1		NRP5-2_ Pine needle granule		Mean of the differences	t	df	p-value
	(Mean)	(Std Dev)	(Mean)	(Std Dev)				
[Before smoking] Pack aroma preference	3.21	0.89	3.00	1.09	0.21	0.7483	11	0.470
[During smoking] Odor and taste	2.42	1.18	2.29	1.18	0.13	0.8971	11	0.389
[During smoking] Harmony between taste and fragrance	3.54	0.75	3.38	0.43	0.17	0.7418	11	0.474
[During smoking] smoothness of throat	3.42	0.87	3.96	1.08	-0.54	-3.2228	11	0.008
[After smoking] Satisfaction feel	3.17	0.69	2.96	0.54	0.21	1.1005	11	0.295
[After smoking] Freshness in taste	3.50	0.95	3.67	1.09	-0.17	-0.8866	11	0.394
[After smoking] Unpleasant remaining odor	2.63	0.74	2.29	0.86	0.33	1.6086	11	0.136

[Table 11]

Comparison of control (NRP5-1, no added granule and flavoring liquid) and test group (NRP5-3, pine needle granule and flavoring liquid as added), and evaluation of cigarette taste or sense properties and halitosis reduction effect								
NRP5-1 vs NRP5-3_ Pine needle granule + Flavoring liquid	NRP5-1		NRP5-3_ Pine needle granule + Flavoring liquid		Mean of the differences	t	df	p-value
	(Mean)	(Std Dev)	(Mean)	(Std Dev)				
[Before smoking] Pack aroma preference	2.67	1.05	3.17	1.23	-0.50	-1.2688	11	0.231
[During smoking] Odor and taste	2.13	1.21	2.67	1.59	-0.54	-2.1695	11	0.053

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(continued)

Comparison of control (NRP5-1, no added granule and flavoring liquid) and test group (NRP5-3, pine needle granule and flavoring liquid as added), and evaluation of cigarette taste or sense properties and halitosis reduction effect								
NRP5-1 vs NRP5-3_ Pine needle granule + Flavoring liquid	NRP5-1		NRP5-3_ Pine needle granule + Flavoring liquid		Mean of the differences	t	df	p-value
	(Mean)	(Std Dev)	(Mean)	(Std Dev)				
[During smoking] Harmony between taste and fragrance	3.67	0.83	2.92	0.67	0.75	3.3166	11	0.007
[During smoking] smoothness of throat	3.75	1.20	3.96	1.30	-0.21	-1.1637	11	0.269
[After smoking] Satisfaction feel	3.46	1.05	2.92	0.73	0.54	2.3147	11	0.041
[After smoking] Freshness in taste	3.79	1.10	3.83	1.42	-0.04	-0.1847	11	0.857
[After smoking] Unpleasant remaining odor	2.67	0.72	2.33	1.29	0.33	1.1725	11	0.266

[Table 12]

Comparison of control (NRP5-1, no added granule and flavoring liquid) and test group (NRP5-4, rosemary granule as added), and evaluation of cigarette taste or sense properties and halitosis reduction effect								
NRP5-1 vs NRP5-4_ Rosemary granule	NRP5-1		NRP5-4_ Rosemary_ granule		Mean of the differences	t	df	p-value
	(Mean)	(Std Dev)	(Mean)	(Std Dev)				
[Before smoking] Pack aroma preference	2.13	1.07	3.46	1.08	-1.33	-2.4795	11	0.031
[During smoking] Odor and taste	1.88	0.86	1.79	1.23	0.08	0.4316	11	0.674
[During smoking] Harmony between taste and fragrance	3.21	0.69	3.50	1.07	-0.29	-1.1683	11	0.267
[During smoking] smoothness of throat	4.04	1.27	4.33	1.42	-0.29	-1.1683	11	0.267
[After smoking] Satisfaction feel	3.17	0.81	3.04	0.78	0.13	1.0000	11	0.339
[After smoking] Freshness in taste	4.08	1.18	4.25	1.44	-0.17	-0.7154	11	0.489
[After smoking] Unpleasant remaining odor	2.50	0.48	2.38	0.88	0.13	0.3974	11	0.699

[Table 13]

Comparison of control (NRP5-1, no added granule and flavoring liquid) and test group (NRP5-5, rosemary granule and flavoring liquid as added), and evaluation of cigarette taste or sense properties and halitosis reduction effect								
NRP5-1 vs NRP5-5_ Rosemary granule + Flavoring liquid	NRP5-1		MRP5-5_ Rosemary granule + Flavoring liquid		Mean of the differences	t	df	p- value
	(Mean)	(Std Dev)	(Mean)	(Std Dev)				
[Before smoking] Pack aroma preference	2.25	0.97	3.25	0.78	-1.00	-2.2978	11	0.042
[During smoking] Odor and taste	2.00	0.60	2.17	0.91	-0.17	-0.6702	11	0.517
[During smoking] Harmony between taste and fragrance	3.17	0.62	3.13	0.64	0.04	0.1670	11	0.870
[During smoking] smoothness of throat	4.00	1.43	4.29	1.41	-0.29	-1.4656	11	0.171
[After smoking] Satisfaction feel	2.96	0.69	2.79	0.75	0.17	1.1728	11	0.266
[After smoking] Freshness in taste	4.13	1.19	3.92	1.44	0.21	1.1005	11	0.295
[After smoking] Unpleasant remaining odor	2.25	0.72	2.29	0.92	-0.04	-0.1723	11	0.866

**Example 24-2: Preparation of cigarette including cigarette filter prepared in Example 23-3**

**[0154]** A cigarette was prepared by connecting the cigarette filter portion 20 of the double complex filter system (108 × 24.2 mm, PD 460 mm H<sub>2</sub>O) prepared in Example 23-3 and the secondary non-flavored filler portion 10 to each other (see FIG. 4).

**Identification of physical properties of cigarette**

**[0155]** The physical properties of the cigarette prepared in Example 24-2 were identified. Based on the experiment result, it was identified that the physical properties of the test group and the control group were comparable to each other (see Table 14).

[Table 14]

2 Identification results of physical properties of cigarette prepared in Example 24-									
Examples			Weight	UPD	EPD	Air dilution percentage	Circum- ference	Out of roundness	Remarks
			mg	mm WG	mm WG	%	mm	%	
1	ACE- Dual	Control	890	59	160	87.2	24.62	96.13	-
2	NG	Test group	909	60	160	87.3	24.62	96.47	Natural plant based granules
3	RE+N G		910	54	149	88.0	24.57	96.10	Rosemary extract, Natural plant based granules

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(continued)

2 Identification results of physical properties of cigarette prepared in Example 24-									
Examples			Weight	UPD	EPD	Air dilution percentage	Circum-ference	Out of roundness	Remarks
			mg	mm WG	mm WG	%	mm	%	
4	PE+NG		909	52	145	88.5	24.60	96.13	Pine needle extract, Natural plant based granules
5	RE+RC		899	55	153	88.6	24.51	96.11	Rosemary extract, Rosemary based oil capsule
6	PE+PC		914	51	150	89.2	24.58	96.27	Pine needle extract, Pine needle based oil capsule
NG: Natural Plant Granule, RE: Rosemary Extract, PE: Pine Extract, RC:									

**[0156]** Rosemary capsule, PC: Pine capsule UPD (Unencapsulated Pressure Drop): Resistance of suction in smoking as measured while a perforation formed in an outer face of the filter was not blocked, EPD (Encapsulated Pressure Drop): Resistance of suction in smoking as measured after a perforation formed in an outer face of the filter was blocked

### Identifying of ingredients in smoke of cigarette

**[0157]** The ingredients in the smoke of the cigarette prepared in Example 24-2 were identified.

**[0158]** Based on the experiment result, it was identified that the ingredients in the smoke of the test group and the control group were similar to each other (see Table 15).

[Table 15]

Identification results of ingredients in smoke of cigarette										
Examples			TPM	Tar	nicotin e	CO	CO <sub>2</sub>	Puff	Moisture	Remarks
			mg/cig	mg/cig	mg/cig	mg/cig	mg/cig	No.	mg/cig	
1	ACE-Dual	Control	1.10	0.96	0.07	0.99	6.22	8.21	0.07	-

(continued)

Identification results of ingredients in smoke of cigarette										
Examples			TPM	Tar	nicotin e	CO	CO <sub>2</sub>	Puff	Moisture	Remarks
			mg/cig	mg/cig	mg/cig	mg/cig	mg/cig	No.	mg/cig	
2	NG	Test group	1.16	1.01	0.07	1.02	6.31	8.38	0.08	Natural plant based granules
3	RE+N G		1.15	1.00	0.08	0.99	6.16	8.28	0.07	Rosemary extract, Natural plant based granules
4	PE+N G		1.17	0.95	0.08	0.99	6.21	8.42	0.13	Pine needle extract , Natural plant based granules
5	RE+R C		1.08	0.94	0.08	0.94	6.08	8.35	0.07	Rosemary extract, Rosemary based oil capsule
6	PE+ PC		1.06	0.93	0.08	0.92	5.99	8.66	0.06	Pine needle extract , Pine needle based oil capsule
NG: Natural Plant Granule, RE: Rosemary Extract, PE: Pine Extract, RC: Rosemary capsule, PC: Pine capsule										

#### Identification of reducing effect for halitosis inducing substance

**[0159]** Using the cigarette prepared in Example 24-2, an experiment was conducted to identify volatile sulfur compounds (VSCs) as the main halitosis inducing substance.

**[0160]** The main ingredient of mouth odor is volatile sulfur compounds (VSCs), and smoking causes the concentration of VSCs in the mouth to rise. The amount of volatile sulfur compounds carried into the smoke is around 1%, but a threshold thereof is very low and the odor thereof is irritating, that is, is rotten onion and garlic odor.

**[0161]** Hydrogen sulfide (H<sub>2</sub>S) and methyl mercaptan (MM) were designated as analysis indicators to analyze halitosis inducing substances. The halitosis collection group included a total of 30 smokers who consumed Rejong Black cigarettes. Halitosis collection method was as follows: the smoker smoked one cigarette in each product and smoked for 2 minutes at 30 minutes intervals and the halitosis was collected (Tedlar Bag, 3L). The halitosis analyzer included a thermal adsorption and desorption system (TD-GC/MS) and PFPD (Purged Flow Photometric Detector); TD/GC-PFPD conforming to an odor process test standard (notified by the National Institute of Environmental Sciences).

**[0162]** Based a result of the experiment, hydrogen sulfide (H<sub>2</sub>S) was detected at a concentration of the threshold or less. Further, methyl mercaptan as the halitosis inducing ingredient was identified to be reduced in a cigarette including the filter having the natural plant material (rosemary and pine needle) added thereto. In particular, when the rosemary extract and rosemary oil capsule were applied thereto, methyl mercaptan decreased by 67.4%, and thus the reduction effect thereof was the highest. When the pine needle extract and pine needle oil capsule were applied thereto, the methyl mercaptan was reduced by 30.7% (see Table 16).

[Table 16]

Identification result of effect of reducing halitosis inducing ingredient methyl mercaptan (MM)					
Examples			Methyl mercaptan		Remarks
			Analysis value (ppb)	Reduction percentage (%)	
1	ACE-Dual	Control	0.22	-	-
2	NG	Test group	0.17	20.4	Natural plant based granules
3	RE+NG		0.18	15.5	Rosemary extract, Natural plant based granules
4	PE+NG		0.15	31.5	Pine needle extract , Natural plant based granules
5	RE+RC		0.07	67.4	Rosemaryextract ,Rosemarybased oil capsule
6	PE+ PC		0.15	30.7	Pine needle extract, Pine needle based oil capsule
NG: Natural Plant Granule, RE: Rosemary Extract, PE: Pine Extract, RC: Rosemary capsule, PC: Pine capsule					

#### Analysis result of carrying of fragrance ingredients related to reduction of halitosis in cigarette smoke

**[0163]** Based on analysis result of fragrance ingredients reducing the methyl mercaptan carried into mainstream smoke of the cigarette prepared in Example 24-2, it was identified that in the cigarettes respectively including respective filters respectively containing rosemary extract and rosemary oil capsule (RE+RC), and pine needle extract and pine needle oil capsule (PE+PC) which reduced the methyl mercaptan as shown in FIG. 16, bonyl acetate, 1,8-cineol, and terpene-based compound limonene having deodorizing effect, and b-caryophyllene were detected at a large amount (see Table 17).

[Table 17]

Analysis result of carrying of fragrance ingredients related to halitosis reduction in cigarette smoke								
Examples			Benzaldehyde	Limonene	1,8-cineol	Bonyl acetate	b-caryophyllene	Remarks
			μg/cig (STD)	μg/cig (STD)	μg/cig (STD)	μg/cig (STD)	μg/cig (STD)	
1	ACE-Dual	Control	n.d	1.3 (0.13)	n.d	n.d	n.d	-



(continued)

Analysis result of carrying of fragrance ingredients related to halitosis reduction in cigarette smoke								
Examples			Benzaldeh de	Limonen e	1,8-ci neol	Bonyl acetate	b- caryophll ene	Remarks
			μg/cig (STD)	μg/cig (STD)	μg/cig (STD)	μg/cig (STD)	μg/cig (STD)	
2	NG	Test group	n.d	1.3 (0.17)	n.d	n.d	n.d	Natural plant based granules
3	RE+NG		n.d	1.7 (0.30)	n.d	n.d	n.d	Rosemary extract, Natural plant based granules
4	PE+NG		n.d	1.8 (0.20)	n.d	n.d	n.d	Pine needle extract , Natural plant based granules
5	RE+RC		5.4 (0.23)	48.7 (4.55)	612.0 (22.7 4)	24.4 (4.66)	11.6 (2.04)	Rosemary extract, Rosemary based oil capsule
6	PE+PC		24.7 (3.18)	361.4 (3.85)	4.4 (0.38)	370.2 (18.25)	2.8 (0.11)	Pine needle extract, Pine needle based oil capsule
NG: Natural Plant Granule, RE: Rosemary Extract, PE: Pine Extract, RC: Rosemary capsule, PC: Pine capsule								

**Analysis result of stinking and toxic ingredients in cigarette smoke**

**[0164]** Based on analysis result of the reduction percentage per tar of stinking and toxic ingredients carried into mainstream smoke of the cigarette prepared in Example 24-2, it was identified that in the cigarettes respectively including respective filters respectively containing rosemary extract and rosemary oil capsule (RE+RC), and pine needle extract and pine needle oil capsule (PE+PC) which reduced the methyl mercaptan as shown in FIG. 16, stinking and toxic ingredients were reduced by 12.4 to 29.2% (see Table 18).

[Table 18]

Analysis result of stinking and toxic ingredients in cigarette smoke

Examples			Stinking ingredient		Toxic ingredients					
			Butyraldehyde		Crotonaldehyde		Isoprene		Benzene	
			Ingredient value μg/cig (STD)	Reduction percentage per tar (%)	Ingredient value μg/cig (STD)	Reduction percentage per tar (%)	Ingredient value μg/cig (STD)	Reduction percentage per tar (%)	Ingredient value μg/cig (STD)	Reduction percentage per tar (%)
1	ACE-Dual	Control	6.11 (0.42)	-	0.60 (0.04)	-	49.5 (2.71)	-	4.87 (0.46)	-
5	RE+RC	Test group	4.25 (0.64)	28.8	0.47 (0.07)	18.6	37.87 (3.73)	21.6	3.36 (0.20)	29.2
6	PE+PC		4.62 (0.64)	21.5	0.50 (0.04)	12.4	36.51 (3.70)	23.4	3.45 (0.18)	26.3

RE: Rosemary Extract, PE: Pine Extract, RC: Rosemary capsule, PC: Pine capsule

#### **Safety evaluation of cigarette including filter containing natural plant materials**

[0165] Using the cigarette prepared in Example 24-2, safety evaluation of the cigarette including the filter containing natural plant materials, that is, pine needle and rosemary was conducted.

#### **Evaluation criteria**

[0166] There is no safety problem of natural plant materials. Natural plant materials are added to non-burnable parts. Unlike a chemical substance composed of a single substance, a composition of the natural plant material is unclear. There is no use case in a cigarette thereof for the present purpose. Therefore, the evaluation was carried out in accordance with an in-house evaluation criteria for safety evaluation of additives to non-burned parts of tobacco.

#### **Evaluation of basic information**

[0167] Basic information about pine needle and rosemary as the natural plant material were evaluated. The two types of the natural plant materials are listed on the KFDA's list of usable food ingredients, and no toxicity such as carcinogenicity or reproductive toxicity of the raw material itself thereof has been reported. Moreover, there are no domestic and foreign regulations on the allowable amount of use thereof. There are no cases of use thereof in the cigarette by major other companies and the present company as the present applicant. However, it was identified that 'Pine Needle oil' and 'Rosemary oil' respectively extracted from pine needle and rosemary were used.

#### **Evaluation of contribution to cigarette smoke**

[0168] The contribution of the natural plant material to the cigarette smoke ingredient of the cigarette including the

filter to which the natural plant material was applied was evaluated via a Purge & Trap analysis method. As a result, toxicity information thereof other than two rosemary ingredients and one pine needle ingredient registered as food additives of the USFDA was insufficient. The toxicity was at a high level (> 5 ppm). The ingredient was not present in the cigarette smoke. It was determined that the contribution thereof to biological activity was required.

### **Biological activity evaluation**

**[0169]** The biological activity of the cigarette smoke ingredient of the cigarette including the filter to which the natural plant material was applied was evaluated. Based on the comparative analysis of the effects of cell function and genetic function by the control (product without granule) and the test group (product with granule), there was no significant difference ( $P < 0.05$ ) between the control and test groups. Therefore, it could be determined that the two types of the granules would have no effect on the increase or decrease in biological activity. FIG. 10 shows the results of the Ames test of the mainstream smoke solid phase fraction (TPM) and the cell function effect evaluation of the mainstream smoke solid phase fraction (TPM) (see FIG. 10).

**[0170]** Pine needle and rosemary natural plant materials are recognized as food raw materials (KFDA), and are limitedly applied to non-burnable areas of cigarettes. Individual ingredients thereof are present at a low concentration of 5 ppm or lower. The contribution thereof to the biological activity of the cigarette smoke is at a negligible level. Thus, when viewed comprehensively, it was determined that the pine needle and rosemary natural plant materials may be used in the filter at a current usage level.

### **Evaluation of storage ability of natural plant material containing cigarette**

**[0171]** Using the cigarette prepared in Example 24-1, the shelf-life or storage ability of the cigarette product having the filter containing the natural plant materials, that is, the pine needle and rosemary was verified.

**[0172]** The experiment was conducted for the purpose of determining the life cycle according to the application of the natural plant material based granules to the cigarette products. Natural plant material based granules 3 mg/mm Tip were applied to the cigarette filter, and the experimental conditions were divided into dry season and rainy season conditions. In case of the dry season condition, the temperature was set to 15 degrees C and humidity was set to 45% RH. In case of the rainy season condition, the temperature was set to 25 degrees C, and humidity was set to 59% RH. The cigarette product of Example 24-1 was prepared. After one month has elapsed, problems in terms of the moisture content of the cigarette product and the change in the shape of the product, and the change of microorganisms therein were not identified. Thus, we verified good storage ability of the cigarette product including the filter to which the natural plant material based granule was applied.

**[0173]** As described above, although the Examples have been described based on the limited drawings, various technical modifications and variations may be made based on the above teachings by the person who has ordinary knowledge in the relevant technical field. For example, appropriate results may be achieved even when the described steps are performed in a different order from the above described order, and/or the described components are combined into a form different from the above described form, and/or the described components are replaced or substituted with other components or equivalents thereto.

**[0174]** Therefore, other embodiments, other Examples, and those equivalent to the scope of the claims are within the scope of the claims to be described later.

### **Descriptions of main parts of the drawings**

**[0175]**

10: Cigarette filler portion 20: Cigarette filter portion  
21: Filler side portion (TE portion) 22: mouth side portion (ME portion) 23: cavity portion  
31: Natural plant based granules 32: Herb extract (flavoring liquid) 33: Herb oil capsule

### **Claims**

1. A cigarette filter comprising:

natural plant based granules composed of a natural plant material and a binder; and  
a filter portion to accommodate therein the natural plant based granules,  
wherein the binder is hydroxy propyl methyl cellulose (HPMC), and a weight ratio of the natural plant material

and the binder is in a range of 95 to 99% : 5 to 1%.

2. The cigarette filter of claim 1, wherein the natural plant material includes at least one selected from the group consisting of rosemary, pine needle, peppermint, spearmint, coffee, pineapple, chamomile, orange, eucalyptus, thyme, geranium, jasmine, rosemary, lavender, lemongrass, pine needle, clover, sage, taxol, bergamot, basil, thyme, valerian, hyssop, tea tree, myrrh, and juniper.
3. The cigarette filter of claim 1, wherein a hardness of the natural plant based granule is in a range of 90.0 to 99.0%.
4. The cigarette filter of claim 1, wherein a size of the natural plant based granule is in a range of 0.25 to 2.0 mm.
5. The cigarette filter of claim 1, wherein a moisture content of the natural plant based granule is in a range of 5 to 12% of a total weight of the natural plant based granules.
6. The cigarette filter of claim 1, wherein the cigarette filter is a single filter or a multiple filter.
7. The cigarette filter of claim 1, wherein the cigarette filter further includes a herb oil capsule containing an oil ingredient extracted from the natural plant material.
8. A cigarette comprising the cigarette filter of one of claims 1 to 7.
9. The cigarette of claim 8, wherein the cigarette reduces methyl mercaptan as a halitosis inducing ingredient.
10. The cigarette of claim 8, wherein the cigarette reduces a cigarette odor inducing ingredient including ammonia, acrolein, crotonaldehyde, methyl ethyl ketone (MEK), 1,3-butadiene, acrylonitrile, benzene, and pyridine by 9 to 55%.
11. A preparation method of a cigarette filter, the method comprising:
  - inserting a binder into a natural plant material to prepare natural plant based granules;
  - drying and classifying the natural plant based granules; and
  - adding the classified natural plant based granules to a cigarette filter, wherein in the preparing of the natural plant based granules, the binder is hydroxy propyl methyl cellulose (HPMC) of a viscosity of 50 cps, wherein a weight ratio of the natural plant material and the binder is in a range of 95 to 99%: 5 to 1%.
12. The method of claim 11, wherein the preparing of the natural plant based granules includes:
  - adding and dissolving 2% of a hydroxy propyl methyl cellulose (HPMC) of a viscosity of 50 cps as a binder to 50% ethanol to produce a binder solution;
  - mixing pulverized natural plant powders and the produced binder solution with each other to form a mixture and stirring the mixture; and
  - shaping and classifying the mixture.
13. The method of claim 11, wherein the drying of the natural plant based granules includes one of freeze-drying, hot air drying, warm air drying, and fluidized bed drying.
14. The method of claim 11, wherein a size of the classified natural plant based granule be in a range of 0.25 to 2.0 mm.
15. The method of claim 11, wherein the adding of the classified natural plant based granules to the cigarette filter includes adding a herb extract to acetate tow using a spray based spraying method or a nozzle based spraying (TJNS).
16. The method of claim 11, wherein the method further includes injecting a herb oil capsule containing an oil ingredient extracted from the natural plant material into the cigarette filter using a capsule injection facility.
17. The method of claim 11, wherein in the adding of the classified natural plant based granules to the cigarette filter, an added amount of the granules is in a range of 0.5 to 4 mg/mm Tip.

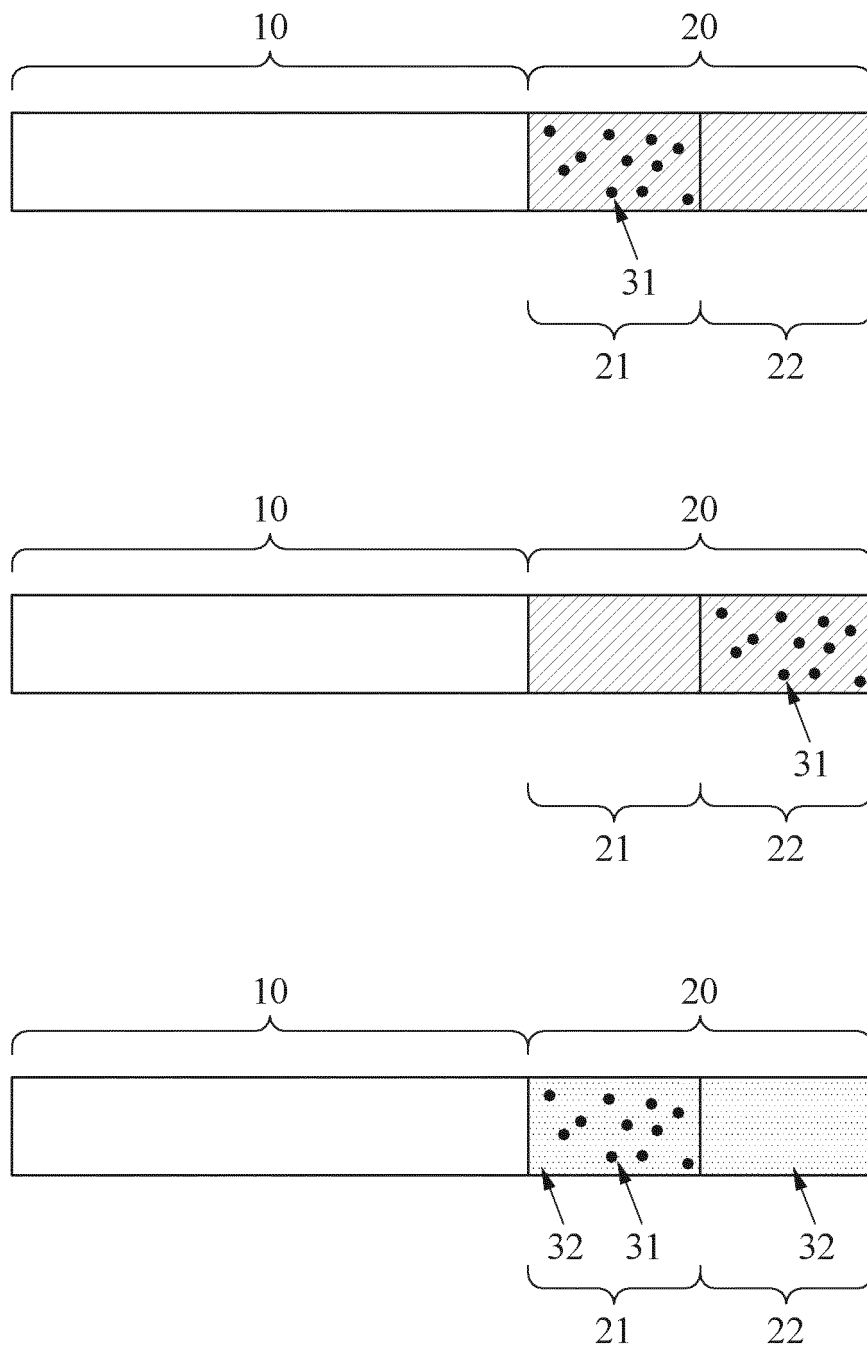


FIG. 1

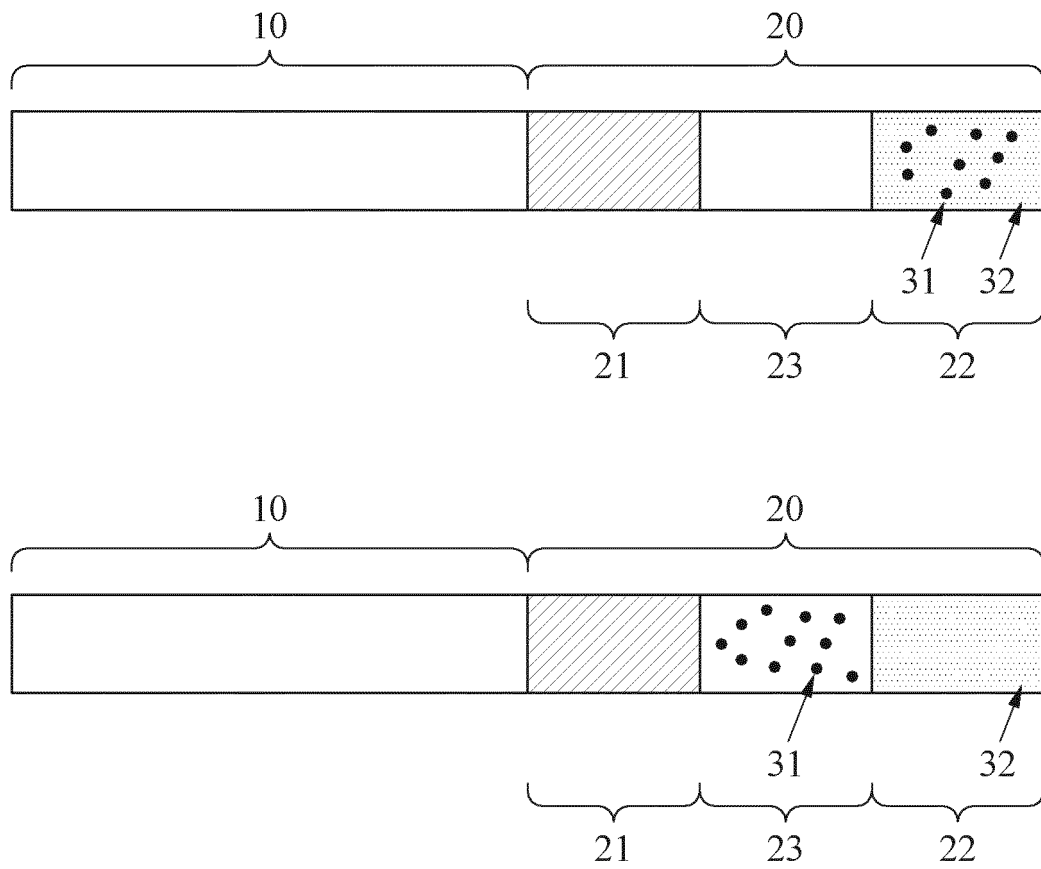


FIG. 2

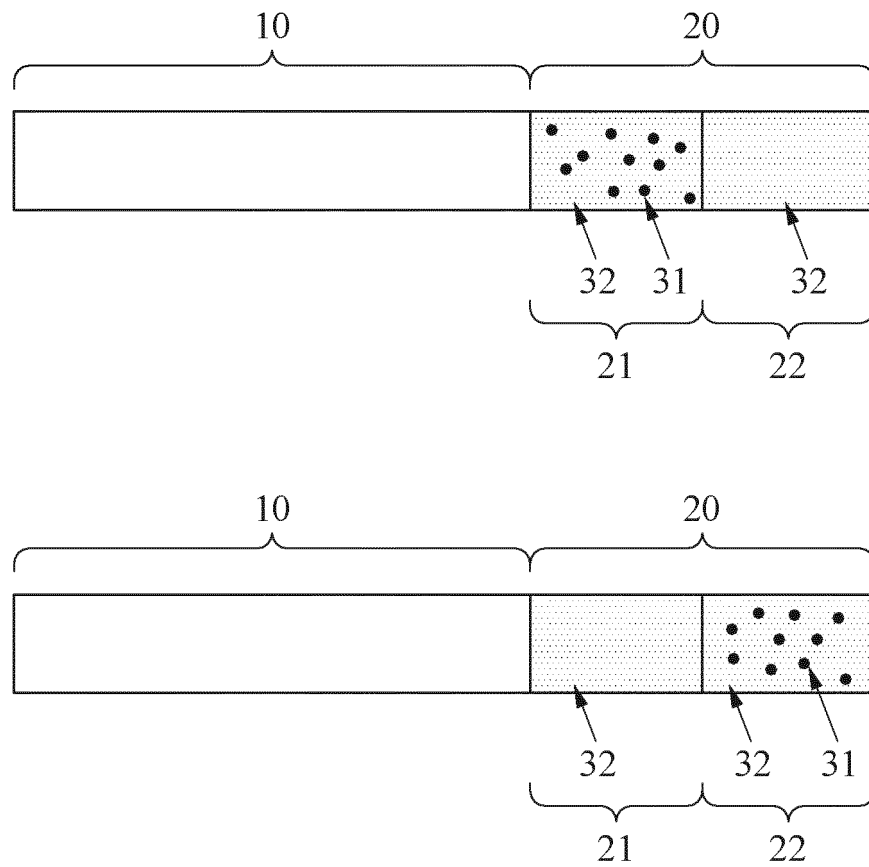


FIG. 3

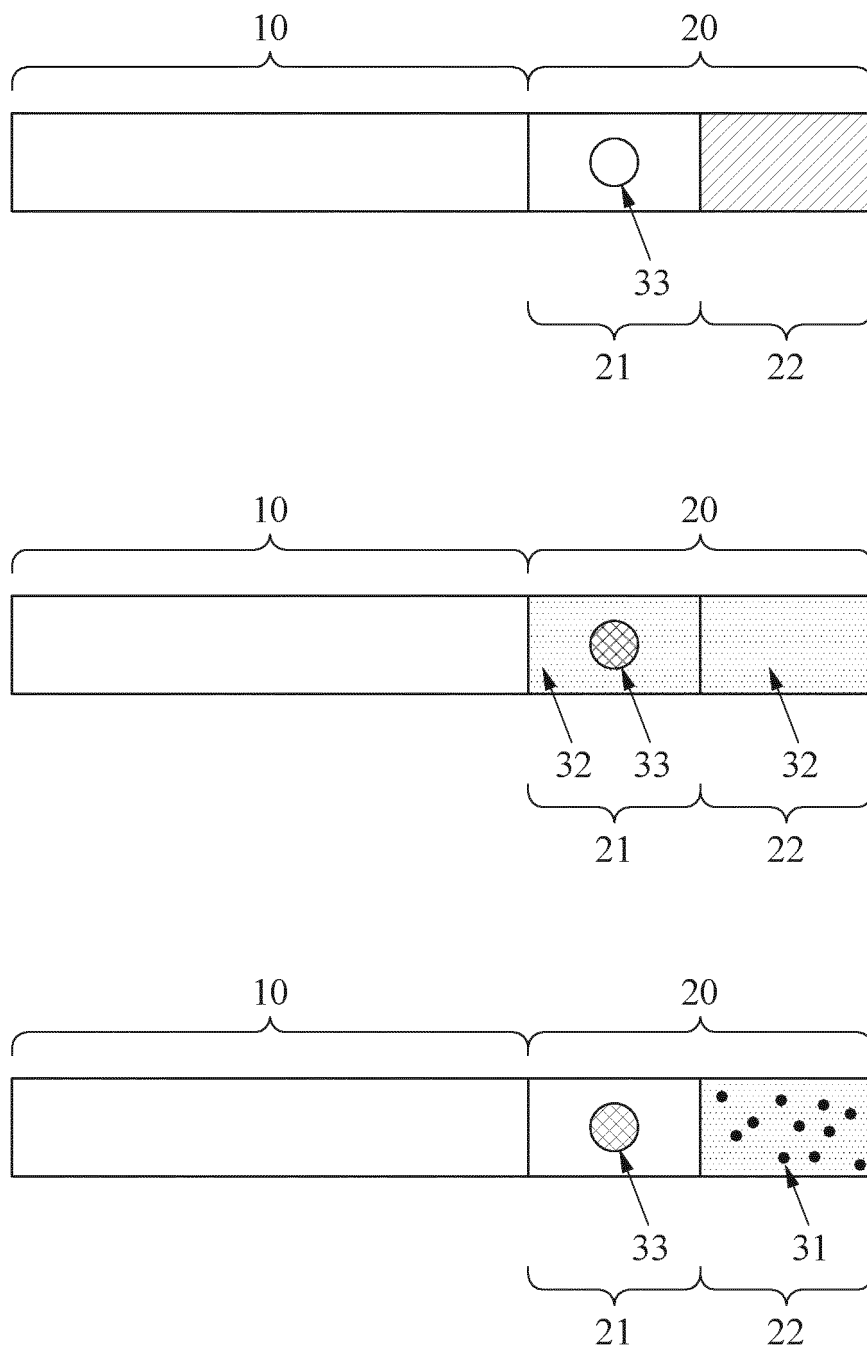


FIG. 4



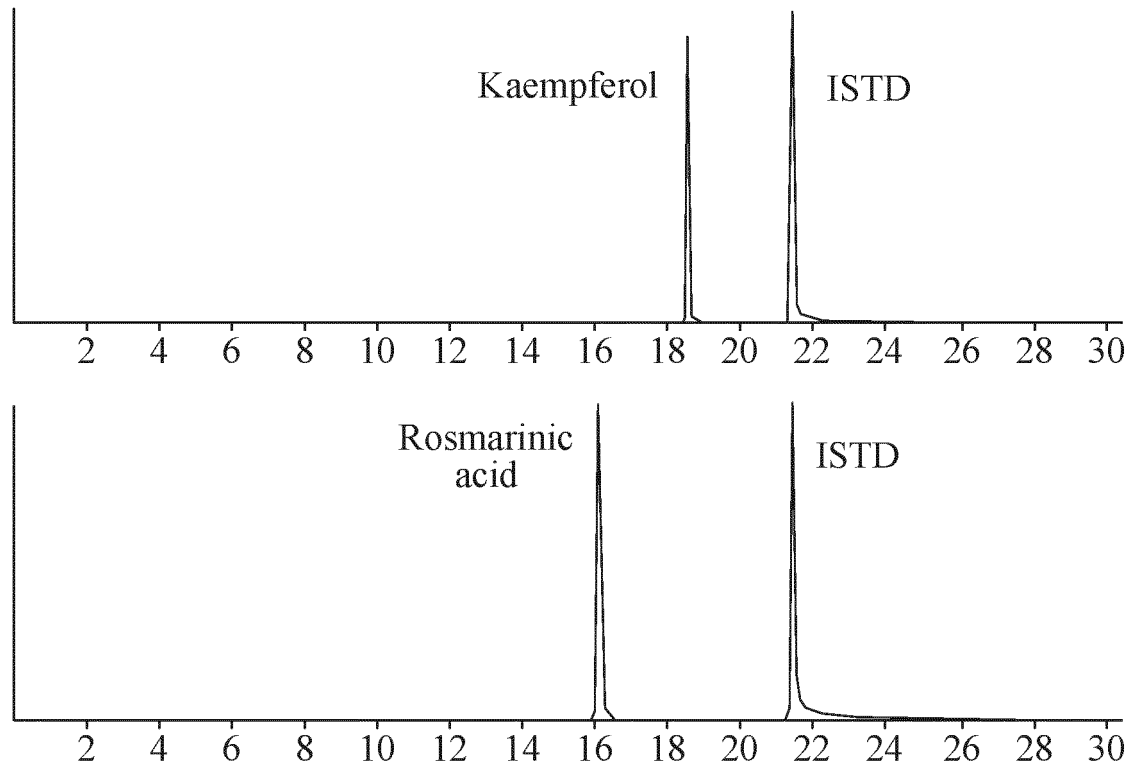


FIG. 5

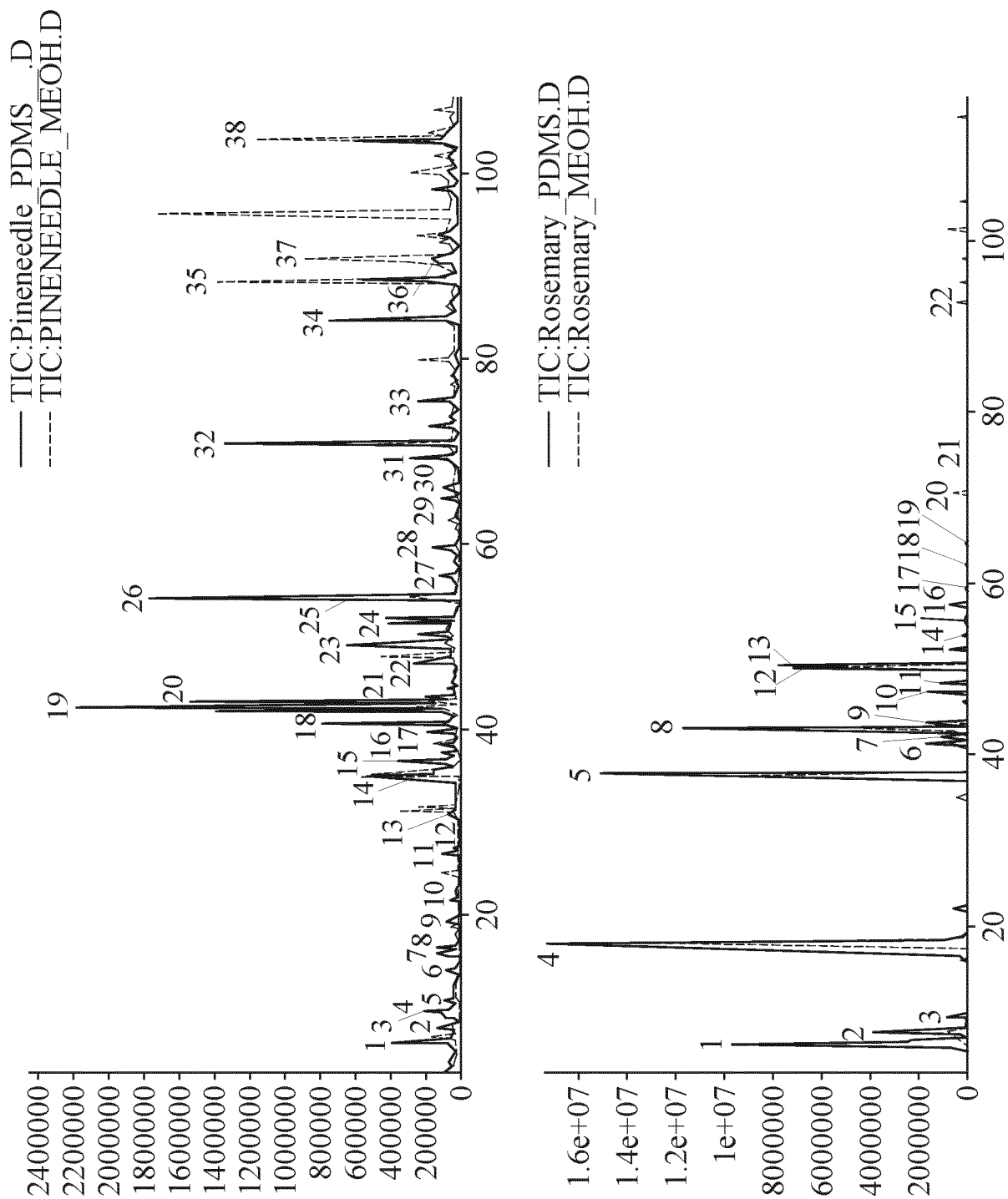


FIG. 6

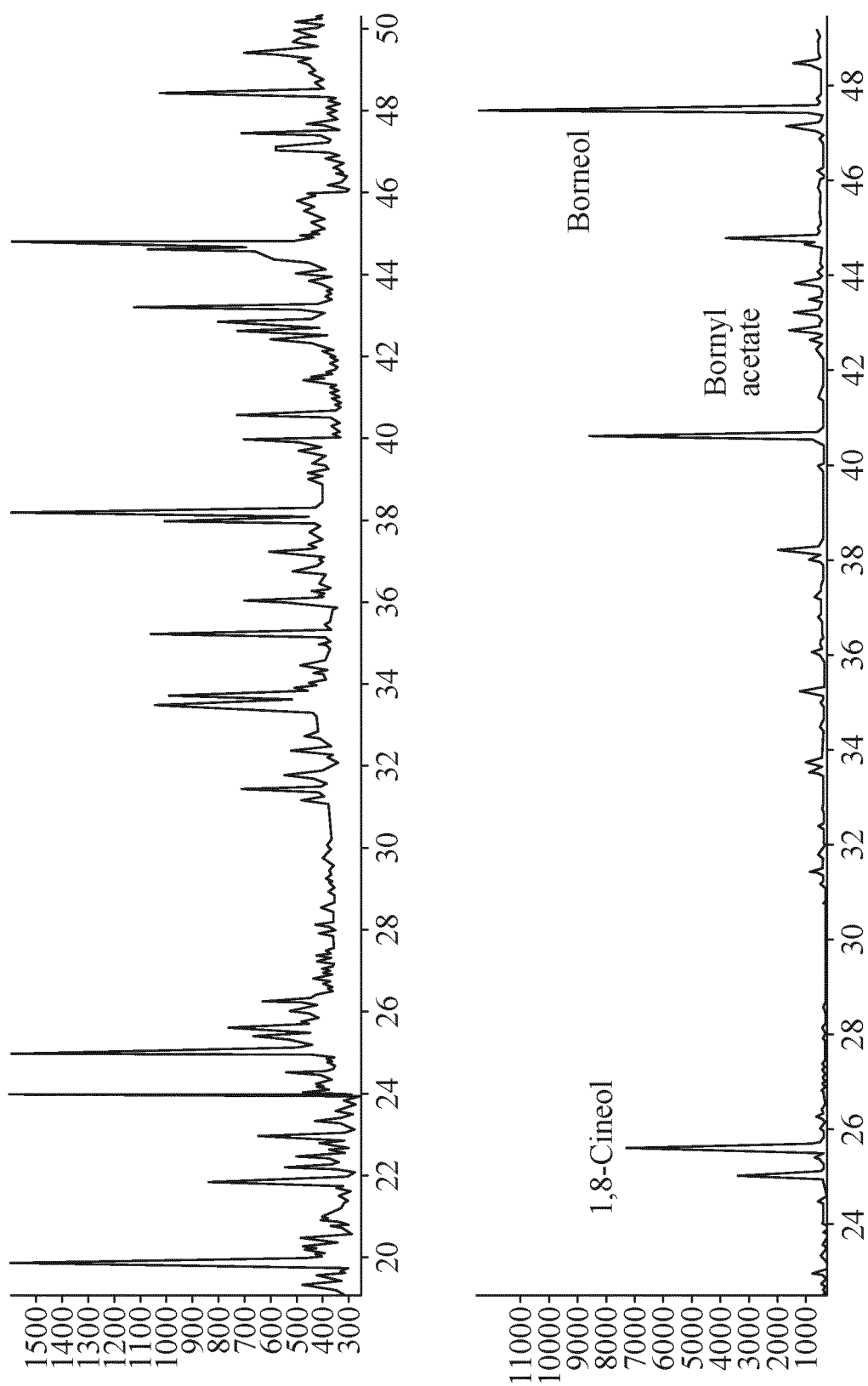


FIG. 7

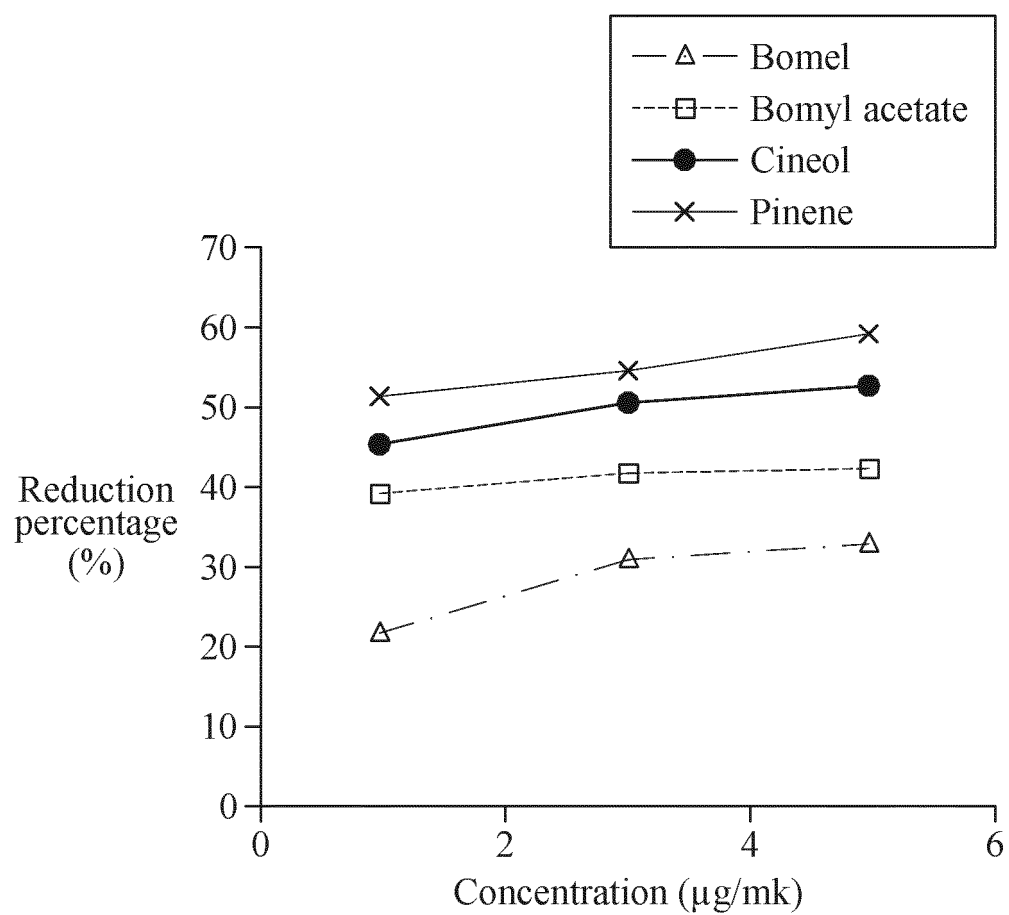


FIG. 8

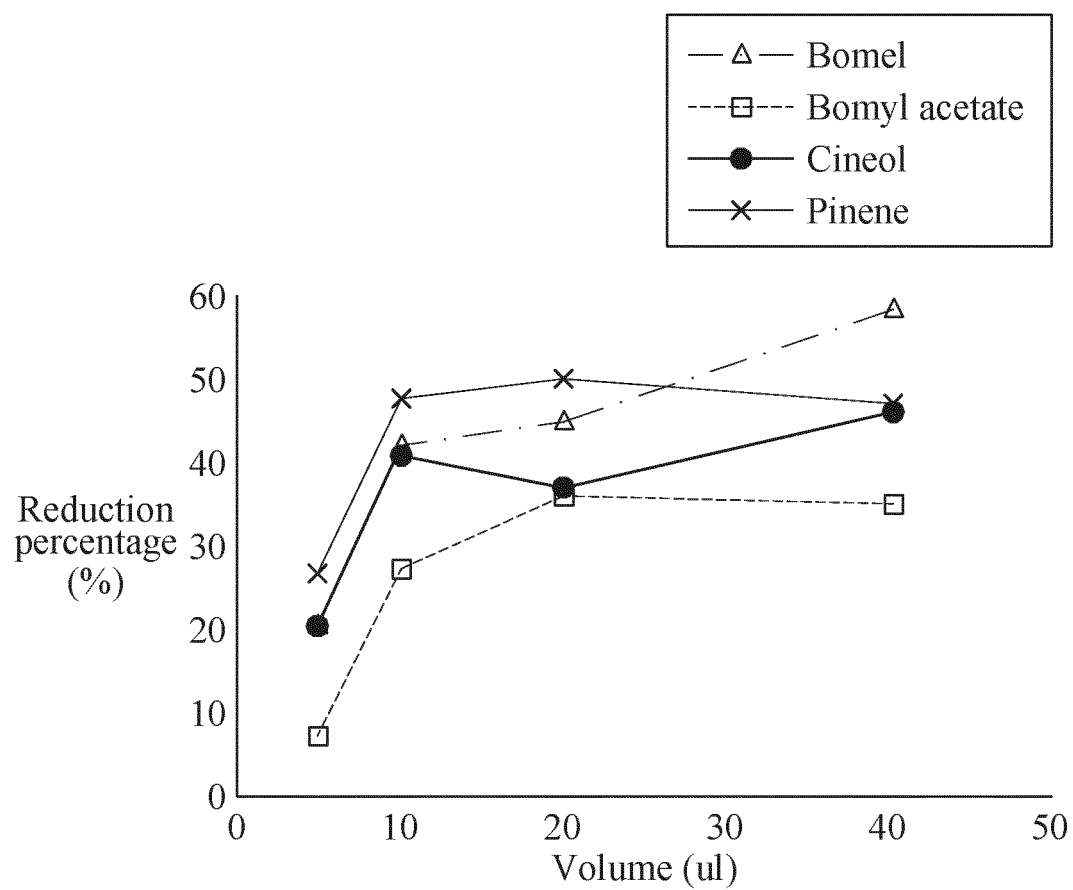


FIG. 9

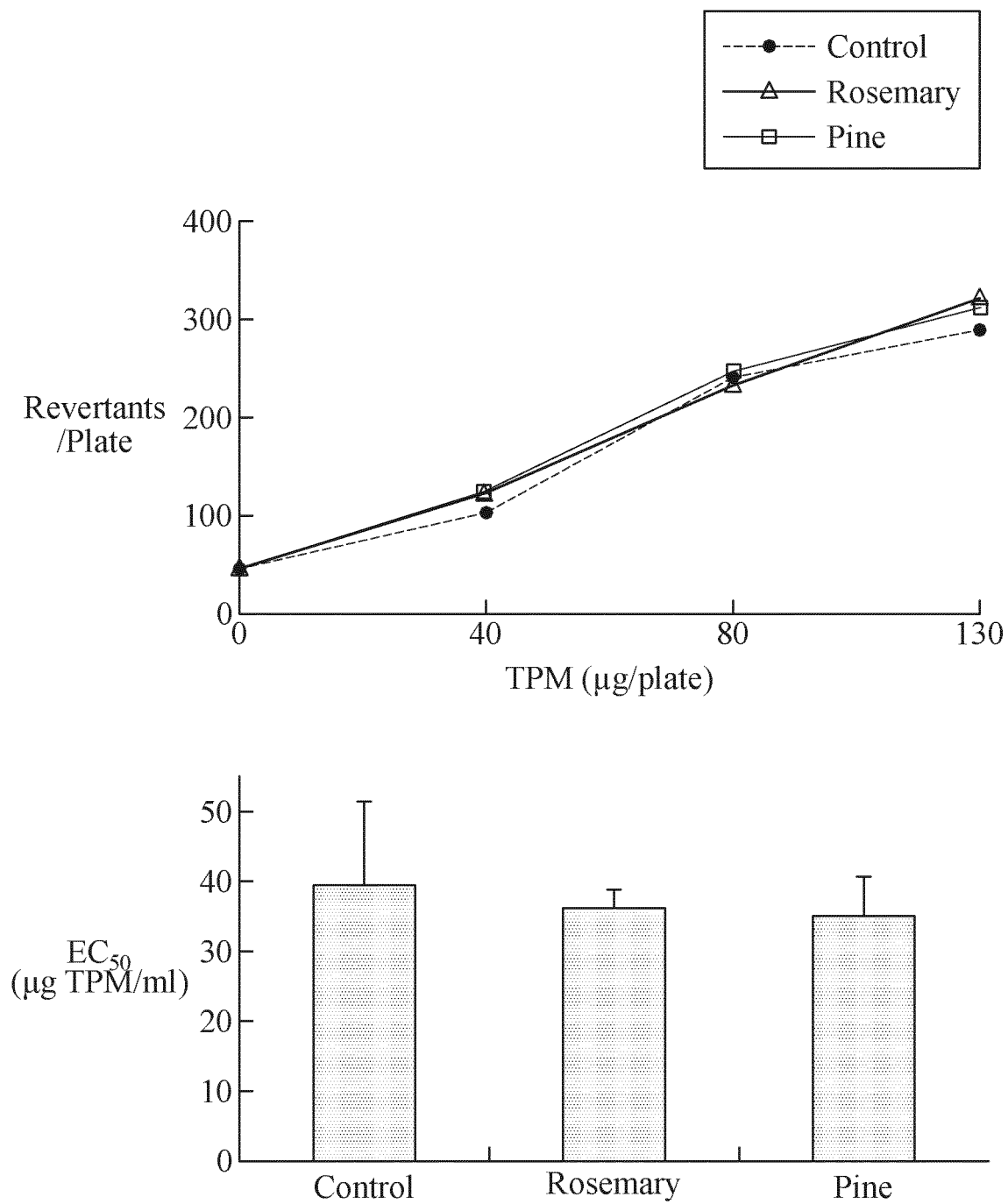


FIG. 10

## INTERNATIONAL SEARCH REPORT

International application No.

PCT/KR2020/012807

## A. CLASSIFICATION OF SUBJECT MATTER

A24D 3/06(2006.01)i; A24D 3/14(2006.01)i; A24D 3/10(2006.01)i; A24D 1/04(2006.01)i; A24D 3/02(2006.01)i;  
A24D 3/04(2006.01)i

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

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

A24D 3/06; A24D 3/02; A24D 3/04; A24D 3/10; A61K 8/00; A61Q 11/00; A24D 3/14; A24D 1/04

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

Korean utility models and applications for utility models: IPC as above

Japanese utility models and applications for utility models: IPC as above

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

eKOMPASS (KIPO internal) & keywords: 천연식물소재(natural plant material), 결합제(binder), 하이드록시프로필메틸 셀  
룰로오스(hydroxypropylmethyl cellulose), 담배필터(cigarette filter)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	KR 10-0984681 B1 (KT & G CORPORATION) 01 October 2010. See paragraphs [0025], [0046], [0047] and [0051]; and claims 1, 3, 10-12, 14 and 16.	1-17
Y	KR 10-2017-0085581 A (NANTONG CELLULOSE FIBERS CO., LTD.) 24 July 2017. See paragraphs [0001], [0002] and [0004]; and claims 1, 7, 19 and 20.	1-17
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A	KR 10-2012-0135425 A (CELANESE ACETATE, LLC) 13 December 2012. See entire document.	1-17

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

* Special categories of cited documents:	"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
"A" document defining the general state of the art which is not considered to be of particular relevance	"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
"D" document cited by the applicant in the international application	"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
"E" earlier application or patent but published on or after the international filing date	"&" document member of the same patent family
"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

12 January 2021

Date of mailing of the international search report

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**INTERNATIONAL SEARCH REPORT**  
**Information on patent family members**

International application No.

**PCT/KR2020/012807**

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**REFERENCES CITED IN THE DESCRIPTION**

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**Non-patent literature cited in the description**

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