

Description

OIL-IN-WATER TYPE PEST INSECT REPELLENT COMPOSITION

Field of the Invention

[0001]

The present invention relates to an oil-in-water type pest insect repellent composition.

Background of the Invention

[0002]

Pest insects, for example, mosquitoes, flies, and other flying pest insects, carry pathogens to animals, such as humans, and are factors which cause infectious diseases and dermatitis. In particular, some mosquitoes are hygienically very harmful insects because they carry pathogens, such as dengue fever, Zika fever, yellow fever, encephalitis, and malaria.

In order to protect themselves from such flying pest insects, for example, Patent Literature 1 (JP-A-2020-109075) discloses, for the purpose of providing an oil-in-water type pest insect repellent composition which has an excellent long-lasting repellent effect against pest insects, particularly flying pest insects, safety, a good feel when applied to the skin, and excellent long-term storage stability, an oil-in-water type pest insect repellent composition and an oil-in-water type pest insect stay inhibition composition, each containing (A)

13 mass% or more and 50 mass% or less of at least one non-volatile liquid oily component selected from the group consisting of silicone oils, ester oils, ether oils, hydrocarbon oils, aliphatic alcohols, and polyhydric alcohols, the non-volatile liquid oily component having a surface tension at 25°C of 40 mN/m or less and a viscosity at 23°C of 400 mPa·s or less as measured with a B-type rotational viscometer, (B) 0.5 mass% or more and 35 mass% or less of hydrophobic particles having a volume median particle size of 0.1 μm or more and 40 μm or less, and (C) 30 mass% or more and 86.5 mass% or less of water.

Summary of the Invention

[0003]

The present invention provides the following [1] to [4].

[1] An oil-in-water type pest insect repellent composition comprising the following components (A) to (E):

(A) 13 mass% or more and 50 mass% or less of at least one non-volatile liquid oily component selected from the group consisting of silicone oils, ester oils, ether oils, hydrocarbon oils, aliphatic alcohols, and polyhydric alcohols;

(B) 0.5 mass% or more and 35 mass% or less of hydrophilic inorganic particles containing silica

particles and/or silicate mineral particles and having a volume median particle size of 1 μm or more and 40 μm or less;

(C) 0.1 mass% or more and 3 mass% or less of at least one selected from the group consisting of polyether-modified silicone, polyglycerin-modified silicone, branched polyglycerol-modified silicone, alkyl glyceryl ether-modified silicone, and carboxy-modified silicone, all of which are liquid at 25°C;

(D) 0.05 mass% or more and 3 mass% or less of a thickener; and

(E) 30 mass% or more and 86.35 mass% or less of water,

wherein the component (A) has a viscosity at 23°C of 1 mPa·s or more and 400 mPa·s or less as measured with a B-type rotational viscometer, and the component (C) is other than the silicone oils as the component (A).

[2] A method for producing an oil-in-water type pest insect repellent composition, the method comprising a step of mixing a mixture containing the following components (B), (C), and (E) with a mixture containing the following components (A) and (D):

(A) at least one non-volatile liquid oily component selected from the group consisting of silicone oils, ester oils, ether oils, hydrocarbon oils, aliphatic alcohols, and polyhydric alcohols;

(B) hydrophilic inorganic particles containing

silica particles and/or silicate mineral particles and having a volume median particle size of 1 μm or more and 40 μm or less;

(C) at least one selected from the group consisting of polyether-modified silicone, polyglycerin-modified silicone, branched polyglycerol-modified silicone, alkyl glyceryl ether-modified silicone, and carboxy-modified silicone, all of which are liquid at 25°C;

(D) a thickener; and

(E) water,

wherein the component (A) has a viscosity at 23°C of 1 mPa·s or more and 400 mPa·s or less as measured with a B-type rotational viscometer, and the component (C) is other than the silicone oils as the component (A), and

wherein in the oil-in-water type pest insect repellent composition, the component (A) is contained in an amount of 13 mass% or more and 50 mass% or less, the component (B) is contained in an amount of 0.5 mass% or more and 35 mass% or less, the component (C) is contained in an amount of 0.1 mass% or more and 3 mass% or less, the component (D) is contained in an amount of 0.05 mass% or more and 3 mass% or less, and the component (E) is contained in an amount of 30 mass% or more and 86.35 mass% or less.

[3] A method for repelling pest insects, comprising applying an oil-in-water type pest insect repellent composition comprising the following components (A) to

(E) to a human skin surface:

(A) 13 mass% or more and 50 mass% or less of at least one non-volatile liquid oily component selected from the group consisting of silicone oils, ester oils, ether oils, hydrocarbon oils, aliphatic alcohols, and polyhydric alcohols;

(B) 0.5 mass% or more and 35 mass% or less of hydrophilic inorganic particles containing silica particles and/or silicate mineral particles and having a volume median particle size of 1 μm or more and 40 μm or less;

(C) 0.1 mass% or more and 3 mass% or less of at least one selected from the group consisting of polyether-modified silicone, polyglycerin-modified silicone, branched polyglycerol-modified silicone, alkyl glyceryl ether-modified silicone, and carboxy-modified silicone, all of which are liquid at 25°C;

(D) 0.05 mass% or more and 3 mass% or less of a thickener; and

(E) 30 mass% or more and 86.35 mass% or less of water,

wherein the component (A) has a viscosity at 23°C of 1 mPa·s or more and 400 mPa·s or less as measured with a B-type rotational viscometer, and the component (C) is other than the silicone oils as the component (A).

[4] A method for repelling pest insects, comprising attaching an oil-in-water type pest insect repellent

composition comprising the following components (A) to (E) to legs of a pest insect to prevent the pest insect from staying on a human skin:

(A) 13 mass% or more and 50 mass% or less of at least one non-volatile liquid oily component selected from the group consisting of silicone oils, ester oils, ether oils, hydrocarbon oils, aliphatic alcohols, and polyhydric alcohols;

(B) 0.5 mass% or more and 35 mass% or less of hydrophilic inorganic particles containing silica particles and/or silicate mineral particles and having a volume median particle size of 1 μm or more and 40 μm or less;

(C) 0.1 mass% or more and 3 mass% or less of at least one selected from the group consisting of polyether-modified silicone, polyglycerin-modified silicone, branched polyglycerol-modified silicone, alkyl glyceryl ether-modified silicone, and carboxy-modified silicone, all of which are liquid at 25°C;

(D) 0.05 mass% or more and 3 mass% or less of a thickener; and

(E) 30 mass% or more and 86.35 mass% or less of water,

wherein the component (A) has a viscosity at 23°C of 1 mPa·s or more and 400 mPa·s or less as measured with a B-type rotational viscometer, and the component (C) is other than the silicone oils as the component (A).

Detailed Description of the Invention

[0004]

The oil-in-water type pest insect repellent composition and the like described in Patent Literature 1 have a good feel when applied to the skin and good long-term storage stability. However, the consideration of the present inventors has revealed that there is room for further improvement in the long-lasting repellent effect.

The present invention relates to an oil-in-water type pest insect repellent composition which has a good feel when applied to the skin, excellent long-term storage stability, and further an excellent long-lasting repellent effect against pest insects, particularly flying pest insects, a method for producing an oil-in-water type pest insect repellent composition, and a method for repelling pest insects.

[0005]

As a result of extensive research, the present inventors have found that the above problem can be solved by an oil-in-water type pest insect repellent composition containing predetermined amounts of a specific non-volatile liquid oily component, hydrophilic inorganic particles containing silica particles and/or silicate mineral particles and having a specific volume median particle size, specific modified silicone which is liquid at 25°C, a thickener, and water.

[0006]

The present invention makes it possible to provide an oil-in-water type pest insect repellent composition which has a good feel when applied to the skin, excellent long-term storage stability, and further an excellent long-lasting repellent effect against pest insects, particularly flying pest insects, a method for producing an oil-in-water type pest insect repellent composition, and a method for repelling pest insects.

[0007]

A preferred embodiment of the present invention will be described below. In the present specification, "from X to Y" indicating the range means "X or more and Y or less." Further, in the present specification, the upper and lower limits of the numerical range can be combined in any way.

[0008]

[Pest insect Repellent Composition]

The pest insect repellent composition of the present invention (hereinafter also referred to simply as "the composition of the present invention") is an oil-in-water type composition containing the following components (A) to (E):

(A) 13 mass% or more and 50 mass% or less of at least one non-volatile liquid oily component selected from the group consisting of silicone oils, ester oils,

ether oils, hydrocarbon oils, aliphatic alcohols, and polyhydric alcohols;

(B) 0.5 mass% or more and 35 mass% or less of hydrophilic inorganic particles containing silica particles and/or silicate mineral particles and having a volume median particle size of 1 μm or more and 40 μm or less;

(C) 0.1 mass% or more and 3 mass% or less of at least one selected from the group consisting of polyether-modified silicone, polyglycerin-modified silicone, branched polyglycerol-modified silicone, alkyl glyceryl ether-modified silicone, and carboxy-modified silicone, all of which are liquid at 25°C;

(D) 0.05 mass% or more and 3 mass% or less of a thickener; and

(E) 30 mass% or more and 86.35 mass% or less of water,

in which the component (A) has a viscosity at 23°C of 1 mPa·s or more and 400 mPa·s or less as measured with a B-type rotational viscometer, and the component (C) is other than the silicone oils as the component (A).

[0009]

The composition of the present invention is an oil-in-water type composition containing specific amounts of components (A) to (E) and having a good feel when applied to the skin, excellent long-term storage stability, and

an excellent long-lasting repellent effect against pest insects, particularly flying pest insects.

[0010]

In the present invention, the "repellent effect against pest insects" includes a stay prevention effect whereby pest insects coming into contact with a target do not stay there but leave away immediately, and may further include one or more of keeping pest insects away from the target and exterminating pest insects.

The composition of the present invention at least exhibits an effect of repelling pest insects in such a manner that even if a pest insect, such as a mosquito, lands on a target to which the composition of the present invention is applied or attached, the pest insect does not stay there but leaves away immediately.

Specifically, the composition of the present invention has an effect of preventing pest insects, such as mosquitoes, from staying in a predetermined area on the skin surface of an animal, such a human, after landing thereon, for a time enough to pierce the animal, specifically one second or more. Such an effect is safe without side effects, such as skin irritation.

In the present invention, the "long-lasting repellent effect against pest insects" means that after the composition of the present invention is applied to a target, the repellent effect described above lasts even after a lapse of time. In the following description, the

long-lasting repellent effect particularly against flying pest insects in the present invention is simply referred to as "the long-lasting repellent effect."

[0011]

The composition of the present invention contains specific amounts of at least one non-volatile liquid oily component selected from the group consisting of silicone oils, ester oils, ether oils, hydrocarbon oils, aliphatic alcohols, and polyhydric alcohols as the component (A), hydrophilic inorganic particles containing silica particles and/or silicate mineral particles and having a volume median particle size of 1 μm or more and 40 μm or less as the component (B), at least one selected from the group consisting of polyether-modified silicone, polyglycerin-modified silicone, branched polyglycerol-modified silicone, alkyl glyceryl ether-modified silicone, and carboxy-modified silicone, all of which are liquid at 25°C, as the component (C), a thickener as the component (D), and water as the component (E).

The composition of the present invention has a good feel when applied to the skin, excellent long-term storage stability, and further an excellent long-lasting repellent effect against pest insects, particularly flying pest insects. The reason for this is unclear but considered as follows.

Flying pest insects, such as mosquitoes, have the characteristic of avoiding staying on the surface where

their legs get wet in order to avoid attracting force generated from the landing surface when the legs get wet. It is considered that when a flying pest insect lands on a skin surface to which the component (A) is applied or attached, its legs get wet, and at that time, the flying pest insect avoids attracting force generated from the landing surface and jumps off without staying at the landing point. In the composition, a portion of the component (C) is adsorbed onto the component (B), thereby improving the feel of the component (B) for the skin. In addition, a portion of the component (C) is mixed with the component (A), thereby imparting thixotropic properties to the component (A), preventing the component (A) from sagging due to gravity, and allowing the composition, when an external force is applied due to the landing of a flying pest insect, to wet and spread on its legs. As a result, it is considered that when the composition is applied to a skin surface, the uniform application of the composition to the skin surface is maintained for a longer period of time, resulting in a better feel and improving the long-lasting repellent effect. Further, due to the inclusion of a specific amount of the component (D) and the like, the composition of the present invention is considered to also have excellent long-term storage stability.

The term "land" in the present invention indicates that a flying pest insect, such as a mosquito, is in

contact with the target for less than one second. In addition, the term "stay" in the present invention indicates that a flying pest insect, such as a mosquito, stays in contact with the target for one second or more.
[0012]

<Component (A): Non-Volatile Liquid Oily Component>

The component (A) contained in the composition of the present invention is at least one non-volatile liquid oily component selected from the group consisting of (a) silicone oils, (b) ester oils, (c) ether oils, (d) hydrocarbon oils, (e) aliphatic alcohols, and (f) polyhydric alcohols.

However, the component (C) is other than the silicone oils as the component (A).

[0013]

The component (A) is preferably liquid at 25°C, more preferably liquid at 20°C, further more preferably liquid at 15°C, and further more preferably liquid at 10°C, from the viewpoint of facilitating application to the skin and improving the long-lasting repellent effect.

The term "liquid" in the liquid oily component means that the viscosity at 23°C as measured with a B-type rotational viscometer is 120 000 mPa·s or less.

The component (A) is preferably a poorly water-soluble or water-insoluble component, from the viewpoint of improving the long-lasting repellent effect. Specifically, the amount of the component (A) dissolved

in 100 g of water at 20°C is preferably 1 g or less, more preferably 0.5 g or less, further more preferably 0.1 g or less, and even more preferably substantially 0 g.

[0014]

The liquid oily component as the component (A) used in the present invention is a non-volatile liquid oily component, from the viewpoint of improving the long-lasting repellent effect. The non-volatile liquid oily component in the present invention refers to a component having a volatilization rate of 50% or less after drying at 1 atm at 25°C for 60 minutes, and preferably a component having a volatilization rate of 50% or less after drying at 1 atm at 25°C for 120 minutes. The evaluation of the volatilization rate is measured in accordance with German test standard DIN 53249, and specifically performed by the following procedures 1 to 4.

1. The weight (P (g)) of a round filter paper with a diameter of 150 mm is measured.

2. A sample of 0.3 g of the component (A) is dropped on the round filter paper using a pipette, and the weight (W₀ (g)) of the filter paper is measured immediately after dropping.

3. The weight of the filter paper is measured by weighing with an accuracy of about 0.001 g at 25°C in 5-minute intervals in the absence of ventilation. After

measuring 2, the weight of the filter paper measured after a lapse of 60 minutes is regarded as W_{60} (g).

4. A value derived from the calculation formula $\{((W_0-P)-(W_{60}-P))/(W_0-P)\} \times 100$ is regarded as the volatilization rate (%).

As the volatilization rate is lower, the liquid oily component remains for a longer period of time when applied to the skin; thus, the long-lasting repellent effect becomes higher.

When two or more liquid oily components are used, the volatilization rate of the liquid oily components refers to that of a mixture of the two or more liquid oily components. Therefore, liquid oily components having a volatilization rate of more than 50% after drying at 1 atm at 25°C for 60 minutes may be used in combination as long as the volatilization rate of the mixture is within the above range.

[0015]

The surface tension at 25°C of the component (A) is preferably 15 mN/m or more, and more preferably 17 mN/m or more, from the viewpoint of availability, and is preferably 40 mN/m or less, more preferably 30 mN/m or less, further more preferably 28 mN/m or less, even more preferably 25 mN/m or less, even more preferably 23 mN/m or less, and even more preferably 21 mN/m or less, from the viewpoint of improving the long-lasting repellent effect. When two or more liquid oily components with

different surface tensions are used, the surface tension refers to that of a mixture of these liquid oily components.

The viscosity at 23°C of the component (A) as measured with a B-type rotational viscometer is 1 mPa·s or more and 400 mPa·s or less, more preferably 300 mPa·s or less, further more preferably 210 mPa·s or less, even more preferably 100 mPa·s or less, even more preferably 60 mPa·s or less, even more preferably 40 mPa·s or less, and even more preferably 30 mPa·s or less. When two or more liquid oily components with different viscosities are used, the viscosity refers to that of a mixture of these liquid oily components.

The viscosity at 23°C of the component (A) as measured with a B-type rotational viscometer is preferably equal to or more than the lower limit value described above, from the viewpoint of suppressing volatility and improving the long-lasting repellent effect, and is preferably equal to or less than the upper limit value described above, from the viewpoint of ensuring long-term storage stability.

The surface tension and viscosity at 23°C as measured with a B-type rotational viscometer of the component (A) are measured by the methods described in the Examples.

[0016]

(a) Silicone Oil

The silicone oil is preferably at least one selected from the group consisting of dimethylpolysiloxane (dimethicone), dimethiconol (dimethylpolysiloxane having a hydroxy group at its end), methylphenylpolysiloxane, and modified silicone, from the viewpoint of improving the long-lasting repellent effect.

Examples of modified silicone include amino-modified silicone (dimethylpolysiloxane having an amino group), amino derivative silicone, fatty acid-modified silicone, alcohol-modified silicone, aliphatic alcohol-modified silicone, epoxy-modified silicone, fluorine-modified silicone, alkyl-modified silicone, and the like.

[0017]

From the viewpoint of improving the long-lasting repellent effect and from the viewpoint of providing a better feel when applied to the skin, preferred among the silicone oils is at least one selected from the group consisting of dimethylpolysiloxane, dimethiconol, methylphenylpolysiloxane, and modified silicone; more preferred is at least one selected from the group consisting of dimethylpolysiloxane and dimethiconol; and further more preferred is dimethylpolysiloxane.

[0018]

Examples of dimethylpolysiloxane include at least one selected from the group consisting of linear dimethylpolysiloxane and cyclic dimethylpolysiloxane. More preferred among these is linear

dimethylpolysiloxane, from the viewpoint of improving the long-lasting repellent effect.

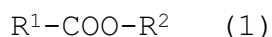
Examples of commercially available products of linear dimethylpolysiloxane include KF-96 Series, manufactured by Shin-Etsu Chemical Co., Ltd.; SH200C Series and "2-1184 Fluid," manufactured by Dow Corning Toray Co., Ltd.; "Silsoft DML," "Element14 PDMS 5-JC," "Element14 PDMS 10-JC," and "Element14 PDMS 20-JC," manufactured by Momentive Performance Materials, Inc.; and the like.

[0019]

(b) Ester Oil

Preferred examples of the ester oil include ester oils of any one of the following general formulas (1) to (3) and a dialkyl carbonate compound of the following general formula (4), from the viewpoint of improving the long-lasting repellent effect.

[0020]



In general formula (1), R^1 represents a linear or branched alkyl group optionally substituted with a hydroxy group and having 7 or more and 23 or less carbon atoms, or an aromatic hydrocarbon group having 6 or more and 24 or less carbon atoms, and R^2 represents a linear or branched alkyl group or alkenyl group having 1 or more and 22 or less carbon atoms.

When R^1 is an alkyl group, the number of carbon atoms is preferably 7 or more, and more preferably 9 or more, from the viewpoint of improving the long-lasting repellent effect, and is preferably 21 or less, and more preferably 17 or less, from the viewpoint of providing a better feel when applied to the skin. When R^1 is an aromatic hydrocarbon group, the number of carbon atoms is preferably 8 or more, and more preferably 10 or more, from the viewpoint of improving the long-lasting repellent effect, and is preferably 22 or less, and more preferably 20 or less, from the viewpoint of providing a better feel when applied to the skin.

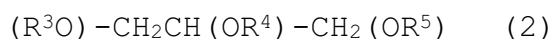
R^2 is preferably a linear or branched alkyl group or alkenyl group having 20 or less, and more preferably 18 or less carbon atoms, from the viewpoint of providing a better feel when applied to the skin. Further, at least one of R^1 and R^2 is preferably a branched alkyl group, from the viewpoint of providing a better feel when applied to the skin.

[0021]

Examples of the ester oil of general formula (1) include at least one selected from the group consisting of myristyl 2-ethylhexanoate, cetyl 2-ethylhexanoate, stearyl 2-ethylhexanoate, isodecyl octanoate, isocetyl octanoate, isononyl isononanoate, isotridecyl isononanoate, cetearyl isononanoate, octyl propyl heptanoate, methyl laurate, isopropyl myristate,

octyldodecyl myristate, isopropyl palmitate, 2-ethylhexyl palmitate, isocetyl palmitate, 2-ethylhexyl stearate, isocetyl stearate, isotridecyl stearate, isopropyl isostearate, octyl isostearate, isocetyl isostearate, isostearyl isostearate, 2-ethylhexyl hydroxystearate, methyl oleate, oleyl oleate, isobutyl oleate, oleyl erucate, alkyl benzoate (number of carbon atoms in alkyl: from 12 to 15), diethylhexyl naphthalene dicarboxylate, and the like.

[0022]



In general formula (2), R^3 , R^4 , and R^5 are each independently a hydrogen atom or a group of the following general formula (2-1), provided that not all of them are hydrogen atoms.

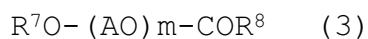


(in the formula, R^6 represents an alkyl group or alkenyl group optionally substituted with a hydroxy group and having 7 or more and 23 or less, preferably 17 or less, carbon atoms.)

Examples of the ester oil of general formula (2) include at least one selected from the group consisting of glyceryl tri-2-ethylhexanoate and glyceryl tricaprylate. These may also be plant-derived ester oils, such as jojoba oil, olive oil, sunflower oil, soybean oil, peanut oil, rapeseed oil, almond oil, palm oil, coconut oil, castor oil, wheat germ oil, grape seed

oil, thistle oil, evening primrose oil, macadamia nut oil, corn germ oil, and avocado oil.

[0023]



In general formula (3), R^7 represents an aromatic hydrocarbon group having 6 or more and 20 or less carbon atoms, R^8 represents an alkyl group or alkenyl group having 1 or more and 23 or less carbon atoms. AO represents an alkyleneoxy group having 2 or more and 4 or less carbon atoms, and m represents an average addition molar number of 1 or more and 50 or less.

R^7 is preferably an aromatic hydrocarbon group having 6 or more carbon atoms, from the viewpoint of improving the long-lasting repellent effect, and preferably 12 or less carbon atoms, and more preferably 10 or less carbon atoms, and further more preferably a benzyl group, from the viewpoint of providing a better feel when applied to the skin.

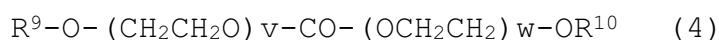
R^8 is preferably an alkyl group having 7 or more carbon atoms, and more preferably 11 or more carbon atoms, from the viewpoint of improving the long-lasting repellent effect, and preferably 21 or less carbon atoms, and more preferably 15 or less carbon atoms, from the viewpoint of providing a better feel when applied to the skin.

The AO group is preferably a propyleneoxy group, from the viewpoint of improving the long-lasting

repellent effect, and m is preferably 1 or more and 10 or less, and more preferably 1 or more and 5 or less, from the viewpoint of improving the long-lasting repellent effect and from the viewpoint of providing a better feel when applied to the skin.

Examples of the ester oil of general formula (3) include at least one selected from the group consisting of an ester of a propylene oxide 3 mol adduct of benzyl alcohol and myristic acid ("Crodamol STS," manufactured by Croda) and an ester of a propylene oxide 3 mol adduct of benzyl alcohol and 2-ethylhexanoic acid ("Crodamol SFX," manufactured by Croda).

[0024]



In general formula (4), R^9 and R^{10} each independently represent an alkyl group or alkenyl group having 6 or more and 22 or less carbon atoms, and v and w each independently represent 0 or an average addition molar number of 1 or more and 50 or less.

R^9 and R^{10} are each preferably an alkyl group having 8 or more carbon atoms, from the viewpoint of improving the long-lasting repellent effect, and preferably having 18 or less, and more preferably 12 or less carbon atoms, from the viewpoint of providing a better feel when applied to the skin.

v and w are each preferably 0 or a number of 1 or more and 5 or less, and more preferably 0, from the

viewpoint of providing a better feel when applied to the skin.

Examples of the dialkyl carbonate compound of general formula (4) include dioctyl carbonate ("Cetiol CC," manufactured by Cognis).

[0025]

Examples of ester oils other than those described above include esters of polyvalent carboxylic acids and alcohols, and esters of polyhydric alcohols, excluding glycerin, and fatty acids.

Specific examples thereof include at least one selected from the group consisting of diisopropyl dimerate, diisopropyl adipate, diethoxyethyl succinate, 2-ethylhexyl succinate, propanediol dicaprate, neopentyl glycol dicaprate, neopentyl glycol di-2-ethylhexanoate, and the like. Preferred among these are esters of neopentyl glycol and fatty acids, and more preferred is at least one selected from the group consisting of neopentyl glycol dicaprate and neopentyl glycol di-2-ethylhexanoate, from the viewpoint of improving the long-lasting repellent effect.

Preferred among the ester oils (b) described above is at least one selected from the group consisting of the ester oil of general formula (1) and esters of neopentyl glycol and fatty acids.

[0026]

(c) Ether Oil

The ether oil is preferably a dialkyl ether compound of the following general formula (5) or a polyoxyalkylene alkyl ether compound of the following general formula (6), from the viewpoint of improving the long-lasting repellent effect.

[0027]

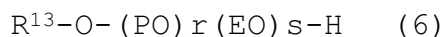


In general formula (5), R^{11} and R^{12} are each independently a linear or branched alkyl group or alkenyl group having 6 or more and 22 or less carbon atoms, or an aromatic hydrocarbon group having 6 or more and 24 or less carbon atoms.

R^{11} and R^{12} are each preferably an alkyl group, from the viewpoint of improving the long-lasting repellent effect. The number of carbon atoms therein is preferably 8 or more, from the viewpoint of improving the long-lasting repellent effect, and is preferably 18 or less, more preferably 16 or less, and further more preferably 12 or less, from the viewpoint of providing a better feel when applied to the skin.

Examples of the dialkyl ether compound of general formula (5) include dihexyl ether, dioctyl ether ("Cetiol OE," manufactured by Cognis), dicaprylyl ether, cetyl-1,3-dimethylbutyl ether ("ASE-166K," manufactured by Kao Corporation), and the like.

[0028]



In general formula (6), R^{13} represents an alkyl group or alkenyl group having 6 or more and 22 or less carbon atoms, PO represents a propyleneoxy group, and EO represents an ethyleneoxy group. r represents an average addition molar number of 0.1 or more and 15 or less, and s represents an average addition molar number of 0 or more and 10 or less. When s is not 0, the addition form of each of PO and EO may be random or block.

The number of carbon atoms in R^{13} is preferably 8 or more, from the viewpoint of improving the long-lasting repellent effect, and is preferably 20 or less, more preferably 18 or less, and further more preferably 12 or less, from the viewpoint of providing a better feel when applied to the skin.

The average addition molar number r is preferably 1 or more, more preferably 2 or more, and further more preferably 3 or more, from the viewpoint of improving the long-lasting repellent effect, and is preferably 13 or less, and more preferably 10 or less, from the viewpoint of providing a better feel when applied to the skin. The average addition molar number s is preferably 5 or less, more preferably 1 or less, and further more preferably 0, from the viewpoint of providing a better feel when applied to the skin.

The polyoxyalkylene alkyl ether compound of general formula (6) is preferably at least one selected from the group consisting of polypropylene glycol, and

polyoxypropylene octyl ether, polyoxypropylene decyl ether, and polyoxypropylene lauryl ether, in which the average addition molar number r of the propyleneoxy group is 3 or more and 10 or less.

Preferred among the ether oils (c) described above is the ether oil of general formula (5), from the viewpoint of improving the long-lasting repellent effect.
[0029]

(d) Hydrocarbon Oil

Examples of the hydrocarbon oil include liquid paraffin, liquid isoparaffin, squalane, isohexadecane, isoeicosane, hydrogenated polyisobutene, light liquid isoparaffin, heavy liquid isoparaffin, α -olefin oligomer, cycloparaffin, and the like.

Preferred among these is at least one selected from the group consisting of liquid paraffin, liquid isoparaffin, and squalane, from the viewpoint of improving the long-lasting repellent effect and from the viewpoint of providing a better feel when applied to the skin.

[0030]

(e) Aliphatic Alcohol

The aliphatic alcohol may be a monovalent chain or cyclic aliphatic alcohol, and is preferably a monovalent chain aliphatic alcohol, from the viewpoint of improving the long-lasting repellent effect. The number of carbon atoms in the aliphatic alcohol is preferably 14 or more,

and more preferably 18 or more, from the viewpoint of improving the long-lasting repellent effect, and is preferably 28 or less, more preferably 24 or less, and further more preferably 22 or less, from the viewpoint of providing a better feel when applied to the skin.

The aliphatic alcohol described above may be linear or branched, and may be saturated or unsaturated, but is preferably a branched aliphatic saturated alcohol, from the viewpoint of improving the long-lasting repellent effect and from the viewpoint of providing a better feel when applied to the skin.

Examples of linear aliphatic alcohols include oleyl alcohol and the like, and examples of branched aliphatic saturated alcohols include butyl octanol, butyl decanol, hexyl decanol, octyl dodecanol, and the like.

Preferred among these are branched aliphatic saturated alcohols, from the viewpoint of improving the long-lasting repellent effect and from the viewpoint of providing a better feel when applied to the skin, and preferred is at least one selected from the group consisting of hexyl decanol and octyl dodecanol.

[0031]

(f) Polyhydric Alcohol

The polyhydric alcohol may be an aliphatic alcohol having 2 or more carbon atoms, an aromatic alcohol, a sugar alcohol having 4 or more carbon atom, or the like, and may be saturated or unsaturated, but is preferably at

least one selected from the group consisting of an aliphatic alcohol having 2 or more and 10 or less carbon atoms, an aromatic alcohol, and a sugar alcohol having 4 or more and 10 or less carbon atoms, from the viewpoint of providing a better feel when applied to the skin.

Among the polyhydric alcohols, examples of the aliphatic alcohol having 2 or more carbon atoms include dihydric alcohols, such as ethylene glycol, propylene glycol, 1,2-butylene glycol, 1,3-butylene glycol, 1,2-pentanediol, 1,2-hexanediol, diethylene glycol, dipropylene glycol, and hexylene glycol.

Examples of the sugar alcohol described above include sorbitol, erythritol, pentaerythritol, xylitol, mannitol, and the like.

[0032]

From the viewpoint of improving the long-lasting repellent effect, preferred among the liquid oily components described above is at least one selected from the group consisting of (a) silicone oils, (b) ester oils, (c) ether oils, and (d) hydrocarbon oils; more preferred is at least one selected from the group consisting of (a) silicone oils, (b) ester oils, and (d) hydrocarbon oils; further more preferred is at least one selected from the group consisting of (a) silicone oils and (b) ester oils; and even more preferred are (a) silicone oils. From the viewpoint of improving the long-lasting repellent effect, preferred among the silicone

oils (a) is at least one selected from the group consisting of dimethylpolysiloxane, dimethiconol, methylphenylpolysiloxane, and modified silicone; more preferred is at least one selected from the group consisting of dimethylpolysiloxane and dimethiconol; further more preferred is dimethylpolysiloxane; and even more preferred is linear dimethylpolysiloxane.

[0033]

(Content of Component (A))

The content of the component (A) in the composition of the present invention is 13 mass% or more, preferably 18 mass% or more, and more preferably 25 mass% or more, from the viewpoint of improving the long-lasting repellent effect. The content of the component (A) is also 50 mass% or less, preferably 40 mass% or less, and more preferably 35 mass% or less, from the viewpoint of providing a better feel when applied to the skin and from the viewpoint of ensuring the long-term storage stability of the composition. The content of the component (A) in the composition of the present invention is from 13 to 50 mass%, preferably from 18 to 40 mass%, and more preferably from 25 to 35 mass%.

[0034]

<Component (B): Hydrophilic Inorganic Particles>

The composition of the present invention contains, as the component (B), hydrophilic inorganic particles containing silica particles and/or silicate mineral

particles and having a volume median particle size of 1 μm or more and 40 μm or less. Since the composition of the present invention contains the component (B), the long-lasting repellent effect due to the component (A) is improved, and the composition has a good feel when applied to the skin.

[0035]

In the present specification, the "hydrophilic inorganic particles" refer to inorganic particles having a wetting tension at 23°C of 70 mN/m or more on the particle surface.

Since the particles of the component (B) are hydrophilic inorganic particles having a specific particle size, the long-lasting repellent effect is improved, and the composition has a good feel when applied to the skin. The component (B) used in the composition of the present invention preferably has a wetting tension at 23°C of 70 mN/m or more, and more preferably 73 mN/m or more, from the viewpoint of improving the long-lasting repellent effect and from the viewpoint of providing a better feel when applied to the skin. The upper limit can be, for example, 100 mN/m or less, and may be 95 mN/m or less. The wetting tension of the particle surface can be measured by the method described in the Examples.

[0036]

The component (B) contains silica particles and/or silicate mineral particles.

From the viewpoint of providing a better feel when applied to the skin, the silicate mineral particles preferably contain at least one selected from the group consisting of talc particles, mica particles, sericite particles, smectite particles, montmorillonite particles, bentonite particles, and kaolin particles; and more preferably at least one selected from the group consisting of talc particles and mica particles.

From the viewpoint of providing a better feel when applied to the skin, the component (B) preferably contains at least one type of particles selected from the group consisting of silica particles, talc particles, and mica particles; more preferably silica particles; and further more preferably porous silica particles.

[0037]

Examples of the shape of the component (B) include a spherical shape, a plate-like shape, a flat shape, a flaky shape, an irregular shape, and the like. More preferred among these is a spherical or flat shape, from the viewpoint of providing a better feel when applied to the skin.

More specifically, when the component (B) is silica particles, their shape is more preferably spherical, and when the component (B) is silicate mineral particles, their shape is more preferably flat.

[0038]

The volume median particle size (D50) of the component (B) is 1 μm or more, preferably 2 μm or more, and more preferably 3 μm or more, and is 40 μm or less, preferably 30 μm or less, more preferably 20 μm or less, further more preferably 15 μm or less, and even more preferably 12 μm or less, from the viewpoint of providing a better feel when applied to the skin.

[0039]

(Content of Component (B))

The content of the component (B) in the composition of the present invention is 0.5 mass% or more, preferably 1 mass% or more, more preferably 2 mass% or more, and further more preferably 4 mass% or more, from the viewpoint of providing a better feel when applied to the skin and from the viewpoint of ensuring long-term storage stability. The content of the component (B) is also 35 mass% or less, preferably 27 mass% or less, more preferably 16 mass% or less, and further more preferably 8 mass% or less, from the viewpoint of improving the long-lasting repellent effect. The content of the component (B) in the composition of the present invention is from 0.5 to 35 mass%, preferably from 1 to 27 mass%, more preferably from 2 to 16 mass%, and further more preferably from 4 to 8 mass%.

The total content of silica particles and silicate mineral particles in the component (B) is preferably 80

mass% or more, more preferably 85 mass% or more, further more preferably 90 mass% or more, and further more preferably 95 mass% or more, and is 100 mass% or less, and preferably 100 mass%, from the viewpoint of improving the long-lasting repellent effect and from the viewpoint of providing a better feel when applied to the skin.

[0040]

<Component (C): Modified Silicone>

The composition of the present invention contains, as the component (C), at least one modified silicone selected from the group consisting of polyether-modified silicone, polyglycerin-modified silicone, branched polyglycerol-modified silicone, alkyl glyceryl ether-modified silicone, and carboxy-modified silicone, all of which are liquid at 25°C. The phrase "liquid at 25°C" refers to a state in which the material has fluidity in a bulk state in an environment of 1 atm and 25°C. Specifically, the phrase "liquid at 25°C" means that the viscosity measured at 25°C using a Brookfield viscometer based on JIS Z8803: v2011 "Methods for viscosity measurement of liquid" is 120 000 mPa·s or less.

In the composition of the present invention, a portion of the component (C) is adsorbed onto the component (B) via a hydrogen bond or the like to cover a portion or all of the component (B), thereby improving the long-lasting repellent effect due to the component (A) and providing a good feel when the composition is

applied to the skin. From the viewpoint of improving the long-lasting repellent effect and from the viewpoint of providing a better feel when applied to the skin, in the component (C) contained in the composition of the present invention, the amount of the component (C) adsorbed onto the component (B) is preferably 1 mass% or more, more preferably 2 mass% or more, and further more preferably 4 mass% or more, and is preferably 15 mass% or less, more preferably 10 mass% or less, and further more preferably 6 mass% or less.

[0041]

The polyether-modified silicone has a structure in which a hydrocarbon group in the side chain and/or end of silicone oil is replaced by a polyether group.

The polyether group in the polyether-modified silicone is preferably a polyethyleneoxy group, a polypropyleneoxy group, or a polyalkyleneoxy group in which ethyleneoxy groups (EO) and propyleneoxy groups (trimethyleneoxy groups or propane-1,2-diylloxy groups; PO) are added in a block or random manner, and more preferably a polyethyleneoxy group. Usable examples of the polyether-modified silicone include compounds in which polyether groups are grafted onto a silicone main chain, and compounds in which silicone and polyether groups are bonded in blocks.

[0042]

The hydrophilic-lipophilic balance (HLB) of the polyether-modified silicone is preferably 1.0 or more, more preferably 2.0 or more, and further more preferably 2.5 or more, and is preferably 20.0 or less, more preferably 18.0 or less, further more preferably 16.0 or less, even more preferably 15.5 or less, and even more preferably 15.0 or less, from the viewpoint of providing a better feel when the composition is applied to the skin.

The HLB value as mentioned herein is a value which indicates the affinity of the polyether-modified silicone for water and oil, and can be calculated from the following formula using the Griffin method:

$$\text{HLB} = 20 \times [(\text{total molecular weight of hydrophilic groups contained in polyether-modified silicone}) / (\text{molecular weight of polyether-modified silicone})]$$

Examples of the hydrophilic groups include hydroxy groups, ethyleneoxy groups, and the like.

[0043]

The viscosity at 25°C of the polyether-modified silicone is preferably 50 mPa·s or more, more preferably 75 mPa·s or more, and further more preferably 100 mPa·s or more, and may be 120 000 or less, 10 000 or less, 4 500 mPa·s or less, 4 000 mPa·s or less, or 3 500 mPa·s or less, from the viewpoint of providing a better feel when the composition is applied to the skin. The viscosity

can be measured based on JIS Z8803: 2011 "Methods for viscosity measurement of liquid" using a Brookfield viscometer at 25°C.

[0044]

Specific examples of the polyether-modified silicone include PEG-3 dimethicone, PEG-9 dimethicone, PEG-9 methyl ether dimethicone, PEG-10 dimethicone, PEG-11 methyl ether dimethicone, PEG-12 dimethicone, PEG/PPG-20/22 butyl ether dimethicone, PEG-32 methyl ether dimethicone, PEG-9 polydimethylsiloxylethyl dimethicone, lauryl PEG-9 polydimethylsiloxylethyl dimethicone, lauryl PEG-10 tris(trimethylsiloxylethyl)dimethicone, PEG/PPG-19/19 dimethicone, cetyl PEG/PPG-10/1 dimethicone, PEG/PPG-30/10 dimethicone, and linear polyether-modified silicone in which dimethyl silicone units and polyether units are alternately copolymerized.

Examples of commercially available products of the polyether-modified silicone include KF series (e.g., KF-6004, KF-6011, KF-6012, KF-6013, KF-6015, KF-6016, KF-6017, KF-6028, KF-6038, KF-6043, and KF-6048) manufactured by Shin-Etsu Chemical Co., Ltd., DOWSIL series (BY25-337, BY25-339, SH3775M, FZ-2203, and ES-5300 Formulation Aid) manufactured by Dow Chemical Japan Ltd., and the like.

[0045]

The polyglycerin-modified silicone refers to silicone having a monovalent polyglyceryl group within

its structure, and examples thereof include polyglyceryl-3 disiloxane dimethicone, polyglyceryl-3 polydimethylsiloxylethyl dimethicone, lauryl polyglyceryl-3 polydimethylsiloxylethyl dimethicone, and the like. In the present specification, branched polyglycerol-modified silicone is excluded from the polyglycerin-modified silicone.

Examples of commercially available products of the polyglycerin-modified silicone include KF series (e.g., KF-6100, KF-6104, KF-6106, and KF-6105) manufactured by Shin-Etsu Chemical Co., Ltd., and the like.

[0046]

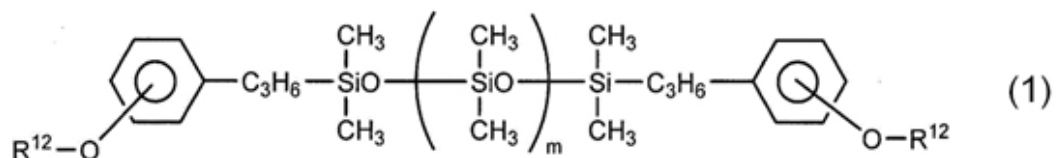
The viscosity at 25°C of the polyglycerin-modified silicone is preferably 500 mPa·s or more, more preferably 1 000 mPa·s or more, further more preferably 2 000 mPa·s or more, and even more preferably 3 000 mPa·s or more, and is preferably 20 000 mPa·s or less, more preferably 10 000 mPa·s or less, and further more preferably 5 000 mPa·s or less, from the viewpoint of providing a better feel when the composition is applied to the skin. The viscosity can be measured based on JIS Z8803: 2011 "Methods for viscosity measurement of liquid" using a Brookfield viscometer at 25°C.

[0047]

Examples of the branched polyglycerol-modified silicone include compounds of the following formula (1) having a structure in which branched polyglycerol chains

are bonded to both ends of dimethylpolysiloxane via a linking group containing an oxyphenylene group.

[0048]

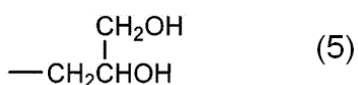
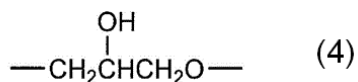
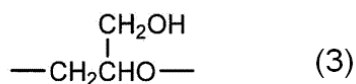
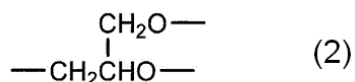


(In formula (1), R^{12} each independently represents a branched polyglycerol chain, and m represents an integer of 0 or more and 10 000 or less.)

[0049]

In formula (1), the bonding mode between the oxygen atom and the trimethylene group ($-\text{C}_3\text{H}_6-$) in the phenylene group portion is not particularly limited, and may be in the ortho, meta, or para position. m represents an integer of 0 or more and 10 000 or less, and preferably 1 or more and 300 or less. R^{12} represents a branched polyglycerol chain, composed of glycerol units of the following structural formulas (2) to (5), having at least one glycerol unit with the branched structure of structural formula (2), and terminating in the glycerol unit of structural formula (5). The average total number of glycerol units bonded in the branched polyglycerol chain is 3 or more and 200 or less, and preferably 3 or more and 30 or less.

[0050]



[0051]

The viscosity at 25°C of the branched polyglycerol-modified silicone is preferably 10 000 mPa·s or more, and more preferably 15 000 mPa·s or more, and is preferably 500 000 mPa·s or less, more preferably 300 000 mPa·s or less, and further more preferably 150 000 mPa·s or less, from the viewpoint of providing a better feel when the composition is applied to the skin. The viscosity can be measured based on JIS Z8803: 2011 "Methods for viscosity measurement of liquid" using a Brookfield viscometer at 25°C.

[0052]

The branched polyglycerol-modified silicone of formula (1) can be produced, for example, by the method described in JP-A-2004-339244, in which 2,3-epoxy-1-propanol is added to modified silicone having a phenyl group substituted with a hydroxy group at both ends in

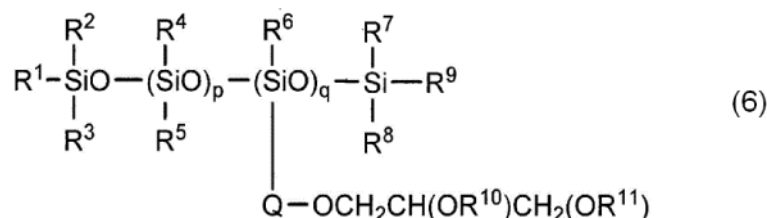
the presence of an acidic or basic catalyst, followed by graft polymerization, or by other commonly known methods.

Examples of commercially available products of the branched polyglycerol-modified silicone include SOFCARE GS-G (manufactured by Kao Corporation) and the like.

[0053]

Examples of the alkyl glyceryl ether-modified silicone include those of the following formula (6).

[0054]



[In formula (6), Q represents a divalent hydrocarbon group having 3 or more and 20 or less carbon atoms; R¹ to R⁹ may be the same or different and each represents a hydrogen atom, a linear or branched hydrocarbon group having 1 or more and 32 or less carbon atoms, or a phenyl group; R¹⁰ and R¹¹ may be the same or different and each represents a hydrogen atom or a linear or branched hydrocarbon group having 1 or more and 32 or less carbon atoms; and a plurality of R¹⁰ and a plurality of R¹¹ in the same molecule each may be different. p represents a number of 1 or more and 500 or less, and q represents a number of 1 or more and 50 or less. p and q each preferably represent a number in which the content of the

alkyl glyceryl ether group $[-Q-OCH_2-CH(OR^{10})-CH_2(OR^{11})]$ in the molecule is 1 mass% or more and 50 mass% or less.]

[0055]

In formula (6), the divalent hydrocarbon group having 3 or more and 20 or less carbon atoms of Q is preferably a linear or branched alkylene group having 3 or more and 20 or less carbon atoms. More specific examples thereof include linear alkylene groups, such as trimethylene, tetramethylene, pentamethylene, hexamethylene, heptamethylene, octamethylene, nonamethylene, decamethylene, undecamethylene, dodecamethylene, tetradecamethylene, hexadecamethylene, and octadecamethylene; branched alkylene groups, such as propylene, 2-methyltrimethylene, 2-methyltetramethylene, 2-methylpentamethylene, and 3-methylpentamethylene; and the like.

[0056]

In the definitions of R^1 to R^{11} , examples of the linear or branched hydrocarbon group having 1 or more and 32 or less carbon atoms include linear alkyl groups, such as methyl, ethyl, propyl, butyl, pentyl, hexyl, octyl, decyl, undecyl, dodecyl, tetradecyl, hexadecyl, octadecyl, eicosyl, docosyl, tetracosyl, hexacosyl, octacosyl, and triacontyl; branched alkyl groups, such as isopropyl, sec-butyl, tert-butyl, neopentyl, 1-ethylpropyl, and 1-heptyldecyl; and the like. In the present specification, R^1 to R^9 are each preferably a

linear or branched alkyl group having 1 or more and 25 or less carbon atoms (in which some of them may be hydrogen atoms), and more preferably a linear or branched alkyl group having 1 or more and 22 or less carbon atoms (in which some of them may be hydrogen atoms). R^{10} and R^{11} are each preferably a hydrogen atom or an alkyl group having 1 or more and 5 or less carbon atoms, and more preferably a hydrogen atom.

[0057]

Furthermore, p and q are each preferably a number in which the content of the alkyl glyceryl ether group $[-Q-OCH_2-CH(OR^{10})-CH_2(OR^{11})]$ is 1 mass% or more and 50 mass% or less, more preferably 5 mass% or more and 40 mass% or less, and further more preferably 10 mass% or more and 30 mass% or less. Specifically, in terms of the availability of organopolysiloxane as a raw material, operability during production, and the like, p is in the range of 1 or more and 500 or less, and preferably 10 or more and 30 or less, and q is in the range of 1 or more and 50 or less, and preferably 1 or more and 30 or less.

[0058]

The viscosity at 25°C of the alkyl glyceryl ether-modified silicone is preferably 1 000 mPa·s or more, more preferably 2 000 mPa·s or more, and further more preferably 3 000 mPa·s or more, and is preferably 20 000 mPa·s or less, more preferably 15 000 mPa·s or less, and further more preferably 10 000 mPa·s or less, from the

viewpoint of providing a better feel when the composition is applied to the skin. The viscosity can be measured based on JIS Z8803: 2011 "Methods for viscosity measurement of liquid" using a Brookfield viscometer at 25°C.

[0059]

Such alkyl glyceryl ether-modified silicone can be produced in accordance with the method described in JP-A-4-134013 or JP-A-2005-194523 by reacting organohydrogenpolysiloxane having at least one silicon-hydrogen bond with corresponding alkenyl glyceryl ether.

Examples of commercially available products of the alkyl glyceryl ether-modified silicone include SI-UGE and SOFCARE RS-U (both manufactured by Kao Corporation), and the like.

[0060]

Examples of the carboxy-modified silicone include compounds having a structure in which a carboxy group is bonded to the side chain and/or end of silicone oil. A carboxy group may be bonded to the main chain of silicone oil via a linking group. Specific examples of the carboxy-modified silicone include carboxydecyl dimethicone, carboxydecyl trisiloxane, bis-carboxydecyl dimethicone, and the like.

The viscosity at 25°C of the carboxy-modified silicone is preferably 500 mPa·s or more, more preferably 1 000 mPa·s or more, and further more preferably 1 500

mPa·s or more, and is preferably 3 500 mPa·s or less, more preferably 3 000 mPa·s or less, and further more preferably 2 500 mPa·s or less, from the viewpoint of providing a better feel when the composition is applied to the skin. The viscosity can be measured based on JIS Z8803: 2011 "Methods for viscosity measurement of liquid" using a Brookfield viscometer at 25°C.

As the carboxy-modified silicone, synthetic products and commercially available products both can be used.

Examples of commercially available products of the carboxy-modified silicone include X-22-3701E, X-22-3710, and X-22-162C, manufactured by Shin-Etsu Chemical Co., Ltd.; DOWSIL BY 16-880 Fluid and DOWSIL ES-5800 Formulation Aid, manufactured by Dow Chemical Japan Ltd.; and the like.

[0061]

These modified silicones may be used singly or in combination of two or more, and are preferably used in combination of two.

[0062]

(Content of Component (C))

The content of the component (C) in the composition of the present invention is 0.1 mass% or more, preferably 0.2 mass% or more, more preferably 0.3 mass% or more, and further more preferably 0.4 mass% or more, from the viewpoint of improving the long-lasting repellent effect and from the viewpoint of providing a better feel when

applied to the skin, and is 3 mass% or less, preferably 2.3 mass% or less, more preferably 1.6 mass% or less, and further more preferably 1.0 mass% or less, from the viewpoint of ensuring long-term storage stability. The content of the component (C) in the composition of the present invention is from 0.1 to 3 mass%, preferably from 0.2 to 2.3 mass%, more preferably from 0.3 to 1.6 mass%, and further more preferably from 0.4 to 1.0 mass%.

[0063]

In the composition of the present invention, the mass ratio of the component (B) to the component (A), $[(B)/(A)]$, is preferably 0.02 or more, more preferably 0.06 or more, and further more preferably 0.1 or more, from the viewpoint of providing a better feel when applied to the skin and from the viewpoint of ensuring long-term storage stability, and is preferably 0.7 or less, more preferably 0.5 or less, and further more preferably 0.3 or less, from the viewpoint of improving the long-lasting repellent effect.

[0064]

In the composition of the present invention, the mass ratio of the component (C) to the component (A), $[(C)/(A)]$, is preferably 0.005 or more, more preferably 0.01 or more, and further more preferably 0.015 or more, from the viewpoint of improving the long-lasting repellent effect and from the viewpoint of ensuring long-term storage stability, and is preferably 0.1 or less,

more preferably 0.08 or less, and further more preferably 0.04 or less, from the viewpoint of providing a better feel when applied to the skin.

[0065]

In the composition of the present invention, the mass ratio of the component (C) to the component (B), $[(C)/(B)]$, is preferably 0.01 or more, more preferably 0.05 or more, further more preferably 0.07 or more, and even more preferably 0.1 or more, from the viewpoint of improving the long-lasting repellent effect, and is preferably 0.9 or less, more preferably 0.7 or less, further more preferably 0.5 or less, and even more preferably 0.3 or less, from the viewpoint of providing a better feel when applied to the skin and from the viewpoint of ensuring long-term storage stability.

[0066]

<Component (D): Thickener>

The composition of the present invention contains a thickener as the component (D). From the viewpoint of improving the long-lasting repellent effect by regulating fluidity after application to the skin, from the viewpoint of well dispersing or dissolving each component to ensure excellent applicability to the skin, and from the viewpoint of ensuring the long-term storage stability of the composition, the thickener is preferably one having appropriate emulsifying ability (i.e., surface active ability). In the present invention, an agent

having emulsifying ability and a thickening effect is regarded as a thickener. Specific examples of the component (D) include water-soluble cationic polymers, anionic polymers, nonionic polymers, and amphoteric polymers or zwitterionic polymers.

[0067]

Specific examples of cationic polymers include hydroxyethyl cellulose having an O-[2-hydroxy-3-(trimethylammonio)propyl] chloride group (polyquaternium-10), vinyl pyrrolidone-dimethylaminomethylethyl methacrylate copolymer diethyl sulfate (polyquaternium-11), a methylvinylimidazolium chloride-vinylpyrrolidone copolymer, and the like.

[0068]

Specific examples of anionic polymers include a carboxyvinyl polymer, carboxymethyl cellulose, carrageenan, xanthan gum, polystyrene sulfonate, agar, ghatti gum, karaya gum, pectin, an alginate salt, an acrylic acid/alkyl methacrylate copolymer, a (sodium acrylate/sodium acryloyldimethyltaurate) copolymer, polyacrylamide, an (ammonium acryloyldimethyltaurate/vinylpyrrolidone) copolymer, an (acrylate/alkyl acrylate (C10-C30)) crosspolymer, an (ammonium acryloyldimethyltaurate/behene-25 methacrylate) crosspolymer, a (hydroxyethyl acrylate/sodium acryloyldimethyltaurate) copolymer, an (acrylamide/ammonium acrylate) copolymer, a (sodium

acrylate/acryloyldimethyltaurine/dimethylacrylamide) crosspolymer, polyacrylate-13, polyacrylate crosspolymer-6, an alkali metal salt of poly(acrylic acid), and hyaluronic acid or an alkali metal salt thereof.

[0069]

Specific examples of nonionic polymers include cellulose ether (hydroxybutyl methylcellulose, hydroxypropyl cellulose, hydroxypropyl methylcellulose, ethyl hydroxyethyl cellulose, hydroxyethyl cellulose, hydroxypropyl methylcellulose stearoxy ether, and the like), propylene glycol alginate, poly(ethylene oxide), polyvinyl alcohol, polyvinylpyrrolidone, hydroxypropyl guar gum, locust bean gum, amylose, hydroxyethyl amylose, starch and starch derivatives, and mixtures thereof.

[0070]

Specific examples of amphoteric polymers or zwitterionic polymers include an octylacrylamide/acrylate/butylaminoethyl methacrylate copolymer, polyquaternium-47, polyquaternium-43, and the like.

[0071]

The components (D) can be used singly or in combination of two or more, and is preferably used in combination of two or more.

From the viewpoint of improving the duration of the pest insect repellent effect by regulating fluidity after application to the skin, from the viewpoint of well

dispersing or dissolving each component to ensure excellent applicability to the skin, and from the viewpoint of ensuring the long-term storage stability of the composition, the component (D) is preferably at least one selected from the group consisting of anionic polymers and nonionic polymers; preferably at least one selected from the group consisting of a carboxyvinyl polymer, an acrylic acid/alkyl methacrylate copolymer, a (sodium acrylate/sodium acryloyldimethyltaurate) copolymer, an (ammonium acryloyldimethyltaurate/beheneth-25 methacrylate) crosspolymer, hydroxypropyl methylcellulose stearoxy ether, and an (acrylate/alkyl acrylate (C10-C30)) crosspolymer; and more preferably at least one selected from the group consisting of an (ammonium acryloyldimethyltaurate/beheneth-25 methacrylate) crosspolymer, hydroxypropyl methylcellulose stearoxy ether, and an (acrylate/alkyl acrylate (C10-C30)) crosspolymer.

Usable examples of commercially available products of the component (D) include "CARBOPOL 980," "CARBOPOL 981," "CARBOPOL 1342," "CARBOPOL ETD2020," "PEMULEN TR-1," and "PEMULEN TR-2," manufactured by Lubrizol Advanced Materials, Inc.; "Aculyn 22," manufactured by the Dow Chemical Company; "Aristoflex AVC," "Aristoflex HMB," and "Aristoflex Velvet," manufactured by Clariant Iberica Production, S.A.; "SANGELOSE 60L," manufactured by Daido Chemical Industry Co., Ltd.; and the like. The thickener

can be dispersed in a dispersion medium and incorporated as a dispersion.

[0072]

The content of the component (D) in the composition of the present invention is 0.05 mass% or more, preferably 0.1 mass% or more, and more preferably 0.2 mass% or more, from the viewpoint of obtaining the above effects, and is preferably 3 mass% or less, more preferably 2 mass% or less, and further more preferably 1.5 mass% or less, from the same viewpoint. The content of the component (D) in the composition of the present invention is from 0.05 to 3 mass%, preferably from 0.1 to 2 mass%, and more preferably from 0.2 to 1.5 mass%.

When the composition of the present invention contains a thickener having surface active ability, the content of other surfactants is preferably 1.5 mass% or less, and more preferably 1 mass% or less.

[0073]

<Component (E): Water>

The composition of the present invention contains water as the component (E). The content of the component (E) in the composition is 30 mass% or more, from the viewpoint of forming an oil-in-water type composition, and is preferably 32 mass% or more, more preferably 36 mass% or more, and further more preferably 40 mass% or more, from the viewpoint of providing a better feel when applied to the skin. The content of the component (E) is

also 86.35 mass% or less, preferably 80 mass% or less, more preferably 75 mass% or less, and further more preferably 70 mass% or less, from the viewpoint of improving the long-lasting repellent effect and from the viewpoint of providing a better feel when applied to the skin. The specific range of the content of the component (E) in the composition is from 30 to 86.35 mass%, preferably from 32 to 80 mass%, more preferably from 36 to 75 mass%, and further more preferably from 40 to 70 mass%.

[0074]

(Surfactant)

The composition of the present invention may further contain a surfactant, from the viewpoint of forming an oil-in-water type composition. Usable surfactants are known surfactants, and any of anionic surfactants, cationic surfactants, ampholytic surfactants, nonionic surfactants, and the like can be used. These may be used singly or in combination of two or more.

When the composition of the present invention contains a surfactant, the content of the surfactant in the composition of the present invention is preferably 0.01 mass% or more, more preferably 0.05 mass% or more, and further more preferably 0.1 mass% or more, from the viewpoint of forming an oil-in-water type composition, and is preferably 5 mass% or less, more preferably 3

mass% or less, and further more preferably 1 mass% or less, from the same viewpoint.

[0075]

<Other Components>

The composition of the present invention may contain components other than those described above within the range that does not impair the effects of the invention. Examples of other components include medicinal and biologically active components such as organic solvents, preservatives, coloring agents, moisturizers, fragrances, pH adjusters, vitamins, blood circulation promoters, active oxygen scavengers, anti-inflammatory agents, skin whitening agents, disinfectants. Each of these components can also be used for other applications; for example, it is possible to use fragrances as disinfectants.

[0076]

The components (A) to (E) contained in the composition of the present invention and components that can be contained in the composition of the present invention may be used singly or in combination of two or more.

[0077]

From the viewpoint of preventing allergies and skin irritation in users, it is preferable that the composition of the present invention contains the component (A) as a repellent active ingredient for pest

insect repellence and does not contain an effective amount of other pest insect repellents.

Even when the composition of the present invention does not contain an effective amount of any of the existing pest insect repellents described later, the composition has a long-lasting repellent effect. In other words, even when the composition of the present invention contains a pest insect repellent other than the component (A) in an amount less than the lower limit of the effective amount, the stay prevention effect of the present invention is exhibited as long as a pest insect lands but does not stay on that place. That is, the composition of the present invention may not substantially contain a pest insect repellent other than the component (A).

[0078]

In the present specification, the phrase that a pest insect repellent other than the component (A) is "not contained in an effective amount" generally means that the content of an existing pest insect repellent other than the component (A) is preferably 15 mass% or less, more preferably 10 mass% or less, further more preferably 5 mass% or less, even more preferably 4 mass% or less, even more preferably 3 mass% or less, and even more preferably 1 mass% or less, in the composition of the present invention.

As for the effective amount of the existing pest insect repellent, for example, the minimum effective amount and the like published by manufacturers and the like of repellent products can be made by reference.

More specifically, the effective amounts of the existing pest insect repellents described below are as follows. The effective amount of DEET: N,N-diethyltoluamide is 4 mass% or more, and preferably 10 mass% or more, the effective amount of Icaridin: 1-methylpropyl-2-(2-hydroxyethyl)-1-piperidine carboxylate is 4 mass% or more, the effective amount of IR3535: 3-(acetylbutyl)aminopropionic acid ethyl ester is 4 mass% or more, and the effective amount of citronella oil is 10 mass% or more.

[0079]

The effective amount of the existing pest insect repellent can also be measured by the repellent evaluation test described below.

(Repellent Evaluation Test)

100 mated female mosquitoes (*Aedes albopictus*) are placed in a plastic cage (30 × 30 × 30 cm: BugDorm-1 cage) covered with a mesh. An arm is inserted into a KUALATEC Super Long Glove (50 cm) (manufactured by AS ONE Corporation, Catalog number: 3-6432-02) with a cut of a rectangular shape with a size of 10 cm in length × 3 cm in width. In order to activate the mosquitoes, breath is blown into the case for 5 seconds. The arm is inserted

into the cage under the conditions of 27°C and a relative humidity (RH) of 60% in a state that nothing is applied to an exposed portion from the cut, and it is confirmed that the mosquitoes land in two locations of the exposed skin portion within 2 minutes and then stay for one second or more. When the mosquitoes do not stay for one second or more, new mosquitoes are prepared.

Hereinafter, staying for one second or more is referred to simply as "stay." The test of an evaluation sample, which is a solution of the pest insect repellent, is performed by adjusting the concentration thereof with ethanol so that the evaluation sample can be applied at 3.3 mg/cm² to the exposed skin portion (10 cm × 3 cm). Using a pipetman, the solution with an adjusted concentration is placed on the exposed portion and applied so as to spread over the entire exposed skin portion. Then, after allowing to stand for 3 minutes, the test is started.

The test is performed by inserting the arm applied with the evaluation sample into the cage for 2 minutes, and counting the number of stays. The test is terminated when mosquitoes have stayed twice in total, and the test is performed by inserting the arm for 2 minutes every 30 minutes until the test is terminated. When the second stay occurs at 30 minutes, the repellent effect duration is determined to be 0 minutes, and when the second stay occurs at 60 minutes, the repellent effect duration is

determined to be 30 minutes. The test is performed on three subjects, and the average repellent effect duration is calculated.

In this test, the concentration of a pest insect repellent at which its repellent effect lasts for an average of more than 2 hours can be taken as the effective concentration (effective amount) of the pest insect repellent.

It is preferable that even if the composition of the present invention does not contain an effective amount of an existing pest insect repellent, it has a long-lasting repellent effect, and rather does not substantially contain pest insect repellents other than the component (A), from the viewpoint of safety. In the composition of the present invention, the content of pest insect repellents other than the component (A) is preferably 0.5 mass% or less, more preferably 0.1 mass% or less, further more preferably 0.01 mass% or less, and even more preferably substantially 0 mass%.

[0080]

Examples of existing pest insect repellents other than the component (A) include known pest insect repellent compounds, such as DEET, Icaridin, dimethyl phthalate, 2-ethyl-1,3-hexanediol, p-menthane-3,8-diol, carane-3,4-diol, di-n-butyl succinate, hydroxyanisole, rotenone, 3-(acetylbutyl)aminopropionic acid ethyl ester, citronellol, eucalyptol, α -pinene, geraniol, citronellal,

camphor, linalool, and 2-undecanone, as well as repellent essential oils, such as citronella oil and lemongrass oil. The "repellent essential oils" refer to, among essential oils obtained by the extraction, distillation, compression, or other means of components contained in plants, those having a pest insect repellent effect.

[0081]

From the viewpoint of environmental compatibility, the composition of the present invention preferably does not substantially contain organic particles having a wetting tension of 70 mN/m or less. The phrase "not substantially contain organic particles having a wetting tension of 70 mN/m or less" means that the content of the organic particles in the composition of the present invention is less than 1.0 mass%, preferably 0.5 mass% or less, more preferably 0.1 mass% or less, and further more preferably 0 mass%.

[0082]

Although the pest insects to be repelled by the composition of the present invention are not particularly limited, the composition of the present invention is more effective against flying pest insects.

The "flying pest insects" refer to, for example, pest insects which approach animals, such as humans, while flying and suck blood from their skin, pest insects which carry pathogenic bacteria or the like while flying

without sucking blood, and pest insects whose own flying gives a sense of discomfort to humans.

Specific examples of the flying pest insects include house mosquitoes, such as *Culex pipiens*, *Culex tritaeniorhynchus*, *Culex pipiens molestus*, and *Culex quinquefasciatus*; mosquitoes, such as *Aedes aegypti*, *Aedes albopictus*, *Anopheles sinensis*, *Aedes togoi*, *Anopheles gambiae*, and *Anopheles stephensi*; chironomids, such as *Chironomus yoshimatsui* and *Prosimulium akamusi*; black flies, such as *Twinnia japonensis*, *Prosimulium yezoense*, and *Simulium aokii*; flies, such as *Musca domestica*, *Muscina stabulans*, *Fannia canicularis*, blowflies, flesh flies, *Delia platura*, *Delia antiqua*, fruit flies, vinegar flies, moth flies, tsetse flies, and *Stomoxys calcitrans*; horseflies, such as *Tabanus stygius*, *Tabanus trigonus*, *Chrysops suavis*, and *Haematopota pluvialis*; biting midges, such as *Leptoconops nipponensis*, *Culicoides amamiensis*, and *Culicoides arakawae*; and bees, such as *Vespa simillima xanthoptera*, *Polistes jokahamae*, and honeybees.

The composition of the present invention has an excellent long-lasting repellent effect particularly against mosquitoes among them. The composition of the present invention has an excellent long-lasting repellent effect against, among the mosquitoes described above, *Culex pipiens*, *Culex tritaeniorhynchus*, *Culex pipiens*

molestus, *Culex quinquefasciatus*, *Aedes aegypti*, and *Aedes albopictus*.

[0083]

[Method for Producing Pest Insect Repellent
Composition]

The composition of the present invention can be obtained by mixing the components (A) to (E).

The order of mixing the components (A) to (E) is not particularly limited as long as an oil-in-water type mixture is obtained. For example, from the viewpoint of adsorbing at least a portion of the component (C) onto the surface of the component (B), the method preferably has a step of mixing a mixture containing the components (B), (C), and (E) (hereinafter referred to as "the mixture (1)") with a mixture containing the components (A) and (D) (hereinafter referred to as "the mixture (2)").

The mixture (1) can be obtained by a step of mixing the components (B), (C), and (E). The order of mixing the components (B), (C), and (E) is not particularly limited; however, it is preferable to mix the components (B) and (E), and then mix the components (B) and (E) with the component (C). From the viewpoint of uniformly adsorbing the component (C) onto the surface of the component (B), it is more preferable to mix the components (B) and (E), and then mix the components (B) and (E) with the component (C) dissolved in a solvent.

The solvent used can be a solvent mixture of water and an organic solvent. The organic solvent is not particularly limited; however, usable examples thereof include alcohols, such as methanol, ethanol, 1-propanol, and 2-propanol. More preferred among these alcohols is at least one selected from the group consisting of ethanol and 2-propanol, and further more preferred is ethanol.

The concentration of the organic solvent in the solvent mixture is preferably 70 V/V% or more, more preferably 80 V/V% or more, and further more preferably 90 V/V% or more, and is preferably 99 V/V% or less, and more preferably 98 V/V% or less, from the viewpoint of uniformly mixing the components (B), (C), and (E).

[0084]

The temperature when mixing the components (B), (C), and (E) is preferably 5°C or more, and more preferably 10°C or more, from the viewpoint of efficiently adsorbing the component (C) onto the component (B), and is preferably 70°C or less, and more preferably 50°C or less, from the viewpoint of preventing the evaporation of the solvent.

The time of mixing the components (B), (C), and (E) is preferably 20 minutes or more, and more preferably 30 minutes or more, from the viewpoint of sufficiently adsorbing the component (C) onto the component (B), and is preferably 3 hours or less, and more preferably 2

hours or less, from the viewpoint of production efficiency.

[0085]

The mixture (2) can be obtained by a step of mixing the components (A) and (D).

The temperature when mixing the components (A) and (D) is preferably 5°C or more, and more preferably 10°C or more, and is preferably 70°C or less, and more preferably 50°C or less, from the viewpoint of uniformly mixing the components (A) and (D).

The time of mixing the components (A) and (D) is preferably 20 minutes or more, more preferably 30 minutes or more, and is preferably 3 hours or less, and more preferably 2 hours or less, from the viewpoint of uniformly mixing the components (A) and (D).

[0086]

The temperature when mixing the mixtures (1) and (2) is preferably 5°C or more, and more preferably 10°C or more, and is preferably 70°C or less, and more preferably 50°C or less, from the viewpoint of uniformly dispersing the oil phase in the aqueous phase.

The time of mixing the mixtures (1) and (2) is preferably 1 minute or more, and more preferably 2 minutes or more, and is preferably 15 minutes or less, and more preferably 10 minutes or less, from the viewpoint of uniformly dispersing the oil phase in the aqueous phase.

[0087]

The rotational speed of the stirring device used to mix the mixtures (1) and (2) is preferably 3 000 rpm or more, and more preferably 4 000 rpm or more, and is preferably 20 000 rpm or less, and more preferably 15 000 rpm or less, from the viewpoint of uniformly dispersing the oil phase in the aqueous phase.

[0088]

A known stirring device can be used in the step of mixing the components (B), (C), and (E), the step of mixing the components (A) and (D), and the step of mixing the mixtures (1) and (2).

[0089]

[Method for Repelling Pest Insects]

The method for repelling pest insects of the present invention is performed by applying the pest insect repellent composition of the present invention to a human skin surface.

Another method for repelling pest insects of the present invention is to attach the pest insect repellent composition of the present invention to the legs of pest insects, particularly flying pest insects, to prevent the pest insects from staying on the human skin.

Hereinafter, these methods are also collectively referred to as "the method of the present invention."

[0090]

In the present specification, the phrase "applying to the skin surface" includes not only directly applying the composition to the skin surface by the hand or the like, but also attaching the composition to the skin surface by spraying or the like.

The amount of the composition applied to the skin surface is preferably 0.1 mg or more, more preferably 0.2 mg or more, and further more preferably 0.25 mg or more, per cm², from the viewpoint of improving the long-lasting repellent effect. The upper limit of the application amount is preferably 10 mg or less, more preferably 8 mg or less, and further more preferably 5 mg or less, per cm², from the viewpoint of suppression of stickiness and economy.

[0091]

Regarding the embodiments described above, the present invention further discloses the following embodiments.

<1>

An oil-in-water type pest insect repellent composition comprising the following components (A) to (E):

(A) 13 mass% or more and 50 mass% or less of at least one non-volatile liquid oily component selected from the group consisting of silicone oils, ester oils, ether oils, hydrocarbon oils, aliphatic alcohols, and polyhydric alcohols;

(B) 0.5 mass% or more and 35 mass% or less of hydrophilic inorganic particles containing silica particles and/or silicate mineral particles and having a volume median particle size of 1 μm or more and 40 μm or less;

(C) 0.1 mass% or more and 3 mass% or less of at least one selected from the group consisting of polyether-modified silicone, polyglycerin-modified silicone, branched polyglycerol-modified silicone, alkyl glyceryl ether-modified silicone, and carboxy-modified silicone, all of which are liquid at 25°C;

(D) 0.05 mass% or more and 3 mass% or less of a thickener; and

(E) 30 mass% or more and 86.35 mass% or less of water,

wherein the component (A) has a viscosity at 23°C of 1 mPa·s or more and 400 mPa·s or less as measured with a B-type rotational viscometer, and the component (C) is other than the silicone oils as the component (A).

<2>

The oil-in-water type pest insect repellent composition according to <1>, wherein the component (A) is at least one selected from the group consisting of silicone oils, ester oils, and hydrocarbon oils.

<3>

The oil-in-water type pest insect repellent composition according to <1> or <2>, wherein a mass ratio

of the component (B) to the component (A), $[(B)/(A)]$, is 0.02 or more and 0.7 or less.

<4>

The oil-in-water type pest insect repellent composition according to any one of <1> to <3>, wherein a mass ratio of the component (C) to the component (A), $[(C)/(A)]$, is 0.005 or more and 0.1 or less.

<5>

The oil-in-water type pest insect repellent composition according to any one of <1> to <4>, wherein the component (B) has a volume median particle size of 2 μm or more, and preferably 3 μm or more, and 30 μm or less, preferably 20 μm or less, more preferably 15 μm or less, and further more preferably 12 μm or less.

<6>

The oil-in-water type pest insect repellent composition according to any one of <1> to <5>, wherein the component (B) is contained in an amount of 1 mass% or more, preferably 2 mass% or more, and more preferably 4 mass% or more, and 27 mass% or less, preferably 16 mass% or less, and more preferably 8 mass% or less.

<7>

The oil-in-water type pest insect repellent composition according to any one of <1> to <6>, wherein the component (C) is contained in an amount of 0.2 mass% or more, preferably 0.3 mass% or more, and more preferably 0.4 mass% or more, and 2.3 mass% or less,

preferably 1.6 mass% or less, and more preferably 1.0 mass% or less.

<8>

The oil-in-water type pest insect repellent composition according to any one of <1> to <7>, wherein a mass ratio of the component (C) to the component (B), $[(C)/(B)]$, is 0.01 or more and 0.9 or less, preferably 0.05 or more, more preferably 0.07 or more, further more preferably 0.1 or more, and is preferably 0.7 or less, more preferably 0.5 or less, and further more preferably 0.3 or less.

[0092]

<9>

A method for producing an oil-in-water type pest insect repellent composition, the method comprising a step of mixing a mixture containing the following components (B), (C), and (E) with a mixture containing the following components (A) and (D):

(A) at least one non-volatile liquid oily component selected from the group consisting of silicone oils, ester oils, ether oils, hydrocarbon oils, aliphatic alcohols, and polyhydric alcohols;

(B) hydrophilic inorganic particles containing silica particles and/or silicate mineral particles and having a volume median particle size of 1 μm or more and 40 μm or less;

(C) at least one selected from the group consisting

of polyether-modified silicone, polyglycerin-modified silicone, branched polyglycerol-modified silicone, alkyl glyceryl ether-modified silicone, and carboxy-modified silicone, all of which are liquid at 25°C;

(D) a thickener; and

(E) water,

wherein the component (A) has a viscosity at 23°C of 1 mPa·s or more and 400 mPa·s or less as measured with a B-type rotational viscometer, and the component (C) is other than the silicone oils as the component (A), and

wherein in the oil-in-water type pest insect repellent composition, the component (A) is contained in an amount of 13 mass% or more and 50 mass% or less, the component (B) is contained in an amount of 0.5 mass% or more and 35 mass% or less, the component (C) is contained in an amount of 0.1 mass% or more and 3 mass% or less, the component (D) is contained in an amount of 0.05 mass% or more and 3 mass% or less, and the component (E) is contained in an amount of 30 mass% or more and 86.35 mass% or less.

<10>

The method for producing an oil-in-water type pest insect repellent composition according to <9>, wherein the component (A) is at least one selected from the group consisting of silicone oils, ester oils, and hydrocarbon oils.

<11>

The method for producing an oil-in-water type pest insect repellent composition according to <9> or <10>, wherein a mass ratio of the component (B) to the component (A), $[(B)/(A)]$, is 0.02 or more and 0.7 or less.

<12>

The method for producing an oil-in-water type pest insect repellent composition according to any one of <9> to <11>, wherein a mass ratio of the component (C) to the component (A), $[(C)/(A)]$, is 0.005 or more and 0.1 or less.

<13>

The method for producing an oil-in-water type pest insect repellent composition according to any one of <9> to <12>, wherein the component (B) has a volume median particle size of 2 μm or more, and preferably 3 μm or more, and 30 μm or less, preferably 20 μm or less, more preferably 15 μm or less, and further more preferably 12 μm or less.

<14>

The method for producing an oil-in-water type pest insect repellent composition according to any one of <9> to <13>, wherein the component (B) is contained in an amount of 1 mass% or more, preferably 2 mass% or more, and more preferably 4 mass% or more, and 27 mass% or less, preferably 16 mass% or less, and more preferably 8 mass% or less.

<15>

The method for producing an oil-in-water type pest insect repellent composition according to any one of <9> to <14>, wherein the component (C) is contained in an amount of 0.2 mass% or more, preferably 0.3 mass% or more, and more preferably 0.4 mass% or more, and 2.3 mass% or less, preferably 1.6 mass% or less, and more preferably 1.0 mass% or less.

<16>

The method for producing an oil-in-water type pest insect repellent composition according to any one of <9> to <15>, wherein a mass ratio of the component (C) to the component (B), $[(C)/(B)]$, is 0.01 or more and 0.9 or less, preferably 0.05 or more, more preferably 0.07 or more, further more preferably 0.1 or more, and is preferably 0.7 or less, more preferably 0.5 or less, and further more preferably 0.3 or less.

[0093]

<17>

A method for repelling pest insects, comprising applying an oil-in-water type pest insect repellent composition comprising the following components (A) to (E) to a human skin surface:

(A) 13 mass% or more and 50 mass% or less of at least one non-volatile liquid oily component selected from the group consisting of silicone oils, ester oils,

ether oils, hydrocarbon oils, aliphatic alcohols, and polyhydric alcohols;

(B) 0.5 mass% or more and 35 mass% or less of hydrophilic inorganic particles containing silica particles and/or silicate mineral particles and having a volume median particle size of 1 μm or more and 40 μm or less;

(C) 0.1 mass% or more and 3 mass% or less of at least one selected from the group consisting of polyether-modified silicone, polyglycerin-modified silicone, branched polyglycerol-modified silicone, alkyl glyceryl ether-modified silicone, and carboxy-modified silicone, all of which are liquid at 25°C;

(D) 0.05 mass% or more and 3 mass% or less of a thickener; and

(E) 30 mass% or more and 86.35 mass% or less of water,

wherein the component (A) has a viscosity at 23°C of 1 mPa·s or more and 400 mPa·s or less as measured with a B-type rotational viscometer, and the component (C) is other than the silicone oils as the component (A).

<18>

The method for repelling pest insects according to <17>, wherein the component (A) is at least one selected from the group consisting of silicone oils, ester oils, and hydrocarbon oils.

<19>

The method for repelling pest insects according to <17> or <18>, wherein a mass ratio of the component (B) to the component (A), $[(B)/(A)]$, is 0.02 or more and 0.7 or less.

<20>

The method for repelling pest insects according to any one of <17> to <19>, wherein a mass ratio of the component (C) to the component (A), $[(C)/(A)]$, is 0.005 or more and 0.1 or less.

<21>

The method for repelling pest insects according to any one of <17> to <20>, wherein the component (B) has a volume median particle size of 2 μm or more, and preferably 3 μm or more, and 30 μm or less, preferably 20 μm or less, more preferably 15 μm or less, and further more preferably 12 μm or less.

<22>

The method for repelling pest insects according to any one of <17> to <21>, wherein the component (B) is contained in an amount of 1 mass% or more, preferably 2 mass% or more, and more preferably 4 mass% or more, and 27 mass% or less, preferably 16 mass% or less, and more preferably 8 mass% or less.

<23>

The method for repelling pest insects according to any one of <17> to <22>, wherein the component (C) is contained in an amount of 0.2 mass% or more, preferably

0.3 mass% or more, and more preferably 0.4 mass% or more, and 2.3 mass% or less, preferably 1.6 mass% or less, and more preferably 1.0 mass% or less.

<24>

The method for repelling pest insects according to any one of <17> to <23>, wherein a mass ratio of the component (C) to the component (B), $[(C)/(B)]$, is 0.01 or more and 0.9 or less, preferably 0.05 or more, more preferably 0.07 or more, further more preferably 0.1 or more, and is preferably 0.7 or less, more preferably 0.5 or less, and further more preferably 0.3 or less.

[0094]

<25>

A method for repelling pest insects, comprising attaching an oil-in-water type pest insect repellent composition comprising the following components (A) to (E) to legs of a pest insect to prevent the pest insect from staying on a human skin:

(A) 13 mass% or more and 50 mass% or less of at least one non-volatile liquid oily component selected from the group consisting of silicone oils, ester oils, ether oils, hydrocarbon oils, aliphatic alcohols, and polyhydric alcohols;

(B) 0.5 mass% or more and 35 mass% or less of hydrophilic inorganic particles containing silica particles and/or silicate mineral particles and having a

volume median particle size of 1 μm or more and 40 μm or less;

(C) 0.1 mass% or more and 3 mass% or less of at least one selected from the group consisting of polyether-modified silicone, polyglycerin-modified silicone, branched polyglycerol-modified silicone, alkyl glyceryl ether-modified silicone, and carboxy-modified silicone, all of which are liquid at 25°C;

(D) 0.05 mass% or more and 3 mass% or less of a thickener; and

(E) 30 mass% or more and 86.35 mass% or less of water,

wherein the component (A) has a viscosity at 23°C of 1 mPa·s or more and 400 mPa·s or less as measured with a B-type rotational viscometer, and the component (C) is other than the silicone oils as the component (A).

<26>

The method for repelling pest insects according to <25>, wherein the component (A) is at least one selected from the group consisting of silicone oils, ester oils, and hydrocarbon oils.

<27>

The method for repelling pest insects according to <25> or <26>, wherein a mass ratio of the component (B) to the component (A), $[(B)/(A)]$, is 0.02 or more and 0.7 or less.

<28>

The method for repelling pest insects according to any one of <25> to <27>, wherein a mass ratio of the component (C) to the component (A), $[(C)/(A)]$, is 0.005 or more and 0.1 or less.

<29>

The method for repelling pest insects according to any one of <25> to <28>, wherein the component (B) has a volume median particle size of 2 μm or more, and preferably 3 μm or more, and 30 μm or less, preferably 20 μm or less, more preferably 15 μm or less, and further more preferably 12 μm or less.

<30>

The method for repelling pest insects according to any one of <25> to <29>, wherein the component (B) is contained in an amount of 1 mass% or more, preferably 2 mass% or more, and more preferably 4 mass% or more, and 27 mass% or less, preferably 16 mass% or less, and more preferably 8 mass% or less.

<31>

The method for repelling pest insects according to any one of <25> to <30>, wherein the component (C) is contained in an amount of 0.2 mass% or more, preferably 0.3 mass% or more, and more preferably 0.4 mass% or more, and 2.3 mass% or less, preferably 1.6 mass% or less, and more preferably 1.0 mass% or less.

<32>

The method for repelling pest insects according to

any one of <25> to <31>, wherein a mass ratio of the component (C) to the component (B), $[(C)/(B)]$, is 0.01 or more and 0.9 or less, preferably 0.05 or more, more preferably 0.07 or more, further more preferably 0.1 or more, and is preferably 0.7 or less, more preferably 0.5 or less, and further more preferably 0.3 or less.

Examples

[0095]

The present invention will be described below with reference to Examples; however, the present invention is not limited to the scope of the Examples. In the present Examples, various measurements and evaluations were performed by the following methods.

[0096]

(Surface Tension of Liquid Oily Component)

The surface tension of the liquid oily component is a surface tension measured by using an automatic surface tensiometer "Tensiometer K100" (manufactured by Kruss) by the Wilhelmy method using a platinum plate in an environment of 25°C.

[0097]

(Viscosity of Liquid Oily Component)

As a B-type rotational viscometer in accordance with JIS K7117-1: 1999, "Viscometer TVB-10," manufactured by Toki Sangyo Co., Ltd., was used. Since components to be measured have very different viscosity values for each

sample, it is difficult to accurately measure all components under a single measurement condition. For this reason, the measurement was performed using two types of rotors. The viscosity was first measured at a rotational speed of 12 rpm using a rotor M2 in an environment of 23°C. At this time, components having a viscosity of 2 500 mPa·s or more were measured again at a rotational speed of 6 rpm to obtain a viscosity value.

On the other hand, components having a viscosity of 20 mPa·s or less were measured again at a rotational speed of 30 rpm using an L adapter, which is a rotor for low viscosity, in an environment of 23°C to obtain a viscosity value.

[0098]

(Wetting Tension of Particle Surface)

Using wetting tension test mixtures of FUJIFILM Wako Pure Chemical Corporation, the measurement was performed in the following manner.

0.2 mL of a wetting tension test mixture No. 70 (surface tension: 70 mN/m) was weighed in a container in an environment of 23°C. Then, 0.01 g of particles to be measured were slowly placed in the mixture.

30 seconds after adding the particles, whether the particles got wet with the mixture was visually observed. When the particles got wet with the mixture No. 70 (surface tension: 70 mN/m), the wetting tension of the particle surface was determined to be 70 mN/m or more.

When the particles got wet with the mixture No. 70 (surface tension: 70 mN/m) described later, it was determined that the wetting tension of the particle surface was 70 mN/m or more, and that the particles were hydrophilic.

The phrase that the particle surface gets wet with the mixture means that the mixture permeates into the particles. The state in which the particles do not get wet with the mixture means that even when the particles and the mixture are mixed with each other, the particles are separated (or repelled) from the mixture.

The wetting tension of each particle surface was measured by performing the above test using each of the following wetting tension test mixtures. For example, the particle surface having a surface tension of 23 mN/m means that in the above test, when using No. 25.4 (surface tension: 25.4 mN/m) as the wetting tension test mixture, the particle surface does not get wet with this mixture, whereas when using No. 22.6 (surface tension: 22.6 mN/m) as the wetting tension test mixture, the particle surface gets wet with this mixture.

<Wetting Tension Test Mixtures of FUJIFILM Wako Pure Chemical Corporation>

No. 73 (surface tension: 73 mN/m), No. 70 (surface tension: 70 mN/m), No. 67 (surface tension: 67 mN/m), No. 64 (surface tension: 64 mN/m), No. 62 (surface tension: 62 mN/m), No. 60 (surface tension: 60 mN/m), No. 58

(surface tension: 58 mN/m), No. 56 (surface tension: 56 mN/m), No. 54 (surface tension: 54 mN/m), No. 52 (surface tension: 52 mN/m), No. 32 (surface tension: 32 mN/m), No. 30 (surface tension: 30 mN/m), No. 27.3 (surface tension: 27.3 mN/m), No. 25.4 (surface tension: 25.4 mN/m), No. 22.6 (surface tension: 22.6 mN/m)

[0099]

(Volume Median Particle Size (D50) of Hydrophilic Inorganic Particles)

Using a "Laser Scattering Particle Size Distribution Analyzer LA-960," manufactured by Horiba, Ltd., distilled water was added to a measurement cell, and each particle size distribution of hydrophilic inorganic particles was measured at a concentration at which the absorbance was in an appropriate range. The volume median particle size (D50) of the hydrophilic inorganic particles was calculated from the obtained particle size distribution.

[0100]

(Evaluation of Long-Lasting Repellent Effect against Pest Insects)

(I) Preparation of *Aedes albopictus*

As *Aedes albopictus*, imagoes grown by breeding eggs of *Aedes albopictus* purchased from Sumika Technoservice Corporation in a cage under the conditions of 27°C and a relative humidity (RH) of 60% were used.

A transparent plastic pan was filled with water in a depth of about 1 cm, a filter paper having the purchased

eggs laid thereon was placed therein, and the eggs were hatched into pupae. Thereafter, the hatched pupae were fed with a tropical fish bait (tetramine) as a food for larvae every day. One week later, the pupae were collected with a dropper and transferred into a 20-mL plastic cup, which was then transferred into a netted cage. The pupae were fed with 10 mass% of sucrose contained in a 25-mL plastic tube as a food for imagoes. After emergence, males and females were bred in the same cage for 5 days for mating. After 5 days of breeding, imagoes were collected using a fluke tube, the males and females were visually separated after anesthesia for 5 minutes on ice, and only the females were collected and used for evaluation.

[0101]

(II) Measurement of Repellent Effect Duration

100 mated female mosquitoes (*Aedes albopictus*) were placed in a plastic cage (30 × 30 × 30 cm: BugDorm-1 cage) covered with a mesh. An arm was inserted into a KUALATEC Super Long Glove (50 cm) (manufactured by AS ONE Corporation, Catalog number: 3-6432-02) with a cut of a rectangular shape with a size of 10 cm in length × 3 cm in width. In order to activate the mosquitoes, breath was blown into the case for 5 seconds. The arm was inserted into the cage under the conditions of 27°C and a relative humidity (RH) of 60% in a state that nothing was applied to an exposed portion from the cut, and it was

confirmed that the mosquitoes landed in two locations of the exposed skin portion within 2 minutes and then stayed for one second or more. When the mosquitoes did not stay for one second or more, new mosquitoes were prepared. Hereinafter, staying for one second or more is referred to simply as "stay." The test of an evaluation sample, which was a solution of the pest insect repellent, was performed by uniformly applying the evaluation sample at 3.3 mg/cm² to the exposed skin portion (10 cm × 3 cm).

The test was performed by inserting the arm applied with the evaluation sample into the cage for 2 minutes, and counting the number of stays. The test was terminated when mosquitoes had stayed twice in total, and the test was performed by inserting the arm for 2 minutes every 30 minutes until the test was terminated. When the second stay occurs at 30 minutes, the repellent effect duration is determined to be 0 minutes, and when the second stay occurs at 60 minutes, the repellent effect duration is determined to be 30 minutes. The test was performed on three subjects, and the average repellent effect duration was calculated.

[0102]

(Evaluation of Feel (Smooth Feeling When Applied to Skin))

Three expert panelists applied 0.02 mL of the composition of each Example to a 3-cm-diameter circle on the inner forearm and spread evenly over 1 minute under

the conditions of 25°C and a relative humidity (RH) of 57%. The smooth feeling during drying and after drying was subjected to sensory evaluation on 5-point scale based on the following criteria, and the average scores of the three panelists were shown in the tables. The evaluation result of No. (4) was regarded as "1."

5: No stickiness, and good and smooth feel.

4: Slight stickiness, but smooth feel.

3: A little stickiness, but stronger smooth feel.

2: Stickiness and weak smooth feel.

1: Stickiness and no smooth feel.

[0103]

(Evaluation of Long-Term Storage Stability)

80 mL of the composition of each Example was placed in a 100-mL glass bottle, which was then sealed and stored at 50°C for 1 month. Thereafter, the presence or absence of separation of the oil phase in each composition was visually observed and expressed based on the following criteria.

A: Not changed.

B: The composition does not remain on the glass bottle surface with which the composition comes into contact due to oil.

C: The oil phase coalesces, and some seepage from the emulsion to the surface is observed.

D: Separation of the oil phase is observed.

[0104]

(Production and Evaluation of Compositions)

No. 1

Components (B1) and (E) were placed in a glass beaker, and while stirring with a stirrer, a component (C1) dissolved in a solvent mixture of water and ethanol (ethanol concentration: 95 V/V%) was added, followed by stirring for 1 hour to prepare an aqueous phase. Components (A1), (D1), and (D2) were added to another glass beaker and stirred for 10 minutes using T.K. Robomix (manufactured by Tokushu Kika Kogyo Co., Ltd.) whose stirring unit was replaced by T.K. Homogenizing Disper Model 2.5 to prepare an oil phase. While the oil phase was gradually added to the aqueous phase, the mixture was pre-emulsified with propeller stirring and then stirred at 8 000 rpm for 3 minutes using T.K. Robomix (manufactured by Tokushu Kika Kogyo Co., Ltd.) whose stirring unit was replaced by T.K. Homogenizing Mixer Mark II Model 2.5 to obtain an oil-in-water type composition. Using the resulting compositions, various evaluations were performed by the methods described above. The results are shown in the tables. The compositions were produced and evaluated at room temperature (25 to 28°C).

[0105]

Nos. 2 to 55 and Nos. (1) to (14)

Composition Nos. 2 to 55 and Nos. (1) to (14) were produced in the same manner as for the composition No. 1,

except that in the production of the composition No. 1, the formulations were changed as shown in the tables. Using the resulting compositions, various evaluations were performed by the methods described above. The results are shown in the tables. The compositions were produced and evaluated at room temperature (25 to 28°C).
[0106]

As shown in the tables, in the composition Nos. 1 to 55, in the component (C), the amount of the component (C) adsorbed onto the component (B) was from 2 to 12 mass%.
[0107]

No. (15)

Ethanol (95 V/V%) was added while stirring with a stirrer to a glass beaker containing a component (E) to prepare an aqueous phase. Components (A1), (b1), (D1), and (D2) were added to the glass beaker, followed by mixing using T.K. Robomix (manufactured by Tokushu Kika Kogyo Co., Ltd.) whose stirring unit was replaced by T.K. Homogenizing Disper Model 2.5 to prepare an oil phase.

While the oil phase was gradually added to the aqueous phase, the mixture was pre-emulsified with propeller stirring and then stirred at 8 000 rpm for 3 minutes using T.K. Robomix (manufactured by Tokushu Kika Kogyo Co., Ltd.) whose stirring unit was replaced by T.K. Homogenizing Mixer Mark II Model 2.5 to obtain an oil-in-water type composition. Using the resulting compositions, various evaluations were performed by the

methods described above. The results are shown in the tables. The compositions were produced and evaluated at room temperature (25 to 28°C).

[0108]

The composition Nos. 1 to 55 listed in the tables are the compositions of the present invention, and the composition Nos. (1) to (15) are compositions outside the scope of the present invention.

The expressions (A) to (E) shown in the tables correspond to the components (A) to (E), respectively, in the present specification. The expressions (a), (b), and (c) correspond to the liquid oily component outside the range of the component (A), the particles outside the range of the component (B), and the modified silicone outside the range of the component (C), respectively, in the present specification. For convenience, the tables list the components (a) to (c) in correspondence with the components (A) to (C), in terms of physical properties and contents in the compositions.

Table 1 shows the components used in Tables 2 to 8. The blending amounts listed in each table are the active ingredient amounts (mass%) of the respective components.

For the content shown in the row of the component (C8) of the composition No. 27, the content of PEG/PPG-19/19 dimethicone (content of component (C)) in the composition is shown in parentheses. Similarly, for Nos. 37, 38, 42, 44, and (7) to (9), the content of the

component (C) or the component outside the range of the component (C) in the composition is shown in parentheses.

"O/W" refers to an oil-in-water type composition, and "W/O" refers to a water-in-oil type composition.

[0109]

[Table 1]

Table 1

Product name	Manufacturer name	Component formulation (mass%)	Classification	Component name	Viscosity [mPa·s] ¹⁾	Surface tension (25°C) [mN/m]	Wetting tension (23°C) [mN/m]	Particle size (D50) [μm]	Structure	Shape	HLB value	Modified silicone classification
SILICONE KF-98A-6cs	Shin-Etsu Chemical Co., Ltd.	100	(A1)	Dimethicone	5	19						
EXCEPARL IPP	Kao Corporation	100	(A2)	Isopropyl palmitate	7	27						
NIKKOL SOLIALANE	Nippon Surfactant Industries Co., Ltd.	100	(A3)	Squalane	42	30						
HCS 160M5	JGC Catalysts and Chemicals Ltd.	100	(B1)	Silica			73 or more	5	Porous	Spherical		
SUNSPHERE NP-30	AGC Si-Tech Co., Ltd.	100	(B2)	Silica			73 or more	3	Non-porous	Spherical		
SUNSPHERE NP-100	AGC Si-Tech Co., Ltd.	100	(B3)	Silica			73 or more	10	Non-porous	Spherical		
SATINER M13	JGC Catalysts and Chemicals Ltd.	100	(B4)	Silica			73 or more	13	Non-porous	Spherical		
SUNSPHERE NP-200	AGC Si-Tech Co., Ltd.	100	(B5)	Silica			73 or more	20	Non-porous	Spherical		
SILICA MICRO BEAD P-4000	JGC Catalysts and Chemicals Ltd.	100	(B6)	Silica			73 or more	30	Non-porous	Spherical		
SUNSPHERE H-51	AGC Si-Tech Co., Ltd.	100	(B7)	Silica			73 or more	5	Porous	Spherical		
GODD BALL AF-16C	Suzuki Yushi Industrial Co. Ltd.	100	(B8)	Silica			73 or more	5	Porous	Spherical		
GODD BALL B-25C	Suzuki Yushi Industrial Co. Ltd.	100	(B9)	Silica			73 or more	9	Porous	Spherical		
SUNSPHERE H-122	AGC Si-Tech Co., Ltd.	100	(B10)	Silica			73 or more	12	Porous	Spherical		
SUNSPHERE H-201	AGC Si-Tech Co., Ltd.	100	(B11)	Silica			73 or more	20	Porous	Spherical		
TALC JA-13R	Asada Milling Co., Ltd.	100	(B12)	Talc			70	6	Non-porous	Flat		
TALC JA-68R	Asada Milling Co., Ltd.	100	(B13)	Talc			70	11	Non-porous	Flat		
MICA Y-1800	Yamaguchi Mica Co., Ltd.	100	(B14)	Mica			70	10	Non-porous	Flat		
GANZPEARL SIG-070	Aica Kogyo Co. Ltd.	100	(b1)	Polymethylmethacrylate			23	7	Non-porous	Spherical		
AEROSIL 200	Nippon Aerosil Co., Ltd.	100	(b2)	Silica			73 or more	0.012	Non-porous	Spherical		
MT-100WP	Tayca Corporation	100	(b3)	Titanium oxide			73 or more	0.015	Non-porous	Spindle		
MZ-500	Tayca Corporation	100	(b4)	Zinc oxide			73 or more	0.025	Non-porous	Spherical		
SILICONE KF-6017	Shin-Etsu Chemical Co., Ltd.	100	(C1)	PEG-10 dimethicone	600						4.5	Polyether-modified
SILICONE KF-6011	Shin-Etsu Chemical Co., Ltd.	100	(C2)	PEG-11 methyl ether dimethicone	130						14.5	Polyether-modified
SILICONE KF-6004	Shin-Etsu Chemical Co., Ltd.	100	(C3)	PEG-32 methyl ether dimethicone	100,000						9	Polyether-modified
SILICONE KF-6015	Shin-Etsu Chemical Co., Ltd.	100	(C4)	PEG-3 dimethicone	150						4.5	Polyether-modified
SILICONE KF-6028	Shin-Etsu Chemical Co., Ltd.	100	(C5)	PEG-9 polydimethylsiloxethyl dimethicone	900						4	Polyether-modified
SILICONE KF-6038	Shin-Etsu Chemical Co., Ltd.	100	(C6)	Lauryl PEG-9 polydimethylsiloxethyl dimethicone	700						3	Polyether-modified
DOWSIL SH 3775 M	Dow Toray Co., Ltd.	100	(C7)	PEG-12 dimethicone	1,600						5	Polyether-modified
		50	(C8)	PEG/PPG-19/19 dimethicone	3,000						3	Polyether-modified
DOWSIL BY 25-337	Dow Toray Co., Ltd.	(40) (10)		((C13-16) isoparaffin) ((C10-13) isoparaffin)								
DOWSIL ES-5300 Formulation Aid	Dow Toray Co., Ltd.	100	(C9)	Lauryl PEG-10 tris(trimethylsiloxy)silyl ethyl dimethicone	1,800						3	Polyether-modified
DOWSIL ES-5300 Formulation Aid	Dow Toray Co., Ltd.	100	(C10)	Carboxydecyl dimethicone	2,000							Carboxyl-modified
SILICONE KF-6104	Shin-Etsu Chemical Co., Ltd.	100	(C11)	Polyglyceryl-3 polydimethylsiloxethyl dimethicone	4,000							Polyglycerin-modified
SOFCARE GS-G	Kao Corporation	100	(C12)	Bis(polyglyceryl-3-oxyphenylpropyl) dimethicone	20,000-100,000							Branched polyglycerol-modified
SOFCARE RS-U	Kao Corporation	100	(C13)	Bisalkyl (C16-18) glyceryl undecyl dimethicone	4000-9000							Alkyl glyceryl ether-modified
DOWSIL FA-4002 ID Silicone Acrylate	Dow Toray Co., Ltd.	30 (70)	(c1)	(Acrylate/polytrimethylsiloxy methacrylate) copolymer (isododecane)	Solid							Silicone dendrimer-modified
DOWSIL EL-8050 ID Silicone Organic Elastomer Blend	Dow Toray Co., Ltd.	15 (85)	(c2)	(Dimethicone/bisobutyl PPG-20) crosspolymer (isododecane)	350,000-580,000 (solid)							Carboxyl-modified
DOWSIL EL-7314 Silicone Elastomer Blend	Dow Toray Co., Ltd.	30 (70)	(c3)	(Hexylsuccinyl dimethicone) crosspolymer (isododecane)	1 million (solid)							Carboxyl-modified
ARISTOFLEX Velvet	Clariant Iberica Production S.A.	100	(D1)	(Ammonium acryloyl/dimethylacrylate/beheneth-25 methacrylate) crosspolymer								
SANGELOSE 60L	Daido Chemical Industry Co., Ltd.	100	(D2)	Hydroxypropyl methylcellulose stearoyl ether								
PEMULEN TR-1	Lubrizol Advanced Materials, Inc.	100	(D3)	(Acrylate/alkyl acrylate (C10-C30)) crosspolymer								

*1: For component (A), viscosity at 23°C measured with a B-type rotational viscometer; for component (C), viscosity at 25°C measured using a Brookfield viscometer based on JIS Z8803:2011 "Methods for viscosity measurement of liquid"

[0110]

[Table 2]

Table 2

Classification	Component name	1	2	3	4	5	6	7	8	9	10	11
(A1)	Dimethicone	30.00	30.00	30.00	30.00	30.00	30.00	30.00	30.00	30.00	30.00	30.00
(B1)	Silica	6.00										
(B2)	Silica		6.00									
(B3)	Silica											
(B4)	Silica			6.00								
(B5)	Silica					6.00						
(B6)	Silica						6.00					
(B7)	Silica							6.00				
(B8)	Silica								6.00			
(B9)	Silica									6.00		
(B10)	Silica										6.00	
(B11)	Silica											6.00
(C1)	PEG-10 dimethicone	0.75	0.75	0.75	0.75	0.75	0.75	0.75	0.75	0.75	0.75	0.75
(D1)	Crosspolymer *1	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30
(D2)	Hydroxypropyl methylcellulose stearoxy ether	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30
(E)	Water	57.65	57.65	57.65	57.65	57.65	57.65	57.65	57.65	57.65	57.65	57.65
Other components	ETHANOL (95V/V%)	5.00	5.00	5.00	5.00	5.00	5.00	5.00	5.00	5.00	5.00	5.00
Total content of components (mass%)		100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00
Mass ratio (B)/(A)		0.20	0.20	0.20	0.20	0.20	0.20	0.20	0.20	0.20	0.20	0.20
Mass ratio (C)/(A)		0.025	0.025	0.025	0.025	0.025	0.025	0.025	0.025	0.025	0.025	0.025
Mass ratio (C)/(B)		0.125	0.125	0.125	0.125	0.125	0.125	0.125	0.125	0.125	0.125	0.125
Amount of (C) adsorbed onto (B) in (C) (mass%)		7.11	7.87	7.92	8.25	8.33	8.12	7.57	7.65	7.88	8.01	8.34
Repellent effect duration	Repellent effect immediately after use	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays
	30 min after use	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays
	60 min after use	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays
	90 min after use	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays
	120 min after use	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays
	150 min after use	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays
	180 min after use	1 stay	0 stays	0 stays	0 stays	1 stay	1 stay	0 stays	0 stays	1 stay	1 stay	0 stays
	210 min after use	1 stay	0 stays	0 stays	0 stays	1 stay	1 stay	1 stay	2 stays	1 stay	1 stay	1 stay
	240 min after use	1 stay	2 stays	2 stays	2 stays			1 stay		1 stay		1 stay
	270 min after use											
	300 min after use											
	330 min after use											
	360 min after use											
	390 min after use											
	420 min after use											
	450 min after use											
	480 min after use											
Feel		3.7	3.3	3.0	2.8	2.7	2.7	3.5	3.7	3.3	3.0	2.8
Long-term storage stability		A	A	A	A	A	A	B	A	B	A	B
Emulsification system		O/W	O/W	O/W	O/W	O/W	O/W	O/W	O/W	O/W	O/W	O/W

*1 (Ammonium acryloyldimethyltaurate/beheneth-25 methacrylate) crosspolymer

[0111]

[Table 3]

Table 3

Classification	Component name	12	13	14	15	16	17	18	19	20
(A1)	Dimethicone	30.00	30.00	30.00	30.00	30.00	30.00	30.00	30.00	30.00
(B1)	Silica	3.00	3.00	3.00	2.00	12.00				
(B4)	Silica						2.00	12.00	2.00	12.00
(B11)	Silica									
(B12)	Talc	3.00								
(B13)	Talc		3.00							
(B14)	Mica			3.00						
(C1)	PEG-10 dimethicone	0.75	0.75	0.75	0.75	0.75	0.75	0.75	0.75	0.75
(D1)	Crosspolymer *1	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30
(D2)	Hydroxypropyl methylcellulose stearoxy ether	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30
(D3)	(Acrylate/alkyl acrylate (C10-30)) crosspolymer									
(E)	Water	57.65	57.65	57.65	61.65	51.65	61.65	51.65	61.65	51.65
Other components	ETHANOL (95V/V%)	5.00	5.00	5.00	5.00	5.00	5.00	5.00	5.00	5.00
Total content of components (mass%)		100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00
Mass ratio (B)/(A)		0.20	0.20	0.20	0.07	0.40	0.07	0.40	0.07	0.40
Mass ratio (C)/(A)		0.025	0.025	0.025	0.025	0.025	0.025	0.025	0.025	0.025
Mass ratio (C)/(B)		0.125	0.125	0.125	0.375	0.063	0.375	0.063	0.375	0.063
Amount of (C) adsorbed onto (B) in (C) (mass%)		7.51	8.34	7.98	4.44	11.24	3.98	10.9	3.67	10.66
Repellent effect duration	Repellent effect immediately after use	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays
	30 min after use	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays
	60 min after use	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays
	90 min after use	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays
	120 min after use	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays
	150 min after use	1 stay	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays
	180 min after use	1 stay	2 stays	2 stays	1 stay	2 stays	1 stay	2 stays	0 stays	0 stays
	210 min after use				1 stay		1 stay		1 stay	2 stays
	240 min after use								1 stay	
	270 min after use									
	300 min after use									
	330 min after use									
	360 min after use									
	390 min after use									
	420 min after use									
	450 min after use									
	480 min after use									
Feel		3.0	3.0	2.8	2.7	4.7	2.3	4.5	2.0	4.5
Long-term storage stability		B	B	A	B	A	B	A	C	B
Emulsification system		O/W	O/W	O/W	O/W	O/W	O/W	O/W	O/W	O/W
*1 (Ammonium acryloyldimethyltaurate/beheneth-25 methacrylate) crosspolymer										

[0112]

[Table 4]

Table 4															
Classification	Component name														
(A1)	Dimethicone		21	22	23	24	25	26	27	28	29	30	31	32	
(B1)	Silica		30.00	30.00	30.00	30.00	30.00	30.00	30.00	30.00	30.00	30.00	30.00	30.00	
(C2)	PEG-11 methyl ether dimethicone		6.00	6.00	6.00	6.00	6.00	6.00	6.00	6.00	6.00	6.00	6.00	6.00	
(C3)	PEG-32 methyl ether dimethicone		0.75												
(C4)	PEG-3 dimethicone			0.75											
(C5)	PEG-9 polydimethylsiloxethyl dimethicone					0.75									
(C6)	Lauryl PEG-9 polydimethylsiloxethyl dimethicone						0.75								
(C7)	PEG-12 dimethicone							0.75							
(C8)	PEG/PPG-19/19 dimethicone ((C13-16) isoparaffin) ((C10-13) isoparaffin)								1.50 (0.75)						
(C9)	Lauryl PEG-10 tris(trimethylsiloxy)silylethyl dimethicone									0.75					
(C10)	Carboxydecyl dimethicone										0.75				
(C11)	Polyglyceryl-3 polydimethylsiloxethyl dimethicone											0.75			
(C12)	Bis(polyglyceryl-3-oxyphenylpropyl) dimethicone												0.75		
(C13)	Bisalkyl (C16-18) glyceryl undecyl dimethicone													0.75	
(D1)	Grosspolymer *1		0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30	
(D2)	Hydroxypropyl methylcellulose stearoxy ether		0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30	
(E)	Water		57.65	57.65	57.65	57.65	57.65	57.65	56.90	57.65	57.65	57.65	57.65	57.65	
Other components	ETHANOL (95V/V%)		5.00	5.00	5.00	5.00	5.00	5.00	5.00	5.00	5.00	5.00	5.00	5.00	
Total content of components (mass%)			100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	
Mass ratio (B)/(A)			0.20	0.20	0.20	0.20	0.20	0.20	0.20	0.20	0.20	0.20	0.20	0.20	
Mass ratio (C)/(A)			0.025	0.025	0.025	0.025	0.025	0.025	0.025	0.025	0.025	0.025	0.025	0.025	
Mass ratio (C)/(B)			0.125	0.125	0.125	0.125	0.125	0.125	0.125	0.125	0.125	0.125	0.125	0.125	
Amount of (C) adsorbed onto (B) in (C) (mass%)			7.18	8.98	9.77	8.34	9.01	8.39	7.87	8.12	8.88	7.57	6.78	6.87	
Repellent effect duration	Repellent effect immediately after use		0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	
	30 min after use		0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	
	60 min after use		0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	
	90 min after use		0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	
	120 min after use		0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	
	150 min after use		0 stays	0 stays	0 stays	0 stays	0 stays	1 stay	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	
	180 min after use		0 stays	0 stays	0 stays	0 stays	0 stays	1 stay	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	
	210 min after use		1 stay	0 stays	0 stays	2 stays	2 stays	1 stay	0 stays	0 stays	1 stay	1 stay	0 stays	0 stays	
	240 min after use		1 stay	2 stays	2 stays				1 stay	1 stay	1 stay	1 stay	2 stays	2 stays	
	270 min after use														
	300 min after use														
	330 min after use														
	360 min after use														
	390 min after use														
	420 min after use														
	450 min after use														
	480 min after use														
Feel			3.5	2.3	2.3	2.5	2.0	2.8	3.3	2.8	2.3	3.2	3.7	3.5	
Long-term storage stability			A	A	A	A	A	A	A	A	A	A	A	A	
Emulsification system			O/W	O/W	O/W	O/W	O/W	O/W	O/W	O/W	O/W	O/W	O/W	O/W	

*1 (Ammonium acryloyldimethyltaurate/beheneth-25 methacrylate) crosspolymer

[0113]

[Table 5]

Table 5

Classification	Component name	33	34	35	36	37	38	39	40	41	42	43	44
(A1)	Dimethicone	30.00	30.00	30.00	30.00	30.00	30.00	30.00	30.00	30.00	30.00	30.00	30.00
(B1)	Silica	6.00	6.00	6.00	6.00	6.00	6.00	6.00	6.00	6.00	6.00	6.00	6.00
(C1)	PEG-10 dimethicone	0.20	1.50							0.75	0.75	0.75	0.75
(C2)	PEG-11 methyl ether dimethicone			0.20	1.50					0.75			
(C8)	PEG/PPG-19/19 dimethicone ((C13-16) isoparaffin) ((C10-13) isoparaffin)					0.40 (0.20)	3.00 (1.50)				1.50 (0.75)		3.00 (1.50)
(C12)	Bis(polyglyceryl-3-oxphenylpropyl) dimethicone							0.20	1.50			0.75	
(D1)	Crosspolymer *1	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30
(D2)	Hydroxypropyl methylcellulose stearoxy ether	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30
(E)	Water	58.20	56.90	58.20	56.90	58.00	55.40	58.20	56.90	56.90	56.15	56.90	55.40
Other components	ETHANOL (95V/V%)	5.00	5.00	5.00	5.00	5.00	5.00	5.00	5.00	5.00	5.00	5.00	5.00
Total content of components (mass%)		100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00
Mass ratio (B)/(A)		0.20	0.20	0.20	0.20	0.20	0.20	0.20	0.20	0.20	0.20	0.20	0.20
Mass ratio (C)/(A)		0.007	0.050	0.007	0.050	0.007	0.050	0.007	0.050	0.050	0.050	0.050	0.075
Mass ratio (C)/(B)		0.033	0.250	0.033	0.250	0.033	0.250	0.033	0.250	0.250	0.250	0.250	0.375
Amount of (C) adsorbed onto (B) in (C) (mass%)		2.53	9.88	2.78	10.01	3.45	11.88	2.37	9.68	7.12	7.54	7.01	9.58
Repellent effect duration	Repellent effect immediately after use	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays
	30 min after use	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays
	60 min after use	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays
	90 min after use	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays
	120 min after use	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays
	150 min after use	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays
	180 min after use	2 stays	0 stays	1 stay	0 stays	2 stays	0 stays	2 stays	0 stays	0 stays	0 stays	0 stays	0 stays
	210 min after use		1 stay		0 stays		0 stays		1 stay	0 stays	0 stays	0 stays	0 stays
	240 min after use		1 stay		2 stays		0 stays		1 stay	0 stays	0 stays	1 stay	0 stays
	270 min after use						1 stay			2 stays	2 stays	1 stay	0 stays
	300 min after use						1 stay						1 stay
	330 min after use												1 stay
	360 min after use												
	390 min after use												
	420 min after use												
	450 min after use												
	480 min after use												
Feel		4.2	3.2	4.0	2.8	3.8	2.8	4.2	3.2	3.5	3.3	3.5	2.7
Long-term storage stability		B	A	B	A	A	A	B	A	A	A	A	A
Emulsification system		OW	OW	OW	OW	OW	OW	OW	OW	OW	OW	OW	OW

*1 (Ammonium acryloyldimethyltaurate/beheneth-25 methacrylate) crosspolymer

[0114]

[Table 6]

Table 6

Classification	Component name	45	46	47	48	49	50	51	52	53	54	55
(A1)	Dimethicone	20.00	45.00							30.00	30.00	30.00
(A2)	Isopropyl palmitate			20.00	30.00	45.00						
(A3)	Squalane						20.00	30.00	45.00			
(B1)	Silica			6.00	6.00	6.00	6.00	6.00	6.00	6.00	6.00	6.00
(C1)	PEG-10 dimethicone	0.75	0.75	0.75	0.75	0.75	0.75	0.75	0.75	0.75	0.75	0.75
(D1)	Crosspolymer *1	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.50		
(D2)	Hydroxypropyl methylcellulose stearoxy ether	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.50	1.00	0.30
(D3)	(Acrylate/alkyl acrylate (C10-30)) crosspolymer											0.30
(E)	Water	67.65	42.65	67.65	57.65	42.65	67.65	57.65	42.65	57.25	57.25	57.65
Other components	ETHANOL (95V/V%)	5.00	5.00	5.00	5.00	5.00	5.00	5.00	5.00	5.00	5.00	5.00
Total content of components (mass%)		100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.30
Mass ratio (B)/(A)		0.30	0.13	0.30	0.20	0.13	0.30	0.20	0.13	0.20	0.20	0.20
Mass ratio (C)/(A)		0.038	0.017	0.038	0.025	0.017	0.038	0.025	0.017	0.025	0.025	0.025
Mass ratio (C)/(B)		0.125	0.125	0.125	0.125	0.125	0.125	0.125	0.125	0.125	0.125	0.125
Amount of (C) adsorbed onto (B) in (C) (mass%)		7.16	7.05	7.1	7.11	7.13	7.05	7.08	7.11	7.08	7.06	7.1
Repellent effect duration	Repellent effect immediately after use	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays
	30 min after use	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays
	60 min after use	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays
	90 min after use	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays
	120 min after use	0 stays	0 stays	1 stay	0 stays	0 stays	1 stay	0 stays	0 stays	0 stays	0 stays	0 stays
	150 min after use	0 stays	0 stays	1 stay	0 stays	0 stays	1 stay	2 stays	0 stays	0 stays	0 stays	0 stays
	180 min after use	2 stays	0 stays		2 stays	0 stays			0 stays	1 stay	0 stays	0 stays
	210 min after use		0 stays			0 stays			0 stays	1 stay	2 stays	2 stays
	240 min after use		0 stays			0 stays			0 stays			
	270 min after use		0 stays			0 stays			1 stay			
	300 min after use		0 stays			0 stays			1 stay			
	330 min after use		0 stays			2 stays						
	360 min after use		0 stays									
	390 min after use		2 stays									
	420 min after use											
	450 min after use											
	480 min after use											
Feel		4.3	3.0	4.0	3.3	2.5	3.8	3.0	2.2	3.2	2.7	3.0
Long-term storage stability		A	B	A	A	B	A	A	B	A	A	B
Emulsification system		OW	OW	OW	OW	OW	OW	OW	OW	OW	OW	OW
*1 (Ammonium acryloyldimethyltaurate/beheneth-25 methacrylate) crosspolymer												

[0115]

[Table 7]

Classification	Component name	(1)	(2)	(3)	(4)	(5)	(6)	(7)	(8)	(9)
(A1)	Dimethicone	30.00	30.00	30.00	30.00	30.00	30.00	30.00	30.00	30.00
(B1)	Silica					0.10	40.00	6.00	6.00	6.00
(b2)	Silica	6.00								
(b3)	Titanium oxide		6.00							
(b4)	Zinc oxide			6.00						
(C1)	PEG-10 dimethicone	0.75	0.75	0.75	0.75	0.75	0.75			
(c1)	(Acrylate/polytrimethylsiloxy methacrylate) copolymer (isododecane)							2.50 (0.75)		
(c2)	(Dimethicone/bisobutyl PPG-20) crosspolymer (isododecane)								5.00 (0.75)	
(c3)	(Hexyl/succinyl dimethicone) crosspolymer (isododecane)									2.50 (0.75)
(D1)	Crosspolymer *1	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30
(D2)	Hydroxypropyl methylcellulose stearoxy ether	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30	0.30
(E)	Water	57.65	57.65	57.65	63.65	63.55	23.65	55.90	53.40	55.90
Other components ETHANOL (95V/V%)		5.00	5.00	5.00	5.00	5.00	5.00	5.00	5.00	5.00
Total content of components (mass%)		100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00
Mass ratio (B)/(A)		0.20	0.20	0.20	-	0.003	1.33	0.20	0.20	0.20
Mass ratio (C)/(A)		0.025	0.025	0.025	0.025	0.025	0.025	0.025	0.025	0.025
Mass ratio (C)/(B)		0.125	0.125	0.125	-	7.500	0.019	0.125	0.125	0.125
Amount of (C) adsorbed onto (B) in (C) (mass%)		7.71	8.76	8.9	-	0.31	20.34	unmeasurable	unmeasurable	unmeasurable
Repellent effect duration	Repellent effect immediately after use	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays
	30 min after use	0 stays	0 stays	0 stays	0 stays	0 stays	1 stay	0 stays	0 stays	0 stays
	60 min after use	0 stays	0 stays	0 stays	2 stays	2 stays	1 stay	0 stays	0 stays	0 stays
	90 min after use	0 stays	1 stay	0 stays				0 stays	0 stays	0 stays
	120 min after use	0 stays	1 stay	1 stay				0 stays	0 stays	0 stays
	150 min after use	0 stays						1 stay	0 stays	1 stay
	180 min after use	1 stay						1 stay	2 stays	1 stay
	210 min after use	1 stay								
	240 min after use									
	270 min after use									
	300 min after use									
	330 min after use									
	360 min after use									
	390 min after use									
	420 min after use									
	450 min after use									
	480 min after use									
Feel		1.2	1.0	1.3	1.0	1.2	5.0	1.8	1.3	1.2
Long-term storage stability		B	B	B	D	D	A	C	D	D
Emulsification system		O/W	O/W	O/W	O/W	O/W	O/W	O/W	O/W	O/W

*1 (Ammonium acryloyldimethyltaurate/beheneth-25 methacrylate) crosspolymer

[0116]

[Table 8]

Table 8

Classification	Component name	(10)	(11)	(12)	(13)	(14)	(15)
(A1)	Dimethicone	30.00	30.00	30.00	10.00	60.00	30.00
(B1)	Silica	6.00	6.00	6.00	6.00	6.00	
(b1)	Polymethylmethacrylate						6.00
(C1)	PEG-10 dimethicone		0.05	5.00	0.75	0.75	
(D1)	Crosspolymer *1	0.30	0.30	0.30	0.30	0.30	0.30
(D2)	Hydroxypropyl methylcellulose stearoxy ether	0.30	0.30	0.30	0.30	0.30	0.30
(E)	Water	58.40	58.35	53.40	77.65	27.65	58.40
Other components	ETHANOL (95V/V%)	5.00	5.00	5.00	5.00	5.00	5.00
Total content of components (mass%)		100.00	100.00	100.00	100.00	100.00	100.00
Mass ratio (B)/(A)		0.20	0.20	0.20	0.60	0.10	0.20
Mass ratio (C)/(A)		0.000	0.002	0.167	0.075	0.013	0.000
Mass ratio (C)/(B)		0.000	0.008	0.833	0.125	0.125	0.000
Amount of (C) adsorbed onto (B) in (C) (mass%)		-	0.52	34.4	7.09	7.06	-
Repellent effect duration	Repellent effect immediately after use	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays
	30 min after use	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays
	60 min after use	0 stays	0 stays	0 stays	0 stays	0 stays	0 stays
	90 min after use	0 stays	0 stays	0 stays	2 stays	0 stays	0 stays
	120 min after use	0 stays	0 stays	0 stays		0 stays	1 stay
	150 min after use	0 stays	0 stays	0 stays		0 stays	1 stay
	180 min after use	2 stays	1 stay	0 stays		0 stays	
	210 min after use			1 stay		0 stays	
	240 min after use			1 stay		0 stays	
	270 min after use					0 stays	
	300 min after use					0 stays	
	330 min after use					0 stays	
	360 min after use					0 stays	
	390 min after use					0 stays	
	420 min after use					0 stays	
	450 min after use					1 stay	
	480 min after use					1 stay	
Feel		1.2	1.0	1.7	4.8	2.3	3.0
Long-term storage stability		D	D	D	A	D	A
Emulsification system		O/W	O/W	W/O	O/W	W/O	O/W

*1 (Ammonium acryloyldimethyltaurate/beheneth-25 methacrylate) crosspolymer

[0117]

Tables 2 to 8 reveal that the compositions of the present invention containing predetermined amounts each of the components (A) to (E) have a smooth and good feel when applied to the skin and excellent long-term storage stability, and further can maintain the long-lasting repellent effect for 2 hours or more. A comparison between, for example, the composition Nos. 1 to 14 and 21 to 44 with the composition No. (15) reveals that due to the use of the component (B) specified in the present invention in combination with the component (C) in place of the hydrophobic organic particles

(polymethylmethacrylate) used in the composition No.(15), the long-lasting repellent effect can be maintained for a longer period of time.

Industrial Applicability

[0118]

The present invention makes it possible to provide an oil-in-water type pest insect repellent composition which has a good feel when applied to the skin and excellent long-term storage stability, and a method for repelling pest insects.

Claims

[Claim 1] An oil-in-water type pest insect repellent composition comprising the following components (A) to (E) :

(A) 13 mass% or more and 50 mass% or less of at least one non-volatile liquid oily component selected from the group consisting of silicone oils, ester oils, ether oils, hydrocarbon oils, aliphatic alcohols, and polyhydric alcohols;

(B) 0.5 mass% or more and 35 mass% or less of hydrophilic inorganic particles containing silica particles and/or silicate mineral particles and having a volume median particle size of 1 μm or more and 40 μm or less;

(C) 0.1 mass% or more and 3 mass% or less of at least one selected from the group consisting of polyether-modified silicone, polyglycerin-modified silicone, branched polyglycerol-modified silicone, alkyl glyceryl ether-modified silicone, and carboxy-modified silicone, all of which are liquid at 25°C;

(D) 0.05 mass% or more and 3 mass% or less of a thickener; and

(E) 30 mass% or more and 86.35 mass% or less of water,

wherein the component (A) has a viscosity at 23°C of 1 mPa·s or more and 400 mPa·s or less as measured with a

B-type rotational viscometer, and the component (C) is other than the silicone oils as the component (A).

[Claim 2] The oil-in-water type pest insect repellent composition according to claim 1, wherein the component (A) is at least one selected from the group consisting of silicone oils, ester oils, and hydrocarbon oils.

[Claim 3] The oil-in-water type pest insect repellent composition according to claim 1 or 2, wherein a mass ratio of the component (B) to the component (A), $[(B)/(A)]$, is 0.02 or more and 0.7 or less.

[Claim 4] The oil-in-water type pest insect repellent composition according to any one of claims 1 to 3, wherein a mass ratio of the component (C) to the component (A), $[(C)/(A)]$, is 0.005 or more and 0.1 or less.

[Claim 5] The oil-in-water type pest insect repellent composition according to any one of claims 1 to 4, wherein a mass ratio of the component (C) to the component (B), $[(C)/(B)]$, is 0.01 or more and 0.3 or less.

[Claim 6] A method for producing an oil-in-water type pest insect repellent composition, the method comprising

a step of mixing a mixture containing the following components (B), (C), and (E) with a mixture containing the following components (A) and (D):

(A) at least one non-volatile liquid oily component selected from the group consisting of silicone oils, ester oils, ether oils, hydrocarbon oils, aliphatic alcohols, and polyhydric alcohols;

(B) hydrophilic inorganic particles containing silica particles and/or silicate mineral particles and having a volume median particle size of 1 μm or more and 40 μm or less;

(C) at least one selected from the group consisting of polyether-modified silicone, polyglycerin-modified silicone, branched polyglycerol-modified silicone, alkyl glyceryl ether-modified silicone, and carboxy-modified silicone, all of which are liquid at 25°C;

(D) a thickener; and

(E) water,

wherein the component (A) has a viscosity at 23°C of 1 mPa·s or more and 400 mPa·s or less as measured with a B-type rotational viscometer, and the component (C) is other than the silicone oils as the component (A), and

wherein in the oil-in-water type pest insect repellent composition, the component (A) is contained in an amount of 13 mass% or more and 50 mass% or less, the component (B) is contained in an amount of 0.5 mass% or more and 35 mass% or less, the component (C) is contained

in an amount of 0.1 mass% or more and 3 mass% or less, the component (D) is contained in an amount of 0.05 mass% or more and 3 mass% or less, and the component (E) is contained in an amount of 30 mass% or more and 86.35 mass% or less.

[Claim 7] A method for repelling pest insects, comprising applying an oil-in-water type pest insect repellent composition comprising the following components (A) to (E) to a human skin surface:

(A) 13 mass% or more and 50 mass% or less of at least one non-volatile liquid oily component selected from the group consisting of silicone oils, ester oils, ether oils, hydrocarbon oils, aliphatic alcohols, and polyhydric alcohols;

(B) 0.5 mass% or more and 35 mass% or less of hydrophilic inorganic particles containing silica particles and/or silicate mineral particles and having a volume median particle size of 1 μm or more and 40 μm or less;

(C) 0.1 mass% or more and 3 mass% or less of at least one selected from the group consisting of polyether-modified silicone, polyglycerin-modified silicone, branched polyglycerol-modified silicone, alkyl glyceryl ether-modified silicone, and carboxy-modified silicone, all of which are liquid at 25°C;

(D) 0.05 mass% or more and 3 mass% or less of a thickener; and

(E) 30 mass% or more and 86.35 mass% or less of water,

wherein the component (A) has a viscosity at 23°C of 1 mPa·s or more and 400 mPa·s or less as measured with a B-type rotational viscometer, and the component (C) is other than the silicone oils as the component (A).

[Claim 8] A method for repelling pest insects, comprising attaching an oil-in-water type pest insect repellent composition comprising the following components (A) to (E) to legs of a pest insect to prevent the pest insect from staying on a human skin:

(A) 13 mass% or more and 50 mass% or less of at least one non-volatile liquid oily component selected from the group consisting of silicone oils, ester oils, ether oils, hydrocarbon oils, aliphatic alcohols, and polyhydric alcohols;

(B) 0.5 mass% or more and 35 mass% or less of hydrophilic inorganic particles containing silica particles and/or silicate mineral particles and having a volume median particle size of 1 μm or more and 40 μm or less;

(C) 0.1 mass% or more and 3 mass% or less of at least one selected from the group consisting of polyether-modified silicone, polyglycerin-modified

silicone, branched polyglycerol-modified silicone, alkyl glyceryl ether-modified silicone, and carboxy-modified silicone, all of which are liquid at 25°C;

(D) 0.05 mass% or more and 3 mass% or less of a thickener; and

(E) 30 mass% or more and 86.35 mass% or less of water,

wherein the component (A) has a viscosity at 23°C of 1 mPa·s or more and 400 mPa·s or less as measured with a B-type rotational viscometer, and the component (C) is other than the silicone oils as the component (A).