Title: COMPOSITION AND METHOD FOR CONTROLLING HARMFUL ARTHROPODS

Abstract: The present invention provides a composition for controlling harmful arthropods having an excellent control efficacy on harmful arthropods. A composition for controlling harmful arthropods comprising an amide represented by a formula (I); wherein each of symbols are the same as defined in the Description; or salts thereof and at least one kind of pyrazole compounds selected from the group (A) consisting of chlorantraniliprole, cyantraniliprole and a compound represented by a formula (II), shows an excellent controlling efficacy on harmful arthropods.
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DESCRIPTION

COMPOSITION AND METHOD FOR CONTROLLING HARMFUL ARTHROPODS

TECHNICAL FIELD

[0001]
The present invention relates to a composition for controlling harmful arthropods and a method for controlling harmful arthropods.

BACKGROUND ART

[0002]

DISCLOSURE of INVENTION

(PROBLEMS TO BE SOLVED BY INVENTION)

[0003]
An object of the present invention is to provide a composition for controlling harmful arthropods having an excellent control efficacy on harmful arthropods.

(MEANS TO SOLVE PROBLEMS)

[0004]
The present inventors have intensively studied to find out a composition for controlling harmful arthropods having an excellent control efficacy on harmful arthropods. As a result, they have found that a composition comprising an amide compound represented by the following formula (I) or salts thereof and
at least one kind of compounds selected from the group consisting of the following group (A) has an excellent controlling effect on harmful arthropods. Thus, the present invention has been completed.

[0005]

Specifically, the present invention includes:

1. A composition for controlling harmful arthropods comprising an amide represented by a formula (I)

\[
\begin{align*}
&\text{R}^2-\text{CH}_2\text{N-C-} \quad (\text{CH}_2\text{H}^n \quad \text{C} \quad \text{R}^1) \\
&\text{O} \quad \text{O}
\end{align*}
\]

wherein

- \( n \) is 3 or 4;
- \( \text{R}^1 \) represents a hydroxyl group, an amino group or a C1-C6 alkoxy group;
- \( \text{R}^2 \) represents an optionally substituted phenyl group, an optionally substituted 1-naphthyl group or an optionally substituted 3-indolyl group, and the phenyl group, the 1-naphthyl group or the 3-indolyl group being represented by the \( \text{R}^2 \) may be substituted on the carbon atoms independently of each other with one or more substituents selected from a halogen atom, a hydroxy group, a nitro group, a C1-C6 alkyl group or a C1-C6 alkoxy group;
- or salts thereof and
- at least one kind of pyrazole compounds selected from the group (A) consisting of chlorantraniliprole, cyantraniliprole and a compound represented by a formula (II).
The composition for controlling harmful arthropods according to [1] wherein a weight ratio of the amide compound or salts thereof to the pyrazole compounds selected from the group (A) is in the range of 100:1 to 1:100.

A method for controlling harmful arthropods which comprises applying an effective amount of the composition for controlling harmful arthropods according to [1] or [2] to harmful arthropods or a place where the harmful arthropods live.

A method for controlling harmful arthropods which comprises applying an effective amount of the composition for controlling harmful arthropods according to [1] or [2] to plant seeds.

The method for controlling harmful arthropods according to [4] wherein the plant seeds are seeds of com, cotton, soybean, beet, rapeseed or rice.

The present invention can control harmful arthropods.

Hereinafter, the present invention is explained in detail. The term "composition for controlling harmful arthropods..."
of the present invention' refers to a composition comprising an amide compound represented by a formula (I):

\[ R^2-\text{CH}_2\text{CH}_2-\overset{\text{H}}{\text{N}}-\text{(CH}_2)_n-\overset{\text{O}}{\text{C}}-R^1 \]  

(I)

wherein

- \( n \) is 3 or 4;
- \( R^1 \) represents a hydroxyl group, an amino group or a C1-C6 alkoxy group;
- \( R^2 \) represents an optionally substituted phenyl group, an optionally substituted 1-naphthyl group or an optionally substituted 3-indolyl group, and the phenyl group, the 1-naphthyl group or the 3-indolyl group being represented by the \( R^2 \) may be substituted on the carbon atoms independently of each other with one or more substituents selected from a halogen atom, a hydroxy group, a nitro group, a C1-C6 alkyl group or a C1-C6 alkoxy group (hereinafter referred as to 'the present amide compound');
- or salts thereof and
- at least one kind of pyrazole compounds selected from the group (A) consisting of chlorantraniliprole, cyantraniliprole and a compound represented by a formula (II) (hereinafter referred as to 'the present pyrazole compounds').

\[ \text{(II)} \]

[0008]

In the formula (I), as the group represented by the \( R^1 \),
the term 'C1-C6 alkoxy group' includes, for example, a methoxy group, an ethoxy group, a propoxy group, a butoxy group, a pentyloxy group, a hexyloxy group, a 1-methylethoxy group, a 2-methylpropoxy group, 3-methylbutoxy group and 4-methylpentyloxy group.

[0009]

In the formula (I), when the phenyl group, the 1-naphthyl group or the 3-indolyl group being represented by the R2 may be substituted on the carbon atoms independently of each other with one or more substituents (preferably one or two substituents and more preferably one substituent), as the substituent,

the term 'halogen atom' includes, for example, a fluorine atom, a chlorine atom, a bromine atom and an iodine atom;

the term 'C1-C6 alkyl group' includes, for example, a methyl group, an ethyl group, a propyl group, a butyl group, a pentyl group, a hexyl group, a 1-methylethyl group, a 2-methylpropyl group, a 3-methylbutyl group and a 4-methylpentyl group;

the term 'C1-C6 alkoxy group' includes, for example, a methoxy group, an ethoxy group, a propoxy group, a butoxy group, a pentyloxy group, a hexyloxy group, a 1-methylethoxy, a 2-methylpropoxy group, a 3-methylbutoxy group and a 4-methylpentyloxy group.

When in the formula (I), the phenyl group, the 1-naphthyl group or the 3-indolyl group being represented by the R2 may be substituted on the carbon atoms simultaneously with each other with two or more substituents selected from the halogen atom, the hydroxyl group, the nitro group, the C1-C6 alkyl group or the C1-C6 alkoxy group, the substituent on each of the carbon
Atoms may be the same or different to each other.

Examples of the present amide compound includes
the amide compound represented by the formula (I) wherein
n is 3, R₁ represents a hydroxyl group, an amino group or a Cl-C6 alkoxy group, and R₂ is an 3-indolyl group;
the amide compound represented by the formula (I) wherein
n is 3 or 4, and R₁ represents a hydroxyl group and R₂ is an 3-indolyl group,
the amide compound represented by the formula (I) wherein
n is 3, R₁ represents a hydroxyl group or a Cl-C2 alkoxy group and R₂ is a phenyl group, a 1-naphthyl group, an 3-indolyl group or a 5-methyl-3-indolyl group, and
the amide compound represented by the formula (I) wherein
n is 4, R₁ represents a hydroxyl group or a Cl-C2 alkoxy group and R₂ is a phenyl group.

The salts of the present amide compound include, for example, inorganic base salts and organic base salts.

The inorganic base salts include, for example, alkali metal salts such as sodium salts and potassium salts, alkaline-earth metal salts such as calcium salts and magnesium salts, and ammonium salts.

The organic base salts include, for example, amine salts such as triethylamine salts, pyridine salts, picoline salts, ethanolamine salts, triethanolamine salts, dicyclohexylamine salts, and N,N'-dibenzylethlenediamine salts.

Next, specific examples of the present amide compound are shown below.

The amide compound represented by the formula (I-a):
\[ R^2-\text{CH}_2\text{CH}_2-N-C\text{(CH}_2\text{)}_r-C-\text{R}^1 \] (I-a)

wherein a combination of \( R^1 \) and \( R^2 \) represents any combination as shown in Table 1.

<table>
<thead>
<tr>
<th>Compound No.</th>
<th>( n )</th>
<th>( R^1 )</th>
<th>( R^2 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>3</td>
<td>OH</td>
<td>phenyl</td>
</tr>
<tr>
<td>2</td>
<td>3</td>
<td>OCH(_3)</td>
<td>phenyl</td>
</tr>
<tr>
<td>3</td>
<td>3</td>
<td>OH</td>
<td>3-indolyl</td>
</tr>
<tr>
<td>4</td>
<td>3</td>
<td>OCH(_3)</td>
<td>3-indolyl</td>
</tr>
<tr>
<td>5</td>
<td>3</td>
<td>OCH(_2\text{CH}_3)</td>
<td>3-indolyl</td>
</tr>
<tr>
<td>6</td>
<td>3</td>
<td>OCH(_2\text{CH}_2\text{CH}_3)</td>
<td>3-indolyl</td>
</tr>
<tr>
<td>7</td>
<td>3</td>
<td>OH</td>
<td>5-methyl-3-indolyl</td>
</tr>
<tr>
<td>8</td>
<td>3</td>
<td>OH</td>
<td>1-naphthyl</td>
</tr>
<tr>
<td>9</td>
<td>3</td>
<td>OCH(_3)</td>
<td>1-naphthyl</td>
</tr>
<tr>
<td>10</td>
<td>3</td>
<td>OCH(_2\text{CH}_3)</td>
<td>1-naphthyl</td>
</tr>
<tr>
<td>11</td>
<td>4</td>
<td>OH</td>
<td>phenyl</td>
</tr>
<tr>
<td>12</td>
<td>4</td>
<td>OCH(_3)</td>
<td>phenyl</td>
</tr>
</tbody>
</table>

The present amide compounds are those described in, for example, JP-11-255607 A and JP-2001-139405 A, and can be prepared, for example, according to the methods described therein.

Also, chlorantraniliprole and cyantraniliprole that used in the present invention are all known compounds, and are described in, for example, "The PESTICIDE MANUAL - 15th EDITION (BCFC published) ISBN 978-1-901396-18-8", pages 175 and 251 respectively. These compounds are either commercially available, or can be prepared by known methods.

The compound represented by the formula (II) is described in JP-2004-238307 A, and can be prepared, for example, according to the methods described therein.
The weight ratio of the present amide compound or salts thereof to the present pyrazole compounds in the composition for controlling harmful arthropods of the present invention includes, but is not limited to, in the range of usually 2 to 10,000,000 parts by weight, preferably 10 to 100,000 parts by weight, more preferably 100 to 10,000 parts by weight, and further preferably 500 to 10,000 parts by weight of the present pyrazole compounds opposed to 1,000 parts by weight of the present amide compound or salts thereof.

Although the composition for controlling harmful arthropods of the present invention may be a mixture as itself of the present amide compound or salts thereof and the present pyrazole compounds, the composition of the present invention is usually prepared by mixing the present amide compound or salts thereof, the present pyrazole compounds and an inert carrier, and if necessary, adding a surfactant or other pharmaceutical additives, and then formulating into the form of oil solution, emulsifiable concentrate, flowable formulation, wettable powder, granulated wettable powder, dust formulation, granules and so on.

Also the composition for controlling harmful arthropods formulated as aforementioned can be used by itself or with an addition of other inert components as agent for controlling harmful arthropods.

In the composition for controlling harmful arthropods of the present invention, a total amount of the present amide compound or salts thereof and the present pyrazole compounds is in the range of usually 0.1% to 99% by weight, preferably 0.2% to 90% by weight, and more preferably 1% to 80% by weight.
Also the composition for controlling harmful arthropods of the present invention may further optionally contain one or more pesticides and/or fungicides other than those mentioned above.

Examples of the inert carrier used in the formulation include an inert solid carrier and an inert liquid carrier.

Examples of a solid carrier used in the formulation include finely-divided power or particles of clay consisting of minerals (for example, kaolin clay, attapulgite clay, bentonite, montmorillonite, acid clay, pyrophyllite, talc, diatomaceous earth, or calcite), natural organic substances (for example, corn cob powder, or walnut shell powder), synthetic organic substances (for example, urea), salts (for example, calcium carbonate, or ammonium sulfate), synthetic inorganic substances (for example, synthetic hydrous silicon oxide) and the others. Examples of a liquid carrier include aromatic hydrocarbons (for example, xylene, alkyl benzene, or methylnaphtalene), alcohols (for example, 2-propanol, ethylene glycol, propylene glycol, or ethylene glycol monoethyl ether), ketones (for example, acetone, cyclohexanone, or isophorone), vegetable oils (for example, soybean oil, or cotton oils), petroleum-derived aliphatic hydrocarbons, esters, dimethylsulf oxide, acetonitrile and water.

Examples of the surfactant include anionic surfactant (for example, alkyl sulfate salts, alkylaryl sulfate salts, dialkyl sulfosuccinate salts, polyoxyethylene alkylaryl ether phosphates, lignin sulfonate, or naphthalenesulfonate formaldehyde polycondensation), nonionic surfactant (for example, polyoxyethylene alkylaryl ether, polyoxyethylene alkyl polyoxypropylene block copolymer, or sorbitan fatty acid ester)
and cationic surfactant (for example, alkyltrimethyl ammonium salts).

Examples of the other pharmaceutical additives include water-soluble polymer (for example, polyvinyl alcohol, or polyvinyl pyrrolidone), polysaccharides (for example, arabic gum, alginic acid and salts thereof, CMC (carboxymethyl-cellulose), or xanthan gum), inorganic substances (for example, aluminum magnesium silicate, or alumina-sol), antiseptic agent, coloring agent, and PAP (isopropyl acid phosphate), and stabilizing agent (for example, BHT).

The composition for controlling harmful arthropods of the present invention can be used for protecting plants from damage due to eating or sucking or the like by harmful arthropods.

The harmful arthropods on which the composition for controlling harmful arthropods of the present invention has a controlling efficacy is exemplified below:

**Hemiptera:**

Delphacidae (for example, *Laodelphax striatellus*, *Nilaparvata lugens*, and *Sogatella furcifera*), Deltocephalidae (for example, *Nephotettix cincticeps*, and *Nephotettix virescens*), Aphididae (for example, *Aphis gossypii*, *Myzus persicae*, *Brevicoryne brassicae*, *Macrosiphum euphorbiae*, *Aulacorthum solani*, *Rhopalosiphum padi*, and *Toxoptera citricidus*), Pentatomidae (for example, *Nezara antennata*, *Riptortus clavetus*, *Leptocorisa chinensis*, *Eysarcoris parvus*, *Halyomorpha mista*, and *Lygus lineolaris*), Aleyrodidae (for example, *Trialeurodes vaporariorum*, *Bemisia tabaci*, and *Bemisia argentifolii*), and the others;
Lepidoptera:

Pyralidae (for example, Chilo suppressalis, Tryporyza incertulas, Cnaphalocrocis medinalis, Notarcha derogata, Plodia interpunctella, Ostrinia furnacalis, Ostrinia nubilaris, Hellula undalis, and Pediasia teterrellus),

Noctuidae (for example, Spodoptera litura, Spodoptera exigua, Pseudaletia separata, Mamestra brassicae, Agrotis ipsilon, Plusia nigrisigna, Trichoplusia spp., Heliothis spp., and Helicoverpa spp.),

Pieridae (for example, Pieris rapae),

Tortricidae (for example, Adoxophyes spp., Grapholita molesta, Leguminivora glycinivorella, Matsumuraeses azukivora, Adoxophyes orana fasciata, Adoxophyes sp., Homona magnanima, Archips fuscocupreanus, and Cydia pomonella),

Gracillariidae (for example, Caloptilia theivora, and Phyllonorycter ringoneella),

Carposinidae (for example, Carposina niponensis),

Lyonetiidae (for example, Lyonetia spp.),

Lymantriidae (for example, Lymantria spp., and Euproctis spp.),

Yponomeutidae (for example, Plutella xylostella),

Gelechiidae (for example, Pectinophora gossypiella, and Phthorimaea operculella),

Arctiidae (for example, Hyphantria cunea),

Tineidae (for example, Tinea translucens), and the others;

Thysanoptera:

Thripidae (for example, Frankliniella occidentalis, Thrips parmi, Scirtothrips dorsalis, Thrips tabaci, Frankliniella intonsa, and Frankliniella fusca), and the others;

Diptera:

Agromyzidae (for example, Hylemya antiqua, Hylemya platura, Agromyza oryzae, Hydrellia griseola, Chlorops oryzae, and
Liriomyza trifolii),
Dacus cucurbitae, Ceratitis capitata, and the others;
Coleoptera:
Epilachna vigintioctopunctata, Aulacophora femoralis,
Phyllostreta striolata, Oulema oryzae, Echinocnemus squameus,
Lissorhoptrus oryzophilus, Anthonomus grandis, Callosobruchus chinensis, Sphenophorus venatus, Popillia japonica, Anomala cuprea, Diabrotica spp., Leptinotarsa decemlineata, Agriotes spp., Lasioderma serricorne and the others;
Orthoptera:
Gryllotalpa africana, Oxya yezoensis, Oxya japonica and the others.

[0021]
The composition for controlling harmful arthropods of the present invention can be used in agricultural lands such as fields, paddy fields, dry paddy fields, lawns and orchards or in non-agricultural lands. Also the composition for controlling harmful arthropods of the present invention can control harmful arthropods that live in agricultural lands in the agricultural lands and the others for cultivating the following "plant" and the others.

[0022]
The plant which can be applied by the composition for controlling for harmful arthropods of the present invention is exemplified below:

Crops:
corn, rice, wheat, barley, rye, oat, sorghum, cotton, soybean,
peanut, buckwheat, beet, rapeseed, sunflower, sugar cane,
tobacco, and the others;
Vegetables:
solanaceous vegetables (for example, eggplant, tomato, pimento,
pepper and potato),
cucurbitaceous vegetables (for example, cucumber, pumpkin,
zucchini, watermelon and melon),
cruciferous vegetables (for example, *Japanese radish*, white
turnip, horseradish, kohlrabi, *Chinese cabbage*, cabbage, leaf
mustard, broccoli, cauliflower, colza),
asteraceous vegetables (for example, burdock, crown daisy,
artichoke and lettuce),
liliaceous vegetables (for example, green onion, onion, garlic
and asparagus),
ammiaceous vegetables (for example, carrot, parsley, celery and
parsnip),
chenopodiaceous vegetables (for example, spinach and *Swiss
chard*),
lamiaceous vegetables (for example, *Perilla frutescens*, mint
and basil),
strawberry, sweet potato, *Dioscorea japonica*, colocasia and the
others;

Fruits:
pomaceous fruits (for example, apple, pear, *Japanese pear,
Chinese quince* and quince),
stone fleshy fruits (for example, peach, plum, nectarine,
*Primus mume*, cherry fruit, apricot and prune),
citrus fruits (for example, *Citrus unshiu*, orange, lemon, lime
and grapefruit),
nuts (for example, chestnut, walnuts, hazelnuts, almond,
pistachio, cashew nuts and macadamia nuts),
berry fruits (for example, blueberry, cranberry, blackberry and
raspberry)
grape, kaki persimmon, olive, *Japanese plum*, banana, coffee,
date palm, coconuts, oil palm and the others;
Trees other than fruit trees:

tea, mulberry,
flowering plant (for example, dwarf azalea, camellia, hydrangea, sasanqua, *Illicium anisatum*, cherry trees, tulip tree, crape myrtle and fragrant olive),
*Sweet viburnum*, *Podocarpus macrophyllus*, Japanese cedar, Japanese cypress, croton, Japanese spindletree and *Photinia glabra*

Lawn:
sods (for example, *Zoysia japonica*, *Zoysia matrella*),
bermudagrasses (for example, *Cynodon dactylon*),
bent glasses (for example, *Agrostis gigantea*, *Agrostis stolonifera*, *Agrostis capillaris*),
blueglasses (for example, *Poa pratensis*, *Poa trivialis*),
festucae (for example, *Festuca arundinacea Schreb.*, *Festuca rubra* L. var. *commutata Gaud.*, *Festuca rubra* L. var. *genuina Hack*),
ryegrassses (for example, *Lolium multiflorum Lam*, *Lolium perenne* L),
*Dactylis glomerata*, *Phleum pratense*);

Others:
flowers (for example, rose, carnation, chrysanthemum, *Eustoma*, gypsophila, gerbera, marigold, salvia, petunia, verbena, tulip, aster, gentian, lily, pansy, cyclamen, orchid, lily of the valley, lavender, stock, ornamental cabbage, primula, poinsettia, gladiolus, cattleya, daisy, cymbidium and begonia),
bio-fuel plants (for example, jatropha, safflower, Camelina, switch grass, Miscanthus giganteus, Phalaris arundinacea, Arundo donax, kenaf, cassava, willow), and ornamental foliage plants, and the others.

Among the above-mentioned plants, preferred examples include corn, cotton, soybean, beet, rapeseed and rice.

The above-mentioned 'plant' includes plants, to which a resistance has been conferred by a classical breeding method or genetic engineering technique.

The composition for controlling harmful arthropods of the present invention is used to control harmful arthropods by applying it to the plant or an area for cultivating the plant. Such plants to be used herein include foliages of plant, flowers of plant, fruits of plant, seeds of plant, or bulbs of plant. The bulbs to be used herein are intended to mean bulb, corm, rootstock, tubera, tuberous root and rhizophore.

The method for controlling harmful arthropods of the present invention comprises applying the composition for controlling harmful arthropods of the present invention.

Examples of the method of applying the composition for controlling harmful arthropods of the present invention include, an application to stems and leaves of plants such as a foliage application; an application to seeds of plants, and an application to area for cultivating plants such as a soil treatment and a submerged application.
Specific examples of the application to stems and leaves of plants such as a foliage application in the present invention include an application to surfaces of plants to be cultivated, for example, by a ground application with a manual sprayer, a power sprayer, a boom sprayer or Pancle sprayer or by an aerial application by using manned or unmanned airplane or helicopter.

Specific examples of the application to seeds of plants in the present invention include an application of the composition for controlling harmful arthropods of the present invention to seeds or bulbs of plants, more specifically, a spray coating treatment on the surface of seeds or bulbs, a smear treatment on the seeds or bulbs of plants, an immersion treatment, a film coating treatment and a pellet coating treatment.

Specific examples of the application to area for cultivating plants such as a soil application and submerged application in the present invention include, a planting hole application, a plant foot application, a row application, an in-furrow application, an overall application, a side ditch application, a nursery box application, a nursery bed application, a nursery soil incorporation, a bed soil incorporation, a paste fertilizer incorporation, a paddy water application, and a submerged application under flooding condition.

When the composition for controlling harmful arthropods of the present invention is applied to plants or area for cultivating plants, the application dose varies depending on
the kinds of plants to be protected, the species or the degree of emergence of harmful arthropods to be controlled, the dosage form, the timing of application, weather conditions, etc., but the total amount of the present amide compound or salt thereof and the pyrazole compounds is in the range of usually from 0.05 to 10,000 g, preferably from 0.5 to 1,000 g per 1,000 m² of the area for cultivating plants.

[0032]

When the composition for controlling harmful arthropods of the present invention is applied to seeds of plants, the application dose varies depending on the kinds of plants to be protected, the species or the degree of emergence of harmful arthropods to be controlled, the dosage form, the timing of application, weather conditions, etc., but the total amount of the present amide compound or salts thereof and the pyrazole compounds is in the range of usually from 0.001 to 100 g, preferably from 0.05 to 50 g per 1 kg of the seeds.

[0033]

The emulsifiable concentrate, the wettable powder or the flowable formulation, etc. of the composition for controlling harmful arthropods of the present invention is usually applied by diluting it with water, and then spreading it. In this case, the total concentration of the present amide compound or salts thereof and the pyrazole compounds is in the range of usually 0.00001 to 10% by weight, and preferably 0.0001 to 5% by weight. The dust formulation or the granular formulation, etc, is usually applied as itself without diluting it.

EXAMPLES

[0034]
The following Examples including Formulation examples and Test examples serve to illustrate the present invention in more detail, which should not intend to limit the present invention. In the Examples, the term "part(s)" means part(s) by weight unless otherwise specified, and "the present amide compound (Compound No. X)" corresponds to "Compound No. X" listed in Table 1, that is, for example, "the present amide compound (Compound No. 4)" refers to Compound No. 4 listed in Table 1.

Formulation examples are shown below.

Formulation example 1

Ten (10) parts of the present amide compound selected from Compound No. 1 to Compound No. 12, 5 parts of chlorantraniliprole, 35 parts of a mixture (weight ratio 1:1) of white carbon and ammonium polyoxyethylene alkyl ether sulfate are mixed with an appropriate amount of water so as to give a total amount of 100 parts, and then the mixture is finely-ground by a wet grinding method to obtain a flowable formulation.

Formulation example 2

Ten (10) parts of the present amide compound selected from Compound No. 1 to Compound No. 12, 5 parts of cyantraniliprole, 35 parts of a mixture (weight ratio 1:1) of white carbon and ammonium polyoxyethylene alkyl ether sulfate are mixed with an appropriate amount of water so as to give a total amount of 100 parts, and then the mixture is finely-ground by a wet grinding method to obtain a flowable formulation.

Formulation example 3
Ten (10) parts of the present amide compound selected from Compound No. 1 to Compound No. 12, 5 parts of a compound represented by a formula (II), 35 parts of a mixture (weight ratio 1:1) of white carbon and ammonium polyoxyethylene alkyl ether sulfate are mixed with an appropriate amount of water so as to give a total amount of 100 parts, and then the mixture is finely-ground by a wet grinding method to obtain a flowable formulation.

[0039]

Formulation example 4

Ten (10) parts of the present amide compound selected from Compound No. 1 to Compound No. 12, 10 parts of chlorantraniliprole, 1.5 parts of sorbitan triooleate, and 28 parts of an aqueous solution containing 2 parts of polyvinyl alcohol are mixed, and then the mixture is finely-ground by a wet grinding method. To this mixture is added an appropriate amount of aqueous solution containing 0.05 parts of xanthane gum and 0.1 parts of magnesium aluminium silicate so as to give a total amount of 90 parts, and then 10 parts of propylene glycol is added thereto. The mixture is stirred to obtain a flowable formulation.

[0040]

Formulation example 5

Ten (10) parts of the present amide compound selected from Compound No. 1 to Compound No. 12, 10 parts of cyantraniliprole, 1.5 parts of sorbitan triooleate, and 28 parts of aqueous solution containing 2 parts of polyvinyl alcohol are mixed, and then the mixture is finely-ground by a wet grinding method. To this mixture is added an an appropriate amount of aqueous solution containing 0.05 parts of xanthane gum and 0.1 parts of magnesium aluminium silicate so as to give a total amount of 90
parts, and then 10 parts of propylene glycol is added thereto. The mixture is stirred to obtain a flowable formulation.

Formulation example 6

Ten (10) parts of the present amide compound selected from Compound No. 1 to Compound No. 12, 10 parts of a compound represented by a formula (II), 1.5 parts of sorbitan trioleate, and 28 parts of an aqueous solution containing 2 parts of polyvinyl alcohol are mixed, and then the mixture is finely-ground by a wet grinding method. To this mixture is added an appropriate amount of aqueous solution containing 0.05 parts of xanthane gum and 0.1 parts of magnesium aluminium silicate so as to give a total amount of 90 parts, and then 10 parts of propylene glycol is added thereto. The mixture is stirred to obtain a flowable formulation.

Formulation example 7

Ten (10) parts of the present amide compound selected from Compound No. 1 to Compound No. 12, 20 parts of chlorantraniliprole, 3 parts of calcium lignin sulfonate, 2 parts of sodium lauryl sulfate, and the rest parts of synthetic hydrated silicon oxide are well mixed while grinding to obtain 100 parts of a wettable powder.

Formulation example 8

Ten (10) parts of the present amide compound selected from Compound No. 1 to Compound No. 12, 20 parts of cyantraniliprole, 3 parts of calcium lignin sulfonate, 2 parts of sodium lauryl sulfate, and the rest parts of synthetic hydrated silicon oxide are well mixed while grinding to obtain 100 parts of a wettable powder.
Formulation example 9

Ten (10) parts of the present amide compound selected from Compound No. 1 to Compound No. 12, 20 parts of a compound represented by a formula (II), 3 parts of calcium lignin sulfonate, 2 parts of sodium lauryl sulfate, and the rest parts of synthetic hydrated silicon oxide are well mixed while grinding to obtain 100 parts of a wettable powder.

Treatment example 1

The flowable formulation prepared in Formulation example 1 is used for a smear treatment in an amount of 500 ml per 100 kg of dried sorghum seeds by using a rotary seed treatment machine (seed dresser, produced by Hans-Ulrich Hege GmbH) to obtain the treated seeds.

The seeds treated with each of the flowable formulations prepared in Formulation examples 2 to 6 are obtained in a manner similar to the above, by using the flowable formulations prepared in Formulation examples 2 to 8 instead of the flowable formulation prepared in Formulation example 1.

Treatment example 2

The flowable formulation prepared in Formulation example 1 is used for a smear treatment in an amount of 40 ml per 10 kg of dried corn seeds by using a rotary seed treatment machine (seed dresser, produced by Hans-Ulrich Hege GmbH) to obtain the treated seeds.

The seeds treated with each of the flowable formulations prepared in Formulation examples 2 to 8 are obtained in a manner similar to the above, by using the flowable formulations prepared in Formulation examples 2 to 6 instead of the flowable
formulation prepared in Formulation example 1.

[0047]

Treatment example 3

The wettable powder prepared in Formulation example 7 is used for powder coating treatment in an amount of 50 g per 10 kg of dried corn seeds to obtain the treated seeds.

The seeds treated with each of the wettable powders prepared in Formulation examples 10 to 15 are obtained in a manner similar to the above, by using the wettable powders prepared in Formulation examples 8 or 9 instead of the wettable powder prepared in Formulation example 7.

[0048]

Treatment example 4

The flowable formulation prepared in Formulation example 1 is used for a smear treatment in an amount of 50 ml per 10 kg of dried soybean seeds by using a rotary seed treatment machine (seed dresser, produced by Hans-Ulrich Hege GmbH) to obtain the treated seeds.

The seeds treated with each of the flowable formulations prepared in Formulation examples 2 to 6 are obtained in a manner similar to the above, by using the flowable formulations prepared in Formulation examples 2 to 8 instead of the flowable formulation prepared in Formulation example 1.

[0049]

Next, the effect of the present invention is shown in test examples.

[0050]

Test example 1

The present amide compound 1.25mg and the present pyrazole compound 12.5mg were mixed and thereto were added 150 microliters of slurry that was prepared by mixing 10 parts of
Color Coat Red (manufactured by Becker Underwood Inc.: coloring agent), 10 parts of CF-CLEAR (manufactured by Becker Underwood Inc.: spreading agent) and an appropriate amount of water so as to give a total amount of 100 parts and the resulting mixtures were then mixed thoroughly, and the total amount of the mixture was added to 25 grains of corn seeds and the resulting mixtures were then agitated to give treated corn seeds. After air drying, the treated corn seeds were seeded into a 160 ml plastic cup in a ratio of 2 grains per the cup.

At 14 days post the seeding, Spodoptera litura at the fourth-instar larval stages were released in a ratio of about 5 heads of insects per the cup, and then the cups were covered with a 540 ml plastic cup (hereinafter, referred to as a 'treated area'). At 3 days post the release, the number of surviving larvae in each cup was observed.

On the other hand, the same seeding was carried out using corn seeds without the above-mentioned treatment (hereinafter, referred to as an 'untreated area'), and at 3 days post the release, the number of surviving larvae in each cup was observed.

Each of mortality of insects in the treated area and the untreated area was calculated by the following equation 1):

\[ \text{Mortality of insects (\%)} = \frac{\text{Number of test insects} - \text{Number of surviving insects}}{\text{Number of test insects}} \times 100 \]

The two duplicate tests were performed. The average value is shown in Table 2.
Table 2

<table>
<thead>
<tr>
<th>Test compounds</th>
<th>Dose (mg/ 25grains)</th>
<th>Mortality of Insects (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Present amide compound (Compound No. 3) + chlorantraniliprole</td>
<td>1.25 + 12.5</td>
<td>100</td>
</tr>
<tr>
<td>Present amide compound (Compound No. 3) + cyantraniliprole</td>
<td>1.25 + 12.5</td>
<td>100</td>
</tr>
<tr>
<td>Present amide compound (Compound No. 12) + chlorantraniliprole</td>
<td>1.25 + 12.5</td>
<td>100</td>
</tr>
<tr>
<td>Present amide compound (Compound No. 12) + cyantraniliprole</td>
<td>1.25 + 12.5</td>
<td>100</td>
</tr>
<tr>
<td>untreated area</td>
<td>-</td>
<td>0</td>
</tr>
</tbody>
</table>

[0053]

Test example 2

The present amide compound 2.5mg and the present pyrazole compound 2.5mg were mixed and thereto were added 150 microliters of slurry that was prepared by mixing 10 parts of Color Coat Red (manufactured by Becker Underwood Inc.: coloring agent), 10 parts of CF-CLEAR (manufactured by Becker Underwood Inc.: spreading agent) and an appropriate amount of water so as to give a total amount of 100 parts and the resulting mixtures were then mixed thoroughly, and the total amount of the mixture was added to 25 grains of corn seeds and the resulting mixtures were then agitated to give treated corn seeds. After air drying, the treated corn seeds were seeded into a 160 ml plastic cup in a ratio of 2 grains per the cup.
At 14 days post the seeding, *Spodoptera litura* at the fourth-instar larval stages were released in a ratio of about 5 heads of insects per the cup, and then the cups were covered with a 540 ml plastic cup (hereinafter, referred to as a 'treated area'). At 3 days post the release, the number of surviving larvae in each cup was observed.

Each of mortality of insects in the treated area was calculated by the following equation 1). The two duplicate tests were performed. The average value is shown in Table 3.

![Table 3](attachment:table3.png)
1. A composition for controlling harmful arthropods comprising an amide represented by a formula (I);

\[
\text{R}^2\text{CH}_2\text{CH}_2\text{N}(-\text{CH}_2)_n\text{C}-\text{R}^1
\]

wherein

- \( n \) is 3 or 4;
- \( \text{R}^1 \) represents a hydroxy group, an amino group or a C1-C6 alkoxy group;
- \( \text{R}^2 \) represents an optionally substituted phenyl group, an optionally substituted 1-naphthyl group or an optionally substituted 3-indolyl group, and the phenyl group, the 1-naphthyl group or the 3-indolyl group being represented by the \( \text{R}^2 \) may be substituted on the carbon atoms independently of each other with one or more substituents selected from a halogen atom, a hydroxy group, a nitro group, a C1-C6 alkyl group or a C1-C6 alkoxy group, or salts thereof and
- at least one kind of pyrazole compounds selected from the group (A) consisting of chlorantraniliprole, cyantraniliprole and a compound represented by a formula (II).

\[
\text{II}\]

2. The composition for controlling harmful arthropods
according to claim 1 wherein a weight ratio of the amide compound or salts thereof to the pyrazole compounds selected from the group \((A)\) is in the range of 100:1 to 1:100.

3. A method for controlling harmful arthropods which comprises applying an effective amount of the composition for controlling harmful arthropods according to claim 1 or 2 to harmful arthropods or a place where the harmful arthropods live.

4. A method for controlling harmful arthropods which comprises applying an effective amount of the composition for controlling harmful arthropods according to claim 1 or 2 to plant seeds.

5. The method for controlling harmful arthropods according to claim 4 wherein the plant seeds are seeds of corn, cotton, soybean, beet, rapeseed or rice.
**INTERNATIONAL SEARCH REPORT**

International application No.  
PCT/JP2012/082120

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### A. CLASSIFICATION OF SUBJECT MATTER

Int.Cl. A01N37/30 (2006.01) i, A01N25/00 (2006.01) i, A01N43/56 (2006.01) i, A01P7/04 (2006.01) i

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

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### B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

Int.Cl. A01N37/30, A01N25/00, A01N43/56, A01P7/04

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

- Published examined utility model applications of Japan 1992-1996
- Published unexamined utility model applications of Japan 1971-2013
- Registered utility model specifications of Japan 1996-2013
- Published registered utility model applications of Japan 1994-2013

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

CA/ REGISTRY (ETN)

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### C. DOCUMENTS CONSIDERED TO BE RELEVANT

<table>
<thead>
<tr>
<th>Category</th>
<th>Citation of document, with indication, where appropriate, of the relevant passages</th>
<th>Relevant to claim No.</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>JP 2001-139405 A (SNOW BRAND SEED CO., LTD.) 2001.05.22, (No Family)</td>
<td>1-5</td>
</tr>
</tbody>
</table>

- Special categories of cited documents:
  - "A" document defining the general state of the art which is not considered to be of particular relevance
  - "E" earlier application or patent but published on or after the international filing date
  - "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
  - "O" document referring to an oral disclosure, use, exhibition or other means
  - "P" document published prior to the international filing date but later than the priority date claimed
  - "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
  - "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
  - "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
  - "&" document member of the same patent family

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

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Date of the actual completion of the international search: 08.02.2013  
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