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(54) Title: MICROCAPSULE

(57) Abstract: This invention relates to a microcapsule comprising: (a) an agriculturally active ingredient in which a retention rate is less than 50% after irradiation with xenon light (290 nm cutoff) at an intensity of 0.68 W/m<sup>2</sup> at 340 nm for 8 hours; and (b) a hydrophobic organic solvent; wherein a weight ratio of the agriculturally active ingredient to the hydrophobic organic solvent is from 10:90 to 70:30. By the microcapsule of the present invention, it is possible to inhibit photodegradation of the agriculturally active ingredient.

## DESCRIPTION

## TITLE OF THE INVENTION

MICROCAPSULE

5

## TECHNICAL FIELD

[0001]

The present invention relates to a microcapsule in which photodegradation of an agriculturally active ingredient 10 has been inhibited.

## BACKGROUND ART

[0002]

Various studies have hitherto been made for the purpose 15 of inhibiting photodegradation of agriculturally active ingredients which are unstable against light. There has been known a microcapsule in which an agriculturally active ingredient, and UV protectants such as titanium dioxide and zinc oxide are contained in an organic solvent (see, for 20 example, Patent Document 1).

[Prior Art Document]

[Patent Document]

[0003]

[Patent Document 1]: JP 11-504030 W

25

## DISCLOSURE OF INVENTION

[Problems to be Solved by the Invention]

[0004]

An object of the present invention is to provide a new microcapsule in which photodegradation of an agriculturally active ingredient has been inhibited.

5 [Means for Solving the Problems]

[0005]

The present inventors have intensively been studied so as to find a microcapsule in which photodegradation of an agriculturally active ingredient has been inhibited, and 10 found that, in a microcapsule comprising:

(a) an agriculturally active ingredient in which a retention rate is less than 50% after irradiation with xenon light (290 nm cutoff) at an intensity of  $0.68 \text{ W/m}^2$  at 340 nm for 8 hours (hereinafter sometimes referred to as a component a); and 15 (b) a hydrophobic organic solvent (hereinafter sometimes referred to as a component b); when a weight ratio of the component a to the component b is within a specific range, it is possible to inhibit photodegradation of the component a.

The present invention includes the followings:

20 [1] A microcapsule comprising:

(a) an agriculturally active ingredient in which a retention rate is less than 50% after irradiation with xenon light (290 nm cutoff) at an intensity of  $0.68 \text{ W/m}^2$  at 340 nm for 8 hours; and

25 (b) a hydrophobic organic solvent;

wherein a weight ratio of the agriculturally active ingredient to the hydrophobic organic solvent is from 10:90

to 70:30.

[2] The microcapsule according to [1], which has an average particle diameter of 5 to 60  $\mu\text{m}$ .

[3] The microcapsule according to [1] or [2], wherein the 5 agriculturally active ingredient is an agriculturally active ingredient which is solid at normal temperature.

[4] The microcapsule according to any one of [1] to [3], wherein the hydrophobic organic solvent(s) is/are one or more hydrophobic organic solvent(s) selected from the group 10 consisting of esters, ketones, aromatic hydrocarbons, and paraffins.

[5] A pesticidal composition in the form of an aqueous suspension in which the microcapsule according to any one of [1] to [4] is suspended in an aqueous continuous phase.

15 [Effects of the Invention]

[0006]

According to the present invention, it is possible to inhibit photodegradation of an agriculturally active ingredient.

20

#### MODE FOR CARRYING OUT THE INVENTION

[0007]

The microcapsule of the present invention (hereinafter sometimes referred to as a microcapsule of the present 25 invention) is a microcapsule containing an agriculturally active ingredient which is significantly unstable against light, such as the component a, and is a microcapsule which

can effectively inhibit photodegradation of the agriculturally active ingredient.

[0008]

As used herein, xenon light means a light source in which a gas filled in a tube is xenon. Usually, xenon light means uniquely the same spectrum. Xenon light (290 nm cutoff) means xenon light having a wavelength within a range of 290 nm or more, and xenon light passed through a borosilicate filter corresponds to this.

A method for the calculation of a retention rate of the agriculturally active ingredient defined above is more specifically as follows. First, 250 mg of the agriculturally active ingredient is dissolved in 50 mL of a solvent such as acetone, which can sufficiently dissolve the agriculturally active ingredient and also has high volatility, and 1 mL of the solution is added to a glass petri dish having a diameter of 6 cm using a one-mark pipette, spread entirely over the petri dish, and then air-dried at room temperature. The petri dish is covered with a lid made of quartz glass and then set in a weathering tester (manufactured by Q-Lab Corporation under the trade name of Q-SUN Xenon Accelerated Weathering Tester, Model Xe-3) equipped with a borosilicate filter (manufactured by Q-Lab Corporation under the trade name of Daylight-BB Optical Filter) attached thereto. After irradiation with xenon light under the conditions of an intensity at 340 nm of 0.68 W/m<sup>2</sup> and a temperature of 35°C (35°C as measured by an insulated black panel thermometer)

for 8 hours, the amount of the agriculturally active ingredient remaining on the petri dish is determined by a known determination method such as high-performance liquid chromatography. It is possible to determine a retention rate by calculating as the weight percentage relative to 5 mg of the agriculturally active ingredient used above.

5 [0009]

The above agriculturally active ingredient used in the microcapsule of the present invention may be any of an 10 insecticidally active component, a fungicidally active component, and a herbicidally active component, and is not particularly limited. The agriculturally active ingredient includes fenpyrazamine, allethrin, and prallethrin. The 15 retention rates of fenpyrazamine, allethrin, and prallethrin after irradiation with xenon light (290 nm cutoff) at an intensity of 0.68 W/m<sup>2</sup> at 340 nm for 8 hours according to the method for calculation above-mentioned, are 28.1%, 3.3% and 6.4%, respectively. It is preferred to use an agriculturally 20 active ingredient which is solid at normal temperature (25°C), for example fenpyrazamine.

[0010]

The microcapsule of the present invention usually contains a component a in the total amount of 1 to 80% by weight, and preferably 10 to 70% by weight.

25 [0011]

In the microcapsule of the present invention, a component b means an organic solvent which is liquid at

normal temperature (25°C) and is immiscible in water.

Specifically, an organic solvent having water solubility of 20% by weight or less at 25°C is used. Examples of the hydrophobic organic solvent include hydrophobic organic solvents such as vegetable oils, esters, ketones, aromatic hydrocarbons, and paraffins, for example, the following hydrophobic organic solvents:

[0012]

Vegetable oils: rapeseed oil, soybean oil, linseed oil, 10 corn oil, and olive oil;

[0013]

Esters: diisobutyl adipate, diisodecyl adipate, dialkyl phthalate (didecyl phthalate, etc.), octyl oleate, lauryl oleate, octyldodecyl oleate, and isopropyl myristate;

[0014]

Ketones: methyl isobutyl ketone, heptanone, octanone, nonanone, cyclohexanone and acetophenone; and

[0015]

Aromatic hydrocarbons: toluene, xylene, 20 phenylxylylethane, 1-phenyl-1-ethylphenylethane, methylnaphthalene, dimethylnaphthalene, triisopropylbiphenyl and dimethylisopropylnaphthalene.

In the present invention, commercially available aromatic hydrocarbon solvents can be used as aromatic hydrocarbons. Examples of the commercially available aromatic hydrocarbon solvents include Hisol SAS-296 (mixture of 1-phenyl-1-xylylethane and 1-phenyl-1-ethylphenylethane,

trade name of JX Nippon Oil & Energy Corporation), Hisol SAS-LH (trade name of JX Nippon Oil & Energy Corporation), CACTUS SOLVENT HP-MN (methylnaphthalene 80%, trade name of JX Nippon Oil & Energy Corporation), CACTUS SOLVENT HP-DMN

5 (dimethylnaphthalene 80%, trade name of JX Nippon Oil & Energy Corporation), CACTUS SOLVENT P-180 (mixture of methylnaphthalene and dimethylnaphthalene, trade name of JX Nippon Oil & Energy Corporation), CACTUS SOLVENT P-200 (mixture of methylnaphthalene and dimethylnaphthalene, trade  
10 name of JX Nippon Oil & Energy Corporation), CACTUS SOLVENT P-220 (mixture of methylnaphthalene and dimethylnaphthalene, trade name of JX Nippon Oil & Energy Corporation), CACTUS SOLVENT PAD-1 (dimethylmonoisopropylnaphthalene, trade name of JX Nippon Oil & Energy Corporation), Solvesso 100 (aromatic  
15 hydrocarbon, trade name of ExxonMobil Chemical Ltd.), Solvesso 150 (aromatic hydrocarbon, trade name of ExxonMobil Chemical Ltd.), Solvesso 200 (aromatic hydrocarbon, trade name of ExxonMobil Chemical Ltd.), Solvesso 150ND (aromatic hydrocarbon, trade name of ExxonMobil Chemical Ltd.),  
20 Solvesso 200ND (aromatic hydrocarbon, trade name of ExxonMobil Chemical Ltd.), Ruetasolv BP 4302 (manufactured by RKS GmbH), NIKANOL (trade name of Fudow Company Limited.), SWASOL 100 (toluene, trade name of Maruzen Petrochemical CO, LTD.), and SWASOL 200 (xylene, trade name of Maruzen  
25 Petrochemical CO, LTD.).

[0016]

Paraffins: normal paraffin, isoparaffin, cycloparaffin,

and liquid paraffin

In the present invention, commercially available paraffin solvents can be used as paraffins. Examples of the commercially available paraffin solvents include NORPAR 13 5 (normal paraffin, trade name of ExxonMobil Chemical Ltd.), NORPAR 15 (normal paraffin, trade name of ExxonMobil Chemical Ltd.), ISOPAR E (isoparaffin, trade name of ExxonMobil Chemical Ltd.), ISOPAR G (isoparaffin, trade name of ExxonMobil Chemical Ltd.), ISOPAR L (isoparaffin, trade name 10 of ExxonMobil Chemical Ltd.), ISOPAR H (isoparaffin, trade name of ExxonMobil Chemical Ltd.), ISOPAR M (isoparaffin, trade name of ExxonMobil Chemical Ltd.), MORESCO-WHITE P-40 (liquid paraffin, trade name of MORESCO Corporation), MORESCO-WHITE P-70 (liquid paraffin, trade name of MORESCO Corporation), MORESCO-WHITE P-200 (liquid paraffin, trade 15 name of MORESCO Corporation), EXXSOL D110 (mixed solvent of paraffin and cycloparaffin, trade name of ExxonMobil Chemical Ltd.), EXXSOL D130 (mixed solvent of paraffin and cycloparaffin, trade name of ExxonMobil Chemical Ltd.), and EXXSOL D160 (mixed solvent of paraffin and cycloparaffin, 20 trade name of ExxonMobil Chemical Ltd.).

[0017]

There is no particular limitation on the component b used in the microcapsule of the present invention, and one or 25 more hydrophobic organic solvent(s) selected from the group consisting of esters, ketones, aromatic hydrocarbons, and paraffins is/are preferably used. Of these, aromatic ketones

such as acetophenone are more preferably used.

[0018]

In the microcapsule of the present invention, a weight ratio of the component a to the component b is from 10:90 to 5 70:30, and preferably from 30:70 to 70:30. The microcapsule of the present invention usually contains the component b in the total amount of 20 to 95% by weight, and preferably 30 to 90% by weight, based on the entire amount of the microcapsule of the present invention.

10 [0019]

It is preferred that the microcapsule of the present invention further contains one or more UV protectants.

Examples of the UV protectants include benzophenone-based ultraviolet absorbers such as 2,4-dihydroxybenzophenone, 2-15 hydroxy-4-methoxybenzophenone, and 2-hydroxy-4-n-octyl-benzophenone; benzotriazole-based ultraviolet absorbers such as 2-(2-hydroxy-5-methylphenyl)benzotriazole, 2-[2-hydroxy-3-(3,4,5,6-tetrahydronaphthalimide-methyl)-5-methylphenyl]benzotriazole, 2-(3-tert-butyl-2-hydroxy-5-methylphenyl)-5-chlorobenzotriazole, 2-(2-hydroxy-5-tert-octylphenyl)benzotriazole, and 2-(2-hydroxy-3,5-di-tert-pentylphenyl)benzotriazole; benzoate-based ultraviolet absorbers such as 2,4-di-tert-butylphenyl 3,5-di-tert-butyl-4-hydroxybenzoate; hydroxyphenyltriazine-based ultraviolet 20 absorber such as 2-(2-hydroxy-4-[1-octyloxycarbonyl]ethoxy]phenyl)-4,6-bis(4-phenylphenyl)-1,3,5-triazine; photostabilizers such as non-sulfonated

lignin (for example, manufactured by MeadWestvaco Corporation under the trade name of INDULIN AT) and a hindered amine-based photostabilizer (HALS); and inorganic UV shielding agents such as titanium oxide.

5       There is no particular limitation on the UV protectant used in the microcapsule of the present invention, and it is preferred to use a UV protectant which is miscible in the component b.

[0020]

10       When the microcapsule of the present invention contains the UV protectant (hereinafter sometimes referred to as a component c), the total content is usually from 1 to 50% by weight, and preferably from 3 to 35% by weight, based on the entire amount of the microcapsule of the present invention,.

15       [0021]

      The microcapsule of the present invention is a microcapsule in which liquid droplets containing a component a, a component b, and more preferably a component c are coated with a resin. The component a is suspended or dissolved in the component b and, when the component c is contained, the component c is suspended or dissolved in the component b.

[0022]

      A film in the microcapsule of the present invention is formed from the resin. The resin includes, for example, thermosetting resins such as a polyurethane resin, a polyurea resin, a urea-formalin resin, a melamine-urea resin, and a

phenol-formalin resin.

Of these resins, a polyurethane resin or a polyurea resin is preferably used.

[0023]

5 The polyurethane resin or polyurea resin used preferably as a resin which forms a film of the microcapsule of the present invention is usually a resin obtained by reacting polyisocyanate with polyol or polyamine.

[0024]

10 The polyisocyanate includes, for example, hexamethylene diisocyanate, adduct of hexamethylene diisocyanate and trimethylolpropane, biuret condensate of three molecules of hexamethylene diisocyanate, adduct of tolylene diisocyanate and trimethylolpropane, isocyanurate condensate of tolylene diisocyanate, isocyanurate condensate of hexamethylene diisocyanate, isocyanurate condensate of isophorone diisocyanate, isocyanate prepolymer in which one isocyanate moiety of hexamethylene diisocyanate composes isocyanurate together with two molecules of tolylene diisocyanate, and the 15 other isocyanate moiety composes isocyanurate together with two molecules of the other hexamethylene diisocyanate, 4,4'-methylenebis(cyclohexyl isocyanate), and trimethyl hexamethylene diisocyanate.

20

The amount of the polyisocyanate used in the present invention is usually determined depending on the amount of the film of the obtained microcapsule. The amount of the film of the obtained microcapsule is usually from 0.1 to 40%

by weight, and preferably from 0.2 to 30% by weight, based on the entire amount of the obtained microcapsule. The amount of the polyisocyanate used in the present invention is usually from 25 to 95% by weight, and preferably from 50 to 5 90% by weight, based on the amount of the film of the obtained microcapsule.

[0025]

The polyol includes, for example, ethylene glycol, propylene glycol, butylene glycol, and cyclopropylene glycol.

10 The polyamine includes, for example, ethylenediamine, hexamethylenediamine, diethylenetriamine, and triethylenetetramine.

15 The amount of the polyol or polyamine used in the present invention is usually determined depending on the amount of the film of the obtained microcapsule. The amount of the polyol used in the present invention is usually from 1 to 80% by weight, and preferably from 3 to 60% by weight, based on the amount of the film of the microcapsule. The amount of the polyamine used in the present invention is 20 usually from 1 to 40% by weight, and preferably from 3 to 20% by weight, based on the amount of the film of the microcapsule.

[0026]

25 A method for producing the microcapsule of the present invention (hereinafter sometimes referred to as a present production method) will be described, but the present production method is not limited only to the following method.

[0027]

The present production method includes the following first to third steps.

[0028]

5 The first step of the present production method is the step of suspending or dissolving a component a in a component b to prepare a suspension or a solution of the component a.

The component a is suspended or dissolved in the component b.

10 When the component a is a component a which is solid at normal temperature (25°C) (hereinafter sometimes referred to as a solid component a), it is possible to take the form of a suspension (hereinafter sometimes referred to as a present suspension a) depending on solubility of the solid component a in the component b and a weight ratio of the solid component a to the component b. The present suspension a can be prepared by finely grinding the solid component a in the component b using a wet grinding mill such as a beads mill. It is also possible to prepare by finely grinding the solid component a with or without adding other components using a dry grinding mill such as a jet mill, and adding the thus obtained finely ground powder to the component b.

15

20

25

The solution in which a component a is dissolved in a component b can be prepared by dissolving the component a in the component b

[0029]

When the solid component a is dispersed (that is,

suspended) in the component b in the form of fine particles, an average particle diameter of the fine particles is usually 15  $\mu\text{m}$  or less, preferably 1 to 10  $\mu\text{m}$ , and more preferably 1 to 5  $\mu\text{m}$ .

5 [0030]

When the microcapsule of the present invention contains the component e, the first step includes the step of respectively suspending or dissolving the component a and the component c in the component b to prepare a suspension or a 10 solution of the component a and the component c.

In the suspension or solution of the component a and the component c, the component a and the component c take any one of the following forms (1) to (4).

(1) Both the component a and the component c are suspended in 15 the component b.

(2) The component c is dissolved in the component b, and the component a is suspended in the component b in which the component c has been dissolved.

(3) The component a is dissolved in the component b, and the component c is suspended in the component b in which the component a has been dissolved. 20

(4) Both the component a and the component c are dissolved in the component b.

The solid component a can take the form (1) or (2) 25 depending on solubility of the solid component a in the component b and a weight ratio of the solid component a to the component b. When the component c is a component c which

is solid at normal temperature (25°C) (hereinafter sometimes referred to as a solid component c), the component c can take the form (1) or (3) depending on solubility of the solid component c to the component b and a weight ratio of the 5 solid component c to the component b.

[0031]

Suspensions of (1) to (3) can be prepared by the same operation as in a method for preparing the present suspension a. In the case of preparing the suspension (1), the solid 10 component a and the solid component c may be finely milled at the same time by a wet grinding mill or a dry grinding mill, or the present suspension a and a suspension in which the solid component c has been suspended in the component b may be separately prepared, followed by mixing, in the method for 15 preparing the present suspension a. In the case of preparing the suspension (2), usually, the component c is dissolved in the component b and then the solid component a is added to the solution. In the case of preparing the suspension (3), usually, the component a is dissolved in the component b and 20 then the solid component c is added to the solution.

The solution (4) can be prepared by dissolving the component a and the component c in the component b.

[0032]

When the resin composing the film is a polyurethane 25 resin or a polyurea resin, usually, polyisocyanate is added in advance to the suspension or solution obtained in the first step.

It is preferred that the suspension or solution, which is obtained in the first step in which the component a and optionally added component c are suspended or dissolved in the component b, is quickly used in the subsequent step.

5 [0033]

The second step of the present production method is the step of mixing the suspension or solution obtained in the first step with water to prepare liquid droplets.

[0034]

10 The amount of water used in the second step is usually within a range of 0.8 to 2 times more than that of the suspension or solution obtained in the first step. Deionized water is preferably used as water used in the second step, and a thickener may be added to water.

15 Examples of the thickener include natural polysaccharides such as gum arabic, xanthan gum, rhamsan gum, locust bean gum, guar gum, carrageenan, welan gum, alginic acid, alginate, and gum tragacanth; vinyl-based polymers such as polyvinyl alcohol, polyvinylpyrrolidone, vinyl acetate copolymer, and sodium polyacrylate; synthetic polymers such as polyoxyalkylene; semi-synthetic polymers such as carboxymethyl cellulose; mineral matter powders such as aluminum silicate, magnesium aluminum silicate, smectite, bentonite, hectorite, synthetic hydrated silicic acid, and dry silica; and alumina sol. It is possible to use, as these thickeners, commercially available products as they are.

25 Examples of commercially available products include KELZAN S

(trade name of CP Kelco, Inc.) as xanthan gum, VEEGUM Granules (trade name of Vanderbilt Company, Inc.) as aluminum silicate, and Aerosil 200 (trade name of Evonik Degussa Corporation) as dry silica.

5 [0035]

In the second step, the method for preparing liquid droplets in water includes, for example, a method in which water is added to the suspension or solution obtained in the first step, followed by stirring using a stirrer. The 10 stirrer used in this case includes, for example, a propeller stirrer, a turbine stirrer, and a high-speed shear stirrer. Specific examples of the stirrer include T.K. Homo Mixer, T.K. Homomic Line Flow, T.K. Pipeline Homo Mixer, and T.K. Filmix manufactured by PRIMIX Corporation; CLEARMIX manufactured by 15 M Technique Co., Ltd.; POLYTRON homogenizer and MEGATRON homogenizer manufactured by KINEMATICA AG; and SUPRATON manufactured by Tsukishima Kikai Co., Ltd.

[0036]

In the second step, usually, liquid droplets are 20 prepared in water, and then polyol or polyamine is added. When polyol is added, a microcapsule with a film formed of a polyurethane resin is obtained. When polyamine is added, a microcapsule with a film formed of a polyurea resin is obtained. The polyol or polyamine may be added before 25 preparing liquid droplets in water, and is preferably added after preparation so as to prevent polyisocyanate during preparation of liquid droplets from reacting with polyol or

polyamine.

[0037]

The third step is the step of forming a film of a resin around the liquid droplets obtained in the second step. In 5 the liquid droplets existing in water obtained in the second step, polyisocyanate is dissolved in the component b.

Therefore, polyisocyanate contained in the liquid droplets react with polyol or polyamine existing in water at an 10 interface of the liquid droplets. As a result, a film of polyurethane or polyurea is formed around the liquid droplets, and thus a microcapsule is obtained as an aqueous suspension.

[0038]

When the resin composing the film is a polyurethane resin, for example, a water dispersion of the liquid droplets 15 obtained in the second step is heated at 40 to 80°C under stirring, followed by maintaining for 0.5 to 48 hours, whereby, a film of the polyurethane resin is formed around the liquid droplets. When the resin composing the film is a polyurea resin, for example, the pH of a water dispersion of 20 the liquid droplets is adjusted within a range of neutral to weak alkaline, followed by maintaining at 0 to 60°C for about 0.5 to 48 hours, whereby, a film of the polyurea resin is formed around the liquid droplets.

[0039]

25 An average particle diameter of the microcapsule of the present invention is almost the same as that of liquid droplets prepared in the second step. The average particle

diameter of liquid droplets prepared in the second step and that of the microcapsule of the present invention are usually from 5 to 60  $\mu\text{m}$ , preferably from 10 to 50  $\mu\text{m}$ , and more preferably from 10 to 45  $\mu\text{m}$ .

5 [0040]

In the present invention, the average particle diameter means a volume median diameter. The volume median diameter refers to a particle diameter at which a cumulative frequency in a volume equivalent frequency distribution is to be 50%, and the volume median diameter can be determined, for example, by wet measurement using a laser diffraction particle size distribution measuring apparatus. More specifically, liquid droplets or microcapsules are dispersed in water and then the volume median diameter is measured using the apparatus. The 10 laser diffraction particle size distribution measuring apparatus includes, for example, Mastersizer 2000 15 (manufactured by Malvern Instruments Ltd.).

[0041]

By the present production method, the microcapsule of 20 the present invention is obtained as a composition in the form of an aqueous suspension.

The microcapsule of the present invention obtained by the present production method can be used as a powder formulation of a microcapsule from a composition in the form 25 of an aqueous suspension by centrifugal separation, filtration, spray drying or the like.

[0042]

To the composition in the form of an aqueous suspension of the microcapsule of the present invention obtained by the present production method, pesticide auxiliary agents which are used in a conventional pesticidal composition in the form 5 of an aqueous suspension is added, whereby, it is possible to use as a pesticidal composition in the form of an aqueous suspension in which the microcapsule of the present invention is suspended in an aqueous continuous phase (hereinafter sometimes referred to as a composition of the present 10 invention). In this case, the microcapsule of the present invention obtained by the present production method is used, for example, as a pesticidal composition in the form of an aqueous suspension, containing the component a in the amount 15 of 0.5 to 25% by weight based on the entire amount of the composition of the present invention.

[0043]

Examples of the pesticide auxiliary agents which may be contained in the composition of the present invention include surfactants, thickeners, defoamers, preservatives, 20 antifreezing agents, pH adjustors and the like.

[0044]

Examples of the surfactants include nonionic surfactants, cationic surfactants, anionic surfactants, and amphoteric surfactants. Examples of nonionic surfactant 25 include polyoxyethylene alkyl ether, polyoxyethylene alkyl aryl ether, polyoxyethylene lanolin alcohol, polyoxyethylene alkyl phenol formalin condensate, polyoxyethylene sorbitan

fatty acid ester, polyoxyethylene glyceryl monofatty acid ester, polyoxypropylene glycol monofatty acid ester, polyoxyethylene sorbitol fatty acid ester, polyoxyethylene castor oil derivative, polyoxyethylene fatty acid ester, 5 higher fatty acid glycerol ester, sorbitan fatty acid ester, sucrose fatty acid ester, polyoxyethylene polyoxypropylene block polymer, polyoxyethylene fatty acid amide, alkylol amide, polyoxyethylenealkylamine, and polyoxyethylene alkanediol. Examples of cationic surfactants include 10 alkylamine hydrochlorides such as dodecylamine hydrochloride; alkyl quaternary ammonium salts such as dodecyltrimethyl ammonium salt, alkyldimethylbenzyl ammonium salt, alkylpyridinium salt, alkylisoquinolinium salt, and dialkylmorpholinium salt; benzethonium chloride, and 15 polyalkyl vinyl pyridinium salt. Examples of anionic surfactants include fatty acid sodium such as sodium palmitate; sodium ether carboxylate such as sodium polyoxyethylene lauryl ether carboxylate; amino acid condensates of higher fatty acid, such as sodium lauroyl sarcosine and sodium N-lauroyl glutamate; higher fatty acid 20 ester sulfonates such as higher alkyl sulfonate and lauric acid ester sulfonic acid salt; dialkyl sulfosuccinates such as dioctyl sulfosuccinate; higher fatty acid amide sulfonates such as oleic acid amide sulfonic acid; alkyl aryl sulfonates such as sodium dodecylbenzene sulfonate and diisopropyl 25 naphthalene sulfonate; higher alcohol sulfuric acid ester salts such as formalin condensate of alkyl aryl sulfonate and

pentadecane-2-sulfate; polyoxyethylene alkyl phosphate esters such as dipolyoxyethylene dodecyl ether phosphate; styrene-maleic acid copolymer; and lignin sulfonate.

Examples of amphoteric surfactants include N-5 laurylalanine, N,N,N-trimethylaminopropionic acid, N,N,N-trihydroxyethylaminopropionic acid, N-hexyl-N,N-dimethylaminoacetic acid, 1-(2-carboxyethyl)pyridinium betaine, and lecithin.

When the composition of the present invention contains 10 the surfactant, the total content is usually from 0.1 to 20% by weight, and preferably from 0.5 to 10% by weight, based on the entire amount of the composition of the present invention.

[0045]

Examples of the thickeners include those exemplified 15 previously.

When the composition of the present invention contains the thickener, the total content is usually from 0.01 to 10% by weight, and preferably from 0.1 to 5% by weight, based on the entire amount of the composition of the present invention.

20 [0046]

Examples of the defoamer include silicone-based defoamers such as ANTIFOAM C EMULSION (trade name of Dow Corning Toray Co., Ltd.), ANTIFOAM CE (trade name of Dow Corning Toray Co., Ltd.), TSA730 (trade name of MOMENTIVE PERFORMANCE MATERIALS JAPAN LLC), TSA731 (trade name of MOMENTIVE PERFORMANCE MATERIALS JAPAN LLC), TSA732 (trade name of MOMENTIVE PERFORMANCE MATERIALS JAPAN LLC), and

YMA6509 (trade name of MOMENTIVE PERFORMANCE MATERIALS JAPAN LLC); and fluorine-based defoamers such as Fluowet PL80 (trade name of Clariant GmbH).

When the composition of the present invention contains 5 the defoamer, the total content is usually from 0.01 to 3% by weight, and preferably from 0.05 to 1% by weight, based on the entire amount of the composition of the present invention.

[0047]

Examples of the preservative include p-hydroxybenzoic 10 acid ester, salicylic acid derivative, proxel (1,2-benzisothiazolin-3-one), and isothiazolin-3-one derivative (for example, BIOHOPE L (trade name of KI Chemical Industry Co., Ltd.)).

When the composition of the present invention contains 15 the preservative, the total content is usually from 0.01 to 5% by weight, and preferably from 0.05 to 3% by weight, based on the entire amount of the composition of the present invention.

[0048]

20 Examples of the antifreezing agent include water-soluble glycols such as ethylene glycol and propylene glycol.

When the composition of the present invention contains the antifreezing agent, the total content is usually from 0.5 to 30% by weight, and preferably from 1 to 20% by weight, 25 based on the entire amount of the composition of the present invention.

[0049]

Examples of the pH adjustor include citric acid monohydrate, sorbic acid, potassium sorbate, disodium hydrogen phosphate, dipotassium hydrogen phosphate, and sodium hydroxide.

5 When the composition of the present invention contains the pH adjustor, the total content is usually from 0.01 to 5% by weight, and preferably from 0.5 to 3% by weight, based on the entire amount of the composition of the present invention.  
[0050]

10 There is no particular limitation on water used in the composition of the present invention, and it is possible to use water used in a conventional pesticidal composition in the form of an aqueous suspension, such as tap water, well water, and deionized water.

15 [0051]

The composition of the present invention usually contains water in the amount of 40 to 95% by weight, and preferably 45 to 90% by weight, based on the entire amount of the composition of the present invention.

20 [0052]

The composition of the present invention can be applied to places such as paddy fields, cultivated lands, orchards, grass plot, and non-agricultural lands in the same manner as in the case of a conventional pesticidal composition in the 25 form of an aqueous suspension. The composition of the present invention is optionally diluted with water and then the obtained water dilution can be applied by a method in

which the composition is applied to plants growing in the above places or the soil in the above places. Examples of the method for applying the water dilution include a soil surface application or foliage application method of the 5 water dilution using a known sprinkler.

It is also possible to use the water dilution in a seed treatment, a seedling raising box treatment and the like.

[0053]

The composition of the present invention can be applied 10 as it is without being diluted with water and, for example, the composition of the present invention is applied along from levee to levee of paddy fields under flooding. Before application, the composition of the present invention is usually mixed by slightly shaking a vessel containing the 15 composition of the present invention.

[Example]

[0054]

The present invention will be described in further detail below by way of Examples, but the present invention is 20 not limited only to these Examples.

[0055]

First, Preparation Examples and Comparative Preparation Examples are shown.

[0056]

25 Preparation Example 1

Fenpyrazamine was dry-milled by a vertical type jet mill (JOM-0101-type jet mill, manufactured by Seishin

Enterprise Co., Ltd.) to prepare fenpyrazamine fine powders having an average particle diameter of 5  $\mu\text{m}$  or less. Ten (10) parts by weight of fenpyrazamine fine powders, 0.2 part by weight of polyisocyanate (manufactured by Sumika Bayer 5 Urethane Co., Ltd. under the trade name of Sumidur L-75), and 19.8 parts by weight of an aromatic hydrocarbon (manufactured by ExxonMobil Chemical Ltd. under the trade name of Solvesso 200ND) were mixed together to obtain a fenpyrazamine suspension. Then, 30 parts by weight of the suspension was 10 added to 33 parts by weight of an aqueous 5% by weight polyvinyl alcohol (manufactured by The Nippon Synthetic Chemical Industry Co., Ltd. under the trade name of GOHSENOL GH-17) solution. This mixture was stirred by a rotor-stator homogenizer (manufactured by KINEMATICA AG under the trade 15 name of POLYTRON homogenizer) thereby emulsifying the fenpyrazamine suspension in the aqueous polyvinyl alcohol solution to obtain a fenpyrazamine emulsion. To the obtained emulsion, 2 parts by weight of an aqueous 1.2% by weight diethylenetriamine (manufactured by Wako Pure Chemical 20 Industries, Ltd.) solution was added, followed by stirring at 60°C for 3 to 4 hours to obtain an aqueous suspension composition of a microcapsule containing fenpyrazamine.

To 14.2 parts by weight of deionized water, 0.4 part by weight of magnesium aluminum silicate (manufactured by 25 Vanderbilt Company, Inc. under the trade name of VEEGUM Granules) was added, followed by stirring at room temperature for 15 minutes. To the solution, 0.2 part by weight of

xanthan gum (manufactured by CP Kelco, Inc. under the trade name of KELZAN S) and 5 parts by weight of propylene glycol were added. This mixture was stirred at 60°C for 60 minutes. The obtained dispersion was cooled to room temperature, and 5 then 0.2 part by weight of an preservative (manufactured by Arch Chemicals, Inc. under the trade name of proxel GXL) was added to the dispersion to give a viscosity modifying liquid. Twenty (20) parts by weight of the viscosity modifying liquid, 15 parts by weight of water, and 65 parts by weight of the 10 above aqueous suspension composition were mixed together to obtain the composition (1) of the present invention, containing 10% by weight of fenpyrazamine. A microcapsule containing fenpyrazamine had an average particle diameter of 40.1 µm.

15 [0057]

#### Preparation Example 2

By performing the same operation as in Preparation Example 1, except that the amount of the fenpyrazamine fine powders prepared in Preparation Example 1 was changed from 10 parts by weight to 5 parts by weight, and the amount of the 20 aromatic hydrocarbon (the same as mentioned above) was changed from 19.8 parts by weight to 24.8 parts by weight, the composition (2) of the present invention, containing 5% by weight of fenpyrazamine was obtained. A microcapsule containing fenpyrazamine had an average particle diameter of 25 44.1 µm.

[0058]

## Preparation Example 3

By performing the same operation as in Preparation Example 1, except that the average particle diameter of the microcapsule containing fenpyrazamine was changed from 40.1 5  $\mu\text{m}$  to 11.1  $\mu\text{m}$  by adjusting a stirring force using the rotor-stator homogenizer (the same as mentioned above), the composition (3) of the present invention, containing 10% by weight of fenpyrazamine was obtained.

[0059]

## 10 Preparation Example 4

By performing the same operation as in Preparation Example 1, except that the average particle diameter of the microcapsule containing fenpyrazamine was changed from 40.1  $\mu\text{m}$  to 4.2  $\mu\text{m}$  by adjusting a stirring force using the rotor-stator homogenizer (the same as mentioned above), the 15 composition (4) of the present invention, containing 10% by weight of fenpyrazamine was obtained.

[0060]

## Preparation Example 5

20 By performing the same operation as in Preparation Example 1, except that diisobutyl adipate (manufactured by Kao Corporation under the trade name of Vinisizer 40) was used in place of the aromatic hydrocarbon (the same as mentioned above), the composition (5) of the present 25 invention, containing 10% by weight of fenpyrazamine was obtained. A microcapsule containing fenpyrazamine had an average particle diameter of 33.2  $\mu\text{m}$ .

[0061]

Preparation Example 6

By performing the same operation as in Preparation Example 1, except that acetophenone (manufactured by Wako Pure Chemical Industries, Ltd.) was used in place of the aromatic hydrocarbon (the same as mentioned above), and the average particle diameter of the microcapsule containing fenpyrazamine was changed from 40.1  $\mu\text{m}$  to 13.3  $\mu\text{m}$  by adjusting a stirring force using the rotor-stator homogenizer (the same as mentioned above), the composition (6) of the present invention, containing 10% by weight of fenpyrazamine was obtained.

[0062]

Preparation Example 7

Ten (10) parts by weight of fenpyrazamine fine powders prepared in Preparation Example 1, 0.2 part by weight of polyisocyanate (the same as mentioned above), 14.8 parts by weight of the aromatic hydrocarbon (the same as mentioned above), and 5 parts by weight of a benzotriazole-based ultraviolet absorber (manufactured by BASF Corporation under the trade name of Tinuvin 571) were uniformly mixed to prepare a fenpyrazamine suspension. Then, 30 parts by weight of the suspension was added to 33 parts by weight of the aqueous 5% by weight polyvinyl alcohol (the same as mentioned above) solution. This mixture was stirred thereby emulsifying the fenpyrazamine suspension in the aqueous polyvinyl alcohol solution to obtain a fenpyrazamine emulsion.

To the obtained emulsion, 2 parts by weight of the aqueous 1.1% by weight diethylenetriamine (the same as mentioned above) solution was added, followed by stirring at 60°C for 3 to 4 hours to obtain an aqueous suspension composition of a 5 microcapsule containing fenpyrazamine. Twenty (20) parts by weight of the viscosity modifying liquid prepared in Preparation Example 1, 15 parts by weight of water and 65 parts by weight of the above aqueous suspensinon composition were mixed together to obtain the composition (7) of the 10 present invention, containing 10% by weight of fenpyrazamine. A microcapsule containing fenpyrazamine had an average particle diameter of 32.0 µm.

[0063]

Preparation Example 8

15        Ten (10) parts by weight of allethrin, 0.2 part by weight of polyisocyanate (manufactured by Sumika Bayer Urethane Co., Ltd. under the trade name of Sumidur L-75), and 19.8 parts by weight of an aromatic hydrocarbon (manufactured by ExxonMobil Chemical Ltd. under the trade name of Solvesso 20 200ND) were mixed together to obtain an allethrin solution. Then, 30 parts by weight of the solution was added to 33 parts by weight of an aqueous 5% by weight polyvinyl alcohol (manufactured by The Nippon Synthetic Chemical Industry Co., Ltd. under the trade name of GOHSENOL GH-17) solution. This 25 mixture was stirred by a rotor-stator homogenizer (manufactured by KINEMATICA AG under the trade name of POLYTRON homogenizer) thereby emulsifying the allethrin

solution in the aqueous polyvinyl alcohol solution to obtain a allethrin emulsion. To the obtained emulsion, 2 parts by weight of an aqueous 1.2% by weight diethylenetriamine (manufactured by Wako Pure Chemical Industries, Ltd.)

5 solution was added, followed by stirring at 60°C for 3 to 4 hours to obtain an aqueous suspension composition of a microcapsule containing allethrin.

Twenty (20) parts by weight of the viscosity modifying liquid prepared in Preparation Example 1, 15 parts by weight of 10 water, and 65 parts by weight of the above aqueous suspension composition were mixed together to obtain the composition (8) of the present invention, containing 10% by weight of allethrin. A microcapsule containing allethrin had an average particle diameter of 12.0  $\mu\text{m}$ .

15 [0064]

#### Preparation Example 9

By performing the same operation as in Preparation Example 8, except that the amount of pinamin prepared in Preparation Example 8 was changed from 10 parts by weight to 20 5 parts by weight, the amount of the aromatic hydrocarbon (the same as mentioned above) was changed from 19.8 parts by weight to 24.8 parts by weight, and the average particle diameter of the microcapsule containing pinamin was changed from 12.0  $\mu\text{m}$  to 39.8  $\mu\text{m}$  by adjusting a stirring force using the rotor-stator homogenizer (the same as mentioned above), the composition (9) of the present invention, containing 5% by weight of allethrin was obtained.

[0065]

Preparation Example 10

By performing the same operation as in Preparation Example 8, except that prallethrin was used in place of 5 allethrin, and the average particle diameter of the microcapsule containing etoc was changed from 12.0  $\mu\text{m}$  to 30.6  $\mu\text{m}$  by adjusting a stirring force using the rotor-stator homogenizer (the same as mentioned above), the composition (10) of the present invention, containing 10% by weight of 10 prallethrin was obtained.

[0066]

Preparation Example 11

By performing the same operation as in Preparation Example 10, except that the average particle diameter of the 15 microcapsule containing prallethrin was changed from 30.6  $\mu\text{m}$  to 12.4  $\mu\text{m}$  by adjusting a stirring force using the rotor-stator homogenizer (the same as mentioned above), the composition (11) of the present invention, containing 10% by weight of prallethrin was obtained.

20 [0067]

Preparation Example 12

By performing the same operation as in Preparation Example 10, except that the average particle diameter of the 25 microcapsule containing prallethrin was changed from 30.6  $\mu\text{m}$  to 5.9  $\mu\text{m}$  by adjusting a stirring force using the rotor-stator homogenizer (the same as mentioned above), the composition (12) of the present invention, containing 10% by

weight of prallethrin was obtained.

Next, Test Example will be shown.

[0068]

Test Example 1

5        The compositions (1) to (12) of the present invention  
were respectively diluted with water so as to make each  
active ingredient concentration to 1,120 ppm. Each of the  
dilutions (100  $\mu$ L) was added to a glass petri dish having a  
diameter of 6 cm, uniformly spread and then air-dried at room  
10      temperature. The petri dish was covered with a lid made of  
quartz glass and placed in a weathering tester (manufactured  
by Q-Lab Corporation under the trade name of Q-SUN Xenon  
Accelerated Weathering Tester, Model Xe-3) equipped with a  
borosilicate filter (manufactured by Q-Lab Corporation under  
15      the trade name of Daylight-BB Optical Filter) attached  
thereto, followed by irradiation with xenon light under the  
conditions of an intensity at 340 nm of 0.68 W/m<sup>2</sup> and a  
temperature of 34°C (34°C as measured by an insulated black  
panel thermometer) for 6 hours. After irradiation,  
20      fenpyrazamine remaining on the petri dish was extracted with  
acetonitrile, followed by quantitative analysis through high-  
performance liquid chromatography or gas chromatography. A  
retention rate was determined by calculating as the weight  
percentage relative to the amount of fenpyrazamine, allethrin  
25      or prallethrin before irradiation.

The results are shown in Table 1.

[0069]

[Table 1]

Composition used	Component a	Component b	Component c	Component a:Component b (weight ratio)	Average particle diameter (μm)	Retention rate (%)
Composition (1) of the present invention	fenpyrazamine	Aromatic hydrocarbon	—	34:66	40.1	31.8
Composition (2) of the present invention	fenpyrazamine	Aromatic hydrocarbon	—	17:83	44.1	2.4
Composition (3) of the present invention	fenpyrazamine	Aromatic hydrocarbon	—	34:66	11.1	13.6
Composition (4) of the present invention	fenpyrazamine	Aromatic hydrocarbon	—	34:66	4.2	4.7
Composition (5) of the present invention	fenpyrazamine	Diisobutyl adipate	—	34:66	33.2	23.0
Composition (6) of the present invention	fenpyrazamine	Acetophenon	—	34:66	13.3	27.5
Composition (7) of the present invention	fenpyrazamine	Aromatic hydrocarbon	Benzotriazole-based ultraviolet absorber	40:60	32.0	71.6
Composition (8) of the present invention	allethrin	Aromatic hydrocarbon	—	34:66	12.0	10.3
Composition (9) of the present invention	allethrin	Aromatic hydrocarbon	—	17:83	39.8	73.0
Composition (10) of the present invention	prallethrin	Aromatic hydrocarbon	—	34:66	30.6	33.1
Composition (11) of the present invention	prallethrin	Aromatic hydrocarbon	—	34:66	12.4	20.2
Composition (12) of the present invention	prallethrin	Aromatic hydrocarbon	—	34:66	5.9	17.1

## CLAIMS

## 1. A microcapsule comprising:

(a) an agriculturally active ingredient in which a retention

5 rate is less than 50% after irradiation with xenon light (290 nm cutoff) at an intensity of 0.68 W/m<sup>2</sup> at 340 nm for 8 hours; and

(b) a hydrophobic organic solvent;

wherein a weight ratio of the agriculturally active

10 ingredient to the hydrophobic organic solvent is from 10:90 to 70:30.

## 2. The microcapsule according to claim 1, which has an

average particle diameter of 5 to 60  $\mu\text{m}$ .

15

3. The microcapsule according to claim 1 or 2, wherein the agriculturally active ingredient is an agriculturally active ingredient which is solid at normal temperature.

20

4. The microcapsule according to any one of claims 1

to 3, wherein the hydrophobic organic solvent(s) is/are one or more hydrophobic organic solvent(s) selected from the group consisting of esters, ketones, aromatic hydrocarbons, and paraffins.

5. A pesticidal composition in the form of an aqueous suspension in which the microcapsule according to any one of claims 1 to 4 is suspended in an aqueous continuous phase.

**INTERNATIONAL SEARCH REPORT**

International application No.  
PCT/JP2013/067587

**A. CLASSIFICATION OF SUBJECT MATTER**

Int.Cl. A01N25/28 (2006.01) i, A01N25/00 (2006.01) i, A01N47/16 (2006.01) i, A01N53/02 (2006.01) i, A01P3/00 (2006.01) i, A01P7/00 (2006.01) i

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

**B. FIELDS SEARCHED**

Minimum documentation searched (classification system followed by classification symbols)

Int.Cl. A01N25/28, A01N25/00, A01N47/16, A01N53/02, A01P3/00, A01P7/00

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 1922-1996  
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)

CAplus/REGISTRY (STN), JSTplus/JMEDplus/JST'580 (JDreamIII), Thomson Innovation

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	JP 62-215505 A (SUMITOMO CHEMICAL COMPANY LIMITED) 1987.09.22, claim 1, page 5 example 13, page 7 test example 1 & DE 3708671 A & FR 2595545 A & GB 2187957 A	1-5
X	JP 2003-040706 A (SUMITOMO CHEMICAL COMPANY LIMITED) 2003.02.13, claim 1-3, paragraph [0026], [0029], [0033], example 1-2, 4-5, 7-8, (no family)	1-5

Further documents are listed in the continuation of Box C.

See patent family annex.

\* 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

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“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

Date of the actual completion of the international search

01.08.2013

Date of mailing of the international search report

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## INTERNATIONAL SEARCH REPORT

International application No.  
PCT/JP2013/067587

## C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	JP 11-504030 A (ZENEKA LIMITED) 1999.04.06, claim 1-3, 8-9, page 14-17 example 1-3, page 8 line 2-8, page 10, line 18-21 & CN 1135160 A & CN 1185090 A & US 5846554 A & US 5993842 A & US 6015571 A & US 6077522 A & US 6149843 A & WO 1995/013698 A1 & WO 1996/033611 A2	1-5
Y	JP 2011-514842 A (SYNGENTA LIMITED) 2011.05.12, page 7 line 13, paragraph [0019] & CN 102083306 A & KR 10-2010-0124284 A & US 2011/0200658 A1 & WO 2009/063257 A2	1-5
A	JP 62-215504 A (SUMITOMO CHEMICAL COMPANY LIMITED) 1987.09.22, claim 1, page 5 example 11, page 6 test example 1 & DE 3708671 A & FR 2595545 A & GB 2187957 A	1-5
A	JP 59-039807 A (SUMITOMO CHEMICAL COMPANY LIMITIED) 1984.03.05, whole document (no family)	1-5
A	US 2003/0134910 A1 (CIBA SPECIALTY CHEMICALS CORPORATION) 2003.07.17, whole document & CN 1438837 A & WO 2002/000023 A1	1-5
A	JP 2002-316902 A (SUMITOMO CHEMICAL COMPANY LIMITED) 2002.10.31, paragraph [0007], [0041] (no family)	1-5

## &lt;Subject of search&gt;

Claims 1-5 relate to a microcapsule comprising an agriculturally active ingredient (a) that is defined by a specific parameter "a retention rate is less than 50% after irradiation with xenon light (290 nm cutoff) at an intensity of 0.68 W/m<sup>2</sup> at 340 nm for 8 hours". The active ingredient (a) of claims 1-5 includes all of compounds having the property.

However, those compounds which are disclosed in the meaning within PCT Article 5 are just some of the claimed compounds. Furthermore, it is impossible for a person skilled in the art to specify the scope of the compounds having the property, because the above mentioned parameter is neither commonly used nor accepted. Therefore, it cannot be considered that these claims are supported by the disclosure of the description in the meaning within PCT Article 6.

Such being the case, the search was made on a microcapsule comprising the compound that are described specifically in the description as agriculturally active ingredient (a), i.e., "fenpyrazamine, allethrin, and prallethrin" (paragraph[0009], page 5, line 14).