

PESTICIDAL COMPOSITION

BACKGROUND OF THE INVENTION

1) Field of the Invention

The present invention relates to a novel
pesticidal composition, which comprises microcapsules,
5 each microencapsulating an organophosphorus compound.

2) Related Art

Pesticidal composition comprising
microcapsules, each microencapsulating an
organophosphorus compound, for example, chlorpyrifos,
10 have been so far used to control wood-injuring insects.
However, pesticidal compositions comprising
microcapsules, each microencapsulating an
organophosphorus compound which is solid at room
temperature, for example, chlorpyrifos, etc. do not have
15 always a long-term storage stability and also have a
deterioration problem of compositions due to crystal
deposit and crystal growth. Their improvement has been
desired.

SUMMARY OF THE INVENTION

20 An object of the present invention is to solve
the above-mentioned problems.

According to the present invention, there is
provided a pesticidal composition, which comprises
microcapsules, each microencapsulating an



organophosphorus compound having a melting point of at least 15°C, and at least one solvent selected from the group consisting of:

- (i) an ester,
- 5 (ii) a ketone,
- (iii) an indane substituted with at least one lower alkyl group,
- (iv) a naphthalene substituted with at least one lower alkyl group, and
- 10 (v) a phenylxylylethane.

The pesticidal composition according to the present invention will be hereinafter referred to as "the present composition".

DETAILED DESCRIPTION OF THE INVENTION

15 The organophosphorus compound for use in the present invention has a melting point of at least 15°C, and includes, for example, the following effective compounds for insecticides, bactericides, fungicides and herbicides:

- 20 (1) O,O-diethyl O-3,5,6-trichloro-2-pyridyl phosphorothioate
(chlorpyrifos)
- (2) O,O-dimethyl O-3,5,6-trichloro-2-pyridyl phosphorothioate
- 25 (chlorpyrifos-methyl)
- (3) O,O-diethyl O-quinoxalin-2-yl phosphorothioate

(quinalphos)

(4) O-(1,6-dihydro-6-oxo-1-phenylpyridazin-3-yl) O,O-diethyl phosphorothioate

(pyridaphenthion)

5 (5) O,O-dimethyl S-(N-methylcarbamoylmethyl) dithiophosphate

(dimethoate)

(6) S- α -ethoxycarbonylbenzyl O,O-dimethyl phosphorodithioate

10 (PAP)

(7) S-6-chloro-2,3-dihydro-2-oxobenzoxazol-3-ylmethyl O,O-diethyl phosphorodithioate

(phosalone)

(8) S-2,3-dihydro-5-methoxy-2-oxo-1,3,4-
15 thiadiazol-3-ylmethyl O,O-dimethyl phosphorodithioate

(DMTP)

(9) 2-chloro-1-(2,4-dichlorophenyl)vinyl dimethyl phosphate

(dimethylvinphos)

20 (10) dimethyl 2,2,2-trichloro-1-hydroxyethylphosphonate

(trichlorfon)

(11) O-ethyl O-4-nitrophenyl phenylphosphonothioate

25 (EPN)

(12) S-benzyl O,O-diisopropyl phosphorothioate

(IBP)

(13) O-2,6-dichloro-p-tolyl O,O-dimethyl
phosphorothioate

(tolclofos-methyl)

(14) O-methyl O-2-nitro-p-tolyl
5 isopropylphosphoroamidothioate

(amiprofos-methyl)

The (i) ester for use as solvent in the present invention includes non-aromatic ester solvents and aromatic ester solvents. In case of non-aromatic
10 ester solvents, those having at least 8 carbon atoms in total are preferable. Non-aromatic ester solvents having at least 8 carbon atoms in total include liquid ester compounds having no aromatic ring structure and having at least 8 carbon atoms in total, for example, aliphatic
15 monocarboxylic acid esters such as capric acid esters (e.g. methyl caprate, etc.), lauric acid esters (e.g. methyl laurate, etc.), myristic acid esters (e.g. isopropyl myristate, etc.), palmitic acid esters (e.g. isopropyl palmitate, etc.), 2-ethylhexanoic acid esters
20 (e.g. cetyl 2-ethylhexanoate, etc.), oleic acid esters (e.g. methyl oleate, isobutyl oleate, etc.) and the like, and aliphatic dicarboxylic acid esters such as adipic acid diesters (e.g. diisobutyl adipate, dihexyl adipate, di-2-ethylhexyl adipate, diisodecyl adipate, etc.),
25 sebacic acid diesters (e.g. dibutyl sebacate, di-2-ethylhexyl sebacate, dioctyl sebacate, etc.), maleic acid diesters (e.g. dibutyl maleate, etc.), fumaric acid diesters (e.g. dibutyl fumarate, etc.), and the like.

Aromatic ester solvents, that is, ester solvents having an aromatic ring structure in the molecule, include ester solvents derived from aromatic acids, such as phthalic acid dialkyl esters, where alkyl groups are, for example, the same or different alkyl groups having 1 to 13 carbon atoms (e.g. dibutyl phthalate, dioctyl phthalate, dinonyl phthalate, didecyl phthalate, diundecyl phthalate, ditridecyl phthalate, etc.), phthalic acid diesters (e.g. butylbenzyl phthalate, dicyclohexyl phthalate, etc.), benzoic acid esters (e.g. butyl benzoate, isoamyl benzoate, etc.), salicylic acid esters (e.g. methyl salicylate, ethyl salicylate, isoamyl salicylate, etc.), and the like, and ester solvents derived from aromatic alcohols, such as benzyl esters (e.g. benzyl acetate, benzyl propionate, benzyl salicylate, etc.) and the like.

The (ii) ketone for use as solvent in the present invention is preferably those having a low water solubility, such as ketone solvents having a water solubility of not more than 7 wt.% at 20°C, for example, methyl isobutyl ketone, methyl isopropyl ketone, etc.

The (iii) indane substituted with at least one lower alkyl group for use as solvent in the present invention is usually indanes substituted with an alkyl group having not more than 4 carbon atoms, such as methy lindane, ethylindane, etc.

The (iv) naphthalene substituted with at least one lower alkyl group for use as solvent in the present

invention is usually naphthalenes substituted with at least one alkyl group having not more than 4 carbon atoms, such as methylnaphthalene, dimethylnaphthalene, ethylnaphthalene, propylnaphthalene, butylnaphthalene,
5 etc.

The (v) phenylxylylethane for use as solvent in the present invention includes, for example, 1-phenyl-1-xylylethane, a mixture of phenylxylylethane isomers, etc.

10 Microcapsulating procedure for use in the preparation of the present composition includes a procedure capable of easily controlling microcapsulating reaction, for example, so far well known interfacial polymerization process, an in-situ process, etc., among
15 which the interfacial polymerization process is preferable for preparing the present composition owing to a wide selection range for microcapsule wall materials.

In the present invention, the microcapsule wall material includes, for example, polyurethane,
20 polyurea, polyamide, polyester, polycarbonate, polysulfonate, polysulfonamide, urea-formalin condensation products, melamine-urea condensation products, phenol-formalin condensation products, acrylic acid ester polymer, methacrylic acid ester polymer, vinyl
25 acetate polymer, styrene polymer, divinylbenzene polymer, ethylene-dimethacrylate polymer, etc.

Microcapsulation can be attained, that is, polyurethane microcapsule walls can be formed, for

example, by interfacial polymerization of polyvalent isocyanate compounds and polyhydric alcohol compounds; polyurea microcapsule walls by interfacial polymerization of polyvalent isocyanate compounds and polyvalent amine compounds or water; polyamide microcapsule walls by interfacial polymerization of polyvalent acid chloride compounds and polyvalent amine compounds; polyester microcapsule walls by interfacial polymerization of polyvalent acid chloride compounds and polyhydric alcohol compounds, and so on, as is known from T. Kondo et al: New microcapsulating technique and its application development and actual application examples (published by Management Development Center, Japan) etc.

According to one embodiment of the present invention, there is provided a composition comprising microcapsules, each microencapsulating chlorpyrifos, which is particularly suitable for controlling wood-injuring insects such as termites, lyctus powder-post beetles, etc.

When the present composition comprising microcapsules, each microencapsulating chlorpyrifos as an organophosphorus compound, is used to control wood-injuring insects, it is preferable from the view point of an increase in the wood-injuring insect control effect and storage stability of composition to prepare microcapsules to make the average microcapsule diameter not more than 30 μm , a ratio of (average diameter/wall thickness) of microcapsules not less than 30, but not

more than 200, and a product of (average diameter x wall thickness) of microcapsules not more than 20 μm^2 .

The average diameter of microcapsules can be measured, for example, by Coulter Counter, Model TA-II
5 (produced by Coulter Electronics Inc.).

The wall thickness of microcapsules can be calculated according to the following formula (I):

$$\text{Wall thickness} = \frac{\text{Average diameter}}{6} \times \frac{\text{Weight of microcapsule wall material}}{\text{Weight of micro-encapsulated components}} \times \frac{\text{Average density of micro-encapsulated components}}{\text{Average density of micro-capsule wall material}} \quad (I)$$

When the present composition is applied to wood surfaces outdoors, color changes are sometimes observed on the applied areas. However, the color changes can be prevented by selecting a microcapsule wall
5 material so as to make a proportion (A) of aromatic ring structure in the microcapsule wall material not more than 40% by weight and a ratio of (A) to (average diameter/wall thickness) of the microcapsules not less than 0, but not more than 0.8. To this effect, the
10 percent-by-weight proportion (A) of aromatic ring structure in the microcapsule wall material may be lowered, for example, by keeping a proportion of a microcapsule wall forming material having the aromatic

ring structure such as phenylene diisocyanate compounds or phenylene diamine compounds lower, when polyurea microcapsule walls are formed by the above-mentioned interfacial polymerization process. Needless to say, the
5 percent-by-weight proportion (A) may be made zero by using a microcapsule wall forming material having no aromatic ring structure at all.

In view of the foregoing requirements for the microcapsule wall forming materials, preferable
10 polyvalent isocyanate compounds for use in the formation of polyurethane microcapsule walls or polyurea microcapsule walls include, for example, hexamethylene diisocyanate, adducts of hexamethylene diisocyanate and trimethylolpropane, biuret condensation products of three
15 molecules of hexamethylene diisocyanate, adducts of tolylene diisocyanate and trimethylolpropane, isocyanurate condensation products of tolylene diisocyanate, isocyanurate condensation products of hexamethylene diisocyanate, isocyanurate condensation
20 products of isophorone diisocyanate, isocyanate prepolymers each constituted of one isocyanurate moiety that consists of one isocyanate part of hexamethylene diisocyanate and two molecules of tolylene diisocyanate and of another isocyanurate moiety that consists of
25 another isocyanate part of the same hexamethylene diisocyanate and two molecules of other hexamethylene diisocyanate, 4,4'-methylenebis (cyclohexyl isocyanate), trimethyl hexamethylene diisocyanate, etc.

Polyhydric alcohol compounds for use in the formation of polyurethane microcapsule walls or polyester microcapsule walls includes, for example, ethylene glycol, propylene glycol, butanediol, hexanediol, 5 diethylene glycol, triethylene glycol, dipropylene glycol, polyethylene glycol, polypropylene glycol, glycerine, resorcin, hydroquinone, trimethylol propane, 1,2,6-hexanetriol, etc.

Polyvalent amine compounds for use in the 10 formation of polyurea microcapsule walls or polyamide microcapsule walls includes, for example, ethylenediamine, butanediamine, hexamethylenediamine, phenylenediamine, toluylenediamine, diethylenetriamine, triethylenetetramine, tetraethylenepentamine, 15 pentaethylenhexamine, 1,3,6-triaminonaphthalene, piperazine, etc.

Polyvalent acid chloride compounds for use in the formation of polyamide microcapsule walls or polyester microcapsule walls includes, for example, 20 sebacoyl dichloride, adipoyl dichloride, azelaoyl dichloride, terephthaloyl chloride, trimesoyl chloride, pimeloyl dichloride, dodecanedioyl dichloride, citroyl dichloride, etc.

Among the above-mentioned solvents for use in 25 the present invention, non-aromatic ester solvents and ketone solvents containing no aromatic ring are preferable for application to wood surfaces outdoors from the view point of preventing color changes.

When the present composition comprising microcapsules, each microencapsulating chlorpyrifos, is used to prevent wood-injuring insects, an effective amount of the composition is applied to wood-injuring insects, habitat and paths of wood-injuring insects or wood. When the present composition is used to control termites, an application dose of the present composition is about 5 to about 50 g/m² in terms of the weight of chlorpyrifos in case of spraying onto soil or concrete surfaces under house. In case of soil barrier treatment by mixing with soil, an application dose of the present composition is about 50 to about 5000 g/m³ in terms of the weight of chlorpyrifos. In case of spraying the present composition to wood surfaces, the dose is about 1 to about 5 g/m² in terms of the weight of chlorpyrifos.

When the present composition comprising microcapsules, each microencapsulating chlorpyrifos, is used to make insect-proof plywoods to control lyctus powder-post beetles (Lyctus brunneus Stephens), the present composition is mixed with an adhesive and veneers are pasted and bonded to one another with the resulting adhesive containing the present composition in an application dose of about 10 to about 1,000 g/m³ of plywood in terms of the weight of chlorpyrifos, followed by pressing with heating, thereby making insect-proof plywoods. The adhesive for use in making the insect-proof plywoods includes, for example, a urea resin adhesive, a melamine-urea adhesive, a modified phenol

resin adhesive, an alkali-phenol resin adhesive, etc.

The present composition comprising microcapsules, each microencapsulating chlorpyrifos, is effective not only for controlling the wood-injuring insects, but also for controlling nuisance injurious insects such as ants, camel crickets, millipedes, centipedes, sowbugs, pill bugs, etc. In this case, the present composition can be sprayed to the nuisance injurious insects, or habitat or paths of nuisance injurious insects in a dose of about 1 to about 50 g/m² in terms of the weight of chlorpyrifos.

The present composition can be formulated in various forms such as suspension concentrate (SC), dusts (DP), wettable powders (WP), granules (GR), etc., among which the suspension concentrate is often used from the view point of easy preparation and good storage stability. The suspension concentrate can be prepared by adding, if necessary, a stabilizer such as a thickening agent, an anti-freezing agent, a preservative, a specific gravity-regulating agent, etc. to a slurry obtained by microcapsulation reaction based on the interfacial polymerization process.

The thickening agent includes, for example, substances which exhibit a thickening effect in water, such as natural polysaccharides, (e.g. xanthane gum, rhamthan gum, locust bean gum, carrageenan, wellan gum, etc.), synthetic polymers (e.g. sodium polyacrylate, etc.), semi-synthetic polymers (e.g. carboxymethyl

cellulose, etc.), fine powders of minerals (e.g. aluminum magnesium silicate, smectite, bentonite, hectorite, silica, etc.), alumina sol, etc. The amount of the thickening agent in the present composition is usually 0
5 to 10% by weight.

The anti-freezing agent includes, for example, ethylene glycol, propylene glycol, glycerine, etc. The amount of the anti-freezing agent in the present composition is usually 0 to 20% by weight.

10 The preservative includes, for example, those which can be used in the ordinary pesticidal formulations such as p-hydroxybenzoic acid esters, salicyclic derivatives, etc. The amount of the preservative in the present composition is usually 0 to 10% by weight.

15 The specific gravity-regulating agent includes, for example, water-soluble salts such as sodium sulfate, etc., water-soluble fertilizer such as urea, etc., and the like. The amount of the specific gravity-regulating agent in the present composition is usually 0
20 to 50% by weight.

The amount of the organophosphorus compound as an active ingredient in the present composition is usually 0.5 to 80% by weight, though it depends on the kind of active ingredient, type of formulation of the
25 present composition, etc., and in case of an aqueous suspension, it is usually 0.5 to 50% by weight.

The above-mentioned solvents (i) to (v) are usually used in an amount of 0.1 to 10 parts by weight,

preferably 0.3 to 5 parts by weight, per one part by weight of the organophosphorus compound as an active ingredient.

EMBODIMENTS OF THE INVENTION

5 The present invention will be described in detail below, referring to Production Examples, Comparative Examples and Test Examples, where the average diameter was measured by a Coulter Counter Model TA-II, and the wall thickness was calculated according to the
10 above-mentioned formula (I).

Production Example 1

3.21 parts by weight of Sumidur N-75
(trademark of a biuret condensation product of hexamethylene diisocyanate containing no aromatic ring in
15 the molecule, made by Sumitomo-Bayer Urethane Co., Japan), 20 parts by weight of diisodecyl adipate and 20 parts by weight of chlorpyrifos were mixed together to obtain a homogeneous mixture, and then the resulting mixture was added to 150 parts by weight of an aqueous
20 solution containing 8% by weight of polyvinyl alcohol and 6% by weight of ethylene glycol and stirred at room temperature by a T.K. autohomomixer (trademark of a homogenizer made by Tokushukika Kogyo Co., Japan) to obtain a dispersion of microfine droplets. Then, the
25 dispersion was gently stirred at 60°C for 24 hours to obtain a slurry containing microcapsules, each

microencapsulating chlorpyrifos with a polyurethane microcapsule wall. Then, 206.7 parts by weight of an aqueous solution containing one part by weight of xanthane gum and 5 parts by weight of aluminum magnesium silicate was added to the resulting slurry to obtain the present pesticidal composition containing 5% by weight of chlorpyrifos.

It was found that the resulting microcapsules had an average diameter of 14.4 μm , a wall thickness of 0.14 μm , an (average diameter/wall thickness) value of 103, and an (average diameter x wall thickness) value of 2.0 μm^2 . A proportion (A) of aromatic ring structure in the microcapsule wall material was 0% by weight, and a ratio of (A) to (average diameter/wall thickness) was 0.

15 Production Example 2

14.07 parts by weight of Sumidur L-75 (trademark of an adduct of tolylene diisocyanate and trimethylolpropane having a molecular weight of 656 and containing 3 benzene rings as aromatic rings in the molecule, made by Sumitomo-Bayer Urethane Co., Japan), 40 parts by weight of diisobutyl adipate and 20 parts by weight of chlorpyrifos were mixed together to obtain a homogeneous mixture, and then the resulting mixture was added to 150 parts by weight of an aqueous solution containing 5% by weight of gum arabic and 6% by weight of ethylene glycol and stirred at room temperature by a T.K. autohomomixer to obtain a dispersion of microfine

droplets. Then, the dispersion was gently stirred at 60°C for 24 hours to obtain a slurry containing microcapsules, each microencapsulating chlorpyrifos with a polyurethane microcapsule wall. Then, 175.93 parts by weight of an aqueous solution containing 2 parts by weight of xanthane gum and 4 parts by weight of aluminum magnesium silicate was added to the resulting slurry to obtain the present pesticidal composition containing 5% by weight of chlorpyrifos.

It was found that the resulting microcapsules had an average diameter of 25.2 μm , a wall thickness of 0.65 μm , an (average diameter/wall thickness) value of 39 and an (average diameter x wall thickness) of 16.4 μm^2 . A proportion (A) of aromatic ring structure in the microcapsule wall material was 28.8% by weight and a ratio of (A) to (average diameter/wall thickness) was 0.74.

Production Example 3

2.34 parts by weight of Sumidur L-75, 10 parts by weight of diisobutyl adipate and 20 parts by weight of chlorpyrifos were mixed together to obtain a homogeneous mixture and then the resulting mixture was added to 150 parts by weight of an aqueous solution containing 8% by weight of polyvinyl alcohol and 6% by weight of ethylene glycol and stirred at room temperature by a T.K. autohomomixer to obtain a dispersion of microfine droplets. Then, the dispersion was gently stirred at

60°C for 24 hours to obtain a slurry containing microcapsules, each microencapsulating chlorpyrifos with a polyurethane microcapsule wall. Then, 217.66 parts by weight of an aqueous solution containing 2 parts by weight of xanthane gum and 4 parts by weight of aluminum magnesium silicate was added to the resulting slurry to obtain the present pesticidal composition containing 5% by weight of chlorpyrifos.

It was found that the resulting microcapsules had an average diameter of 10.9 μm , a wall thickness of 0.11 μm , an (average diameter/wall thickness) value of 99 and an (average diameter x wall thickness) value of 1.2 μm^2 . A proportion (A) of aromatic ring structure in the microcapsule wall material was 28.8% by weight and a ratio of (A) to (average diameter/wall thickness) was 0.29.

Production Example 4

6.31 parts by weight of Sumidur L-75, 40 parts by weight of diisobutyl adipate and 20 parts by weight of chlorpyrifos were mixed together to obtain a homogeneous mixture, and then the mixture was added to 150 parts by weight of an aqueous solution containing 8% by weight of polyvinyl alcohol and stirred at room temperature with a T.K. autohomomixer to obtain a dispersion of microfine droplets. Then, the dispersion was gently stirred at 60°C for 24 hours to obtain a slurry containing microcapsules, each microencapsulating chlorpyrifos with

a polyurea microcapsule wall. Then, 183.69 parts by weight of an aqueous solution containing 2 parts by weight of xanthane gum and 4 parts by weight of aluminum magnesium silicate was added to the resulting slurry to
5 obtain the present pesticidal composition containing 5% by weight of chlorpyrifos.

It was found that the resulting microcapsules had an average diameter of 4.9 μm , a wall thickness of 0.06 μm , an (average diameter/wall thickness) value of
10 82, and an (average diameter x wall thickness) value of 0.3 μm^2 . A proportion (A) of aromatic ring structure in the microcapsule wall material was 32.9% by weight and a ratio of (A) to (average diameter/wall thickness) was 0.40.

15 Production Example 5

4.39 parts by weight of Sumidur L-75, 20 parts by weight of diisobutyl adipate and 20 parts by weight of chlorpyrifos were mixed together to obtain a homogeneous mixture, and then the mixture was added to 150 parts by
20 weight of an aqueous solution containing 8% by weight of polyvinyl alcohol and 6% by weight of ethylene glycol and stirred at room temperature with a T.K. autohomomixer to obtain a dispersion of microfine droplets. Then, the dispersion was gently stirred at 60°C for 24 hours to
25 obtain a slurry containing microcapsules, each microencapsulating chlorpyrifos with a polyurethane microcapsule wall. Then, 205.61 parts by weight of an

aqueous solution containing 2 parts by weight of xanthane gum and 4 parts by weight of aluminum magnesium silicate was added to the resulting slurry to obtain the present pesticidal composition containing 5% by weight of
5 chlorpyrifos.

It was found that the resulting microcapsules had an average diameter of 6.6 μm , a wall thickness of 0.09 μm , an (average diameter/wall thickness) value of 73, and an (average diameter x wall thickness) value of
10 0.59 μm^2 . A proportion (A) of aromatic ring structure in the microcapsule wall material was 28.8% by weight, and a ratio of (A) to (average diameter/wall thickness) was 0.39.

Production Example 6

15 1.95 parts by weight of Sumidur L-75, 25 parts by weight of dibutyl fumarate and 20 parts by weight of chlorpyrifos were mixed together to obtain a homogeneous mixture, and then the mixture was added to 150 parts by weight of an aqueous solution containing 5% by weight of
20 gum arabic and 6% by weight of ethylene glycol and stirred at room temperature with a T.K. autohomomixer to obtain a dispersion of microfine droplets. Then, the dispersion was gently stirred at 60°C for 24 hours to obtain a slurry containing microcapsules, each
25 microencapsulating chlorpyrifos with a polyurethane microcapsule wall. Then, 203.05 parts by weight of an aqueous solution containing 2 parts by weight of xanthane

gum and 4 parts by weight of aluminum magnesium silicate was added to the resulting slurry to obtain the present pesticidal composition containing 5% by weight of chlorpyrifos.

5 It was found that the resulting microcapsules had an average diameter of 22.6 μm , a wall thickness of 0.12 μm , an (average diameter/wall thickness) value of 188, and an (average diameter x wall thickness) value of 2.7 μm^2 . A proportion (A) of aromatic ring structure in
10 the microcapsule wall material was 28.8% by weight and a ratio of (A) to (average diameter/wall thickness) of 0.15.

Production Example 7

 4.13 parts by weight of Sumidur HT (trademark
15 of an adduct of hexamethylene diisocyanate and trimethylolpropane having no aromatic ring in the molecule, made by Sumitomo-Bayer Urethane Co., Japan), 20 parts by weight of methyl oleate and 20 parts by weight of chlorpyrifos were mixed together to obtain a
20 homogeneous mixture, and then the mixture was added to 150 parts by weight of an aqueous solution containing 8% by weight of polyvinyl alcohol and 6% by weight of ethylene glycol and stirred at room temperature by a T.K. autohomomixer to obtain a dispersion of microfine
25 droplets. Then, the dispersion was gently stirred at 60°C for 24 hours to obtain a slurry containing microcapsules, each microencapsulating chlorpyrifos with

a polyurethane microcapsule wall. Then, 205.87 parts by weight of an aqueous solution containing 2 parts by weight of xanthane gum and 4 parts by weight of aluminum magnesium silicate was added to the resulting slurry to
5 obtain the present pesticidal composition containing 5% by weight of chlorpyrifos.

It was found that the resulting microcapsules had an average diameter of 11.7 μm , a wall thickness of 0.14 μm , an (average diameter/wall thickness) value of 84
10 and an (average diameter x wall thickness) value of 1.6 μm^2 . A proportion (A) of aromatic ring structure in the microcapsule wall material was 0% by weight and a ratio of (A) to (average diameter/wall thickness) was 0.

Production Example 8

15 2.32 parts by weight of Millionate MR-400 (trademark of polyethylene polyphenyl polyisocyanate, containing one benzene ring as an aromatic ring per methylene phenyl isocyanate unit, made by Nihon Polyurethane Kogyo K.K., Japan), 20 parts by weight of
20 diisobutyl adipate and 20 parts by weight of chlorpyrifos were mixed together to obtain a homogeneous mixture, and then the mixture was added to 150 parts by weight of an aqueous solution containing 8% by weight of polyvinyl alcohol and 6% by weight of ethylene glycol and stirred
25 at room temperature by a T.K. autohomomixer to obtain a dispersion of microfine droplets. Then, the dispersion was gently stirred at 60°C for 24 hours to obtain a

slurry containing microcapsules, each microencapsulating chlorpyrifos with a polyurethane microcapsule wall. Then, 207.66 parts by weight of an aqueous solution containing 2 parts by weight of xanthane gum and 4 parts
5 by weight of aluminum magnesium silicate was added to the resulting slurry to obtain the present pesticidal composition containing 5% by weight of chlorpyrifos.

It was found that the resulting microcapsules had an average diameter of 12.1 μm , a wall thickness of
10 0.13 μm , an (average diameter/wall thickness) value of 93 and an (average diameter x wall thickness) value of 1.6 μm^2 . A proportion (A) of aromatic ring structure in the microcapsule wall material was 45.6% by weight and a ratio of (A) to (average diameter/wall thickness) was
15 0.49.

Production Example 9

6.21 parts by weight of Sumidur L-75, 20 parts by weight of diisobutyl adipate and 20 parts by weight of chlorpyrifos were mixed together to obtain a homogeneous
20 mixture, and then the mixture was added to 150 parts by weight of an aqueous solution containing 5% by weight of gum arabic and 6% by weight of ethylene glycol and stirred at room temperature by a T.K. autohomomixer to obtain a dispersion of microfine droplets. Then, the
25 dispersion was gently stirred at 60°C for 24 hours to obtain a slurry containing microcapsules, each microencapsulating chlorpyrifos with a polyurethane

microcapsule wall. Then, 203.79 parts by weight of an aqueous solution containing 2 parts by weight of xanthane gum and 4 parts by weight of aluminum magnesium silicate was added to the resulting slurry to obtain the present
5 pesticidal composition containing 5% by weight of chlorpyrifos.

It was found that the resulting microcapsules had an average diameter of 47.5 μm , a wall thickness of 0.88 μm , an (average diameter/wall thickness) value of 54
10 and an (average diameter x wall thickness) value of 41.8 μm^2 . A proportion (A) of aromatic ring structure in the microcapsule wall material was 28.8% by weight and a ratio of (A) to (average diameter/wall thickness) was 0.53.

15 Production Example 10

One part by weight of Sumidur L-75, 40 parts by weight of diisobutyl adipate and 20 parts by weight of chlorpyrifos were mixed together to obtain a homogeneous mixture, and then the mixture was added to 150 parts by
20 weight of an aqueous solution containing 5% by weight of gum arabic and 6% by weight of ethylene glycol and stirred at room temperature by a T.K. autohomomixer to obtain a dispersion of microfine droplets. Then, the dispersion was gently stirred at 60°C for 24 hours to
25 obtain a slurry containing microcapsules, each microencapsulating chlorpyrifos with a polyurethane microcapsule wall. Then, 189 parts by weight of an

aqueous solution containing 2 parts by weight of xanthane gum and 4 parts by weight of aluminum magnesium silicate was added to the resulting slurry to obtain the present pesticidal composition containing 5% by weight of
5 chlorpyrifos.

It was found that the resulting microcapsules had an average diameter of 25.9 μm , a wall thickness of 0.05 μm , an (average diameter/wall thickness) value of 518 and an (average diameter x wall thickness) value of
10 $1.3 \mu\text{m}^2$. A proportion (A) of aromatic ring structure in the microcapsule wall material was 28.8% by weight and a ratio of (A) to (average diameter/wall thickness) was 0.056.

Production Example 11

15 22.59 parts by weight of Sumidur L-75, 5 parts by weight of diisobutyl adipate and 20 parts by weight of chlorpyrifos were mixed together to obtain a homogeneous mixture, and then the mixture was added to 150 parts by weight of an aqueous solution containing 8% by weight of
20 polyvinyl alcohol and 6% by weight of ethylene glycol and stirred at room temperature by a T.K. autohomomixer to obtain a dispersion of microfine droplets. Then, the dispersion was gently mixed at 60°C for 24 hours to obtain a slurry containing microcapsules, each
25 microencapsulating chlorpyrifos with a polyurethane microcapsule wall. Then, 202.41 parts by weight of an aqueous solution containing 2 parts by weight of xanthane

gum and 4 parts by weight of aluminum magnesium silicate was added to the resulting slurry to obtain the present pesticidal composition containing 5% by weight of chlorpyrifos.

5 It was found that the resulting microcapsules had an average diameter of 19.9 μm , a wall thickness of 2.0 μm , an (average diameter/wall thickness) value of 10 and an (average diameter x wall thickness) value of 39.8 μm^2 . A proportion (A) of aromatic ring structure in the
10 microcapsule wall material was 28.8% by weight and a ratio of (A) to (average diameter/wall thickness) was 2.88.

Production Example 12

2.19 parts by weight of Sumidur L-75, 40 parts
15 by weight of dioctyl sebacate and 20 parts by weight of chlorpyrifos were mixed together to obtain a homogeneous mixture, and the mixture was added to 150 parts by weight of an aqueous solution containing 5% by weight of gum arabic and 6% by weight of ethylene glycol and stirred at
20 room temperature by a T.K. autohomomixer to obtain a dispersion of microfine droplets. Then, the dispersion was gently stirred at 60°C for 24 hours to obtain a slurry containing microcapsules, each microencapsulating chlorpyrifos with a polyurethane microcapsule wall.
25 Then, 187.81 parts by weight of an aqueous solution containing 2 parts by weight of xanthane gum and 4 parts by weight of aluminum magnesium silicate was added to the

slurry to obtain the present pesticidal composition containing 5% by weight of chlorpyrifos.

It was found that the resulting microcapsules had an average diameter of 26.6 μm , a wall thickness of 0.11 μm , an (average diameter/wall thickness) value of 242 and an (average diameter x wall thickness) value of 2.9 μm^2 . A proportion (A) of aromatic ring structure in the microcapsule wall material was 28.8% by weight and a ratio of (A) to (average diameter/wall thickness) was 0.12.

Production Example 13

2 parts by weight of Sumidur L-75, 0.2 part by weight of sebacoyl dichloride, 20 parts by weight of diisobutyl adipate and 20 parts by weight of chlorpyrifos were mixed together to obtain a homogeneous mixture, and then the mixture was added to 150 parts by weight of an aqueous solution containing 8% by weight of polyvinyl alcohol and 6% by weight of hexamethylene diamine and stirred at room temperature by a T.K. autohomomixer to obtain a dispersion of microfine droplets. Then, the dispersion was gently stirred at room temperature for 4 hours and then at 60°C for 20 hours to obtain a slurry containing microcapsules, each microencapsulating chlorpyrifos with a polyurea-polyamide microcapsule wall. Then, 207.08 parts by weight of an aqueous solution containing 2 parts by weight of xanthane gum and 4 parts by weight of aluminum magnesium silicate was added to the

resulting slurry to obtain the present pesticidal composition containing 5% by weight of chlorpyrifos.

It was found that the resulting microcapsules had an average diameter of 10.5 μm , a wall thickness of 0.09 μm , an (average diameter/wall thickness) value of 117 and an (average diameter x wall thickness) value of 0.9 μm^2 . A proportion (A) of aromatic ring structure in the microcapsule wall material was 28.8% by weight and a ratio of (A) to (average diameter/wall thickness) was 0.25.

Production Example 14

20 parts by weight of chlorpyrifos, 20 parts by weight of dibutyl phthalate and 3.24 parts by weight of Sumidur L-75 were mixed together to obtain a homogeneous mixture. Then, the mixture was added to 150 parts by weight of an aqueous solution containing 8% by weight of polyvinyl alcohol and 6% by weight of ethylene glycol and stirred at room temperature by a T.K. autohomomixer to obtain a dispersion of microfine droplets. Then, the dispersion was gently stirred at 60°C for 24 hours to obtain a slurry containing microcapsules, each microencapsulating chlorpyrifos with a polyurethane microcapsule wall. Then, 206.76 parts by weight of an aqueous solution containing 2 parts by weight of xanthane gum and 4 parts by weight of aluminum magnesium silicate was added to the resulting slurry to obtain the present pesticidal composition containing 5%

by weight of chlorpyrifos.

It was found that the resulting microcapsules had an average diameter of 5.1 μm , a wall thickness of 0.05 μm , an (average diameter/wall thickness) value of 102 and an (average diameter x wall thickness) value of 0.26 μm^2 . A proportion (A) of aromatic ring structure in the microcapsule wall material was 28.8% by weight and a ratio of (A) to (average diameter/wall thickness) was 0.28.

10 Production Example 15

20 parts by weight of chlorpyrifos, 20 parts by weight of diisodecyl phthalate and 3.52 parts by weight of Sumidur L-75 were mixed together to obtain a homogeneous mixture. Then, the mixture was added to 150 parts by weight of an aqueous solution containing 8% by weight of polyvinyl alcohol and 6% by weight of ethylene glycol and stirred at room temperature by a T.K. autohomomixer to obtain a dispersion of microfine droplets. Then, the dispersion was gently stirred at 60°C for 24 hours to obtain a slurry containing microcapsules, each microencapsulating chlorpyrifos with a polyurethane microcapsule wall. Then, 206.48 parts by weight of an aqueous solution containing 2 parts by weight of xanthane gum and 4 parts by weight of aluminum magnesium silicate was added to the resulting slurry to obtain the present pesticidal composition containing 5% by weight of chlorpyrifos.

It was found that the resulting microcapsules had an average diameter of 13.1 μm , a wall thickness of 0.14 μm , an (average diameter/wall thickness) value of 94 and an (average diameter x wall thickness) value of 1.83 μm^2 . A proportion (A) of aromatic ring structure in the microcapsule wall material was 28.8% by weight and a ratio of (A) to (average diameter/wall thickness) was 0.31.

Production Example 16

20 parts by weight of chlorpyrifos and 20 parts by weight of methylnaphthalene were mixed together with heating at 60°C, and then 4 parts by weight of Sumidur L-75 was added thereto and mixed to obtain a homogeneous mixture. Then, the mixture was added to 150 parts by weight of an aqueous solution containing 5% by weight of polyvinyl alcohol and 6% by weight of ethylene glycol and stirred at room temperature with a T.K. autohomomixer to obtain a dispersion of microfine droplets. Then, the dispersion was gently stirred at 60°C for 24 hours to obtain a slurry containing microcapsules, each microencapsulating chlorpyrifos with a polyurethane microcapsule wall. Then, 206 parts by weight of an aqueous solution containing one part by weight of xanthane gum and 5 parts by weight of aluminum magnesium silicate was added to the resulting slurry to obtain the present pesticidal composition containing 5% by weight of chlorpyrifos.

It was found that the resulting microcapsules had an average diameter of 12.2 μm , a wall thickness of 0.13 μm , an (average diameter/wall thickness) value of 94 and an (average diameter x wall thickness) value of 1.6 μm^2 . A proportion (A) of aromatic ring structure in the microcapsule wall material was 28.8% by weight and a ratio of (A) to (average diameter/wall thickness) was 0.31.

Production Example 17

10 20 parts by weight of chlorpyrifos and 20 parts by weight of dimethylnaphthalene were mixed together with heating at 60°C, and then 5 parts by weight of Sumidur L-75 was added thereto and mixed to obtain a homogeneous mixture. Then, the mixture was added to 150
15 parts by weight of an aqueous solution containing 5% by weight of gum arabic and 6% by weight of ethylene glycol, and stirred at room temperature by a T.K. autohomomixer to to obtain a dispersion of microfine droplets. Then, the dispersion was gently stirred at 60°C for 24 hours to
20 obtain a slurry containing microcapsules, each microencapsulating chlorpyrifos with a polyurethane microcapsule wall. Then, 205 parts by weight of an aqueous solution containing one part by weight of xanthane gum and 5 parts by weight of aluminum magnesium
25 silicate was added to the resulting slurry to obtain the present pesticidal composition containing 5% by weight of chlorpyrifos.

It was found that the resulting microcapsules had an average diameter of 18.6 μm , a wall thickness of 0.31 μm , an (average diameter/wall thickness) value of 60, and an (average diameter x wall thickness) value of 5.8 μm^2 . A proportion (A) of aromatic ring structure in the microcapsule wall material was 28.8% by weight and a ratio of (A) to (average diameter/wall thickness) was 0.48.

Production Example 18

10 20 parts by weight of chlorpyrifos and 10 parts by weight of methyldane were mixed together with heating at 60°C, and then 5 parts by weight of Millionate MR-400 was added thereto and mixed to obtain a homogeneous mixture. Then, the mixture was added to 150
15 parts by weight of an aqueous solution containing 8% by weight of polyvinyl alcohol and 6% by weight of ethylene glycol and stirred at room temperature by a T.K. autohomomixer to obtain a dispersion of microfine droplets. Then, the dispersion was gently stirred at
20 60°C for 24 hours to obtain a slurry containing microcapsules, each microencapsulating chlorpyrifos with a polyurethane microcapsule wall. Then, 215 parts by weight of an aqueous solution containing one part by weight of xanthane gum and 5 parts by weight of aluminum
25 magnesium silicate was added to the resulting slurry to obtain the present pesticidal composition containing 5% by weight of chlorpyrifos.

It was found that the resulting microcapsules had an average diameter of 5.1 μm , a wall thickness of 0.16 μm , an (average diameter/wall thickness) value of 32 and an (average diameter x wall thickness) value of 0.8 μm^2 . A proportion (A) of aromatic ring structure in the microcapsule wall material was 45.6% by weight and a ratio of (A) to (average diameter/wall thickness) was 1.43.

Production Example 19

10 20 parts by weight of chlorpyrifos and 30 parts by weight of phenylxylylethane were mixed together with heating at 60°C, and then 10 parts by weight of Millionate MR-400 was added thereto and mixed to obtain a homogeneous mixture. Then, the mixture was added to 150
15 parts by weight of an aqueous solution containing 5% by weight of gum arabic and 12% by weight of ethylene glycol and stirred at room temperature by a T.K. autohomomixer to obtain a dispersion of microfine droplets. Then, the dispersion was gently stirred at 60°C for 24 hours to
20 obtain a slurry containing microcapsules, each microencapsulating chlorpyrifos with a polyurethane microcapsule wall. Then, 190 parts by weight of an aqueous solution containing 2 parts by weight of xanthane gum and 4 parts by weight of aluminum magnesium silicate
25 was added to the resulting slurry to obtain the present pesticidal composition containing 5% by weight of chlorpyrifos.

It was found that the resulting microcapsules had an average diameter of 18.6 μm , a wall thickness of an 0.65 μm , an (average diameter/wall thickness) value of 29 and an (average diameter x wall thickness) value of 5 12.1 μm^2 . A proportion (A) of aromatic ring structure in the microcapsule wall material was 45.6% by weight and a ratio of (A) to (average diameter/wall thickness) was 1.57.

Production Example 20

10 20 parts by weight of chlorpyrifos and 20 parts by weight of phenylxylylethane were mixed together with heating at 60°C, and one part by weight of Sumidur N-75 (trademark of biuret condensation product of hexamethylene diisocyanate having no aromatic ring in the 15 molecule, made by Sumitomo-Bayer Urethane Co., Japan) was added thereto and mixed to obtain a homogeneous mixture. Then, the mixture was added to 150 parts by weight of an aqueous solution containing 5% by weight of gum arabic and 6% by weight of ethylene glycol and stirred at room 20 temperature by a T.K. autohomomixer to obtain a dispersion of microfine droplets. Then, the dispersion was gently stirred at 60°C for 24 hours to obtain a slurry containing microcapsules, each microencapsulating chlorpyrifos with a polyurethane microcapsule wall.

25 Then, 190 parts by weight of an aqueous solution containing 2 parts by weight of xanthane gum and 4 parts by weight of aluminum magnesium silicate was added to the

resulting slurry to obtain the present pesticidal composition containing 5% by weight of chlorpyrifos.

It was found that the resulting microcapsules had an average diameter of 20.9 μm , a wall thickness of 0.20 μm , an (average diameter/wall thickness) value of 105, and an (average diameter x wall thickness) value of 4.2 μm^2 . A proportion (A) of aromatic ring structure in the microcapsule wall material was 0% by weight and a ratio of (A) to (average diameter/wall thickness) was 0.

10 Production Example 21

20 parts by weight of tolchlofos-methyl and 80 parts by weight of methylnaphthalene were mixed together with heating at 60°C and then 10 parts by weight of Sumidur L-75 was added thereto and mixed to obtain a homogeneous mixture. Then, the mixture was added to 150 parts by weight of an aqueous solution containing 10% by weight of polyvinyl alcohol and 10% by weight of ethylene glycol and stirred at room temperature by a T.K. autohomomixer to obtain a dispersion of microfine droplets. Then, the dispersion was gently stirred at 60°C for 24 hours to obtain a slurry containing microcapsules, each microencapsulating tolchlofos-methyl with a polyurethane microcapsule wall. Then, 140 parts by weight of an aqueous solution containing 0.8 part by weight of xanthane gum and 1.6 parts by weight of aluminum magnesium silicate was added to the resulting slurry to obtain the present pesticidal composition

containing 5% by weight of tolchlofos-methyl.

It was found that the resulting microcapsules had an average diameter of 8.7 μm , a wall thickness of 0.1 μm , an (average diameter/wall thickness) value of 87
5 and an (average diameter x wall thickness) value of 0.9 μm^2 . A proportion (A) of aromatic ring structure in the microcapsule wall material was 28.8% by weight and a ratio of (A) to (average diameter/wall thickness) was 0.33.

10 Production Example 22

20 parts by weight of tolchlofos-methyl and 70 parts by weight of methylindane were mixed together with heating at 60°C, and then 8 parts by weight of Millionate MR-400 was added thereto and mixed to obtain a
15 homogeneous mixture. Then, the mixture was added to 150 parts by weight of an aqueous solution containing 10% by weight of polyvinyl alcohol and 10% by weight of ethylene glycol and stirred at room temperature by a T.K. autohomomixer to obtain a dispersion of microfine
20 droplets. Then, the dispersion was gently stirred at 60°C for 24 hours to obtain a slurry containing microcapsules, each microencapsulating tolchlofos-methyl with a polyurethane microcapsule wall. Then, 152 parts by weight of an aqueous solution containing 0.8 part by
25 weight of xanthane gum and 1.6 parts by weight of aluminum magnesium silicate was added to the resulting slurry to obtain the present pesticidal composition

containing 5% by weight of tolchlofos-methyl.

It was found that the resulting microcapsules had an average diameter of 9.4 μm , a wall thickness of 0.14 μm , an (average diameter/wall thickness) value of 67, and an (average diameter x wall thickness) value of 1.3 μm^2 . A proportion (A) of aromatic ring structure in the microcapsule wall material was 45.6% by weight and a ratio of (A) to (average diameter/wall thickness) was 0.68.

10 Production Example 23

20 parts by weight of chlorpyrifos and 20 parts by weight of methylisobutylketone were mixed together with heating at 60°C, and then 3.5 parts by weight of Sumidur L-75 was added thereto and mixed to
15 obtain a homogeneous mixture. Then, the mixture was added to 150 parts by weight of an aqueous solution containing 5% by weight of polyvinyl alcohol and 6% by weight of ethylene glycol and stirred at room temperature by a T.K. autohomomixer to obtain a dispersion of
20 microfine droplets. Then, the dispersion was gently stirred at 60°C for 24 hours to obtain a slurry containing microcapsules, each microencapsulating chlorpyrifos with a polyurethane microcapsule wall. Then, 206.5 parts by weight of an aqueous solution
25 containing one part by weight of xanthane gum and 5 parts by weight of aluminum magnesium silicate was added to the resulting slurry to obtain the present pesticidal

composition containing 5% by weight of chlorpyrifos.

It was found that the resulting microcapsules had an average diameter of 4.4 μm , a wall thickness of 0.04 μm , an (average diameter/wall thickness) value of 110, and an (average diameter x wall thickness) value of 0.18 μm^2 . A proportion (A) of aromatic ring structure in the microcapsule wall material was 28.8% by weight and a ratio of (A) to (average diameter/wall thickness) was 0.26.

10 Production Example 24

20 parts by weight of chlorpyrifos and 30 parts by weight of methyl isobutyl ketone were mixed together with heating at 60°C, and then 10 parts by weight of Millionate MR-400 was added thereto and mixed to obtain a homogeneous mixture. Then, the mixture was added to 150 parts by weight of an aqueous solution containing 5% by weight of gum arabic and stirred at room temperature by a T.K. autohomomixer to obtain a dispersion of microfine droplets. Then, the dispersion was gently stirred at 60°C for 24 hours to obtain a slurry containing microcapsules, each microencapsulating chlorpyrifos with a polyurea microcapsule wall. Then, 190 parts by weight of an aqueous solution containing one part by weight of xanthane gum and 5 parts by weight of aluminum magnesium silicate was added to the resulting slurry to obtain the present pesticidal composition containing 5% by weight of chlorpyrifos.

It was found that the resulting microcapsules had an average diameter of 20.5 μm , a wall thickness of 0.65 μm , an (average diameter/wall thickness) value of 32, and an (average diameter x wall thickness) value of 5 13.3 μm^2 . A proportion (A) of aromatic ring structure in the microcapsule wall material was 45.6% by weight and a ratio of (A) to (average diameter/wall thickness) was 1.43.

Production Example 25

10 20 parts by weight of chlorpyrifos and 15 parts by weight of methyl isobutyl ketone were mixed together with heating at 60°C, and then 5 parts by weight of Sumidur HT was added thereto and mixed to obtain a homogeneous mixture. Then, the mixture was added to 150 15 parts by weight of an aqueous solution containing 8% by weight of polyvinyl alcohol and 6% by weight of ethylene glycol and stirred at room temperature by a T.K. autohomomixer to obtain a dispersion of microfine droplets. Then, the dispersion was gently stirred at 20 60°C for 24 hours to obtain a slurry containing microcapsules, each microencapsulating chlorpyrifos with a polyurethane microcapsule wall. Then, 210 parts by weight of an aqueous solution containing one part by weight of xanthane gum and 5 parts by weight of aluminum 25 magnesium silicate was added to the resulting slurry to obtain the present pesticidal composition containing 5% by weight of chlorpyrifos.

It was found that the resulting microcapsules had an average diameter of 5.1 μm , a wall thickness of 0.08 μm , an (average diameter/wall thickness) value of 64, and an (average diameter x wall thickness) value of 0.41 μm^2 . A proportion (A) of aromatic ring structure in the microcapsule wall material was 0% by weight and a ratio of (A) to (average diameter/wall thickness) was 0.

Production Example 26

20 parts by weight of tolchlofos-methyl and 80 parts by weight of methyl isobutyl ketone were mixed together with heating at 60°C, and then 10 parts by weight of Sumidur L-75 was added thereto and mixed to obtain a homogeneous mixture. Then, the mixture was added to 150 parts by weight of an aqueous solution containing 10% by weight of polyvinyl alcohol and stirred at room temperature by a T.K. autohomomixer to obtain a dispersion of microfine droplets. Then, the dispersion was gently mixed at 60°C for 24 hours to obtain a slurry containing microcapsules, each microencapsulating tolchlofos-methyl with a polyurea microcapsule wall. Then, 140 parts by weight of an aqueous solution containing 0.8 part by weight of xanthane gum and 1.6 parts by weight of aluminum magnesium silicate was added to the resulting slurry to obtain the present pesticidal composition containing 5% by weight of tolchlofos-methyl.

It was found that the resulting microcapsules had an average diameter of 6.4 μm , a wall thickness of

0.07 μm , an (average diameter/wall thickness) value of 91 and an (average diameter x wall thickness) value of 0.45 μm^2 . A proportion (A) of aromatic ring structure in the microcapsule wall material was 32.9% by weight and a
5 ratio of (A) to (average diameter/wall thickness) was 0.36.

Production Examples of comparative compositions for Test Examples, which follow, are given below as Comparative Examples.

10 Comparative Example 1

1.92 parts by weight of Sumidur N-75, 20 parts by weight of hexane and 20 parts by weight of chlorpyrifos were mixed together and the mixture was added to 150 parts by weight of an aqueous solution
15 containing 8% by weight of polyvinyl alcohol and 6% by weight of ethylene glycol and stirred at room temperature by a T.K. autohomomixer to obtain a dispersion of microfine droplets. Then, the dispersion was gently stirred at 60°C for 24 hours to obtain a slurry, and then
20 208.08 parts by weight of an aqueous solution containing one part by weight of xanthane gum and 5 parts by weight of aluminum magnesium silicate was added to the resulting slurry to obtain a comparative pesticidal composition containing 5% by weight of chlorpyrifos.

25 Comparative Example 2

0.95 part by weight of Millionate MR-400 and

20 parts by weight of chlorpyrifos were mixed together to obtain a homogeneous mixture, and then the mixture was added to 150 parts by weight of an aqueous solution containing 8% by weight of polyvinyl alcohol and stirred
5 at room temperature by a T.K. autohomomixer to obtain a dispersion of microfine droplets. Then, the dispersion was gently stirred at 60°C for 24 hours to obtain a slurry containing microcapsules, each microencapsulating chlorpyrifos with a polyurea microcapsule wall. Then,
10 229.05 parts by weight of an aqueous solution containing 2 parts by weight of xanthane gum and 4 parts by weight of aluminum magnesium silicate was added to the resulting slurry to obtain a comparative pesticidal composition containing 5% by weight of chlorpyrifos.

15 It was found that the resulting microcapsules had an average diameter of 8.3 μm , a wall thickness of 0.1 μm , an (average diameter/wall thickness) value of 83 and an (average diameter x wall thickness) value of 0.83 μm^2 . A proportion (A) of aromatic ring structure in the
20 microcapsule wall material was 45.6% by weight and a ratio of (A) to (average diameter/wall thickness) value was 0.55.

Comparative Example 3

4 parts by weight of Sumidur L-75, 20 parts by
25 weight of xylene and 20 parts by weight of chlorpyrifos were mixed together with heating at 60°C and the mixture was added to 150 parts by weight of an aqueous solution

containing 5% by weight of gum arabic and 6% by weight of ethylene glycol and stirred at room temperature with a T.K. autohomomixer to obtain a dispersion of microfine droplets. Then, the dispersion was gently stirred at
5 60°C for 24 hours to obtain a slurry containing microcapsules, each microencapsulating chlorpyrifos with a polyurethane microcapsule wall. Then, 206 parts by weight of an aqueous solution containing 2 parts by weight of xanthane gum and 4 parts by weight of aluminum
10 magnesium silicate was added to the resulting slurry to obtain a comparative pesticidal composition containing 5% by weight of chlorpyrifos.

It was found that the resulting microcapsules had an average diameter of 19.1 μm , a wall thickness of
15 0.23 μm , an (average diameter/wall thickness) value of 83 and an (average diameter x wall thickness) value of 4.4 μm^2 . A proportion (A) of aromatic ring structure in the microcapsule wall material was 28.8% by weight and a ratio of (A) to (average diameter/wall thickness) was
20 0.35.

Test Examples will be given below.

Test Example 1

The present pesticidal compositions obtained in Production Examples 1, 3, 4, 7, 9, 12 and 16 were made
25 into aqueous 0.25 wt.% dilute solutions, respectively, and a sufficient amount (6 ml) each of the aqueous dilute solutions was sprayed to ten workers of Formosan

subterranean termites from a height of 60 cm. A 100% mortality was observed one day thereafter in all the cases.

Test Example 2

5 The present pesticidal compositions obtained in Production Examples 14, 15 and 23 were made into aqueous 1 wt.% dilute solutions, respectively, and a sufficient amount (6 ml) each of the aqueous dilute solutions was sprayed to ten workers of Formosan
10 subterranean termites from a height of 60 cm. A 100% mortality was observed one day thereafter in all the cases.

Test Example 3

 About 10 g each of the present pesticidal
15 compositions obtained in Production Examples 1 to 5 and 14 to 26 and the comparative pesticidal compositions obtained in Comparative Examples 1 and 2 were put in polyethylene bottles, respectively, and preserved in a refrigerator (refrigerator inside temperature: about
20 5°C) for two weeks, and then the respective pesticidal compositions were microscopically observed. In all the cases of the present pesticidal compositions, no crystal deposits were observed at all, whereas in cases of the comparative pesticidal compositions obtained in
25 Comparative Examples 1 and 2, a large amount of crystal deposits were observed.

That is, in all the cases of the present
pesticidal compositions obtained in Production Examples 1
to 5 and 14 to 26, no crystal deposits were observed at
all, showing a distinguished storage stability, whereas
5 in cases of the comparative pesticidal composition
obtained in Comparative Example 1 using hexane as a
solvent and the comparative pesticidal composition
obtained in Comparative Example 2 using no solvent,
crystal deposits were observed.

10 Test Example 4

0.8 Gram each of the present pesticidal
compositions obtained in Production Examples 1, 3, 4, 5,
6, 7, 14, 23, 25, 26 and the comparative pesticidal
composition obtained in Comparative Example 2 were
15 applied to filter papers, respectively, air-dried and
exposed to sunlight for 3 days. In all the cases of the
present pesticidal compositions obtained in Production
Examples 1, 3, 4, 5, 6, 7, 14, 23, 25 and 26, no
substantial color change was observed, whereas in case of
20 the comparative pesticidal composition obtained in
Comparative Example 2, yellowing was clearly observed.

Test Example 5

3 Grams each of the present pesticidal
compositions obtained in Production Examples 1, 2, 3, 8,
25 12 and 15 was applied onto glass Petri dishes,
respectively, air-dried and kept at 60°C for one week.

Then, the amounts of the active ingredient (chlorpyrifos) remaining on the Petri dishes were measured by a gas chromatography to determine the residual percentage of the active ingredient on the basis of the applied amount.

5 Results are given in the following Table 1.

Table 1

Test composition	Residual percentage of active ingredient
Production Ex. 1	100
" 2	98
" 3	94
" 8	96
" 12	98
" 15	98

As shown in Table 1, the present pesticidal compositions obtained in Production Examples 1, 2, 3, 8, 12 and 15 are effective for controlling pests for a long period of time, because vaporization of the active
5 ingredient is controlled in all the cases.

Test Example 6

The present pesticidal compositions obtained in Production Examples 16-22 and the comparative

pesticidal composition obtained in Comparative Example 3 were made into aqueous 1 wt.% dilute solutions, respectively, and the aqueous dilute solutions were given by one drop onto slide glasses, respectively, air-dried
5 and preserved in a thermostat at 40°C for 30 days. Then, the state of microcapsules was microscopically observed. In all the cases of the present pesticidal compositions obtained in Production Examples 16-22, neither breakage of microcapsule walls nor crystal deposits was observed
10 at all, whereas in case of the pesticidal composition obtained in Comparative Example 3 shrinkage and breakage of microcapsule walls were observed.

Test Example 7

The present pesticidal compositions obtained
15 in Production Examples 23-26 and the comparative pesticidal composition obtained in Comparative Example 3 were made into aqueous 1 wt.% dilute solutions and the aqueous solutions were given by one drop onto slide glasses, respectively, air-dried and preserved in a
20 thermostat at 60°C for one week. Then, the state of microcapsules was microscopically observed. In all the cases of the present pesticidal compositions obtained in Production Examples 23-26, neither breakage of microcapsule walls nor crystal deposits was observed at
25 all, whereas in the comparative pesticidal composition obtained in Comparative Example 3 shrinkage and breakage of microcapsule walls were observed.

Test Example 8

The present pesticidal compositions obtained in Production Examples 1, 4, 12, 14, 16 and 23 were made into aqueous 0.1 wt.% solutions, respectively, and 6 ml
5 each of the aqueous dilute solutions was sprayed onto soil surfaces. The sprayed soil were preserved at 60°C for respective predetermined periods of time, and then 10
workers of Formosan subterranean termites were liberated on the respective soil surfaces to determine a mortality
10 (%) 3 days thereafter. Results are shown in Table 2.

Table 2

Test composition	Preservation period at 60°C	Mortality (%)
Production Ex. 1	Initial	100
	One month	100
	3 months	100
Production Ex. 4	Initial	100
	One month	100
	3 months	100
Production Ex. 12	Initial	100
	One month	100
	3 months	100
Production Ex. 14	Initial	100
	One month	100
	3 months	100
Production Ex. 16	Initial	100
	One month	100
	3 months	100
Production Ex. 23	Initial	100
	One month	100
	3 months	100



WHAT IS CLAIMED IS:

1. A pesticidal composition, which comprises microcapsules, each microencapsulating an organophosphorus compound having a melting point of at least 15°C and at least one solvent selected from the group consisting of:
 - (i) an ester,
 - (ii) a ketone,
 - (iii) an indane substituted with at least one lower alkyl group,
 - (iv) a naphthalene substituted with at least one lower alkyl group, and
 - (v) a phenylxylylethane.
2. A pesticidal composition according to Claim 1, wherein the solvent is (i) an ester.
3. A pesticidal composition according to Claim 2, wherein the (i) ester is a non-aromatic ester having at least 8 carbon atoms in total.
4. A pesticidal composition according to Claim 2, wherein the (i) ester is an aromatic ester.
5. A pesticidal composition according to Claim 4, wherein the (i) ester is a phthalic acid ester.
6. A pesticidal composition according to Claim 1, wherein the solvent is a (ii) ketone.
7. A pesticidal composition according to Claim 1, wherein the solvent is a (iii) indane substituted with at least one lower alkyl group.
8. A pesticidal composition according to Claim 1,



wherein the solvent is a (iv) naphthalene substituted with at least one lower alkyl group.

9. A pesticidal composition according to Claim 1, wherein the solvent is a (v) phenylxylylethane.

10. A pesticidal composition according to Claim 1, wherein the organophosphorus compound is chlorpyrifos.

11. A pesticidal composition according to Claim 10, wherein the microcapsules have an average diameter of not more than 30 μm , an (average diameter/wall thickness) value of not less than 30, but not more than 200, and an (average diameter x wall thickness) value of not more than 20 μm^2 .

12. A pesticidal composition according to Claim 1, 10 or 11, wherein the microcapsules have a proportion (A) of aromatic ring structure in microcapsule wall material being not more than 40% by weight and a ratio of (A) to (average diameter/wall thickness) being not less than 0, but not more than 0.8.

13. A method for controlling wood-injuring insects, which comprises applying an effective amount of a pesticidal composition of Claim 10, 11 or 12 to wood-injuring insects, habitat or paths of wood-injuring insects or wood.

14. A pesticidal composition according to any one of Claims 2 to 9, wherein the organophosphorus compound is chlorpyrifos.

ABSTRACT OF THE DISCLOSURE
PESTICIDAL COMPOSITION

A pesticidal composition comprising micro-capsules, each microencapsulating an organophosphorus compound having a melting point of at least 15°C and at least one solvent selected from the group consisting of:

- (i) an ester,
- (ii) a ketone,
- (iii) an indane substituted with at least one lower alkyl group,
- (iv) a naphthalene substituted with at least one lower alkyl group, and
- (v) a phenylxylylethane,

has a good storage stability.

organophosphorus compound having a melting point of at least 15°C, and at least one solvent selected from the group consisting of:

- (i) an ester,
- (ii) a ketone,
- (iii) an indane substituted with at least one lower alkyl group,
- (iv) a naphthalene substituted with at least one lower alkyl group, and
- (v) a phenylxylylethane,

wherein the amount of the solvent is 0.1 to 10 parts by weight per one part by weight of the organophosphorus compound.

The pesticidal composition according to the present invention will be hereinafter referred to as "the present composition".

DETAILED DESCRIPTION OF THE INVENTION

The organophosphorus compound for use in the present invention has a melting point of at least 15°C, and includes, for example, the following effective compounds for insecticides, bactericides, fungicides and herbicides:

- (1) O,O-diethyl O-3,5,6-trichloro-2-pyridyl phosphorothioate
(chlorpyrifos)
- (2) O,O-dimethyl O-3,5,6-trichloro-2-pyridyl phosphorothioate
(chlorpyrifos-methyl)
- (3) O,O-diethyl O-quinoxalin-2-yl phosphorothioate

REPLACEMENT SHEET

03 JUL 1998



WHAT IS CLAIMED IS:

1. A pesticidal composition, which comprises microcapsules, each microencapsulating an organophosphorus compound having a melting point of at least 15°C and at least one solvent selected from the group consisting of:
- (i) an ester,
 - (ii) a ketone,
 - (iii) an indane substituted with at least one lower alkyl group,
 - (iv) a naphthalene substituted with at least one lower alkyl group, and
 - (v) a phenylxylylethane,
- wherein the amount of the solvent is 0.1 to 10 parts by weight per one part by weight of the organophosphorus compound.
2. A pesticidal composition according to Claim 1, wherein the solvent is (i) an ester.
3. A pesticidal composition according to Claim 2, wherein the (i) ester is a non-aromatic ester having at least 8 carbon atoms in total.
4. A pesticidal composition according to Claim 2, wherein the (i) ester is an aromatic ester.
5. A pesticidal composition according to Claim 4, wherein the (i) ester is a phthalic acid ester.
6. A pesticidal composition according to Claim 1, wherein the solvent is a (ii) ketone.
7. A pesticidal composition according to Claim 1, wherein the solvent is a (iii) indane substituted with at least one lower alkyl group.
8. A pesticidal composition according to Claim 1,

REPLACEMENT SHEET

03 JUL 1998