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PESTICIDAL COMPOSITION AND METHOD FOR
CONTROLLING PESTS USING THE SAME

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 compositions comprising
microcapsules, each microencapsulating an
organophosphorus compound, for example, chlorpyrifos,
10 have been so far used to control wood-injuring insects.
However, when pesticides comprising microcapsules, each
microencapsulating an organophosphorus compound which is
solid at room temperature, for example, chlorpyrifos,
etc. are used outdoors, color change is sometimes
15 observable in pesticide-sprayed areas, such as woods,
etc. and improvement has been desired.

SUMMARY OF THE INVENTION

As a result of extensive studies, the present
inventors found a novel pesticidal composition free from
20 the color change problem.

According to the present invention, there is
provided a pesticidal composition comprising
microcapsules, each microencapsulating an
organophosphorus compound having a melting point of at

least 15°C, 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) of the microcapsules being not less than 0, 5 but not more than 0.8 (the present pesticidal composition will be hereinafter referred to as "the present composition").

DETAILED DESCRIPTION OF THE INVENTION

The organophosphorus compound for use in the 10 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:

- 15 (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)
- 20 (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)
- 25 (5) O,O-dimethyl S-(N-methylcarbamoylmethyl) dithiophosphate

- (dimethoate)
- (6) S- α -ethoxycarbonylbenzyl O,O-dimethyl
phosphorodithioate
- (PAP)
- 5 (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-
thiadiazol-3-ylmethyl O,O-dimethyl phosphorodithioate
- 10 (DMTP)
- (9) 2-chloro-1-(2,4-dichlorophenyl) vinyl
dimethyl phosphate
- (dimethylvinphos)
- (10) dimethyl 2,2,2-trichloro-1-
15 hydroxyethylphosphonate
- (trichlorfon)
- (11) O-ethyl O-4-nitrophenyl
phenylphosphonothioate
- (EPN)
- 20 (12) S-benzyl O,O-diisopropyl
phosphorothioate
- (IBP)
- (13) O-2,6-dichloro-p-tolyl O,O-dimethyl
phosphorothioate
- 25 (tolclofos-methyl)
- (14) O-methyl O-2-nitro-p-tolyl
isopropylphosphoroamidothioate
- (amiprofos-methyl)

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
5 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
10 wall material includes, for example, polyurethane, polyurea, polyamide, polyester, polycarbonate, polysulfonate, polysulfonamide, urea-formalin condensation products, melamine-urea condensation products, phenol-formalin condensation products, acrylic
15 acid ester polymer, methacrylic acid ester polymer, vinyl acetate polymer, styrene polymer, divinylbenzene polymer, ethylene-dimethacrylate polymer, etc.

Polyurethane microcapsule walls can be formed, that is, microcapsulation can be attained, for example,
20 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
25 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.

5 In the present invention, it is necessary that a proportion (A) of aromatic ring structure in the microcapsule wall material is not more than 40% by weight and a ratio of (A) to (average diameter/wall thickness) of the microcapsules is not less than 0, but not more
10 than 0.8, that is,

(A) ≤ 40 wt.%, and

$$0 \leq \frac{(A)}{\text{Average diameter/wall thickness of microcapsules}} \leq 0.8$$

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For example, the value A can be lowered, for example, by keeping a proportion of a microcapsule wall forming material having the aromatic ring structure such as phenylene diisocyanate or phenylenediamine lower when
20 polyurea microcapsule walls, etc. are formed by the above-mentioned interfacial polymerization process, and consequently the above-mentioned ratio can be satisfied. Needless to say, the value A can be made zero by using a microcapsule wall forming material having no aromatic
25 ring structure at all, and consequently the above-mentioned ratio can be satisfied.

In view of the foregoing requirements for the microcapsule wall forming materials, preferable 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
5 molecules of hexamethylene diisocyanate, adducts of
tolylene diisocyanate and trimethylolpropane,
isocyanurate condensation products of tolylene
diisocyanate, isocyanurate condensation products of
hexamethylene diisocyanate, isocyanurate condensation
10 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
15 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
20 formation of polyurethane microcapsule walls or polyester
microcapsule walls includes, for example, ethylene
glycol, propylene glycol, butanediol, hexanediol,
diethylene glycol, triethylene glycol, dipropylene
glycol, polyethylene glycol, polypropylene glycol,
25 glycerine, resorcin, hydroquinone, trimethylol propane,
1,2,6-hexanetriol, etc.

Polyvalent amine compounds for use in the
formation of polyurea microcapsule walls or polyamide

microcapsule walls includes, for example,
ethylenediamine, buthanediamine, hexamethylenediamine,
phenylenediamine, toluylenediamine, diethylenetriamine,
triethylenetetramine, tetraethylenepentamine,
5 pentaetnylenehexamine, 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,
10 sebacoyl dichloride, adipoyl dichloride, azelaoyl
dichloride, terephtaloyl chloride, trimesoyl chloride,
pimeloyl dichloride, dodecanedioyl dichloride, citroyl
dichloride, etc.

According to one embodiment of the present
15 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.

20 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
25 and storage stability of compositions to prepare
microcapsules so as to make the average microcapsule
diameter not more than 30 μm , a ratio (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 (produced by Coulter Electronics Inc.).

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

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

When the present composition comprising microcapsules, each microencapsulating chlorpyrifos, is used to control termites, an effective dose of the present composition is applied directly to termites, or to habitat of or paths of termites. An application dose of the composition is about 5 to about 50 g/m^2 in case of spraying onto soil or concrete surfaces under house floors, about 50 to about 5000 g/m^3 in case of soil barrier treatment by mixing with soil, or about 1 to about 5 g/m^2 in case of wood treatment by spraying to wood surfaces, 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
5 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 amount of about 10 to about 1,000 g/m³ of plywood in terms of the weight of chlorpyrifos, followed
10 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 alkaline phenol resin adhesive, etc.

15 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,
20 centipedes, sowbugs, pill bugs, etc. In this case, the present composition can be sprayed directly to the nuisance injurious insects or habitat or paths of nuisance injurious insects in an amount of about 1 to about 50 g/m² in terms of the weight of chlorpyrifos.

25 The present composition can be formulated into 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
5 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,
10 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
15 magnesium silicate, smectite, bentonite, hectorite, silica, etc.), alumina sol, etc. The amount of the thickening agent in the present composition is usually 0 to 10% by weight.

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

The preservative includes, for example, those which are used in the ordinary pest-controlling agent
25 preparations 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.

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 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 present composition, etc., and in case of suspension concentrate, it is usually 0.5 to 50% by weight.

EMBODIMENTS OF THE INVENTION

The present invention will be described in detail below, referring to Production Example, Comparative Example and Test Examples.

Production Example 1

1.41 parts by weight of Sumidur L-75 (trademark of adduct of tolylene diisocyanate and trimethylolpropane, made by Sumitomo-Bayer Urethane Co., Japan; molecular weight: 656; 3 benzene rings are contained as aromatic rings in the molecule) and 20 parts by weight of chlorpyrifos were mixed together with heating to obtain a homogeneous mixture. Then, the resulting mixture was immediately 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 (trademark of a homogenizer made by Tokushukika Kogyo Co., Japan) to obtain a dispersion of microfine droplets.

5 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, 228.59 parts by weight of an aqueous solution containing 2 parts by weight of xanthane

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

It was found that the resulting microcapsules

15 had an average diameter of 13.9 μm , a wall thickness of 0.15 μm , an (average diameter/wall thickness) value of 93, and an (average diameter x wall thickness) value of 2.1 μm^2 , and furthermore that the proportion (A) of aromatic ring structure in the microcapsule wall material

20 was 28.8% by weight and the ratio of (A) to (average diameter/wall thickness) was 0.31.

Production example of a comparative composition for Test Example, which follows, is given below as Comparative Example.

25 Comparative Example 1

0.95 part by weight of Millionate MR-400 (trademark of polyethylene polyphenyl polyisocyanate made

by Nihon Polyurethane Kogyo K.K., Japan; one benzene ring is contained as an aromatic ring per methylene phenyl isocyanate unit in the molecule) and 20 parts by weight of chlorpyrifos were mixed together with heating to
5 obtain a homogeneous mixture, and then the mixture was immediately 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 T.K. autohomomixer to obtain a dispersion
10 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, 229.05 parts by weight of an aqueous solution
15 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 composition containing 5% by weight of encapsulated chlorpyrifos.

It was found that the resulting microcapsules
20 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 $\times$ wall thickness) value of 0.83 μm^2 , and further that the proportion (A) of aromatic ring structure in the microcapsule wall material was 45.6% by
25 weight and the ratio of (A) to (average diameter/wall thickness) was 0.55.

Test Example 1

0.8 g each of the compositions, obtained in Production Example 1 and Comparative Example 1 were applied to filter papers, respectively, air-dried, and exposed to sun light for 3 days. No color change was
5 observed in case of the present composition of Production Example 1, whereas color change to yellow was clearly observed in case of the comparative composition of Comparative Example 1.

Test Example 2

10 A sufficient amount (about 6 ml) of an aqueous 0.25 wt.% dilute solution of the present composition of Production Example 1 was directly sprayed to ten workers of Formosan subterranean termites from a 60 cm-distant position and a 100% mortality was observed one day
15 thereafter.

The present composition causes less color change to applied area even if applied outdoors.



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, 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) of the microcapsules being not less than 0, but not more than 0.8.
2. A pesticidal composition according to Claim 1, wherein the organophosphorus compound is chlorpyrifos.
3. A pesticidal composition according to Claim 2, wherein the composition is used to control wood-injuring insects.
4. A pesticidal composition according to Claim 3, 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 $\times$ wall thickness) value of not more than 20 μm^2 .
5. A method for controlling wood-injuring insects, which comprises applying an effective amount of the composition of Claim 2 or 4 to wood-injuring insects, or habitat or paths of wood-injuring insects.
6. A method for controlling termites, which comprises applying an effective amount of the composition of Claim 2 or 4 to termites or habitat or paths of termites.



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7. A method for controlling nuisance injurious insects, which comprises applying an effective amount of the composition of Claim 2 to nuisance injurious insects or habitat or paths of nuisance injurious insects.