FORM 1

COMMONWEALTH OF AUSTRALIA

PATENTS ACT 1952

APPLICATION FOR A STANDARD PATENT

610009

We,

SUMITOMO CHEMICAL COMPANY, LIMITED

of 5-33, KITAHAMA-4-CHOME
CHUO-KU
OSAKA
JAPAN

hereby apply for the grant of a standard patent for an invention entitled:

~~AN INSECTICIDAL~~ TRANSPARENT EMULSION OF PYRETHROIDAL INSECTICIDE WITH A SURFACE ACTIVE AGENT AND POLAR SOLVENT

which is described in the accompanying complete specification

Details of basic application(s):

Number of basic application	Name of Convention country in which basic application was filed	Date of basic application
63-196622	JP	05 AUG 88

Our address for service is care of GRIFFITH HACK & CO., Patent Attorneys, 601 St. Kilda Road, Melbourne 3004, Victoria, Australia.

DATED this 1st day of August 1989

SUMITOMO CHEMICAL COMPANY, LIMITED

GRIFFITH HACK & CO.

TO: The Commissioner of Patents.

APPLICATION ACCEPTED AND AMENDMENTS ALLOWED 15.2.91

M 011228 030889

AUSTRALIAN PATENT OFFICE

AUSTRALIA DMO:AS P11270 **B**

PATENTS ACT 1952

APPLICATION BY ASSIGNEE OF INVENTOR

DECLARATION IN SUPPORT OF AN APPLICATION FOR A PATENT no. 39253/89

NAME OF APPLICANT

In support of an application made by:

SUMITOMO CHEMICAL COMPANY, LIMITED

TITLE

for a patent for an invention entitled:

AN INSECTICIDAL TRANSPARENT EMULSION

FULL NAME AND ADDRESS OF SIGNATORY

I, Yoshihiko NISHIZAWA

of c/o Sumitomo Chemical Company, Limited,

5-33, Kitahama-4-chome, Chuo-ku, Osaka, Japan

do solemnly and sincerely declare as follows:

1. I am authorised by the above mentioned applicant for the patent to make this declaration on its behalf.

FULL NAME AND ADDRESS OF INVENTOR(S)

2. The name and address of each actual inventor of the invention is as follows:

Tadahiro MATSUNAGA, 1-7, Semiyama, Fukiaicho, Chuo-ku, Kobe-shi, Japan

Kazunobu DOHARA, 10-3-331, Sonehigashinocho-2-chome, Toyonaka-shi, Japan

SEE NOTES OVER

3. The facts upon which the applicant is entitled to make this application are as follows:

The applicant is the assignee of the invention from the inventors.

DELETE PARAGRAPHS 3 AND 4 FOR NON-CONVENTION APPLICATION

4. The basic application(s) as defined by Section 141 of the Act was (were) made as follows:

Country Japan on 5th August 1988

in the name(s) SUMITOMO CHEMICAL COMPANY, LIMITED

and in on

in the name(s)

PLACE AND DATE OF SIGNING

5. The basic application(s) referred to in the preceding paragraph was (were) the first application(s) made in a Convention country in respect of the invention the subject of this application.

Declared at Osaka, Japan

this 20th day of October 1989

Signed Y. Nishizawa

Position YOSHIHIKO NISHIZAWA REPRESENTATIVE DIRECTOR

GRIFFITH HACK & CO

PATENT AND TRADE MARK ATTORNEYS

MELBOURNE · SYDNEY · PERTH

(12) PATENT ABRIDGMENT (11) Document No. AU-B-39253/89
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(54) Title
INSECTICIDAL EMULSION CONTAINING PYRETHRIN(S)

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(57) Claim

1. An insecticidal transparent emulsion comprising

(A) at least one pyrethroidal insecticide selected from the group consisting of

3-phenoxybenzyl chrysanthemate,

3-allyl-2-methyl-4-oxocyclopent-2-enyl chrysanthemate,

3-allyl-2-methyl-4-oxocyclopent-2-enyl 2,2,3,3-tetramethylcyclopropanecarboxylate,

2-methyl-4-oxo-3-(2-propynyl)cyclopent-2-enyl chrysanthemate and

α-cyano-3-phenoxybenzyl chrysanthemate, or

a mixture of at least one pyrethroidal insecticide selected from the group consisting of

3-phenoxybenzyl chrysanthemate,

3-allyl-2-methyl-4-oxocyclopent-2-enyl

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chrysanthemate,

3-allyl-2-methyl-4-oxocyclopent-2-enyl 2,2,3,3-tetramethylcyclopropanecarboxylate,

2-methyl-4-oxo-3-(2-propynyl)cyclopent-2-enyl chrysanthemate and

α-cyano-3-phenoxybenzyl chrysanthemate

and at least one pyrethroidal insecticide selected from the group consisting of

α-cyano-3-phenoxybenzyl 2,2,3,3-tetramethyl-cyclopropanecarboxylate,

3,4,5,6-tetrahydrophthalimidemethyl chrysanthemate,

3-phenoxybenzyl 3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropanecarboxylate and

1-ethynyl-2-methyl-2-pentenyl chrysanthemate, as an active ingredient,

(B) a polar solvent-containing mixed surface active agent containing calcium dodecylbenzenesulfonate, polyoxyethylene styrenated phenol having an HLB of 12-16 and propylene glycol, and

(C) water, the content of (B) in the emulsion being equal to or more than that of (A) and 6% by weight or less.

AUSTRALIA

610009

PATENTS ACT 1952

Form 10

COMPLETE SPECIFICATION

(ORIGINAL)

FOR OFFICE USE

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Priority:

Related Art:

TO BE COMPLETED BY APPLICANT

Name of Applicant: SUMITOMO CHEMICAL COMPANY, LIMITED

Address of Applicant: 5-33, KITAHAMA-4-CHOME
CHUO-KU
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JAPAN

Actual Inventor:

Address for Service: GRIFFITH HACK & CO.,
601 St. Kilda Road,
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Australia.

Complete Specification for the invention entitled:
TRANSPARENT EMULSION OF PYRETHROIDAL INSECTICIDE WITH A SURFACE ACTIVE AGENT AND POLAR SOLVENT
The following statement is a full description of this invention including the best method of performing it known to me:-

The present invention relates to an insecticidal transparent emulsion prepared by solubilizing a water-insoluble pyrethroidal insecticidal component in water with the aid of a particular mixed surface active agent, said insecticidal transparent emulsion being transparent and homogeneous as well as superior in the solution state and the stability of the active ingredient over a wide temperature range.

Pyrethroidal insecticides, because of their low toxicity to mammals, have been widely used in household spray insecticides.

Since the pyrethroidal insecticides are insoluble in water, they usually are dissolved in an organic solvent such as kerosene or aromatic solvents at first and (1) the resulting solution is directly sprayed with a sprayer, (2) the resulting solution is formed into an aerosol and sprayed with a jet gas, etc., or (3) the resulting solution is formed into an emulsion with an emulsifier, diluted with water and sprayed.

JP-B-58-29761 and JP-B-60-54928 disclosed a domestical application that a water-based solubilized-type insecticide is sprayed for killing insects.

When insecticides are sprayed indoors, however, insecticidal preparations which contain a large quantity of organic solvents and are used as in the

above item (1) are not only unpleasant to sprayers but also undesirable in terms of safety and environmental hygiene. Aerosols which contain an inflammable gas in addition to organic solvents and are used as in the above item (2) are disadvantageous because they are inflammable at the time of use and are difficult to waste after the use. Further, the life of the emulsion as used in the item (3) after diluted with water is only about several hours at longest, so that the emulsion suffers from creaming and separation of oily layer, and cannot maintain the homogeneity over a long period of time. Because of this, the emulsion is usually used immediately after diluted with water. Further, there would be accompanied offensive odors of non-polar solvents such as aromatic solvents, kerosenes, etc. used, adverse effects on the sprayed surface that these solvents exert and change of the sprayed surface into white by the emulsifier.

The conventionally known water-based solubilized-type insecticides, which has been domestically and horticulturally used to control insect pests, come to lose the stable solution state by the environmental temperature change during the storage and produce precipitates. Thus their performances as insecticides are not reliable when the environmental temperature sharply moves up and down. These insecticides, therefore, have a problem in durability of sufficient quality to put them to practical use, and so may not always be said

to be satisfactory.

According to the present invention, there is provided an insecticidal transparent emulsion comprising

(A) at least one pyrethroidal insecticide selected from the group consisting of

3-phenoxybenzyl chrysanthemate,

3-allyl-2-methyl-4-oxocyclopent-2-enyl chrysanthemate,

3-allyl-2-methyl-4-oxocyclopent-2-enyl 2,2,3,3-tetramethylcyclopropanecarboxylate,

2-methyl-4-oxo-3-(2-propynyl)cyclopent-2-enyl chrysanthemate and

α-cyano-3-phenoxybenzyl chrysanthemate, or

a mixture of at least one pyrethroidal insecticide selected from the group consisting of

3-phenoxybenzyl chrysanthemate,

3-allyl-2-methyl-4-oxocyclopent-2-enyl chrysanthemate,

3-allyl-2-methyl-4-oxocyclopent-2-enyl 2,2,3,3-tetramethylcyclopropanecarboxylate,

2-methyl-4-oxo-3-(2-propynyl)cyclopent-2-enyl chrysanthemate and

α-cyano-3-phenoxybenzyl chrysanthemate

and at least one pyrethroidal insecticide selected from the group consisting of

α-cyano-3-phenoxybenzyl 2,2,3,3-tetramethyl-cyclopropanecarboxylate,

3,4,5,6-tetrahydrophthalimidemethyl chrysanthemate,

3-phenoxybenzyl 3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropanecarboxylate and

1-ethynyl-2-methyl-2-pentenyl chrysanthemate, as an active ingredient,

(B) a polar solvent-containing mixed surface active agent containing at least one metal alkylbenzenesulfonate, at least one nonionic surface active agent having an HLB (hydro- and lipophilicity balance) of 10 to 18 and at least one polar solvent, and

(C) water,

the content of (B) in the emulsion being 6% by weight or less, and the content of (A) in the emulsion being equal to or less than the content of (B).

In the present invention, the mixed surface active agent containing a polar solvent refers to those containing at least one metal alkylbenzenesulfonate, at least one nonionic surface active agent having an HLB of 10 to 18 and at least one polar solvent. The metal alkylbenzenesulfonate is not critical. The number of carbon atoms of the alkyl group is not critical either, but it is preferably 8 to 13, more preferably 10 to 12. The metal of the metal salt is not critical, and it includes for example sodium and calcium.

The nonionic surface active agents used are those containing 6 to 40 moles of ethylene oxide, wherein the ethylene oxide is added so that the agent has an HLB of 10 to 18. The HLB is preferably 12 to 16.

Specific examples of the agent are polyoxyethylene styrenated phenol, polyoxyethylene alkylphenyl ether, polyoxyethylene alkyl ether, etc. Specific examples of the polar solvent are propylene glycol, butyl glycol, butyl diglycol, isopropyl alcohol, ethanol, methoxybutanol, etc.

The mixing weight ratio between the metal alkylbenzenesulfonate, the nonionic surface active agent and the polar solvent is not critical, and it is preferably 25-40 : 40-55 : 5-30, more preferably 28-37 : 43-50 : 15-23. The mixing weight ratio of the metal alkylbenzenesulfonate to the nonionic surface active agent is preferably 1:2 to 1:1.

The insecticidal transparent emulsion of the present invention contains the foregoing polar solvent-containing mixed surface active agent in a weight equal to or more than that of the pyrethroidal insecticide which is an active ingredient, and besides in an amount of 6% by weight or less of the emulsion. Preferably, the emulsion contains the polar solvent-containing mixed surface active agent in a weight 3 to 6 times that of the pyrethroidal insecticide.

Specific examples of the pyrethroidal insecticide used in the present invention are 3-phenoxybenzyl chrysanthemate (phenothrin), 3-allyl-2-methyl-4-oxocyclopent-2-enyl chrysanthemate (allethrin), 3-allyl-2-methyl-4-oxocyclopent-2-enyl 2,2,3,3-tetramethylcyclopropanecarboxylate (terallethrin), 2-methyl-

4-oxo-3-(2-propynyl)cyclopent-2-enyl chrysanthemate (prallethrin), α-cyano-3-phenoxybenzyl chrysanthemate (cyphenothrin) α-cyano-3-phenoxybenzyl 2,2,3,3-tetramethylcyclopropanecarboxylate (fenpropathrin), 3,4,5,6-tetrahydrophthalimidemethyl chrysanthemate (tetramethrin), 3-phenoxybenzyl 3-(2,2-dichlorovinyl)-2,2-dimethyl-cyclopropanecarboxylate (permethrin), 1-ethynyl-2-methyl-2-pentenyl chrysanthemate (empenthrin) and their isomers such as geometrical isomers and optical isomers.

The insecticidal transparent emulsion of the present invention may contain, if necessary, a synergist such as piperonyl butoxide (hereinafter referred to as PBO), octachlorodipropyl ether, etc., whereby the activity is expected to be strengthened.

Further, the stability of the active ingredient can be maintained by optionally adding an antioxidant [e.g. 2,6-di-tert-butyl-4-methylphenol (BHT), 2,2'-methylenebis(4-methyl-6-tert-butylphenol), n-octadecyl 3-(3',5'-di-tert-butyl-4'-hydroxyphenyl)-propionate], salts (e.g. sodium benzoate, ammonium benzoate), etc. Further, the insecticidal transparent emulsion of the present invention can prevent water-mold by adding disinfectant such as Proxel® GXL (manufactured by ICI Americans Inc.). Moreover it can also be used for horticultural purposes together with fungicides.

For producing the insecticidal transparent emulsion of the present invention, it is most general to

prepare a concentrate by mixing the pyrethroidal insecticide which is an active ingredient, the polar solvent-containing mixed surface active agent and if necessary, oil-soluble additives (e.g. BHT), and dilute the resulting concentrate with water (Process A).

In an alternative process (Process B), a concentrate is prepared by mixing the pyrethroidal insecticide as an active ingredient, the polar solvent-containing mixed surface active agent, oil-soluble additives (e.g. BHT) which is added as need arises and a substance for increasing the weight of the concentrate, for example a polar solvent, water or a mixture of the both, and then the resulting concentrate is diluted with water.

In Process A, the concentrate has a high viscosity, and when it is weighed, calculation of the amount to be weighed is troublesome. According to Process B, by contrast, the weighing operation can be improved.

For example, when the insecticidal transparent emulsion containing 0.2 % of allethrin and 0.2% of phenothrin is produced, a concentrate is first produced by mixing 5 parts by weight of each of allethrin and phenothrin, 40 parts by weight of the polar solvent-containing mixed surface active agent, 1 part by weight of BHT and any of propylene glycol, a propylene glycol/water (1:1 by weight) mixture and water in a quantity sufficient for making the total weight 100 parts, and

then 4 parts by weight of the concentrate is mixed with 96 parts by weight of water to prepare the desired insecticidal transparent emulsion.

When the insecticidal transparent emulsion of the present invention thus prepared is domestically used, it is effective to fill the emulsion in a small hand sprayer and directly spray onto the body of flying insects (e.g. flies, mosquitoes) and crawling insects (e.g. cockroaches), or apply to the refuge of the crawling insects. The insecticidal transparent emulsion of the present invention is also useful to exterminate bedbugs, fleas, lice, etc.

The concentrate of the insecticidal transparent emulsion obtained by the foregoing Process B can be used without diluted with water for ULV spraying (ultra low volume spraying).

The insecticidal transparent emulsion of the present invention contains the pyrethroidal insecticides, active ingredients, in an amount of preferably 0.02 to 2% by weight, more preferably 0.05 to 1% by weight.

The present invention will be illustrated in more detail with reference to the following examples, but it is not limited to these examples.

Example 1

Insecticidal transparent emulsions having a composition shown in Table 1 were prepared using Hymal

1119, 1141, 1156 or 1159 (a product of Matsumoto Yushi Seiyaku Co., Ltd.; a mixture of calcium dodecylbenzene-sulfonate, polyoxyethylene styrenated phenol having an HLB of 12 to 16 and propylene glycol) as the polar solvent-containing mixed surface active agent.

Table 1

Formulation example No.	Polar solvent-containing mixed surface active agent* (% w/w)	Active ingredient (% w/w)	Additives (% w/w)
1	0.8	Tetramethrin 0.1 d-Phenothrin 0.1	
2	0.8	d-Allethrin 0.1 Permethrin 0.1	
3	0.8	Terallethrin 0.2	
4	0.8	d-Allethrin 0.1 d-Phenothrin 0.1	
5	1.5	d-Allethrin 0.25 d-Phenothrin 0.25	
6	2.0	d-Allethrin 0.25 d-Phenothrin 0.25	
7	3.0	d-Allethrin 0.25 d-Phenothrin 0.25	
8	3.0	d-Allethrin 0.5 d-Phenothrin 0.5	
9	0.8	d-Phenothrin 0.2	BHT 0.01
10	0.8	d-Tetramethrin 0.1 d-Phenothrin 0.1	BHT 0.01

- Cont'd -

Table 1 (Cont'd)

11	0.8	Prallethrin PBO	0.05 0.15	BHT	0.01
12	0.8	d-Allethrin Phenpropathrin	0.1 0.1	BHT	0.01
13	0.8	d-Allethrin d-Phenothrin	0.1 0.1	BHT Propylene glycol	0.01 0.01
14	0.8	d-Allethrin d-Phenothrin	0.1 0.1	Isopropyl alcohol	0.01
15	0.8	d-Allethrin	0.2	Ammonium benzoate	0.7
16	0.8	d-Allethrin d-Cyphenothrin	0.1 0.1	Ammonium benzoate	0.7
17	0.8	d-Allethrin d-Phenothrin	0.1 0.1	BHT Propylene glycol Proxel GXL	0.02 0.98 0.1
18	0.8	d-Cyphenothrin	0.2		
19	0.8	d-Cyphenothrin	0.2	BHT	0.02
20	0.35	Prallethrin	0.05		
21	0.70	Prallethrin	0.1		
22	0.70	Prallethrin	0.1	BHT	0.01
23	0.70	Prallethrin	0.1	Proxel GXL	0.2

(Cont'd)

Table 1 (Cont'd)

24	0.70	Prallethrin	0.05	BHT Proxel GXL	0.01 0.2
25	1.40	Prallethrin	0.2		
26	1.80	Prallethrin PBO	0.1 0.3		

*Used polar solvent containing mixed surface active agent

No. 1 - 17 Hymal 1119
No. 18, 19 Hymal 1141
No. 20 - 25 Hymal 1156
No. 26 Hymal 1159

In Formulation examples 1 to 8, firstly a single and mixed active ingredients were each mixed with Hymal 1119 in a ratio shown in Table 1 while heating to about 40°C with stirring. After the solution phase had become uniform, the concentrates were diluted with water to the respective active ingredient concentrations shown in Table 1 to obtain uniform and transparent insecticidal transparent emulsions.

In Formulation examples 9 to 12, a single active ingredient, alone or mixed with PBO, and mixed active ingredients were each mixed with Hymal 1119 and BHT in a ratio shown in Table 1 while heating to about 40°C with stirring. After the solution phase had become uniform, the concentrates were diluted with water to the respective active ingredient concentrations shown in Table 1 to obtain uniform and transparent insecticidal transparent emulsions.

In Formulation example 13, d-allethrin and d-phenothrin which are an active ingredient, Hymal 1119, BHT and propylene glycol were mixed in a ratio shown in Table 1 while heating to about 40°C with stirring. After the solution phase had become uniform, the concentrate was diluted with water to the active ingredient concentration shown in Table 1 to obtain a uniform and transparent insecticidal transparent emulsion.

In Formulation example 14, a uniform and transparent insecticidal transparent emulsion was obtained in the same manner as in Formulation example 13

except that isopropyl alcohol was used in place of propylene glycol, and BHT was not added.

In Formulation examples 15 and 16, a single active ingredient and a mixture of two active ingredients were each mixed with Hymal 1119 in a ratio shown in Table 1 while heating to about 40°C with stirring. Thus, concentrates having a uniform solution phase were obtained. The concentrates were then diluted with a 0.7% aqueous ammonium benzoate solution to the active ingredient concentrations shown in Table 1 to obtain uniform and transparent insecticidal transparent emulsions.

In Formulation example 17, a mixture of d-allethrin and d-phenothrin which is an active ingredient, Hymal 1119, BHT and Proxel GXL were mixed in a ratio shown in Table 1 while heating to about 40°C with stirring. After the solution phase had become uniform, the concentrate was diluted with water to the active ingredient concentration shown in Table 1 to obtain a uniform and transparent insecticidal transparent emulsion.

In Formulation examples 18 and 19, a single active ingredient d-Cyphenothrin, Hymal 1141 and BHT were mixed in a ratio shown in Table 1 while heating to about 40°C with stirring. After the solution phase had become uniform, the concentrate was diluted with water to the active ingredient concentration shown in Table 1 to obtain a uniform and transparent insecticidal

transparent emulsion.

In Formulation examples 20 to 25, a single active ingredient Prallethrin, Hymal 1156, BHT and Proxel GXL were mixed in a ratio shown in Table 1 while heating to about 40°C with stirring. After the solution phase had become uniform, the concentrate was diluted with water to the active ingredient concentration shown in Table 1 to obtain a uniform and transparent insecticidal transparent emulsion.

In Formulation example 26, a mixture of Prallethin and PBO which is an active ingredient and Hymal 1159 were mixed in a ratio shown in Table 1 while heating to about 40°C with stirring. After the solution phase had become uniform, the concentrate was diluted with water to the active ingredient concentration shown in Table 1 to obtain a uniform and transparent insecticidal transparent emulsion.

Formulation examples in which surface active agents not included in the scope of the present invention were used, are shown in Table 2 as comparative examples.

In Comparative formulation examples A to E, a single or mixed active ingredients and surface active agents not included in the scope of the present invention were mixed in ratios shown in Table 2 while heating to about 40°C with stirring. The resulting concentrates were diluted with water to the active ingredient concentrations shown in Table 2 to obtain comparative preparations.

Table 2

Comparative formulation example	Surface active agent, etc. (% w/w)		Active ingredient (% w/w)	
A	Polyoxyethylene polypropylene glycol monooleate (HLB, 18.5)	0.2	Tetramethrin	0.1
	Polyoxyethylene (10 moles)·styrenated phenol	0.4	d-Resmethrin	0.1
B	Polyoxyethylene(40 moles)·castor oil	1.0	Phenothrin	0.3
	Polyoxyethylene·styrenated phenol (HLB, 14.0)	0.5		
C	Polyoxyethylene(10 moles)·styrenated phenol	0.4	Tetramethrin	0.1
	Polyoxyethylene stearic acid ester (HLB, 15.6)	0.2	d-Phenothrin	0.1
D	Polyoxyethylene polypropylene glycol monooleate (HLB, 18.5)	0.5	Tetramethrin	0.1
			d-Resmethrin	0.1
	Polyoxyethylene(20 moles)·sorbitan monostearate	0.5	d-Allethrin	0.1
E	Polyoxyethylene polypropylene glycol monooleate (HLB, 18.5)	0.2	d-Allethrin	0.2
	Polyoxyethylene(40 moles)·castor oil	0.2		

Example 2

The insecticidal transparent emulsions and comparative emulsions prepared in Example 1 were stored in the following different conditions and observed for the solution state : (1) Two weeks' storage at different temperatures, 10°C, 25°C and 40°C in a constant-temperature vessel or room, and (2) 2 weeks' storage at a temperature of -20°C and then 24 hours' standing at 25°C or shaking subsequent to the standing. Further, after the test emulsions had been stored under a severe condition of 60°C x 2 weeks, the percentages of the residual active ingredient were measured by gas chromatography as follows.

To 1 g of the sample was added 10 or 20 ml of a 0.1%(w/v) acetone solution of an internal standard substance, and the mixture was concentrated under reduced pressure. The residue was then dissolved in 2 ml of acetone. The resulting test solution was quantitatively analyzed by gas chromatography according to the internal standard substance method with an FID detector. The measurement conditions were as follows.

Column: 5% SE-30 (100-200 mesh)

Carrier gas: Nitrogen (flow rate, 50 ml/min)

Ingredient to be analyzed	Internal standard substance	Temperature of column (°C)	Temperature of gasification room (°C)
Tetramethrin (or d-tetramethrin) and d-phenothrin	Triphenylmethane	180°C	230°C
Terallethrin	Same as above	Same as above	Same as above
d-Allethrin and d-phenothrin	Triphenyl phosphate	220°C	270°C
d-Allethrin	Same as above	Same as above	Same as above
d-Allethrin and d-cyphenothrin	Same as above	Same as above	Same as above
d-Cyphenothrin	Same as above	Same as above	Same as above
Prallethrin and PBO	Diphenyl phthalate	200°C	250°C
Prallethrin	Same as above	Same as above	Same as above
d-Allethrin and permethrin	Triphenyl phosphate*	200°C*	270°C*
	Diphenyl phthalate**	215°C**	270°C**

Note: * Conditions at the analysis of d-allethrin.
** Conditions at the analysis of permethrin.

The results are shown in Table 3. The solution state is shown by the following symbols.

○:Transparent

△:Translucent

×:Opaque (white turbid) or formation of precipitates.

Table 3

Test example	Formulation examples shown in Tables 1 and 2	Initial solution state	Solution state after 2 weeks							Percentage of residual active ingredient after 2 weeks at 60°C (%)
			10°C	25°C	40°C	-20°C→25°C		60°C→25°C		
						Standing	Shaking	Standing	Shaking	
1	1	○	○	○	○	○	○	○	○	Tetramethrin 92 d-Cyphenothrin 100
2	2	○	○	○	○	○	○	○	○	d-Allethrin 98 Permethrin 100
3	3	○	○	○	○	○	○	○	○	Terallethrin 103
4	7	○	○	○	○	○	○	○	○	d-Allethrin 97 d-Phenothrin 99
5	8	○	○	○	○	○	○	○	○	d-Allethrin 98 d-Phenothrin 100
6	10	○	○	○	○	○	○	○	○	d-Tetramethrin 96 d-Phenothrin 98
7	11	○	○	○	○	○	○	○	○	Prallethrin 97 PBO 99
8	13	○	○	○	○	○	○	○	○	d-Allethrin 102 d-Phenothrin 101
9	15	○	○	○	○	○	○	○	○	d-Allethrin 97

- Cont'd -

Table 3 (Cont'd)

10	16	O	O	O	O	O	O	O	O	d-Allethrin 97 d-Cyphenothrin 100
11	17	O	O	O	O	O	O	O	O	d-Allethrin 99 d-Phenothrin 99
12	18	O	O	O	O	O	O	O	O	d-Cyphenothrin 100
13	19	O	O	O	O	O	O	O	O	d-Cyphenothrin 100
14	20	O	O	O	O	O	O	O	O	Prallethrin 98
15	21	O	O	O	O	O	O	O	O	Prallethrin 98
16	22	O	O	O	O	O	O	O	O	Prallethrin 99
17	23	O	O	O	O	O	O	O	O	Prallethrin 100
18	24	O	O	O	O	O	O	O	O	Prallethrin 97
19	25	O	O	O	O	O	O	O	O	Prallethrin 98

- Cont'd -

Table 3 (Cont'd)

20	26	○	○	○	○	○	○	○	○	Prallethrin 100 PBO 98
21	A	△	△	×	×	×	△	×	×	
22	B	○	×	△	△	△	△	△	△	
23	C	△	×	×	×	△	△	×	×	
24	D	○	×	△	△	×	×	△	△	
25	E	○	×	△	△	×	×	△	△	

Example 3

Twenty adults per group (sex ratio = 1 : 1) of housefly (Musca domestica) were liberated in a 0.34 m^3 glass test chamber, and a prescribed amount of the insecticidal transparent emulsion prepared according to Formulation example was sprayed onto the adults by means of a trigger sprayer (Canyon CHS-3B; produced by Canyon Co., Ltd.). After spraying, the number of knocked-down insects was counted with the lapse of time, and after 20 minutes, the whole test insects were recovered in a clean cup. After giving water and baits, the cup was transferred to an observation room, and a mortality after 24 hours was examined. The KT_{50} value was calculated according to the Bliss' probit method. This test was repeated three time to five times.

The results are shown in Table 4.

Table 4

Test agent		Amount sprayed	Knock-down ratio (%, min)								KT_{50} (min)	Mortality (after 24 hours) (%)
Active ingredient (% w/w)	Polar solvent-containing mixed surface active agent (% w/w)		0.7	1	1.5	2	3	5	7	10		
d-Tetramethrin d-Phenothrin	0.2 0.2	0.7 g	1	12	30	43	66	84	95	97	2.3	100
d-Tetramethrin d-Phenothrin	0.2 0.2	1.4 g	9	26	50	65	83	95	98	100	1.6	100
Oil agent (control)* Pyrethrin	- -	0.7 ml 1.4 ml	7 17	22 25	40 37	45 43	48 65	65 80	81 93	88 97	2.7 2.0	63 40

* The oil agent as a control was prepared by dissolving natural pyrethrin in a solvent comprising Nisseki fog solvent (kerosene for insecticides; produced by Nippon Sekiyu Kagaku Co., Ltd.) so that the content of the pyrethrin was 0.1 % w/w, as converted to a pure active ingredient.

The insecticidal transparent emulsion of the present invention is stable in a solution state to temperature change which is an important quality, so that it may be said to have solved the long-standing problem.

THE CLAIMS DEFINING THE INVENTION ARE AS FOLLOWS:

1. An insecticidal transparent emulsion comprising

(A) at least one pyrethroidal insecticide selected from the group consisting of

3-phenoxybenzyl chrysanthemate,

3-allyl-2-methyl-4-oxocyclopent-2-enyl chrysanthemate,

3-allyl-2-methyl-4-oxocyclopent-2-enyl 2,2,3,3-tetramethylcyclopropanecarboxylate,

2-methyl-4-oxo-3-(2-propynyl)cyclopent-2-enyl chrysanthemate and

α-cyano-3-phenoxybenzyl chrysanthemate, or

a mixture of at least one pyrethroidal insecticide selected from the group consisting of

3-phenoxybenzyl chrysanthemate,

3-allyl-2-methyl-4-oxocyclopent-2-enyl chrysanthemate,

3-allyl-2-methyl-4-oxocyclopent-2-enyl 2,2,3,3-tetramethylcyclopropanecarboxylate,

2-methyl-4-oxo-3-(2-propynyl)cyclopent-2-enyl chrysanthemate and

α-cyano-3-phenoxybenzyl chrysanthemate

and at least one pyrethroidal insecticide selected from the group consisting of

α-cyano-3-phenoxybenzyl 2,2,3,3-tetramethyl-cyclopropanecarboxylate,

3,4,5,6-tetrahydrophthalimidemethyl chrysanthemate,

3-phenoxybenzyl 3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropanecarboxylate and

1-ethynyl-2-methyl-2-pentenyl chrysanthemate, as an active ingredient,

(B) a polar solvent-containing mixed surface active agent containing calcium dodecylbenzenesulfonate, polyoxyethylene styrenated phenol having an HLB of 12-16 and propylene glycol, and

(C) water, the content of (B) in the emulsion being equal to or more than that of (A) and 6% by weight or less.

2. An insecticidal transparent emulsion according to Claim 1, wherein the active ingredient is at least one pyrethroidal insecticide selected from the group consisting of

3-phenoxybenzyl chrysanthemate,

3-allyl-2-methyl-4-oxocyclopent-2-enyl chrysanthemate,

3-allyl-2-methyl-4-oxocyclopent-2-enyl 2,2,3,3-tetramethylcyclopropanecarboxylate,

2-methyl-4-oxo-3-(2-propynyl)cyclopent-2-enyl chrysanthemate and

α-cyano-3-phenoxybenzyl chrysanthemate.

3. An insecticidal transparent emulsion according to Claim 1, wherein the weight ratio between calcium dodecylbenzenesulfonate, polyoxyethylene styrenated phenol having an HLB of 12-16 and propylene glycol is 25-40 : 40-55 : 5-30.

4. An insecticidal transparent emulsion according to Claim 1, wherein the active ingredient is at least one pyrethroidal insecticide selected from the group consisting of

3-phenoxybenzyl chrysanthemate,

3-allyl-2-methyl-4-oxocyclopent-2-enyl chrysanthemate,

3-allyl-2-methyl-4-oxocyclopent-2-enyl 2,2,3,3-tetramethylcyclopropanecarboxylate,

2-methyl-4-oxo-3-(2-propynyl)cyclopent-2-enyl chrysanthemate and

α-cyano-3-phenoxybenzyl chrysanthemate

wherein the weight ratio between calcium dodecylbenzensulfonate, polyoxyethylene styrenated phenol having an HLB of 12-16 and propylene glycol is 25-40 : 40-55 : 5-30.

DATED THIS 5TH DAY OF FEBRUARY 1991
SUMITOMO CHEMICAL COMPANY LIMITED
By its Patent Attorneys:
GRIFFITH HACK & CO.
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