

METHOD OF CONTROLLING COCKROACHES USING A COMPOSITION

5. The present invention relates to a method for controlling cockroaches which comprises applying to cockroaches or to their habitat a microencapsulated cockroach-controlling composition wherein an insecticide having a 3-phenoxybenzyl group and phenylxylylethane are encapsulated in microcapsules having polyurethane walls which have an average particle diameter of not more than 80 μm (microns), a wall thickness of not more than 0.3 μm and a value of the ration of the average particle diameter to the wall thickness of 100-357.

10. Controlling cockroaches has been mainly carried out by a residual spray method.

15. At present, formulations such as emulsifiable concentrates, solubilized emulsion concentrates and oil solutions which contain, for example, an organophosphorus insecticide or a pyrethroidal insecticide as an active ingredient are used for the residual spray.

20. Residual efficacy and safety are especially desired factors for formulations used as residual sprays. If the residual efficacy is to be enhanced with the conventional formulations such as emulsifiable concentrates, solubilized emulsions concentrates and oil solutions, a high application dosage is required, which may bring about problems of safety. Thus, formulations which are safer and have greater efficacy are 25. increasingly demanded.

30. Under the circumstances, research and development into the so-called microencapsulation technique in which an active ingredient is enclosed within a wall material have become intensive. As microencapsulated

- insecticides, those which contain organophosphorus insecticide as an active ingredient are disclosed in Japanese Patent Kokai No. 62-161706 and those which contain pyrethroidal insecticide as an active ingredient are disclosed in Japanese Patent Kokoku No. 55-38325.
- 5.

In some cases, microencapsulation of insecticides might be effective for improving residual efficacy.

- However, residual efficacy of microencapsulated insecticides varies depending upon the particle diameter of the microcapsule and the wall thickness. Optimum particle diameter and wall thickness should be chosen in order to maintain residual efficacy for a prolonged period of time. The optimum range naturally varies depending on the varieties of insect pest to be controlled and on the insecticides.
- 10.
- 15.

In general, microencapsulation of insecticides having a polyurethane wall is performed by interfacial polymerization using a polyfunctional isocyanate.

- 20.
- The interfacial polymerization method essentially requires a step of homogeneously mixing an active ingredient to be enclosed in capsules with the polyfunctional isocyanate.

- In the case when the active ingredient is a pyrethroidal insecticide, an organic solvent may be added as a so-called cosolvent when the insecticide is mixed with a polyfunctional isocyanate which is sometimes high in viscosity, since this insecticide is generally high in viscosity and sometimes is in crystalline form.
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- Japanese Patent Kokoku No. 55-38325 mentions "For example, as usual organic solvents, there may be selected from hydrocarbons such as xylene, toluene, hexane and heptane, chlorinated hydrocarbons such as

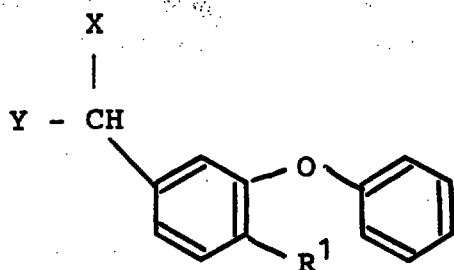
1 carbon tetrachloride and chloroform, ketones such as
2 methylisobutyl ketone, methyl ethyl ketone and
3 cyclohexanone and esters such as diethyl phthalate and
4 n-butyl acetate."

5 However, these known techniques are not
6 necessarily satisfactory when some pyrethroidal
7 insecticides are used as an active ingredient for
8 controlling cockroaches.

9 The inventors have made intensive studies on the
10 optimum ranges of particle diameter and wall thickness
11 of microcapsules and on selection of optimum organic
12 solvents when an insecticide having a 3-phenoxybenzyl
13 group is encapsulated as a core material within a
14 polyurethane wall and then this is used for controlling
15 cockroaches.

16 As a result, it has been found that some
17 insecticides having a 3-phenoxybenzyl group when
18 microencapsulated as a core material within a
19 polyurethane wall using phenylxylylethane as a
20 cosolvent have residual efficacy for a prolonged period
21 of time for controlling cockroaches.

22 The present invention provides a method for
23 controlling cockroaches which comprises applying to
24 cockroaches or to their habitat a microencapsulated
25 composition wherein 1-phenyl-1-xylylethane and at least
26 one of the insecticidal compounds defined below are
27 encapsulated in microcapsules having a polyurethane
28 wall, the microcapsules having an average particle
29 diameter of not more than 80 μm , a wall thickness of
30 not more than 0.3 μm and a value of the ratio of the
31 average particle diameter to the wall thickness of
32 100-357, the said insecticidal compound being selected
33 from compounds represented by the formula:

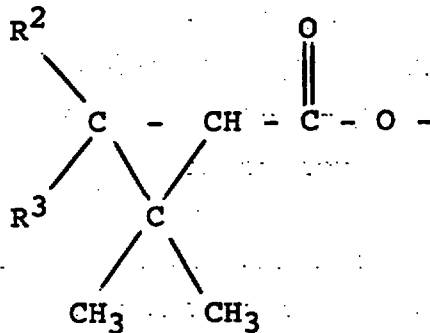


(I)

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wherein R^1 represents a hydrogen atom or a fluorine atom, X represents a hydrogen atom or a cyano group, and Y represents a group represented by the formula:

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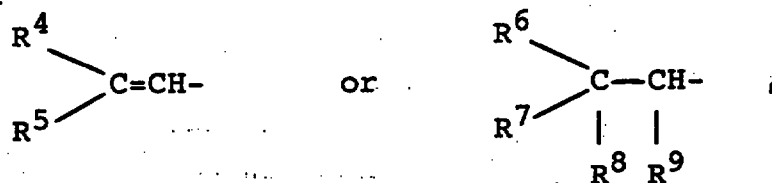


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wherein R^2 represents a hydrogen atom or a methyl group; when R^2 is a hydrogen atom, R^3 represents a group of the formula

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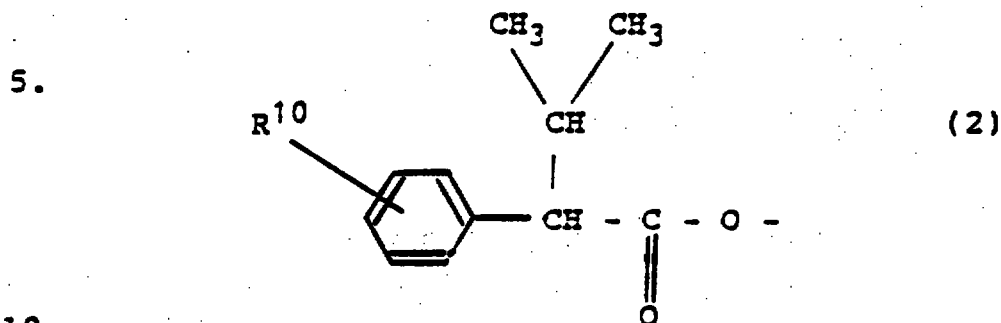


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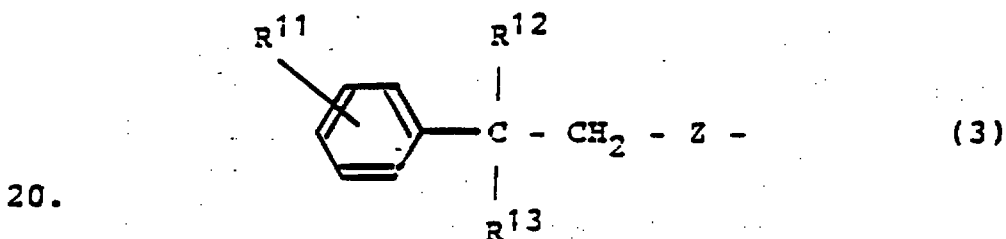
in which R^4 represents a methyl group, a chlorine atom, a bromine atom or a fluorine atom, R^5 represents a methyl group, a trifluoromethyl group, a chlorine atom, a bromine atom or a fluorine atom and R^6 , R^7 , R^8 and R^9 are the same or different and each represents a chlorine atom, a bromine atom or a fluorine atom, and

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when R^2 is a methyl group, R^3 represents a methyl group, a group represented by the formula:



wherein R^{10} represents a chlorine atom, a bromine atom, a fluorine atom, a trifluoromethoxy group, a difluoromethoxy group or a 3,4-methylenedioxy group, or a group represented by the formula:



wherein Z represents an oxygen atom or $-CH_2-$ group, R^{11} represents a C_1-C_5 alkoxyl group, a chlorine atom, a bromine atom or a fluorine atom, R^{12} represents a hydrogen atom or a C_1-C_5 alkyl group (e.g. a C_1-C_2 alkyl group) or a trifluoromethyl group and R^{13} represents a C_1-C_5 alkyl group or a trifluoromethyl group.

30. Cockroaches against which the present compositions may be used include, for example, American cockroach (Periplaneta americana), brown cockroach (Periplaneta brunnea), smokybrown cockroach (Periplaneta fuliginosa)

Nauphaeta cinerea and German cockroach (Blattella germanica).

5. Phenylxylylethane has, we believe, not been used as a cosolvent for preparing homogeneous solution of active ingredient (I) and polyfunctional isocyanate when a polyurethane wall is prepared by an interfacial polymerization method.

10. As cosolvents, there may be used various organic solvents as disclosed in Japanese Patent Kokoku No. 55-38325, but those which have a high flash point and suffer less from an irritating smell are preferred from a viewpoint of safety and user acceptance.

15. Table 1 shows the flash point and degree of smell of various organic solvents disclosed in Japanese Patent Kokoku No. 55-38325 and phenylxylylethane used in the present invention. Clearly, phenylxylylethane is so high in flash point and so weak in smell that it is especially suitable for use as a cosolvent. Furthermore, when phenylxylylethane is compared with diethyl phthalate, the former is preferred to the latter on cost and safety.

TABLE 1

	<u>Name of organic solvent</u>	Flash point	<u>Degree of smell</u>
		<u>(°C)</u>	
	Xylene	25	Strong
25.	Toluene	4	do
	Hexane	-26	do
	Heptane	-4	do
	Carbon tetrachloride	-	do
	Chloroform	-	do
30.	Methylisobutyl ketone	23	do
	Methyl ethyl ketone	-1	do
	Diethyl phthalate	152	Weak

n-Butyl acetate	22	Middle
Phenylxylylethane	152	Weak

5. Surprisingly, the microencapsulated composition of the active ingredient (I) prepared using phenylxylylethane as a cosolvent has residual efficacy for controlling cockroaches which is superior to that of the microencapsulated composition of the active ingredient (I) prepared using other organic solvents
10. such as methyl isobutyl ketone and cyclohexanone or without using any organic solvent.

15. When the microencapsulated composition of the active ingredient (I) having polyurethane wall is used for controlling cockroaches, residual efficacy against cockroaches is able to be retained for a prolonged period of time only by microencapsulating the active ingredient (I) and phenylxylylethane in a microcapsule having a polyurethane wall which has an average particle diameter of not more than 80 μm , a wall thickness of not more than 0.3 μm and a value of the ratio of the average particle diameter to the wall thickness
20. of 100-357.

25. The microencapsulation is performed, for example, by the method in which a hydrophobic solution containing a polyfunctional isocyanate, active ingredient (I) and phenylxylylethane is dispersed in an aqueous solution containing a water-soluble polymer as a dispersing agent in the form of droplets and then a polymerization reaction with a polyhydric alcohol having at least two hydroxy groups is allowed to run. After the encapsulation reaction is over, the resulting capsule suspension as such is diluted with water so as to obtain a desired concentration and, if necessary, a suspension
- 30.

stabilizer is added to obtain a stable slurry formulation.

5. As the polyhydric alcohols having at least two OH groups, mention may be made of, for example, ethylene glycol, propylene glycol, butylene glycol, hexanediol, heptanediol, dipropylene glycol, triethylene glycol, glycerin, resorcinol and hydroquinone. As the polyfunctional isocyanate, mention may be made of, for example, toluene diisocyanate, hexamethylene diisocyanate, adducts of toluene diisocyanate and trimethylolpropane, self-condensates of hexamethylenediisocyanate, SUMIDUR L (Registered Trade Mark) (made by Sumitomo-Bayer Urethane Co. Ltd.) and SUMIDUR N (Registered Trade Mark) (made by Sumitomo-Bayer Urethane Co. Ltd.).

10. The dispersing agents used for dispersing the hydrophobic solution containing the active ingredient (I), phenylxylylethane and polyfunctional isocyanate include, for example, natural polysaccharides such as gum arabic, semi-synthetic polysaccharides such as carboxymethyl cellulose and methyl cellulose, synthetic polymers such as polyvinyl alcohol and fine mineral powders such as magnesium aluminium silicate. They may be used alone or in combinations of two or more. When dispersibility is weak, this may be improved by adding

15. a known surfactant such as given in H. Horiguchi, "Synthetic Surface Active Agent".

20. As suspension stabilizers for capsule slurry, the water-soluble polymers enumerated above as dispersing agents may be used as such, but, if necessary, there may be used, as thickening agents, one or more of

25. natural polysaccharides such as xanthan gum and locust bean gum, semi-synthetic polysaccharides such as carboxymethyl cellulose, synthetic polymers such as

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sodium polyacrylate and fine mineral powders such as magnesium aluminium silicate.

5. The active ingredient (I) used in the present invention includes geometrical isomers and further includes optical isomers resulting from the presence of asymmetric carbon and further includes mixtures thereof.

Typical examples of the active ingredients (I) are as follows.

10. (RS)- α -cyano-3-phenoxybenzyl (RS)-2-(4-chlorophenyl)-3-methylbutyrate (fenvalerate),
 (S)- α -cyano-3-phenoxybenzyl (S)-2-(4-chlorophenyl)-3-methylbutyrate (esfenvalerate),
 (RS)- α -cyano-3-phenoxybenzyl 2,2,3,3-tetramethyl-
 15. cyclopropanecarboxylate (fenpropathrin),
 3-Phenoxybenzyl (1R)-cis, trans-chrysanthemate (d-phenothrin),
 (RS)- α -cyano-3-phenoxybenzyl (1R)-cis,trans-chrysanthemate (cyphenothrin),
 20. 3-Phenoxybenzyl (1RS)-cis,trans-3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropanecarboxylate (permethrin),
 α -Cyano-3-phenoxybenzyl (1R)-cis,trans-3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropanecarboxylate
 25. (cypermethrin),
 α -Cyano-3-phenoxybenzyl (1R)-cis,trans-3-(2,2-dibromovinyl)-2,2-dimethylcyclopropanecarboxylate (deltamethrin),
 2-(4-Ethoxyphenyl)-2-methylpropyl 3-phenoxybenzyl
 30. ether (ethofenprox), and
 (S)- α -cyano-3-phenoxybenzyl (1R)-cis-2,2-dimethyl-3-(1,2,2,2-tetrabromoethyl) cyclopropanecarboxylate (tralomethrin).

If necessary, synergists such as piperonyl butoxide and stabilizers such as BHT (2,6-di-t-butyl-4-methylphenyl) may also be optionally added.

5. When the amount of phenylxylylethane added is too low, development of efficacy is insufficient and when too much is added, the concentration of the active ingredient decreases. Therefore, the amount of phenylxylylethane added is usually 0.2-5 parts by weight per part by weight of active ingredient (I).

10. The average particle diameter of the microcapsules is determined by the type of dispersing agent and the concentration of the dispersing agent used for dispersion and on the degree of mechanical agitation during dispersion. For measurement of average particle
15. diameter, a Coulter (Registered Trade Mark) counter, Model TA-II (available from Nikkaki) may be used, for example.

20. The wall thickness of a microcapsule varies depending on the volume ratio of the core material to the wall material and is obtained from the following approximate equation.

$$\text{Thickness} = \frac{W_w}{W_c} \times \frac{p_c}{p_w} \times \frac{d}{6}$$

25. wherein

d: Average particle diameter of microcapsules
Wc: Weight of core material
Ww: Weight of wall material
30. pw: Density of wall material
pc: Density of core material

The wall thickness in the present invention is calculated using the above equation.

The present composition, before being used, is usually diluted from several times to several hundred

times with water, although it depends on the nominal content of the active ingredient, and then the diluted composition is applied by a conventional sprayer. Dosage which varies depending on the particular active ingredient is usually 10-1000 mg/m² in terms of the active ingredient.

The present invention will be explained in more detail by the following Examples, Comparative Examples and Test Examples.

EXAMPLE 1

Ten gram of "SUMIDUR" L (as indicated herebefore) and 100 g of "HISOL" SAS-296 (Registered Trade Mark) (1-phenyl-1-xylylethane made by Nippon Petrochemicals Co.) were added to 100 g of cyphenothrin and stirred until a uniform solution was obtained. This solution was added to 350 g of an aqueous solution containing 5% by weight of gum arabic as a dispersing agent and stirring was carried out for several minutes by means of a "T.K. autohomomixer" (commercial name, Tokushukika Kogyo Co.) at room temperature until microdrops were formed. The stirrer speed was 8000 rpm. Then, to the dispersed solution was added 10 g of ethylene glycol and the reaction was allowed to proceed with gentle stirring in a constant temperature bath at 60°C for 24 hours to obtain a suspension of the microencapsulated composition.

Water was added to the suspension to make a total weight of 1000 g to obtain a slurry of microcapsules wherein 10% by weight of cyphenothrin is encapsulated (Present composition (1)).

The resulting microcapsule had an average particle diameter of 20 μ m, a wall thickness of 0.11 μ m and a ratio of average particle diameter/wall thickness of

182.

EXAMPLE 2

5. Example 1 was repeated except that the amount of "SUMIDUR" L (as indicated hereinbefore) was changed to 6 g, thereby obtaining a slurry of microcapsules wherein 10% by weight of cyphenothrin is encapsulated (present composition (2)).

10. The resulting microcapsules had an average particle size of 19 μm , a wall thickness of 0.06 μm and a ratio of average particle diameter/wall thickness of 317.

EXAMPLE 3

15. Example 1 was repeated except that the amount of "SUMIDUR" L (as indicated before) was changed to 8 g and the speed of the T.K. autohomomixer (as indicated above) was 6500 rpm, thereby to obtain a slurry of microcapsules wherein 10% by weight of cyphenothrin is encapsulated (present composition (3)).

20. The resulting microcapsules had an average particle diameter of 30 μm , a wall thickness of 0.14 μm and a ratio of average particle diameter/wall thickness of 214.

EXAMPLE 4

25. Example 1 was repeated except that the amount of "SUMIDUR" L (as indicated before) was changed to 5 g and the speed of the T.K. homomixer (as indicated before) was changed to 4700 rpm, thereby to obtain a slurry of microcapsules wherein 10% by weight of cyphenothrin is encapsulated (present composition (4)).

30. The resulting microcapsules had an average particle diameter of 50 μm , a wall thickness of 0.14 μm and a ratio of average particle diameter/wall thickness of 357.

EXAMPLE 5

Example 4 was repeated except that the speed of the T.K. homomixer (indicated above) was changed to 4000 rpm, thereby to obtain a slurry of microcapsules wherein 10% by weight of cyphenothrin is encapsulated (present composition (5)).

The resulting microcapsules had an average particle diameter of 70 μm , a wall thickness of 0.20 μm and a ratio of average particle diameter/wall thickness of 350.

EXAMPLE 6

Eight gram of "SUMIDUR" L (as indicated before) and 100 g of "HISOL" SAS-296 (as indicated before) were added to 100 g of fenvalerate and stirred until a uniform solution was obtained. This solution was added to 350 g of an aqueous solution containing 5% by weight of gum arabic as a dispersing agent, followed by stirring for several minutes with a T.K. homomixer (as indicated before) at a speed of 4700 rpm at room temperature until microdrops were formed. Then, to the dispersed solution was added 10 g of ethylene glycol and the reaction was allowed to proceed with gentle stirring in a constant temperature bath at 60°C for 24 hours to obtain a suspension of microencapsulated composition.

Water was added to the suspension to make a total weight of 1000 g to obtain a slurry of microcapsules wherein 10% by weight of fenvalerate is encapsulated (present composition (6)).

The resulting microcapsules had an average particle diameter of 50 μm , a wall thickness of 0.22 μm and a ratio of average particle diameter/wall thickness of 227.

EXAMPLE 7

5. Seven gram of "SUMIDUR" L (as indicated before) and 20 g of "HISOL" SAS-296 (as indicated before) were added to 100 g of d-phenothrin and stirred until a uniform solution was obtained. This solution was added to 200 g of an aqueous solution containing 5% by weight of gum arabic as a dispersing agent, followed by stirring for several minutes with a T.K. homomixer (as indicated before) at 6400 rpm at room temperature until microdrops were formed. Then, to the dispersed solution was added 12 g of ethylene glycol and the reaction was allowed to proceed with gentle stirring in a constant temperature bath at 70°C for 18 hours to obtain a suspension of microencapsulated composition.

10. To the suspension was added an aqueous solution containing 0.6% by weight of xanthan gum to make a total weight of 500 g to obtain a slurry of microcapsules wherein 20% by weight of d-phenothrin is encapsulated (present composition (7)).

15. The resulting microcapsules had an average particle diameter of 30 μm , a wall thickness of 0.2 μm and a ratio of average particle diameter/wall thickness of 150.

EXAMPLE 8

25. Twelve gram of "SUMIDUR" L (as indicated before) and 50 g of "HISOL" SAS-296 (as indicated before) were added to 100 g of cyphenothrin and stirred until a uniform solution was obtained. This solution was added to 300 g of an aqueous solution containing 5% by weight of gum arabic as a dispersing agent, followed by stirring for several minutes with a T.K. homomixer (as indicated before) at 8500 rpm at room temperature until microdrops were formed. Then, to the dispersed

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solution was added 15 g of ethylene glycol and the reaction was allowed to proceed with gentle stirring in a constant temperature bath at 50°C for 40 hours to obtain a suspension of microencapsulated composition.

5. To the suspension was added an aqueous solution containing 20% by weight of neutralized "AGRISOL" FL-100F (Registered Trade Mark) (Kao Soap Co. Ltd.) to make a total weight of 1000 g to obtain a slurry of microcapsules wherein 10% by weight of cyphenothrin is encapsulated (present composition (8)).

10. The resulting microcapsules had an average particle diameter of 20 μm , a wall thickness of 0.18 μm and a ratio of average particle diameter/wall thickness of 111.

EXAMPLE 9

15. Eight gram of "SUMIDUR" L (as indicated before) and 150 g of "HISOL" SAS-296 (as indicated before) were added to 50 g of fenvalerate and stirred until a uniform solution was obtained. This solution was added to 350 g of an aqueous solution containing 5% by weight of gum arabic as a dispersing agent, followed by stirring for several minutes with a T.K. homomixer (as indicated before) at 6500 rpm at room temperature until microdrops were formed. Then, to the dispersed solution was added 20 g of propylene glycol and the reaction was allowed to proceed with gentle stirring in a constant temperature bath at 60°C for 24 hours to obtain a suspension of microencapsulated composition.

25. To the suspension was added an aqueous solution containing 0.4% by weight of xanthan gum and 0.8% by weight of magnesium aluminium silicate to make a total weight of 1000 g to obtain a slurry of microcapsules wherein 5% by weight of fenvalerate is encapsulated
- 30.

(present composition (9)).

The resulting microcapsules had an average particle diameter of 30 μm , a wall thickness of 0.13 μm and a ratio of average particle diameter/wall thickness of 231.

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EXAMPLE 10

Example 8 was repeated except that 7 g of "SUMIDUR" N (as indicated before) was used in place of 12 g of "SUMIDUR" L (as indicated before) and the stirrer speed was changed to 4700 rpm, thereby to obtain a slurry of microcapsules wherein 10% by weight of cyphenothrin is encapsulated (present composition (10)).

10.

The resulting microcapsules had an average particle diameter of 50 μm , a wall thickness of 0.28 μm and a ratio of average particle diameter/wall thickness of 179.

15.

EXAMPLE 11

Example 8 was repeated except that 4 g of "SUMIDUR" L (as indicated before) and 4 g of "SUMIDUR" T-80 (toluene diisocyanate made by Sumitomo-Bayer Urethane Co. Ltd.) were used in place of 12 g of the "SUMIDUR" L (as indicated before), thereby to obtain a slurry of microcapsules wherein 10% by weight of cyphenothrin is encapsulated (present composition (11)).

20.

25.

The resulting microcapsules had an average particle diameter of 21 μm , a wall thickness of 0.16 μm and a ratio of average particle diameter/wall thickness of 131.

30.

EXAMPLE 12

Ten gram of "SUMIDUR" L (as indicated before) and 150 g of "HISOL" SAS-296 (as indicated before) were

- added to 100 g of permethrin and stirred until a uniform solution was obtained. This solution was added to 400 g of an aqueous solution containing 5% by weight of gum arabic and 3% by weight of ethylene glycol, followed by stirring for several minutes with a T.K. homomixer (as indicated before) at 4700 rpm at room temperature until microdrops were formed. Then, the reaction was allowed to proceed with gentle stirring in a constant temperature bath at 70°C for 18 hours to obtain a suspension of microencapsulated composition.

- To the suspension was added water to make a total weight of 1000 g and this suspension was diluted twice with an aqueous solution containing 0.4% by weight of xanthan gum and 1.0% by weight of magnesium aluminium silicate to obtain slurry of microcapsules wherein 5% by weight of permethrin is encapsulated (present composition (12)).

- The resulting microcapsules had an average particle diameter of 50 μm , a wall thickness of 0.23 μm and a ratio of average particle diameter/wall thickness of 217.

EXAMPLE 13

- Six gram of "SUMIDUR" L (as indicated before) and 80 g of "HISOL" SAS-296 (as indicated before) were added to 100 g of cypermethrin to obtain a uniform solution. This solution was added to 400 g of an aqueous solution containing 10% by weight of polyvinyl alcohol as a dispersing agent, followed by stirring for several minutes with a T.K. homomixer (as indicated before) at 2700 rpm until microdrops were formed. Then, to the dispersed solution was added 8 g of ethylene glycol and the reaction was allowed to proceed with gentle stirring in a constant temperature bath at

60°C for 24 hours to obtain a suspension of microencapsulated composition.

5. Water was added thereto to make a total weight of 1000 g to obtain a slurry of microcapsules wherein 10% by weight of cypermethrin is encapsulated (present composition (13)).

10. The resulting microcapsules had an average particle diameter of 16 μm , a wall thickness of 0.07 μm and a ratio of average particle diameter/wall thickness of 229.

COMPARATIVE EXAMPLE 1

15. Twenty-two gram of "SUMIDUR" L (as indicated before) and 100 g of "HISOL" SAS-296 (as indicated before) were added to 100 g of cyfenothrin to make a uniform solution. This solution was added to 400 g of an aqueous solution containing 10% by weight of polyvinyl alcohol as a dispersing agent, followed by stirring for several minutes with a T.K. homomixer (as indicated before) at 3500 rpm until microdrops were
20. formed. Then, to the dispersed solution was added 20 g of ethylene glycol and the reaction was allowed to proceed with gentle stirring in a constant temperature bath at 60°C for 24 hours to obtain a suspension of microencapsulated composition.

25. Water was added thereto to make a total weight of 1000 g to obtain a slurry of microcapsules wherein 10% by weight of cyphenothrin is encapsulated (comparative composition (1)).

30. The resulting microcapsules had an average particle diameter of 10 μm , a wall thickness of 0.12 μm and a ratio of average particle diameter/wall thickness of 83.

COMPARATIVE EXAMPLE 2

- Example 1 was repeated except that the amount of "SUMIDUR" L (as indicated before) was changed to 3.5 g and the speed of the T.K. homomixer was changed to 4700 rpm, thereby to obtain a slurry of microcapsules wherein 10% by weight of cyphenothrin is encapsulated (comparative composition (2)).
- 5.

- The resulting microcapsules had an average particle diameter of 50 μm , a wall thickness of 0.1 μm and a ratio of average particle diameter/wall thickness of 500.
- 10.

COMPARATIVE EXAMPLE 3

- Six gram of "SUMIDUR" L (as indicated before) was added to 100 g of cyphenothrin to make a uniform solution. This solution was added to 400 g of an aqueous solution containing 10% by weight of polyvinyl alcohol as a dispersing agent, followed by stirring for several minutes with a T.K. homomixer (as indicated before) at 2700 rpm until microdrops were formed. Then, to the dispersed solution was added 6 g of ethylene glycol and reaction was allowed to proceed with gentle stirring in a constant temperature bath at 60°C for 24 hours to obtain a suspension of microencapsulated composition.
- 15.
- 20.

- Water was added to the suspension to make a total weight of 1000 g to obtain a slurry of microcapsules wherein 10% by weight of cyphenothrin is encapsulated (comparative composition (3)).
- 25.

- The resulting microcapsules had an average particle diameter of 15 μm , a wall thickness of 0.11 μm and a ratio of average particle diameter/wall thickness of 136.
- 30.

COMPARATIVE EXAMPLE 4

5. Example 6 was repeated except that methyl isobutyl ketone was used in place of the "HISOL" SAS-296 (as indicated before), thereby to obtain a slurry of microcapsules wherein 10% by weight of fenvalerate is encapsulated (comparative composition (4)).

The resulting microcapsules had an average particle diameter of 50 μm a wall thickness of 0.21 μm and a ratio of average particle diameter/wall thickness of 238.

10. COMPARATIVE EXAMPLE 5

15. Example 6 was repeated except that acetophenone was used in place of the "HISOL" SAS-296 (as indicated before), thereby to obtain a slurry of microcapsules wherein 10% by weight of fenvalerate is encapsulated (comparative composition (5)).

The resulting microcapsules had an average particle diameter of 50 μm , a wall thickness of 0.23 μm and a ratio of average particle diameter/wall thickness of 217.

20. COMPARATIVE EXAMPLE 6

25. Example 6 was repeated except that cyclohexanone was used in place of the "HISOL" SAS-296 (as indicated before), thereby to obtain a slurry of microcapsules wherein 10% by weight of fenvalerate is encapsulated (comparative composition (6)).

The resulting microcapsules had an average particle diameter of 50 μm , a wall thickness of 0.22 μm and a ratio of average particle diameter/wall thickness of 227.

30. COMPARATIVE EXAMPLE 7

Four gram of "SUMIDUR" L (as indicated before) was added to 100 g of heated fenvalerate to make a uniform

- solution. Immediately, this solution was added to 350 g of a heated aqueous solution containing 5% by weight of gum arabic as a dispersing agent, followed by stirring for several minutes with a T.K. homomixer (as indicated before) at 4800 rpm until microdrops were formed. Then, to the dispersed solution was added 6 g of ethylene glycol and the reaction was allowed to proceed with gentle stirring in a constant temperature bath at 60°C for 24 hours to obtain a suspension of microencapsulated composition.

Water was added to the suspension to make a total weight of 1000 g to obtain a slurry of microcapsules wherein 10% by weight of fenvalerate is encapsulated (comparative composition (7)).

- The resulting microcapsules had an average particle diameter of 50 μm , a wall thickness of 0.23 μm and a ratio of average particle diameter/wall thickness of 217.

TEST EXAMPLE 1

- Each of the present composition (6) and comparative compositions (4)-(6) was put in a 100 cc beaker and an olfactory test was conducted on whether or not the compositions smelt of a solvent. Ten subjects were employed, to whom no information was given as to the name of the solvents used in the preparation of microcapsules. The results are shown in Table 2.

TABLE 2 (Test on smell of a solvent)

<u>Test composition</u>	<u>Number of persons (subjects) who sensed smell of a solvent</u>
Present composition (6)	1
Comparative composition (4)	10
Comparative composition (5)	10
Comparative composition (6)	10

- That is, all persons sensed the smell of a solvent for comparative compositions in which methyl isobutyl ketone, acetophenone or cyclohexanone was used as a solvent while only one person sensed the smell of a solvent for the present composition (6) in which phenylxylylethane was used.
- 5.

TEST EXAMPLE 2

- Each of the test compositions enumerated in Table 3 was diluted twenty times with water and each of the diluted composition was uniformly sprayed on a plywood panel 15 cm x 15 cm at a rate of 50 ml/m². After the treated plywood panel was dried for 2 hours, a plastic ring 13 cm in diameter and 5 cm in height (for prevention of escape; inner surface was coated with butter) was placed on the treated panel and a group of ten German cockroaches were confined to contact with the treated panel for 2 hours. The cockroaches were transferred to a plastic cup with water and diet, and mortality was observed after 3 days, this is reported in Table 3 as the week zero mortality.
- 10.
- 15.
- 20.

Furthermore, mortality after lapses of 2, 4 and 8 weeks was determined using the same sprayed surfaces.

- Thus after the weeks zero test the treated panel is then kept in air for two weeks and the above exposure of cockroaches to the panel is repeated i.e. the cockroaches are confined in contact with the treated panel for 2 hours, then removed to a separate feeding cup and fed for three days and mortality observed again. This gives the two weeks mortality value. The same procedure is followed to give the 4 and 8 week values.
- 25.
- 30.

Table 3 Mortality of German cockroach (Repeated 3 times)

Test composition	Average particle diameter (μm)	Wall thickness (μm)	Average particle diameter / wall thickness	Time (week) for treatment with the test composition and mortality (%) after 72 hours			
				0	2	4	8
The present composition (1)	20	0.11	182	100	100	100	100
" (2)	19	0.06	317	100	100	100	90
" (3)	30	0.14	214	100	100	100	94
" (4)	50	0.14	357	100	100	100	83
" (5)	70	0.20	350	100	100	100	80
Comparative composition (1)	10	0.12	83	100	100	70	50
" (2)	50	0.10	500	100	67	47	47
" (3)*	15	0.11	136	100	87	69	52

* No phenylxylylethane was used.

TEST EXAMPLE 3

Each of the test compositions enumerated in Table 4 was diluted forty times with water and each dilution was applied onto a plywood panel 15 cm x 15 cm at a rate of 50 ml/m². After the treated panel was dried for 2 hours, a plastic ring 13 cm in diameter and 5 cm in height (for prevention of escape; inner surface was coated with butter) was placed on the panel and a group of ten German cockroaches were confined to contact with the treated panel for 2 hours. The cockroaches were transferred into a plastic cup with water and diet, and mortality was observed after 3 days.

Furthermore, mortality after treatment for 2, 4 and 8 weeks was obtained using the same applied surfaces, the same procedure being used as described in Test Example 2.

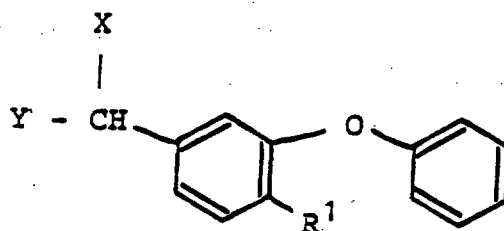
Table 4 Mortality of German cockroach (repeated 3 times)

Test composition	Average particle diameter (μm)	Wall thickness (μm)	Average particle diameter /wall thickness	Solvent used	Time (week) for treatment with composition and mortality (%) after 72 hours			
					0	2	4	8
The present composition (6)	50	0.22	227	Phenylxylyl-ethane	100	100	100	80
Comparative composition (4)	50	0.21	238	Methyl iso-butyl ketone	100	90	67	53
" (5)	50	0.23	217	Acetophenone	100	93	83	53
" (6)	50	0.22	227	Cyclohexanone	100	100	70	67
" (7)	50	0.23	217	-	100	67	50	43

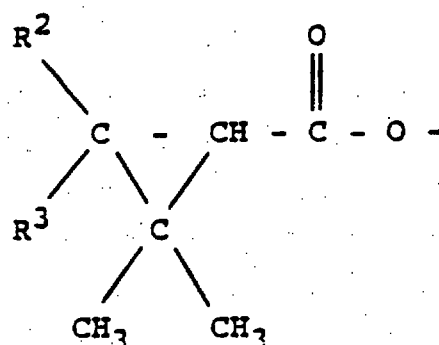


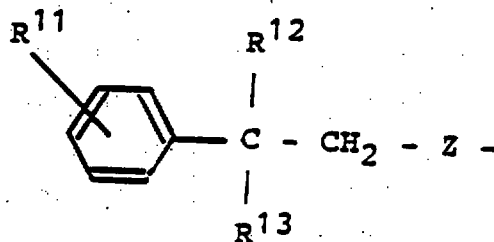
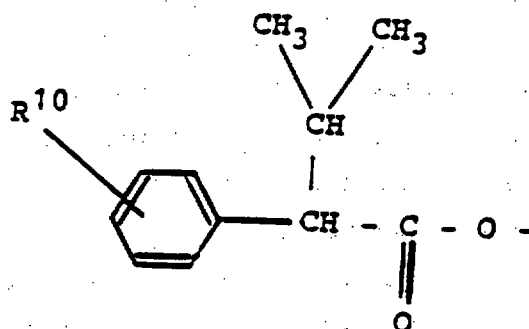
CLAIMS

1. A method for controlling cockroaches which comprises applying to cockroaches or to their habitat a microencapsulated composition wherein 1-phenyl-1-xylylethane and at least one of the insecticidal compounds defined below are encapsulated in microcapsules having a polyurethane wall, the microcapsules having an average particle diameter of not more than 80 um, a wall thickness of not more than 0.3 um and a value of the ratio of the average particle diameter to the wall thickness of 100-357, the said insecticidal compound being selected from compounds represented by the formula:

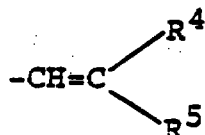


wherein R^1 represents a hydrogen or a fluoride atom, X represents a hydrogen atom or a cyano group, and Y represents a group of the formula

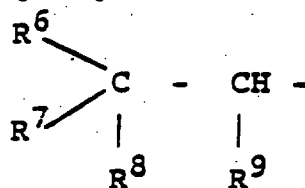




in which, R^2 represents a hydrogen atom or a methyl group; and when R^2 represents a hydrogen atom, R^3 represents a group of the formula



or a group of the formula



in which R^4 represents a methyl group, a chlorine atom, a bromine atom or a fluorine atom, R^5 represents a methyl group, a trifluoromethyl group, a chlorine atom, a bromine atom or a fluorine atom, and R^6 , R^7 , R^8 and R^9 are the same or different and represent a chlorine atom, a bromine atom or a fluorine atom, and when R^2



1 represents a methyl group, R^3 represents a methyl
2 group, R^{10} represents a chlorine atom, a bromine atom,
3 a fluorine atom, a trifluoromethoxy group, a
4 difluoromethoxy group or a 3,4-methylenedioxy group, Z
5 represents an oxygen atom or a $-CH_2-$ group, R^{11}
6 represents a C_1-C_5 alkoxy group, a chlorine atom, a
7 bromine atom or a fluorine atom, R^{12} represents a
8 hydrogen atom or a C_1-C_5 alkyl group, and R^{13}
9 represents a C_1-C_5 alkyl group or a trifluoromethyl
10 group.

11

12 2. A method as claimed in Claim 1, in which the
13 insecticidal compound is (RS)- α -cyano-3-phenoxybenzyl
14 (1R)-cis, trans-chrysanthemate.

15

16 3. A method as claimed in Claim 1, in which the
17 insecticidal compound is (RS)- α -cyano-3-phenoxybenzyl
18 (RS)-2-(4-chlorophenyl)-3-methylbutyrate.

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20 4. A method as claimed in Claim 1, in which the
21 insecticidal compound is 3-phenoxybenzyl (1R)-cis,
22 trans-chrysanthemate.

23

24 5. A method as claimed in Claim 1, in which the
25 insecticidal compound is 3-phenoxybenzyl (1RS)-cis,
26 trans-2-(2,2-dichlorovinyl)-2,2-dimethylcyclopropane-
27 carboxylate.

28

29 6. A method as claimed in anyone of Claims 1 to
30 5 in which the amount of phenylxylylethane is 0.2-5
31 parts by weight per part by weight of the insecticidal
32 compound.

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* C L M S *

1 7. A method as claimed in Claim 1 substantially
2 as specifically described herein with reference to
3 anyone of Examples 1-13.

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