Anti-termite compositions of an organo-phosphorus insecticide and an octachloro-4-oxaheptane

The present invention relates to a termite controlling composition which contains as the active ingredient specific organic phosphorus compounds and 1,1,1,2,6,7,7,7-octachloro-4-oxaheptane.

As the active ingredients for termite controlling compositions use has been made so far mainly of chlorinated hydrocarbon insecticides such as chlordane, heptachlor, dieldrin, aldrin, lindane and chloronaphthalene and inorganic compounds such as arsenic compounds, fluorine compounds and boron compounds. However, the use of these insecticides is being limited or prohibited due to their high and sustained toxicities for vertebrates including human beings.

Therefore, recently, organic phosphate insecticides such as phoxim, chlorpyrifos, fenthion and fenitrothion as well as carbamate insecticides such as BPMC (Baycarb), propoxur (Baygon) and carbaryl have been used either solely or in combination. Baycarb and Baygon are registered Trade Marks.

One of the important properties that antitermite agents should be provided with is the weathering resistance or weatherability. Fundamentally, damage caused by termites is ~~are~~ found to take place in woodwork under the eave exposed to the rain water and under the floor, and kitchen, bathroom, toilet, etc. where water is used. Therefore, the anti-termite agents must retain the enhanced effect in such locations for a prolonged period of time. For this reason, it is required that the antitermite agents be free from

elution and evaporation of active ingredient, or possess the weathering resistance, in moist or humid locations. In addition, such properties are important from the standpoints of safety and hygiene or of prevention of environmental pollution, as well.

In general, organic phosphate insecticides are quick-acting and exhibit high termiticidal property. However, they are readily decomposed by water, heat, ultraviolet rays, etc., and therefore, present the problem in terms of weathering resistance in the practical use.

The present inventors, after intensive research, found that 1,1,1,2,6,7,7,7-octachloro-4-oxaheptane, when used in combination with the specific organic phosphorus compounds, shows exceptionally improved penetrating property toward wood, etc. and also provides the active ingredients having penetrated into wood, etc. with exceptional resistance to elution and evaporation, thus demonstrating excellent termite preventive property over a prolonged period of time, and have come to complete the termites controlling agents being provided with the practically satisfactory weathering resistance.

1,1,1,2,6,7,7,7-Octachloro-4-oxaheptane employed in this invention has been known as the synergist for pyrethroid insecticides, and has been formulated in conjunction with pyrethroids and used in insecticidal preparations for household insect pests intended for use in the control of flies, mosquitos, etc., but has not been known as the active ingredient for termite controlling agents at all.

Although 1,1,1,2,6,7,7,7-octachloro-4-oxaheptane itself shows a limited degree of termiticidal property, its effect is not necessarily adequate, and the above-mentioned weathering resistance of it is far from being practically satisfactory.

The present inventors discovered that 1,1,1,2,6,7,7,7-octachloro-4-oxaheptane, conventionally known as the synergist for pyrethroid insecticides but not at all as the

active ingredient for termite-controlling agents demonstrates an outstandingly high degree of termite preventive property over a prolonged period of time, when used in conjunction with the specific organic phosphorus compounds, and the discovery was followed by intensive investigation with an view to the application to agents for controlling termites, which has culminated in the present invention.

Although many insecticidal organic phosphorus compounds have so far been known, the organic phosphorus compounds which are to be used, in conjunction with 1,1,1,2,6,7,7,7-octachloro-4-oxaheptane, for termite controlling composition according to the present invention are phoxim, chlorpyrifos, fenitrothion, prothiofos and isofenphos. These organic phosphorus compounds have the following chemical names, respectively;

phoxim : O,O-diethyl O-(α-cyanobenzylideneamino) phosphorothioate

chlorpyrifos : O,O-diethyl O-3,5,6-trichloro-2-pyridyl phosphorothioate

fenitrothion : O,O-dimethyl O-(4-nitro-m-tolyl)phosphorothioate

prothiofos : 2,4-dichlorophenyl ethyl s-propylphosphorothiolothionate

isofenphos : O-ethyl O-2-isopropoxycarbonylphenyl isopropylphorsphoramidothioate

Thus, the present invention is directed toward termite controlling composition which comprise at least one of the organic phosphorus compounds consisting of phoxim, chlorpyrifos, fenitrothion, prothiofos and isofenphos and 1,1,1,2,6,7,7,7-octachloro-4-oxaheptane.

The incorporated amounts of the 1,1,1,2,6,7,7,7-octachloro-4-oxaheptane and the organic phosphorus compound which are used in the termite controlling compositions according to the present invention may be arbitrarily selected, depending for example upon whether or not the termite controlling compositions contains as the active ingredients any compounds other than 1,1,1,2,6,7,7,7-octachloro-4-oxaheptane and the organic phosphorus compound,

if the termite controlling composition is intended for use as a preventive agent, eradicating agent or preventive/eradicating agent against termites, whether the object to be treated is wood or soil, and which treatment process is to be adopted out of the coating, spraying, immersion, impregnation, dusting and mixing processes. From a practical point of view, however, the mixing ratio of the organic phosphorus compound to 1,1,1,2,6,7,7,7-octachloro-4-oxaheptane is 1 : 0.05 to 25 on a weight basis, preferably 1 : 0.1 to 10. With reference to the proportions in which these are incorporated in the whole composition, 1,1,1,2,6,7,7,7-octachloro-4-oxaheptane is preferably at 0.01 to 50 weight % and more preferably at 0.2 to 50 weight %, with the organic phosphorus compound being desirably at 0.01 to 50 weight % and more desirably at 0.2 to 50 weight %.

Also, it is more effective to incorporate simultaneously any termiticidal component other than 1,1,1,2,6,7,7,7-octachloro-4-oxaheptane and organic phosphorus compound into the termite controlling compositions according to the present invention. For example, chlorinated hydrocarbon insecticides such as chlordane and chloronaphthalene and carbamate insecticides such as BPMC and propoxur are particularly suitable, but the termiticidal component is not limited to these insecticides.

In general, the living environment of termites is an environment suited for the growth of wood rotting fungi, and the damage by termites often proceeds in parallel with wood rotting. Therefore, it is useful to incorporate a wood-rotting preventive agent into termite controlling agents. In the termite controlling composition according to the present invention, too, it is effective to incorporate wood-rotting preventive agents together with 1,1,1,2,6,7,7,7-octachloro-4-oxaheptane and the organic phosphorus compounds.

Examples of the preventive agents against wood rotting fungi which can be incorporated into the termite controlling composition according to the present invention in conjunction

with 1,1,1,2,6,7,7,7-octachloro-4-oxaheptane and organic phosphorus compounds, include, but are not restricted to;

Pentachlorophenyl laurate, 2,4,6-tribromophenol, 9-chlorophenyl-3'-iodopropargyl formal, diiodomethyl-4-methyl-phenyl sulfone, N,N-dimethyl-N'-dichlorofluoromethylthio-N-phenylsulfamide, bis(tributyltin)oxide (TBTO), tributyltin phthalate (TBTP), aluminum salt of N-nitroso-N-cyclohexyl-hydroxylamine (NCH-AL), N-cyclohexyl-N-methoxy-2,5-dimethyl-furane-3-carboxylic acid amide (general name: furmecyclox) and creosote oil. Such wood rotting preventive agents can be incorporated solely or in combination with not less than two kinds of them, as well. It is particularly preferred to incorporate TBTO, TBTP or NCH-Al in combination with furmecyclox. In the termites controlling composition according to the present invention, also, organic solvent can be employed together with 1,1,1,2,6,7,7,7-octachloro-4-oxaheptane and organic phosphorus compounds, as the case may be. As the organic solvent which is usable in the termite controlling compositions according to the present invention, there may be mentioned those being normally employed as such kind of solvent and diluent, for example, aromatic hydrocarbons, alicyclic hydrocarbons, aliphatic hydrocarbons, alcohols, glycol ethers, ketones and esters, and these solvents are utilized solely or in combination.

In the termite controlling compositions according to the present invention, it is possible to incorporate, in addition to the wood-rotting preventive agents and solvents, antimold agents for wood use and also penetrating agents, fixing agents, surface active agents, coloring matter, etc.

The termite controlling compositions according to the present invention are used, as a wood preservative, in the treatment of wood materials such as plywood, hard board and particle board, and as a soil treatment agent, and so forth.

The termite controlling composition according to the present invention are processed for application into

preparations such as oil-borne preparation, emulsifiable concentrate, granules, powder, paste, wettable powder, foam preparation, paint, etc.

The examples are given in the following to describe the agents for controlling termites according to the present invention as well as their effect.

Test Example 1

Wood pieces were treated with an oil-borne preparation prepared by dissolving 0.2 part by weight of 1,1,1,2,6,7,7,7-octachloro-4-oxaheptane and 0.2 part by weight of phoxim in 99.6 parts by weight of an aromatic hydrocarbon at the application rate of 110 g/m^2, and using the test specimens thus prepared, 150 heads of Coptotermes formosanus Shiraki were bred by the procedure conforming to the general test specified under the Japan Wood Preserving Association (a corporate juridical person) Standard No. 11. The test results are shown in Table 1. The results indicate that the termite controlling composition according to the present invention is superior to 1,1,1,2,6,7,7,7-octachloro-4-oxaheptane or phoxim employed alone.

Test Procedure

1) Test materials

A wood piece to be subjected to testing was made of a sapwood of Japanese red pine, with a number of growth rings of 3 to 5 per 10 mm, and was biaxially straight-grained, with each surface being smoothly and precisely plane-finished.

The wood pieces were dried in a dryer thermostat-controlled at the temperature of 60 ± 2°C for 24 hours.

2) Test specimens

A test specimen was of the cuboid with the dimension of [20(L) ± 0.5] × [10(R) ± 0.5] × [10(T) ± 0.5] mm.

The test specimens were prepared in two kinds, i.e., the treated and non-treated test specimens. The treated test specimens were those having been treated with a sample with or without subsequent exposure to weathering as set forth below.

A number of test specimens was 5 each for the treated and non-treated test specimens.

Preparation of the treated test specimen was in accordance with the following:

Test specimens, after being applied with a sample at a rate of 110 ± 10 g/m^2 by use of a brush and being allowed to stand in a room for not less than 20 days, were divided into two groups each of 5 test specimens to be, and not to be, exposed subsequently to weathering, respectively.

The test specimens having the elapsed time of not less than 20 days were exposed to weathering.

The weathering consisted of repeating the cycle of alternate wetting and vapourizing 10 times.

The wetting procedure: The test specimen, after being immersed in fresh water of room temperature for 30 seconds, was placed in a desiccator containing water in the bottom, and allowed to stand in a room thermostat-controlled at the temperature of 26 ± 2°C for 4 hours.

The vapourizing procedure: The test specimen having the wetting procedure completed was immediately placed in a recycling, hot-air chamber termostat-controlled at the temperature of 40 ± 2°C, and allowed to stand therein for 20 hours.

The treated test specimens with exposure to weathering, test specimens without exposure to weathering and non-treated test specimens were dried at the temperature of 60 ± 2°C until a constant weight is reached. The test specimens having reached constant weight were allowed to stand in a desiccator for about 30 minutes and weighed to precision of 0.01 g (W_1).

3) Breeding container

A breeding container consisted of a cylinder made of plastics having 8 cm of diameter and 6 cm of length, with one end being closed with ca. 5-mm thick, hardened anhydrite, and 10 to 15 pieces of such breeding containers were placed in a covered container having wet cotton spread

in advance to about 2 cm of thickness.

4) Test method

One piece of the treated test specimen or the untreated test specimen, with its straight-grained surface down, was laid horizontally on the anhydrite in the breeding container and 150 termite workers and 15 termite soldiers taken out of the nest at random were placed in it.

The covered container was allowed to stand in a dark place at the temperature of 28 ± 2°C to conduct breeding.

On the day when all the termite workers were killed, the test specimen was taken out of the breeding container, freed carefully of matters adhered on the surface, and dried in a dryer thermostat-controlled at the temperature of 60 ± 2°C until a constant weight is reached. The test specimen having reached constant weight was transferred into a desiccator, allowed to stand for about 30 minutes and weighed to a precision of 0.01 g to determine the weight (W_2).

The number of days required 100% of death rate of termite workers was recorded.

5) Indication of the test results

The rate of reduction in weight for the test specimen was calculated by the following equation, whereby as to the non-treated test specimens, the average rate of reduction in weight was determined for 5 specimens:

Rate of reduction in weight (%) = $(W_1 - W_2)/W_1 \times 100$

Test Example 2

Wood pieces (sapwood of Japanese red pine) measuring 20 mm by 10 mm by 10 mm) were treated with an oil-borne preparation prepared by dissolving 0.2 part by weight of 1,1,1,2,6,7,7,7-octachloro-4-oxaheptane and 0.2 part by weight of chlorpyrifos in 99.6 parts by weight of an aromatic hydrocarbon at the application rate of 110 g/m^2, and using the test speciments thus prepared, 150 heads of Coptotermes formosanus Shiraki were bred by the same procedure as in Test Example 1. The test results are shown in Table 1.

The results indicate that the agent for controlling termites according to the present invention is superior to 1,1,1,2,6,7,7,7-octachloro-4-oxaheptane or chloropyrifos employed alone.

Test Example 3

Wood pieces (sapwood of Japanese red pine) measuring 20 mm by 10 mm by 10 mm were treated with an oil-borne preparation prepared by dissolving 0.2 part by weight of 1,1,1,2,6,7,7,7-octachloro-4-oxaheptane and 0.2 part by weight of fenitrothion in 99.6 parts by weight of an aromatic hydrocarbon at the application rate of 110 g/m^2, and using the test specimens thus prepared, 150 heads of Coptotermes formosanus Shiraki were bred by the same procedure as in Test Example 1. The test results are shown in Table 1. The results indicate that the agent for controlling termites according to the present invention is superior to 1,1,1,2,6,7,7,7-octachloro-4-oxaheptane or fenitrothion employed alone.

Test Example 4

Wood pieces (sapwood of Japanese red pine) measuring 20 mm by 10 mm by 10 mm were treated with an oil-borne preparation prepared by dissolving 0.2 part by weight of 1,1,1,2,6,7,7,7-octachloro-4-oxaheptane and 0.2 part by weight of prothiofos in 99.6 parts by weight of an aromatic hydrocarbon at the application rate of 110 g/m^2, and using the test specimens thus prepared, 150 heads of Coptotermes formosanus Shiraki were bred by the same procedure as Test Example 1. The test results are shown in Table 1. The results indicate that the agent for controlling termites according to the present invention is superior to 1,1,1,2,6,7,7,7-octachloro-4-oxaheptane or prothiofos employed alone.

Test Example 5

Wood pieces (sapwood of Japanese red pine) measuring 20 mm by 10 mm by 10 mm were treated with an oil-borne preparation prepared by dissolving 0.2 part by weight of 1,1,1,2,6,7,7,7-octachloro-4-oxaheptane and 0.2 part by weight of

isofenphos in 99.6 parts by weight of an aromatic hydrocarbon at the application rate of 110 g/m^2, and using the test specimens thus prepared, 150 heads of Coptotermes formosanus Shiraki were bred by the same procedure as in Test Example 1. The test results are shown in Table 1. The results indicate that the agent for controlling termites is superior to 1,1,1,2,6,7,7,7-octachloro-4-oxaheptane and isofenphos employed alone.

Reference Example 1

An oil-borne preparation prepared by dissolving 0.4 part by weight of 1,1,1,2,6,7,7,7-octachloro-4-oxaheptane in 99.6 parts by weight of an aromatic hydrocarbon was treated on wood pieces at the application rate of 110 g/m^2, and using the test specimens thus prepared, 150 heads of Coptotermes formosanus Shiraki were bred by the same procedure as in Test Example 1. The test results are shown in Table 1.

Reference Example 2

An oil-borne preparation prepared by dissolving 0.4 part by weight of phoxim in 99.6 parts by weight of an aromatic hydrocarbon was treated on wood pieces at the application rate of 110 g/m^2, and using the test specimens thus prepared, 150 heads of Coptotermes formosanus Shiraki were bred by the same procedure as in Test Example 1. The test results are shown in Table 1.

Reference Example 3

An oil-borne preparation prepared by dissolving 0.4 part by weight of chlorpyrifos in 99.6 parts by weight of an aromatic hydrocarbon was treated on wood pieces at the application rate of 110 g/m^2, and using the test specimens thus prepared, 150 heads of Coptotermes formosanus Shiraki were bred by the same procedure as in Test Example 1. The test results are shown in Table 1.

Reference Example 4

An oil-borne preparation prepared by dissolving 0.4 part by weight of fenitrothion in 99.6 parts by weight of

an aromatic hydrocarbon was treated on wood pieces at the application rate of 110 g/m^2, and using the test specimens thus prepared, 150 heads of Coptotermes formosanus Shiraki were bred by the same procedure as in Test Example 1. The test results are shown in Table 1.

Reference Example 5

An oil-borne preparation prepared by dissolving 0.4 part by weight of prothiofos in 99.6 parts by weight of an aromatic hydrocarbon was treated on wood pieces at the application rate of 110 g/m^2, and using the test specimens thus prepared, 150 heads of Coptotermes formosanus Shiraki were bred by the same procedure as in Test Example 1. The test results are shown in Table 1.

Reference Example 6

An oil-borne preparation prepared by dissolving 0.4 part by weight of isofenphos in 99.6 parts by weight of an aromatic hydrocarbon was treated on wood pieces at the application rate of 110 g/m^2, and using the test specimens thus prepared, 150 heads of Coptotermes formosanus Shiraki were bred by the same procedure as in Test Example 1. The test results are shown in Table 1.

Table 1. Test results on the termite preventive effectiveness

	Test Example 1		Test Example 2		Test Example 3		Test Example 4		Test Example 5	
1,1,1,2,6,7,7,7-octa-chloro-4-oxaheptane	0.2		0.2		0.2		0.2		0.2	
Phoxim	0.2		-		-		-		-	
Chlorpyrifos	-		0.2		-		-		-	
Fenitrothion	-		-		0.2		-		-	
Prothiofos	-		-		-		0.2		-	
Isofenphos	-		-		-		-		0.2	
Weathering procedure	None	10 times	None	10 times	None	10 times	None	10 times	None	10 times
Number of days required until the death rate reached 100%	1	1	2	2	3	3	2	2	3	3
Rate of reduction in weight for test specimen (%)	0	0	0	0.2	0	0.4	0	0.2	0	0.2

The amounts of the ingredients are given in parts by weight

	Reference Example 1		Reference Example 2		Reference Example 3		Reference Example 4		Reference Example 5		Reference Example 6	
1,1,1,2,6,7,7,7-Octa-chloro-4-oxaheptane	0.4		-		-		-		-		-	
Phoxim	-		0.4		-		-		-		-	
Chlorpyrifos	-		-		0.4		-		-		-	
Fenitrothion	-		-		-		0.4		-		-	
Prothiofos	-		-		-		-		0.4		-	
Isofenphos	-		-		-		-		-		0.4	
Weathering procedure	None	10 times	None	10 times	None	10 times	None	10 times	None	10 times	None	10 times
Number of days required until the death rate reached 100%	6	10	4	7	4	7	5	8	4	8	5	8
Rate of reduction in weight for test specimen (%)	1.0	2.0	0.2	0.5	0.2	0.5	1.0	2.0	0.2	0.7	0.5	2.0

The amounts of the ingredients are given in parts by weight

Example 1

By dissolving 1.0 part by weight of 1,1,1,2,6,7,7,7-octachloro-4-oxaheptane, 1.0 part by weight of phoxim, 0.3 part by weight of aluminum salt of N-nitroso-N-cyclohexyl-hydroxylamine and 0.2 part by weight of N-cyclohexyl-N-methoxy-2,5-dimethylfurane-3-carboxylic acid amide in 97.5 parts by weight of an aromatic hydrocarbon, the termite controlling composition according to the present invention was prepared.

Stakes (sapwood of Japanese red pine) for test uses measuring 30 mm by 30 mm by 350 mm were coated applied on the surfaces with the present termite controlling composition (at the application rate of 200 g/m^2) to make treated test specimens. Using the test specimens, a test on effectiveness was carried out at the experiment site located at Fukiagehama, Kagoshima Prefecture, where Coptotermes formosanus Shiraki is found to live and build the nest, by the procedure conforming to the outdoor test specified under the method of testing the effectiveness of a termite controlling composition for coating and immersion uses (JTCAS-No. 9) as established by the Japan Termite Control Association (a corporate juridical person).

Testing method

Treated test specimens were arranged on a circumference of a radius of 1 meter with the nest as the center and at an equal distance of 70 cm.

5 of non-treated test specimens were arranged each on the circumferences of a radius of 10 cm with each of the treated test specimens arranged mentioned above as the center and at an equal distance.

Test specimens, with their fiber directions kept vertical, were buried 30 cm deep beneath the ground at the specified positions.

Time duration of test was 2 years, whereby, when one year has elapsed, the treated test specimens were drawn out for inspection on damage development; testing was stopped

with the ones found to be damaged, while testing was continued for subsequent one year with the ones found not to be damaged.

When three months have elapsed after initiation, inspection was made on the non-treated test specimens for any damage development.

Even 2 years later, the test specimens treated with the present termite controlling agent were found to be free from attack by termites and also of rotting. On the other hand, the non-treated test specimens, after a lapse of half a year, were found to be severely attacked by termites, with the result that the rate of attack by termites reached 90%. The results of the test on effectiveness indicate that the present termite preventive agent possesses the excellent termite preventive performance and weathering resistance and is useful as a termite preventive agent for coating and immersion uses.

Example 2

By dissolving 30 parts by weight of 1,1,1,2,6,7,7,7-octachloro-4-oxaheptane, 20 parts by weight of phoxim, 5 parts by weight of polyoxyethylene nonylphenyl ether and 5 parts by weight of calcium dodecylbenzenesulfonate in 40 parts by weight of an aromatic hydrocarbon, an emulsifiable concentrate was prepared. A test on effectiveness was carried out at the experiment site located at Fukiagehama, Kagoshima Prefecture, by the procedure conforming to the outdoor test specified under the method of testing the effectiveness of a soil treatment agent (JTCAS-No. 9) as established by the Japan Termite Control Association (a corporate juridical person).

Even 1 year later, stakes (measuring 30 mm by 30 mm by 350 mm, sapwood of Japanese red pine) for test uses having been buried in the soil section treated with a 50-fold diluted solution of the present emulsifiable concentrate (at the application rate of 5 ℓ/m^2) were found be free from attack by termites, whereas stakes for test uses

buried in the non-treated soil section, after a lapse of half a year, were entirely attacked by termites, and had to be replaced with new stakes for test uses half a year later. The results of the test on effectiveness indicate that the present soil treatment agent possesses the excellent termite preventive performance and weathering resistance and is useful as a soil treatment agent.

* C L M S *

Claims

1. A termite controlling composition which comprises at least one of the organic phosphorus compounds consisting of phoxim, chlorpyriphos, fenitrothion, prothiofos and isofenphos, and 1,1,1,2,6,7,7,7-octachloro-4-oxaheptane.

2. A composition according to Claim 1, wherein the organic phosphorus compound is phoxim.

3. A composition according to Claim 1, wherein the mixing ratio of the organic phosphorus compound to 1,1,1,2,6,7,7,7-octachloro-4-oxaheptane is 1 : 0.05 to 25 on a weight basis.

4. A composition according to Claim 1, wherein the mixing ratio of the organic phosphorus compound to 1,1,1,2,6,7,7,7-octachloro-4-oxaheptane is 1 : 0.1 to 10 on a weight basis.

5. Wood material treated with the termite controlling composition which comprises at least one of the organic phosphorus compounds consisting of phoxim, chlorpyriphos, fenitrothion, prothiofos and isofenphos, and 1,1,1,2,6,7,7,7-octachloro-4-oxaheptane.

6. A termite controlling composition substantially as hereinbefore described with reference to any of the Test Examples and/or the Examples.