

ANTI-TERMITE COMPSOSITIONS
CONTAINING 8-HYDROXYQUINOLINE
DERIVATIVES AND THEIR USE.

5 This invention relates to an anti-termitic composition,
and more particularly to an anti-termitic composition which
has little toxic effect on human system and manifests suffi-
ciently persistent efficacy.

10 In most areas having a warm climate, wooden structures
are susceptible of termitic injuries. In the case of cable
buried directly in soil, not only outer layers of synthetic
resin and rubber but also metallic sheaths of aluminum and
iron have a possibility of sustaining holes formed by the
action of formic acid excreted by termites.

15 As a measure to protect structures and underground
cables against such termitic damages, the practice of apply-
ing an antitermitic agent to the surface of structures and
incorporating the anti-termitic agent into rubber and
synthetic resin used in the outer layers of cables has found
20 widespread acceptance.

Heretofore, organic chlorinated compounds have been predominantly used as anti-termite agents. These compounds, however, have a disadvantage that since they persist in their undecomposed form, they are suffered to accumulate in living organisms and deposit on environs of sites of application of such agents to jeopardize the health of workers exposed to the agents and bring about environmental pollution.

Lewis et al, "Hydroxyquinoline and Naphthol Termiticides", published in pages 818-822 of the October 1978 issue of the "Entomological Society of America," discloses the results of the test conducted on anti-termite property of 8-hydroxyquinoline and 1-naphthol incorporated in soil.

From the results, it is noted that 8-hydroxyquinoline showed outstanding anti-termite property and manifested very little toxic effect. This compound, however, has a disadvantage that it boils at a relatively low temperature of 266.6°C (752 mmHg) and is slightly soluble even in water (solubility in water 0.05 at 18°C and 0.35 at 90°C) and, therefore, fails to provide the desired anti-termite property for a long period.

This invention, therefore, seeks to provide an anti-termite composition which neither pollutes the environment nor adversely affects the health of workers exposed to the composition.

This invention also seeks to provide an anti-termite composition which provides a high anti-termite effect for a long time.

According to this invention, we provide a method of treating a substrate against termite damage comprising applying to and/or incorporating in said substrate an anti-termite composition containing a metal chelate with 8-hydroxyquinoline as an active component thereof.

The invention will now be further described by way of example with reference to the accompanying drawings, wherein:-

FIG. 1 is a cross section illustrating an apparatus to be used for testing the effect of the active composition in the anti-termitic composition of the present invention.

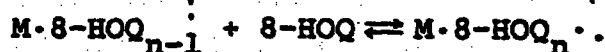
FIG. 2 is a graph showing the relation between the stability

5 constant and the anti-termitic effect of a metal chelate with 8-hydroxyquinoline. Fig. 3 is a cross section illustrating an apparatus to be used for testing the effect of a synthetic resin sheet incorporating the active component of the present invention.

10 The anti-termitic composition of this invention contains a metal chelate with 8-hydroxyquinoline as an active component thereof.

Examples of the metal moiety of the metal chelate with 8-hydroxyquinoline include Sn, Ba, Sr, Ca, Mg, Fe, Cu, Ni, Zn, Mn
15 and Co. Among other metals, Sn, Ba, Sr, Ca, Mg, and Fe are particularly suitable for the purpose of this invention.

Although metal chelates with 8-hydroxyquinoline generally possess very low dissociation constants in water, they slightly undergo dissociation and assume the following states
20 of equilibrium.



(wherein M stands for a metal ion, 8-HOQ for an 8-hydroxy-
25 quinoline, and n for a positive integer, which apply hereinafter).

The equilibrium constants $k_1, k_2, \dots, k_n$, therefore,

(14)

assume the following values.

$$k_1 = \frac{[M \cdot 8HOQ]}{[M] [8-HOQ]}$$

$$k_2 = \frac{[M \cdot 8HOQ_2]}{[M \cdot 8-HOQ] [8-HOQ]}$$

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⋮

$$k_n = \frac{[M \cdot 8HOQ_n]}{[M \cdot 8-HOQ_{n-1}] [8-HOQ]}$$

10 (wherein the brackets [] designate the concentration of the compound indicated).

The overall stability constant, K, is expressed as follows.

$$15 \quad k_1 \cdot k_2 \dots k_n = K = \frac{[M \cdot 8-HOQ_n]}{M [8-HOQ]^n}$$

The anti-termite effect of the metal chelate with 8-hydroxyquinoline of the present invention issues preponderantly from the 8-hydroxyquinoline produced in consequence of the dissociation of the metal chelate with 8-hydroxyquinoline.

20 The notable persistency of this anti-termite effect is mainly ascribable to the very high stability constant of the metal chelate.

The anti-termite composition of this invention, therefore, can be made to manifest a powerful anti-termite effect
25 evenly throughout the entire length of its life from the time of application by incorporating in the composition a plurality of metal chelates with 8-hydroxyquinoline having dissimilar

dissociation constants and further by incorporating therein 8-hydroxyquinoline.

Most metal chelates with 8-hydroxyquinoline have very high decomposition temperatures and do not experience any sharp
5 loss of weight at temperatures below 300°C. They have substantially no toxic effect upon human system. When they are to be incorporated in synthetic resin or rubber materials they do not impair the working environment.

A metal chelate with 8-hydroxyquinoline manifests its anti-
10 termitic effect when it is contained in the material in a concentration of only 0.1% by weight. This effect increases in proportion as the content of this metal chelate in the material increases. When the content exceeds 10% by weight, however, the anti-termitic effect is no longer increased
15 appreciably. Particularly when the metal chelate is contained in a material of synthetic resin or rubber, such a high content of the metal chelate goes to degrading the properties of the carrier. Thus, the upper limit of the metal chelate
content in the anti-termitic composition is desired to be not
20 more than 10% by weight. The range of the content of this metal chelate is desired to be from 0.5 to 5% by weight, preferably from 1 to 4% by weight.

The anti-termitic composition of this invention is generally formed by mixing a metal chelate with 8-hydroxy-
25 quinoline, or a mixture of two or more metal chelates with 8-hydroxyquinoline, or a mixture of a metal chelate with 8-hydroxyquinoline with 8-hydroxyquinoline, and a material of synthetic resin or _____

rubber.

A polyvinyl chloride resin composition and a polyethylene composition are particularly suitable examples of the synthetic resin to be used as the material.

- 5 Nylon is not a suitable synthetic resin for use as the carrier because it has an anti-termite property in itself.

In the inclusion of the active component in the synthetic resin as the material, the dispersibility of the active component in the material can be improved by having the active
10 component treated in advance with a coupling agent.

The anti-termite effect of the composition of this invention is heightened by using the active component having its dispersibility in the carrier improved as described above.

Examples of the coupling agent include silane coupling
15 agents and titanate coupling agents. Examples of the silane coupling agents include, for example, vinyltriethoxysilane, vinyltrimethoxysilane, vinyltris(methoxyethoxy)silane, vinyltrichlorosilane, γ -chloropropyltrimethoxysilane, γ -mercapto-
propyltrimethoxysilane, γ -glycidopropyltrimethoxysilane,
20 γ -aminopropyltriethoxysilane, N- β -(aminoethyl)- γ -aminopropyl-
trimethoxysilane, γ -ureidopropyltriethoxysilane, γ -methacryl-
oxypropyltrimethoxysilane and the like.

And, examples of the titanate coupling agents include, for example, isopropyltriisostearoyltitanate, isopropyl-
25 capryloyltitanate, isopropyl-diisostearoylcumylphenyltitanate;
isopropyltridodecylbenzenesulfonyltitanate, isopropyltris-
(dioctylpyrophosphate)titanate, tetrakis(2,2-diethylloxymethyl-

1-butyl)bis(ditridecylphosphite)titanate, tris(dioctylpyrophosphate)ethylenetitanate and the like.

Besides the above coupling agents, it is effective to use fatty acids such as stearic acid, palmitic acid and
5 others.

The amount of the coupling agent or fatty acids to be advantageously used is in the range of from 2 to 3% by weight based on the combined amount of the active component.

Examples of the form in which the anti-termitic
10 composition of this invention is advantageously used include a paint prepared by dispersing the active component(s) in a suitable binder and a blend prepared by mixing the active component(s) with soil.

Now, the present invention will be described more specifically below with reference to working examples.
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Example 1:

From 100 cc of acetone, 5 g of a varying metal chelate with 8-hydroxyquinoline indicated in Table I was repeatedly extracted.

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Table I

Sample No.	8-Hydroxyquinoline metal chelate
1	Ba-chelate with 8-hydroxyquinoline
2	Sr-chelate with 8-hydroxyquinoline
3	Ca-chelate with 8-hydroxyquinoline
4	Mg-chelate with 8-hydroxyquinoline
5	Fe-chelate with 8-hydroxyquinoline

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In each resultant extract was immersed two filter papers,

removed from the extract to dry, and then each filter paper was placed in respective petri dish 1 illustrated in Fig. 1. On the residue in the petri dish, 100 termites were placed and covered with a lid 3. The length of time required for all the termites to be killed was clocked. The relation between the stability constant and the time for 100% death obtained of each sample is shown in FIG. 2.

Example 2:

The ingredients of a varying sample indicated in Table 2 (From No. 14 to No. 18 are comparative tests) were kneaded in an electric hot roll and then molded through a press to produce a sheet 5 cm x 5cm in width and 0.5 mm in thickness.

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Table 2
(After 3 months)

Sample No.	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
PVC, P = 1300 *1	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100
DOP *2	40	40	40	40	40	40	40	40	40	40	40	40	40	40	40	40	40	40
Tribasic lead sulfate	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3	3
Barium stearate	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
Calcium carbonate (powder)	25	25	25	25	25	25	25	25	25	25	25	25	25	25	25	25	25	25
Ca-chelate with 8-hydroxyquinoline	5						2.5	2.5	2.5		2	0.2	17					
Sr-		5																
Mg-			5															
Fe-				5				2.5										
Cu-					5		2.5			2.5	2							
Ni-						5												
8-hydroxyquinoline									2.5	2.5	1						5	
Chlordane														5				
Chloropyrphos *3															5			
1-naphthol																5		

Notice:

- *1 Trade mark TOYO SODA CO.
- *2 Dioctyl phthalate
- *3 O,0-diethyl-O-3,5,6-tricloro-2-pyridyl phosphoro thioate

The sheet thus obtained was set in position in an incubator 4 illustrated in FIG. 3.

The incubator 4 held water in the bottom thereof.

A gypsum partition plate 7 provided with holes 6 for passage of termites was disposed inside the incubator 4 over the bath of water 5.

On the gypsum plate 7 was placed a tray 7a, and in the tray 7a were placed in order a sample sheet 8a, a pine plate 9a, a sample sheet 8b and a pine plate 9b.

10 The test was conducted by placing 400 termites in the incubator 4 and leaving them therein under a lid 10 for three months and, at the end of the standing, examining the pine plates for marks of bites by termites. The results of this test are shown in Table 3.

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Table 3

Index of damage	Sample No.																	
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
	Original sheet																	
	2	1	1	3	6	5	2	1	0	0	1	5	0	0	0	1	0	8
Sheet heated at 90°C for 8 days	4	3	2	5	5	4	3	4	4	5	4	5	0	6	7	10	4	10
Sheet heated at 90°C for 20 days	4	4	4	4	3	4	3	3	2	2	4	6	1	7	8	8	7	10

In this table, the damage index represents the average of the points of evaluation rated by the following scale.

Index of damage	Appearance of injury	Degree of injury
0:	Absolute absence of visible sign of termitic excretion or bite	No termitic bite
00:	Presence of sign of termitic excretion to sample and absolute absence of visible sign of termitic bite	
1:	Presence of sign of short termitic bites at leading ends of sharp corners of sheets	Traces of bites
2:	Presence of sign of long termitic bites at leading ends of sharp corners of sheets	
3:	Presence of sign of short termitic bites around sharp corners of sheets	Slight injury
4:	Presence of sign of long termitic bites around sharp corners of sheets	
5:	Presence of sign of short termitic bites in lateral cut sections of sheets	Heavy injury
6:	Presence of sign of long termitic bites in lateral cut sections of sheets	
7:	Presence of sign of shallow termitic bites into lateral cut sections of sheets	Serious injury
8:	Presence of sign of deep termitic bites into lateral cut sections of sheets	
9:	Presence of sign of termitic bites in flat surface of sheets	
10:	Presence of sign of deep termitic bites in flat surface of sheets	
*: Short - not more than 5 mm		
Long - over 5 mm		



CLAIMS

1. A method of treating a substrate against termitic damage comprising applying to and/or incorporating in said substrate an anti-termitic composition containing a metal chelate with 8-hydroxyquinoline as an active component thereof.
2. A method according to claim 1, wherein said metal chelate with 8-hydroxyquinoline consists of 8-hydroxyquinoline and one or more metals selected from Sn, Ba, Sr, Ca, Mg, and Fe.
3. A method according to claim 1 or 2, wherein said composition contains at least two metal chelates with 8-hydroxyquinoline, having dissimilar dissociation constants in aqueous solution, as active components thereof.
4. A method according to claim 1, 2 or 3, wherein said composition contains a metal chelate with 8-hydroxyquinoline and 8-hydroxyquinoline as active components thereof.
5. A method according to claim 4, wherein said composition contains at least two metal chelates with 8-hydroxyquinoline, having dissimilar dissociation constants in aqueous solution, and 8-hydroxyquinoline as active components thereof.
6. A method according to any one of the preceding claims wherein said metal chelate with 8-hydroxyquinoline is contained in an amount of 0.1 to 10% by weight of the composition.
7. A method according to any one of the preceding claims wherein said active component is dispersed in a



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synthetic resin composition.

8. A method according to claim 7, wherein said
synthetic resin composition is from a polyvinyl chloride
5 resin composition or polyethylene.

9. A method according to any one of the preceding
claims wherein said active component is treated with a
coupling agent in advance.

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10. A method of treating a substrate against
termite damage according to claim 1 substantially as
described herein and exemplified.

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11. A composition containing a metal chelate with 8-
hydroxyquinoline as an active component thereof for use in
treating a substrate against termite damage.

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