Efficacy of chlorothalonil as a wood preservative against the Formosan subterranean termite

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Abstract

Chlorothalonil (CTL, tetrachloroisophthalonitrile) was both deterrent and toxic to Formosan subterranean termites, *Coptotermes formosanus* Shiraki (Isoptera: Rhinotermitidae), in laboratory tests using southern yellow pine wafers treated with CTL in oil (AWPA P9 Type A), CTL/chlorpyrifos in oil, or CTL in xylene. The wafers were conditioned by evaporative aging at 40°C for 4 weeks and exposed to termite attack in a modified ASTM 4-week (no-choice) test. Termites were also exposed to CTL in the xylene carrier and solvent-treated pine wafers in a 4-week two-choice test for feeding deterrence. CTL retentions were assayed post-test by x-ray fluorescence, and an average 61 percent decrease in CTL concentration was found from the pretest nominal retentions. In the no-choice test, CTL retentions of 0.13 to 0.15 pcf (assayed post-test) limited wood weight loss from termite feeding to 6 to 13 percent, and retentions of 0.26 to 0.39 pcf CTL resulted in only 3 to 4 percent wood weight loss. In the two-choice test, CTL retentions ≥0.06 pcf deterred termite feeding in comparison to solvent controls, and the highest tested retention of 0.38 pcf limited wood weight loss to 1.5 percent. Termite mortality was positively correlated with CTL retention. In this study, assayed CTL retentions ≥ 0.26 pcf restricted wood weight loss from Formosan subterranean termite feeding to less than 5 percent. A heavy oil carrier was not essential for CTL efficacy.

Chlorothalonil (CTL, tetrachloroisophthalonitrile) is a fungicide used extensively in agriculture. In the United States, it is registered for use on turfgrass and over 35 food crops. CTL is also used to control mildew in paints and coatings, and as a marine antifouling agent (18). The excellent fungicidal properties of CTL, relatively low mammalian toxicity (acute oral and dermal LD_{50} 10,000 mg/kg in the rat), and favorable environmental characteristics (18) have encouraged its development as a wood preservative.

The most immediate application of CTL in wood preservation may be for protection against mold and sapstain fungi, either alone or in combination with other fungicides (11-14). With fungicidal efficacy and solubility characteristics comparable to pentachlorophenol (12), CTL formulations in heavy oil (AWPA P9 Type A) (5) are under development for use in utility poles and pilings.

In Hawaii and other tropical regions, termites are as great a threat to utility poles and other wood in service as decay fungi. Where the Formosan subterranean termite, *Coptotermes formosanus* Shiraki (Isoptera: Rhinotermitidae), occurs, it is a more important cause of utility pole failure than fungal decay (7,17). The current study was initiated to evaluate the efficacy of CTL alone and in combination with an insecticide (chlorpyrifos) in protecting wood from attack by the Formosan subterranean termite. Three pressure-impregnated formulations were evaluated in a modified ASTM D 3345-74 (1) or AWPA E1-72 (3)

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laboratory test: CTL in heavy oil, CTL + chlorpyrifos (1/16, w/w) in heavy oil, and CTL in xylene.

Experimental procedure

Wood treatment

The AWPA P9 Type A oil (5) formulations of CTL and CTL/chlorpyrifos (5% + 0.3%) (ISK Biotech Corp., Mentor, Ohio) were diluted in toluene to achieve target CTL retentions of 0.1, 0.2, 0.4, or 0.8 pcf in southern yellow pine (*Pinus* spp.) wafers (ca. 2 g, 1 by 1 by 0.25 in., flat sawn). Previous work has demonstrated an excellent correlation between the retention of chlorothalonil in AWPA P9 Type A oil solvents calculated from treating solution uptake and the assayed retention determined by both x-ray fluorescence and lime-ignition assays (10). The pressure treatment schedule was as follows: 1) 30-minute vacuum (25 in. Hg); 2) 60-minute pressure (100 psi); and 3) 4-hour soak (ambient conditions). Oil solvent control wafers were prepared in similar fashion. After pressure treatment, all wafers were allowed to equilibrate in the treating solution under ambient conditions for at least 10 minutes. The wafers were then removed from the solution, blotted dry, and immediately weighed.

In order to evaluate the effects of CTL independent of the oil solvent system, wafers were also impregnated to the same target retentions with a solution of CTL in paraxylene, followed by rapid evaporation in a forced draft oven set at 100°C. The solvent and drying conditions were chosen to minimize blooming, an undesirable concentration of chlorothalonil on the surface of the wood specimens. Previous work has shown that this procedure results in an acceptable CTL gradient of approximately 2:1 between the exterior and interior equal-volume portions of 0.75-inch pine sapwood cubes (10). Xylene solvent control wafers and untreated controls were also included in the test design.

No-choice termite test

Formosan subterranean termite workers (pseudergates, or undifferentiated individuals older than the third instar) and soldiers were collected from an active field colony on the Manoa campus of the University of Hawaii, using a trapping technique (16). Termites were collected immediately before their use in laboratory assays.

The no-choice laboratory bioassay was similar to ASTM D 3345-74 (reapproved 1986) (1), except that termite mortality was recorded and the ovendry weight loss in the test wafers was used to measure damage, rather than visual estimation. The test containers were 8 cm in diameter by 10 cm high screw-top plastic jars, each containing 150 g washed and ovendried crushed coral sand (sieved to pass a U.S. 14-mesh screen) and 30 ml of distilled water. One test wafer was placed on the surface of the damp sand, and 400 termites (360 workers and 40 soldiers) were added to each jar. Five replicates of each formulation at each retention (three formulations × four retentions) and oil, xylene, and untreated controls were included. Jar lids were loosely replaced and the jars were incubated in an unlighted temperature-controlled cabinet (29° ± 0.5°C) for 4 weeks (28 days), then dismantled, and termite mortality and ovendry wood weights recorded.

An unexpected problem was encountered during the test when all termites exposed to the treated wafers (including the oil and xylene solvent controls) died within 2 weeks, apparently due to toxicity of the residual solvents. At the conclusion of this 4-week test, no measurable feeding had occurred on any of the treated wafers. A weathering procedure was then employed to remove residual solvent (oil or xylene) from the test wafers, and these same wafers were again exposed to termite feeding in a second 4-week test. Thus, the complete weathering sequence for all wafers used in the no-choice tests was as follows: 1) evapo-

TABLE 1. — *Formosan subterranean termite mortality and weight loss of chlorothalonil-treated pine wafers in a 4-week no-choice laboratory test.*

Formulation[a]	Retention Nominal[b]	Retention Post-test[c]	Termite mortality[d]	Wood weight loss[d]	
	(pcf)	(pcf)	(%)	(mg)	(%)
CTL/oil	0.81	0.26	87 ± 7BC	81 ± 10A	3.5 ± 0.2
	0.41	0.14	84 ± 7B	180 ± 13AB	8.0 ± 0.2
	0.20	0.10	32 ± 3D	530 ± 29C	24.6 ± 0.6
	0.10	0.08	21 ± 3D	876 ± 53D	41.7 ± 1.3
CTL/CPY/oil	0.81	0.26	100 ± 0A	65 ± 5A	2.8 ± 0.1
	0.41	0.13	80 ± 8B	133 ± 29A	6.0 ± 0.6
	0.21	0.08	37 ± 2D	520 ± 33C	24.0 ± 0.6
	0.10	0.05	24 ± 4D	929 ± 83D	42.5 ± 1.6
CTL/xylene	0.94	0.39	97 ± 3AB	103 ± 9A	4.3 ± 0.2
	0.45	0.15	70 ± 13B	299 ± 62B	13.4 ± 1.2
	0.21	0.08	24 ± 2D	865 ± 50D	39.9 ± 0.8
	0.11	0.05	18 ± 2D	1125 ± 31F	52.8 ± 0.7
Oil control	- -	- -	33 ± 4D	614 ± 68C	28.0 ± 1.4
Xylene control	- -	- -	22 ± 3D	1091 ± 83EF	51.8 ± 1.0
Untreated control	- -	- -	18 ± 2D	971 ± 44DE	45.5 ± 4.3

[a] CTL/oil = chlorothalonil in AWPA P9 Type A oil; CTL/CPY/oil = chlorothalonil + chlorpyrifos (1/16, w/w) in P9 Type A oil; CTL/xylene = chlorothalonil in xylene.
[b] Average chlorothalonil retention based on treating solution uptake.
[c] Average post-test chlorothalonil retention assayed by x-ray fluorescence.
[d] Mean (± SEM) of five replicates. Means within a column followed by different capital letters are significantly different at the 0.05 level.

TABLE 2. — *Formosan subterranean termite mortality and weight loss of chlorothalonil-treated (xylene carrier) and control pine wafers (each approx. 2 g) in a 4-week two-choice laboratory test.*

CTL retention[a]		Termite mortality[d]	Percent wood weight loss[e]		
Nominal[b]	Post-test[c]		Treatment	Solvent control	t-test probability
------ (pcf) ------		(%)			
0.94	0.38	30.00 ± 2.88A	1.48 ± 0.05	36.03 ± 1.21	0.0003
0.45	0.14	17.05 ± 1.42B	3.10 ± 0.21	44.68 ± 0.85	0.0001
0.21	0.06	11.55 ± 0.41B	9.36 ± 0.50	37.83 ± 0.41	0.0001
0.11	0.02	14.10 ± 0.35B	19.84 ± 0.97	27.74 ± 0.99	0.2035
0.00	--	15.75 ± 0.81B	Untreated: 33.93 ± 1.29	Solvent-treated: 17.76 ± 0.72	0.0107

[a] Chlorothalonil in xylene carrier.
[b] Average chlorothalonil retention based on treating solution uptake.
[c] Average post-test chlorothalonil retention assayed by x-ray fluorescence.
[d] Mean (± SEM) of five replicates. Mean mortality values followed by different capital letters are significantly different at the 0.05 level.
[e] Mean total wood consumption (sum of both wafers in each replicate) was: 0.94 pcf, 813 ± 28 mg; 0.45 pcf, 1013 ± 16 mg; 0.21 pcf, 1020 ± 16 mg; 0.11 pcf, 1028 ± 9 mg; 0.00 pcf, 1103 ± 13 mg.

ration of solvent in a fumehood (ambient conditions) for 2 weeks, xylene samples were also dried in a vacuum oven (40°C) for 5 days; 2) ovendrying (90°C) for 1 day; 3) exposure to damp sand and termites for 4 weeks (29°C); 4) ovendrying (90°C) for 1 day; 5) evaporative aging in a forced air draft oven (40°C) for 4 weeks; and 6) ovendrying (90°C) for 3 days, before the second 4-week exposure to termites.

Following the second (and successful) 4-week termite exposure, the wood samples were again ovendried (90°C) for 3 days and weighed, and the overall CTL retention of each wafer was assayed by x-ray fluorescence on the whole ground wafer using an ASOMA spectrometer (2). When calculating the CTL retention for wafers treated with the CTL/chlorpyrifos formulation, the contribution of the chlorpyrifos chlorine to the assayed CTL retention was assumed to be insignificant.

Termite mortality (percentage mortality transformed by the arcsine of the squareroot) and wood weight loss data were subjected to analysis of variance (ANOVA) and means separated by Duncan's Multiple-Range Test, $\alpha = 0.05$ (15).

Two-choice termite test

Two-choice termite feeding assays, in which termite workers were presented simultaneously with both a CTL-treated and solvent-treated (control) pine wafer, were performed to determine the degree of repellency or feeding deterrence associated with CTL treatment. Because the results of the no-choice tests indicated that the oil solvent was itself deterrent to the insects (Table 1), the two-choice test was limited to the xylene formulation. The experimental setup was otherwise identical to the no-choice test, except that two wafers were placed on the surface of the damp sand, on opposite sides of the jar. Five replicates of each of the four CTL retentions were paired with xylene-treated control wafers, and xylene-treated wafers were also paired with untreated control wafers.

Since the pine wafers used in the two-choice test had not been conditioned by a prior exposure to termites and damp sand for 4 weeks (although weathering was otherwise identical to that described for the no-choice test samples), a preliminary bioassay was conducted and indicated the presence of lethal levels of xylene in the solvent-control wafers. The wafers were then additionally weathered prior to the two-choice test by 1) autoclaving followed by ovendrying (90°C) for 5 days; 2) soaking in distilled water (ambient conditions) for 20 hours; and 3) autoclaving, followed by ovendrying (90°C) for 2 days. After the 4-week test, the wood samples were ovendried (90°C) for 4 days and weighed, and CTL retentions were assayed by x-ray fluorescence (2).

Weight loss of treatment and control wafers was compared by paired comparison t-tests, and transformed percentage termite mortality data were subjected to an ANOVA with means separated by Duncan's Multiple-Range Test, $\alpha = 0.05$ (15).

Results and discussion

Our subsequent experience with these solvent systems (9) suggests that exposure of the test wafers to moist heat during the first unsuccessful termite assay was most important in removing the residual oil and xylene solvents. Although no leaching procedure was performed with the wafers used in the no-choice tests, the initial exposure to termites in damp sand for 4-weeks (29°C) was apparently conducive to leaching. The wafers (xylene carrier) used in the two-choice test were conditioned prior to test by autoclaving and a 20-hour soak at room temperature. This exceeded the 2-hour soak recommended in AWPA soil-block tests (4) by a factor of 10. The 4-week evaporative aging period to which these wafers were exposed was twice the length of the 2-week AWPA volatility test recommended for soil blocks (5).

The lengthy weathering sequence to which the test wafers were ultimately exposed led to an average 61 percent (Table 1) reduction in CTL retentions. Similar reductions in CTL content have also been noted in soil blocks after extensive leaching and fungal exposure (10), but not in tests with larger wood specimens (19), suggesting that the high surface-to-volume ratio of the small test wafers may facilitate leaching.

Both termite mortality and feeding on the treated wood were correlated with CTL retention (Table 1). With all three CTL formulations, termite feeding on wood with assayed retentions of 0.05 to 0.10 pcf (corresponding to 0.10 to 0.21 pcf nominal) did not differ significantly from the solvent controls. CTL retentions of 0.13 to 0.15 pcf (0.41 to 0.45 pcf nominal) limited wood weight loss from termite feeding to 6 to 13 percent. Retentions of 0.26 to 0.39 pcf (0.81 to 0.94 pcf nominal) CTL resulted in only 3 to 4 percent wood weight loss. Nominal retentions of 0.10 to 0.84 pcf have proved effective (average AWPA termite ratings of 9.4 and 10.0) for at least 2.5 years in aboveground, termite-specific field trials in Hawaii (12).

A heavy oil solvent alone is deterrent to termites and contributed to CTL efficacy in the no-choice test. The choice test (Table 2) indicated that the xylene solvent treatment was also deterrent, possibly due to extraction of feeding stimulants from the wood. However, this effect was not apparent in the no-choice test, where the insects were forced to feed or die, and slightly higher CTL retentions were required with the xylene formulation to suppress termite feeding.

Addition of a small amount of chlorpyrifos to the oil formulation resulted in higher termite mortality at the highest preservative retention, but had no significant effect on the amount of damage resulting from termite feeding during the 4-week test. This is consistent with the results of laboratory evaluations of similar CTL and CTL/chlorpyrifos formulations against other termite species (6). However, we did not assay the leached, exposed blocks for chlorpyrifos content and it is likely that the chlorpyrifos residues were greatly reduced by the strenuous conditioning regime. Although chlorpyrifos undergoes exothermic decomposition at approximately 130°C (8), heating the treated wafers for 3 days in a 90°C oven may have resulted in some degradation.

In the two-choice test, termites preferred to feed on untreated wood rather than wood treated with the xylene solvent. However, all but the lowest CTL retention of 0.02 pcf (0.1 pcf nominal) also deterred termite feeding, in comparison to feeding on the solvent-treated control wafers (Table 2). A small amount of feeding (1.5% weight loss) occurred even at the highest retention tested (0.38 pcf or 0.8 pcf nominal), and this feeding was sufficient to cause significant termite mortality (30%) even though an alternative food source was available to the insects.

Our results demonstrate that chlorothalonil-treated wood, even without the presence of a heavy oil carrier, is both toxic and deterrent to Formosan subterranean termites, and that the magnitude of these effects is positively correlated with CTL retentions. Although further work is needed to determine the significance of the decrease in CTL content in the test wafers noted during this study, assayed CTL retentions of 0.26 pcf held wood weight loss from termite feeding to under 5 percent, and thus may represent reasonable minimum retentions for field evaluations.

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