

環動昆

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Effects of Cuticular Coating on the Behavior and Survival of Termites

Ahdin GASSA¹⁾, SHOZO TAKAHASHI¹⁾ and Hidetoshi OKAMOTO²⁾

1) Pesticide Research Institute, Faculty of Agriculture, Kyoto University, Kyoto 606, Japan

2) United Graduate School of Agricultural Sciences, Ehime University, Matsuyama, Ehime 790, Japan

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体表付着物がシロアリの行動ならびに生存に及ぼす影響 Ahdin GASSA¹⁾・高橋正三¹⁾・岡本秀俊²⁾(¹⁾京都大学農学部農業研究施設, ²⁾愛媛大学大学院連合農学研究科)

微粉物質の昆虫体表付着による致死効果については古くから知られている。微粉物質と流動パラフィンのシロアリ職蟻および兵蟻に対する影響について検討した。これらの物質の付着した職蟻および兵蟻は動きが鈍くなり数時間で死亡した。シロアリの兵蟻は体表に付着物を有する同種の職蟻に遭遇すると、職蟻に対し攻撃行動をとり、刺咬行動のちこれらを死亡させた。微粉の職蟻体表への付着は、特に節間膜が顕著で、希エタノールで洗浄しても剥落しなかった。一方、流動パラフィン付着で死亡した職蟻の腹部は膨満し、節間も伸びていた。比較のために、チャバネゴキブリ1齢若虫に対しても微粉物質と流動パラフィン処理を行ったところ、シロアリ類の場合と同様な致死効果と処理個体における形態の変化が認められた。

The lethal effects of powder coating on the body surface of insects has long been known. In tests made in small petri dishes using powdered material and liquid paraffin, most termite workers and soldiers became motionless and then died after a few hours. When a conspecific soldier encountered a treated worker, the soldier attacked with open mandibles and bit the worker to death. Charcoal coating was lethal to the termites and also elicited biting from conspecific soldiers. The reason for coating causes death is not yet clear but strong adherence of the powder particles to the abdominal intersegmental membrane was noted even after washing with diluted ethanol. Bodies of termites killed by liquid paraffin treatment, in contrast, had swollen abdomens. Dead bodies of German cockroach nymphs showed open abdominal segments and adherence of charcoal in intersegmental membranes.

Key Words : Termite, Cuticular coating, *Coptotermes formosanus*, *Reticulitermes speratus*

Introduction

Two species of termites, *Coptotermes formosanus* SHIRAKI and *Reticulitermes speratus* (KOLBE) are widely distributed in the western part of Japan and are known for the damage they cause to wooden structures. The former species usually maintain their nests under the ground where they can easily attack decayed wood. Various chemical treatments have been tested to prevent and control termite infestation.

The lethal effect of powder coating on the body surface of insects was first recognized many years ago (YASUE, 1949). The effects of powders or powdered materials on termites, cockroaches, and mites was reported more recently by YAMAMOTO and OKAMOTO (1993). Their experimental results indicated that the

workers of *C. formosanus* were killed shortly after a treatment which covered the body surface with powdered natural and artificial zeolite, powdered clay minerals, talc, volcanic ash, coal ash or silica gel. All adults of the German cockroach, *Blattella germanica*, treated with 4 kinds of zeolites died within 2 to 3 days, but half of those treated with sepiolite survived for 3 days.

The lethal effects of charcoal and liquid paraffin were compared with that caused by talc coating. Charcoal was chosen as a fine nonmineral powder and paraffin was tested as they have affinities to termite cuticular wax. We observed a behavioral change in the termite workers that lasted a short time after treatment with the powdered materials, charcoal and talc, and with liquid paraffin, followed by a lethal

Table 1 Survival time of workers of *Coptotermes formosanus* SHIRAKI, *Reticulitermes speratus* (KOLBE) and *Hodotermopsis japonica* HOLMGREN after cuticular coating with various materials¹⁾

Treatment		Survival time of worker (min.)		
Material	Duration (min.)	<i>C. formosanus</i>	<i>R. speratus</i>	<i>H. japonica</i>
Charcoal	1	60. 06 (1. 01 hr)	69. 22 (1. 15 hr)	4. 95
	3	45. 00	59. 24	3. 49
	5	43. 08	45. 28	2. 40
Talc	1	244. 20 (4. 07 hr)	262. 01 (4. 37 hr)	6230. 40 (103. 84 hr)
	3	217. 02 (3. 62 hr)	229. 24 (3. 82 hr)	5751. 00 (95. 85 hr)
	5	186. 00 (3. 10 hr)	200. 00 (3. 33 hr)	4464. 60 (74. 41 hr)
Liquid paraffin (Waken)	1	292. 20 (4. 87 hr)	335. 04 (5. 58 hr)	7135. 80 (118. 93 hr)
	3	199. 40 (3. 32 hr)	186. 06 (3. 10 hr)	6248. 40 (104. 14 hr)
	5	164. 40 (2. 74 hr)	182. 04 (3. 03 hr)	4731. 00 (78. 85 hr)
Liquid paraffin (Merck)	1	421. 20 (7. 02 hr)	433. 09 (7. 22 hr)	7429. 80 (123. 83 hr)
	3	323. 40 (5. 39 hr)	391. 02 (6. 52 hr)	6274. 80 (104. 58 hr)
	5	181. 80 (3. 03 hr)	314. 04 (5. 23 hr)	4882. 80 (81. 38 hr)
n-Heptadecane	1	319. 20 (5. 32 hr)	333. 00 (5. 55 hr)	7457. 40 (124. 29 hr)
	3	301. 80 (5. 03 hr)	315. 00 (5. 25 hr)	7450. 80 (124. 18 hr)
	5	261. 00 (4. 35 hr)	305. 04 (5. 08 hr)	6076. 80 (101. 28 hr)
Control		3312. 00 (55. 20 hr)	3894. 00 (64. 90 hr)	10062. 00 (167. 70 hr)

¹⁾ Average of 10 replications. Intact individuals were used in each test.

effect. A similar effect was observed in first instar nymphs of the German cockroach with this treatment.

Materials and Methods

Insects

Workers and soldiers of *C. formosanus* were supplied from a stock culture of the termite in the insectarium of the Wood Research Institute, Kyoto University, Uji, and the colony of *R. speratus* was collected from the Botanical Garden of the Faculty of Science, Kyoto University, Kyoto. Workers and soldiers of *Hodotermopsis japonica* HOLMGREN were supplied from a stock culture in the Department of Biology, College of Arts and Sciences, the University of Tokyo. Each of these colonies was

Table. 2 Survival time of nymphs of *Blattella germanica* (L.) after cuticular coating with various materials¹⁾

Treatment		Survival time of <i>B. germanica</i> (min.)
Material	Duration (min.)	
Charcoal	1	33. 38
	3	23. 00
	5	21. 05
Talc	1	572. 40 (9. 54 hr)
	3	444. 00 (7. 40 hr)
	5	180. 00 (3. 00 hr)
Liquid paraffin (Waken)	1	4385. 70 (73. 09 hr)
	3	4231. 20 (70. 52 hr)
	5	554. 16 (9. 24 hr)
Liquid paraffin (Merck)	1	4630. 50 (77. 17 hr)
	3	4164. 75 (69. 41 hr)
	5	2978. 00 (49. 63 hr)
n-Heptadecane	1	2410. 60 (40. 18 hr)
	3	2728. 92 (45. 48 hr)
	5	390. 00 (6. 50 hr)
Control		6790. 80 (113. 18 hr)

¹⁾Average of 10 replications. Intact individuals were used in each test.

maintained in a plastic container at $25 \pm 2^\circ\text{C}$ in the Pesticide Research Institute, Faculty of Agriculture, Kyoto University, Kyoto. Nymphs of the German cockroach were obtained from a stock culture in the Pesticide Research Institute.

Chemicals

Charcoal (particle size composition: more than $297 \mu\text{m}$ —40%, 63 – $297 \mu\text{m}$ —minimum 50%, less than $63 \mu\text{m}$ —maximum 10%), talc (particle size composition: 2 – $20 \mu\text{m}$ —82%, 20 – $44 \mu\text{m}$ —4%, less than $2 \mu\text{m}$ —14%), liquid paraffin and n-heptadecane (m.p. 22°C) were purchased from Wako Pure Chemical Industries, Ltd. Liquid paraffin for spectroscopy by Merck was also used.

Cuticular coating of termites and nymphs of German cockroach

All experiments were done at $25 \pm 2^\circ\text{C}$ and moisture was unadjusted. Workers of the three termite species were released into individual small petri dishes (3.0 cm in diameter) which contained 100 mg of charcoal or talc. For the liquid paraffin tests, 100 mg of the paraffin was applied to a filter paper (Toyo Roshi Kaisha, Ltd., No. 2) and placed at the bottom of the petri dish. The released workers were allowed to walk in the petri dish for 1 min, 3 min, or 5 min and then were transferred to a new petri dish for behavioral observation. Termite soldiers and the first instar nymphs of the German cockroach were similarly treated. In each treatment 10 insects were used.

Behavior of soldiers to the treated workers

After the workers and soldiers were treated in the petri dish with powder materials or liquid paraffin and transferred to a new petri dish (3.0 cm in diameter), a conspecific soldier was added to observe the behavior toward the treated individual.

Observation of cuticle of dead workers

Dead bodies of termites with talc and charcoal coating were shaken in 1 ml of 50%

aqueous ethanol for 3 min by hand. Most of the talc and charcoal were washed off of the body surface but remained at segmental membranes. The abdominal surface of the abdomen was cut in Ringer solution and was observed with a binocular microscope.

Results

Behavior of workers after treatment with various materials

After treatment with powder material and liquid paraffin, movement of the workers slowed and they became motionless after several minutes. Charcoal adherence to inter-segmental membranes of workers was first observed and then spread all over the body surface. Workers of *C. formosanus* died within

about 1 hour after treatment with charcoal, which was the most effective among the materials used. Treatment with talc or liquid paraffin showed a similar effect but workers took longer to die. Survival time following treatment of workers of *C. formosanus*, *R. speratus* and *H. japonica*, and the first instar nymphs of the German cockroach are shown in Tables 1 and 2. The effects of treatment of soldiers of *C. formosanus* and *R. speratus* were similar (Table 3).

Behavior of soldiers toward treated conspecific workers and soldiers

Following treatment of workers and soldiers and their transfer to a new petri dish, a conspecific soldier was introduced. This soldier recognized the cuticular coated workers or

Table 3 Survival time of soldiers of *Coptotermes formosanus* SHIRAKI and *Reticulitermes speratus* (KOLBE) after cuticular coating with various materials

Treatment		Survival time of soldier (min.)	
Material	Duration (min.)	<i>C. formosanus</i> ¹⁾	<i>R. speratus</i> ²⁾
Charcoal	1	126. 00 (2. 10 hr)	78. 38 (1. 31 hr)
	3	70. 20 (1. 17 hr)	69. 54 (1. 16 hr)
	5	64. 20	67. 40 (1. 12 hr)
Talc	1	525. 60 (8. 76 hr)	448. 80 (7. 48 hr)
	3	477. 60 (7. 96 hr)	410. 40 (6. 84 hr)
	5	400. 20 (6. 67 hr)	370. 20 (6. 17 hr)
Liquid paraffin (Waken)	1	522. 00 (8. 70 hr)	443. 40 (7. 39 hr)
	3	490. 20 (8. 17 hr)	438. 60 (7. 31 hr)
	5	449. 40 (7. 49 hr)	417. 00 (6. 95 hr)
Liquid paraffin (Merck)	1	729. 00 (12. 15 hr)	541. 80 (9. 03 hr)
	3	553. 80 (9. 23 hr)	453. 00 (7. 53 hr)
	5	553. 20 (9. 22 hr)	385. 20 (6. 42 hr)
n-Heptadecane	1	699. 00 (11. 65 hr)	487. 20 (8. 12 hr)
	3	652. 20 (10. 87 hr)	303. 00 (5. 05 hr)
	5	303. 60 (5. 06 hr)	280. 80 (4. 68 hr)
Control		5405. 00 (90. 08 hr)	1710. 00 (28. 50 hr)

¹⁾Average of 10 replications. Intact individuals were used in each test.

²⁾Average of 5 replications. Intact individuals were used in each test.

Table 4 Induction of aggressive behavior in soldiers of *Coptotermes formosanus* SHIRAKI toward conspecific workers and soldiers treated with various materials¹⁾

Material	Treatment		Worker		Soldier	
	Duration (min.)		Time before attack (min.)	Survival time (min.)	Time before attack (min.)	Survival time (min.)
Charcoal	1		1. 56	29. 23	2. 01	54. 53
	3		0. 61	23. 42	1. 50	44. 42
	5		0. 63	18. 09	1. 55	31. 49
Talc	1		6. 05	69. 24 (1. 15 hr)	2. 40	70. 72 (1. 18 hr)
	3		3. 14	61. 17 (1. 01 hr)	1. 50	62. 65 (1. 04 hr)
	5		2. 23	33. 02	1. 57	34. 59
Liquid paraffin (Waken)	1		3. 07	50. 31	4. 14	69. 24 (1. 15 hr)
	3		2. 43	36. 24	3. 36	61. 17 (1. 02 hr)
	5		1. 14	32. 56	1. 57	33. 02
Liquid paraffin (Merck)	1		2. 08	64. 08 (1. 07 hr)	3. 07	77. 85 (1. 30 hr)
	3		1. 35	41. 03	2. 43	45. 82
	5		1. 31	27. 06	1. 48	32. 56
n-Heptadecane	1		2. 01	90. 07 (1. 50 hr)	3. 08	116. 06 (1. 93 hr)
	3		1. 02	75. 31 (1. 25 hr)	2. 35	109. 59 (1. 83 hr)
	5		0. 54	61. 29 (1. 02 hr)	1. 03	95. 52 (1. 59 hr)
Control ²⁾		—	2136. 00 (35. 06 hr)	—	2805. 00 (46. 75 hr)	

¹⁾Average of 10 replications. Intact individuals were used in each test.

²⁾A pair of a worker and a soldier.

soldier and began to attack the individual with open mandibles; some often continued biting until the opponent died or was decapitated (Fig. 1). Tables 4–6 show survival time of workers after introduction of a soldier and the time before the latter attacked the treated individual. The aggressive behavior of soldiers toward the conspecific workers was elicited regardless of any materials used for the treatment. The survival time was much shorter than that caused by coating itself.

Observation of cuticle of dead worker

Dead bodies of charcoal or talc-treated workers of *C. formosanus* and *R. speratus* were washed with 50% aqueous ethanol. Charcoal and talc remained adhering to the intersegmental membranes (Fig. 2). Bodies of those

killed by the liquid paraffin treatment showed swollen abdomens (Fig. 3). A similar effect was observed by treatment with a paraffin compound, n-heptadecane. To compare the lethal effect on other insects, the first instar of the German cockroach was treated with charcoal. Again, charcoal adhered mainly to the intersegmental membranes of the abdomen after washing with diluted ethanol (Fig. 4).

Discussion

The lethal effects of powdered natural minerals on the body surface of termites was suggested to result from desiccation and loss of water by integument abrasion (YAMAMOTO and OKAMOTO, 1993). Charcoal or liquid paraffin coating also was lethal to the termites but this

Table 5 Induction of aggressive behavior in soldiers of *Reticulitermes speratus* (KOLBE) toward conspecific workers treated with various materials¹⁾

Material	Treatment		Time before attack (min.)	Survival time of worker (min.)
	Duration (min.)			
Charcoal	1		1. 58	21. 17
	3		1. 10	18. 54
	5		0. 29	14. 40
Talc	1		5. 22	72. 33 (1. 20 hr)
	3		4. 23	50. 10
	5		2. 12	31. 35
Liquid paraffin (Waken)	1		3. 05	55. 05
	3		2. 38	41. 15
	5		2. 28	35. 38
Liquid paraffin (Merck)	1		2. 58	67. 35 (1. 12 hr)
	3		2. 15	66. 38 (1. 10 hr)
	5		1. 55	63. 47 (1. 05 hr)
n-Heptadecane	1		2. 14	88. 58 (1. 47 hr)
	3		1. 22	81. 36 (1. 35 hr)
	5		1. 19	67. 15 (1. 12 hr)
Control ²⁾			—	1302. 17 (21. 72 hr)

¹⁾Average of 10 replications. Intact individuals were used in each test.

²⁾A pair of a worker and a soldier.

Table 6 Induction of aggressive behavior in soldiers of *Hodotermopsis japonica* HOLMGREN toward conspecific workers treated with various materials¹⁾

Material	Treatment		Time before attack (min.)	Survival time of worker (min.)
	Duration (min.)			
Charcoal	1		1. 25	25. 32
Liquid paraffin (Waken)	1		1. 48	45. 50
Control ²⁾			—	134. 68

¹⁾Average of 10 replications. Intact individuals were used in each test.

²⁾A pair of a worker and a soldier.

would not likely be due to desiccation. Intersegmental membranes are considered to be very important for insect movement (WIGGLESWORTH, 1975) and for oviposition by the female locust (VINCENT, 1976). They are extended and contracted in movement and respi-

ration. Strong adherence of charcoal to the abdominal intersegmental membrane was observed even after washing with diluted ethanol. Although talc particles are finer than charcoal, charcoal showed higher acute termiticidal activity. Liquid paraffin treatment, in contrast,

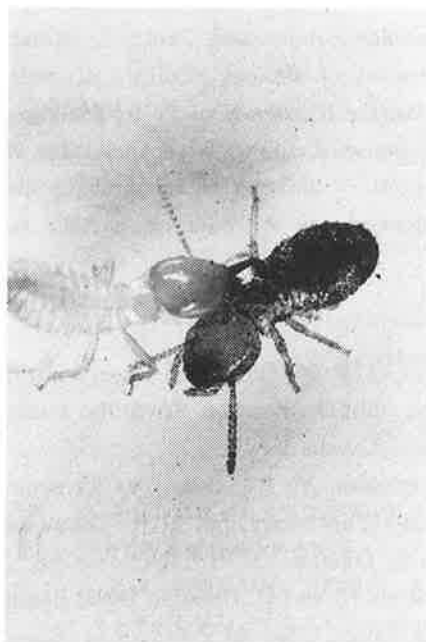


Fig. 1 Biting of a charcoal-coated worker by a conspecific soldier of *Coptotermes formosanus*.



Fig. 2 Adherence of charcoal on abdominal surface of *Reticulitermes speratus* worker. After charcoal treatment, the body was washed with diluted ethanol and dissected in Ringer solution.

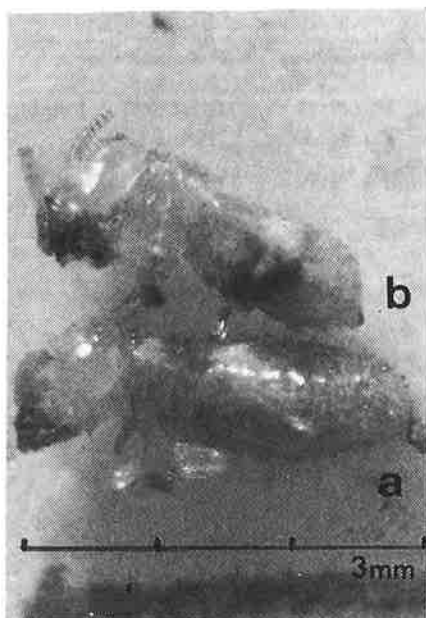


Fig. 3 Effects of liquid paraffin treatment. (a) Swollen abdomen of a *Reticulitermes speratus* worker. (b) Control (Dead body without treatment)



Fig. 4 Adherence of charcoal on the abdominal surface of a first-instar nymph of *Blattella germanica*. After charcoal treatment, the body was washed with diluted ethanol and dissected in Ringer solution.

caused the extension of the abdominal cuticle and this resulted in impeding of movement. Observations confirmed that disturbance of articulation of the body segments due to the adherence of charcoal, talc, and liquid paraffin caused the deaths.

When a soldier encountered a conspecific worker coated with powder or liquid paraffin, he recognized the worker not as a conspecific worker but as a different species. The cuticular wax of termites is composed of a mixture of hydrocarbons. Coating of the worker body surface with foreign hydrocarbons not only was lethal to the workers themselves but imbued them with a kairomonal activity that elicited biting from conspecific soldiers.

Cause of death in termite workers due to extension of the intersegmental membranes and inducing biting in soldiers toward conspecific workers coated with materials are peculiar phenomena. To clarify the mechanism of the behavior change in termite workers and soldiers, further studies are on the way.

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Application of a Synthetic Pyrethroid, Tefuramethrin, as a Vaporific Mothproofing Agent

Hisashi TAKEGAWA and Yoshio KATSUDA

*Research Laboratory, Dainihon Jochugiku Co. Ltd.,
Toyonaka, Osaka 561, Japan*

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新ピレスロイド・テフラメトリンの蒸散性衣料防虫剤への適用 武川 恒・勝田純郎
(大日本除虫菊(株)中央研究所)

近年、刺激臭がなく温血動物にも極めて安全性の高い蒸散性ピレスロイド、エムベントリンを有効成分とする衣料防虫剤が普及した。しかし、エムベントリンは銅との反応性があり、銅を含む染料で処理された衣料品や装飾品とエムベントリン製剤が接触すると、反応して黄緑色のしみを生じる(黄変現象)。その対策として、黄変防止剤や安定剤が検討されたが、満足な結果は得られなかった。今回著者らは、5-propargyl-2-furylmethyl 2,2,3,3-tetramethyl cyclopropanecarboxylateをエムベントリンに対して一定の割合で混合すると、エムベントリンと銅との反応が抑制されることを明らかにし、本化合物をテフラメトリン (ISO申請中) と命名した。また、テフラメトリンは上記の特徴をもつと同時に、常温揮散性を示し、衣料害虫に対する防虫効力は、エムベントリンより若干低いものの実用性はきわめて高い。すなわち、テフラメトリンの単剤あるいはエムベントリンとの一定比率での混合製剤は、銅を含む衣料品や装飾品との反応の心配がなく、高い防虫効果をもつ蒸散性衣料防虫剤として極めて有望である。

Application of a new synthetic pyrethroid, 5-propargyl-2-furylmethyl 2,2,3,3-tetramethyl cyclopropanecarboxylate, as a vaporific mothproofing agent was examined. In recent years, empenethrin has been widely used as an active ingredient in vaporific mothproofing agents because of its low toxicity to mammals and non-irritant odor, but it has the disadvantage of reacting with copper (Cu) contained in dye-stuffs used in clothes and in their ornaments to produce a degraded compound forming yellow-greenish stains. Though newly synthesized vaporific pyrethroids or additive compounds that inhibit the reaction between empenethrin and copper have been required to correct this defective property of empenethrin, none of them have become available that meet the requirements. In the course of this study, we found that the reaction between empenethrin and copper was inhibited by mixing 5-propargyl-2-furylmethyl 2,2,3,3-tetramethyl cyclopropanecarboxylate with empenethrin at a certain mixing ratio, and we named the compound

tefuramethrin (in ISO application). Also, in addition to the characteristics mentioned above, it has a high vapor pressure and proof-efficacy on fabric pests, but is less effective at a minimum dose than empenthrin. Application of tefuramethrin or its mixed formulations with empenthrin at a certain mixing ratio, as a vaporific mothproofing agent, promises to provide a novel type of agent with highly insecticidal activity as well as no undesirable reactions with copper in clothes and in their ornaments.

Key Words : Tefuramethrin, Empenthrin, Mothproofing, Fabric pests

Introduction

Natural pyrethrins derived from pyrethrum flowers (*Chrysanthemum cinerariaefolium* vis.), are characterized by highly insecticidal activity and low mammalian toxicity. Since the structural identifications of the active ingredients in natural pyrethrins in the 1950's (KATSUDA *et al.*, 1958; KATSUDA *et al.*, 1959), many synthetic pyrethroids have been developed by modifying the original structures, and some of them have become a major group of insecticides for the control of household insects as well as agricultural pests.

One of these synthetic pyrethroids, empenthrin, which was originally synthesized by KITAMURA *et al.* (1980), proved to have a higher vapor pressure than any other conventional pyrethroids and a highly volatile effect with strong insecticidal activity against houseflies and fabric pests. Empenthrin-based products were first used commercially as a vaporific mothproofing agent by Dainihon Jochugiku Co., Ltd. in 1983. In Japan, three sublimating compounds, *i. e.* camphor, *p*-dichlorobenzene and naphthalene, have been used as vaporific mothproofing agents for a long time, despite the fact that these compounds have disadvantages such as strong, characteristic odors and deformation of some kinds of plastics. Moreover the combined use of these sublimating mothproofing agents within the small space of

a wardrobe causes liquefaction of the agents, and the clothes in the wardrobe can be stained by the liquefied agents. Therefore, empenthrin, an odorless vaporific insecticide, has been considered to be a promising alternative in the mothproofing field, so that there has been a strong demand for empenthrin in the field.

However, it should be also noted that empenthrin reacts with copper (Cu)-containing materials, to produce a degraded compound, a shiny yellowish oil. Some kinds of dyes and ornaments containing copper react with empenthrin to leave a stain on the clothes and on their ornaments. Though newly synthesized vaporific pyrethroids or additive compounds that inhibit the reaction between empenthrin and copper, have been required to avert the problems of empenthrin reacting with copper-containing materials, none are available.

In the course of this study, we found that the reaction between empenthrin and copper can be inhibited by the addition of 5-propargyl-2-furylmethyl 2,2,3,3-tetramethyl cyclopropane-carboxylate to empenthrin at a certain mixing ratio, and we named the compound tefuramethrin (Fig. 1). Tefuramethrin is a synthetic pyrethroid developed by KATSUDA and OHNO (1971). We also proved tefuramethrin to have vaporific insecticidal activities on houseflies and fabric pests.

In this paper, the remarkable effectiveness of tefuramethrin in inhibiting the reaction be-

tween empenethrin and copper is described, and the insecticidal activities of tefuramethrin toward houseflies and fabric pests are also reported.

Materials and Methods

Test insects

A susceptible strain of housefly (*Musca domestica* L.) was reared at $25 \pm 2^\circ\text{C}$ and maintained in a 14L-10D photoperiod at $60 \pm 10\%$ RH. Adult females of 3 to 5 days post-emergence were used for the efficacy tests.

Casemaking clothes moths (*Tinea translucens* MEYRICK) were reared at $25 \pm 2^\circ\text{C}$ and $60 \pm 10\%$ RH. Larvae of 35 to 40 days post-oviposition were used for the subsequent efficacy tests. Black carpet beetles (*Attagenus unicolor japonicus* REITTER) and varied carpet beetles (*Anthrenus verbasci* L.) were reared under the same conditions, and larvae of 6 to 12 months old were used in the efficacy tests.

Test compounds

Tefuramethrin (in ISO application) ;

5-propargyl-2-furymethyl 2,2,3,3-tetramethyl cyclopropanecarboxylate.

Empenthrin ; (RS)-1-ethynyl-2-methylpent-2-enyl(1R)-cis, trans-chrysanthemate (Vaporthrin[®]).

Reactivity tests between the test compounds and copper (Cu)

One milliliter of an acetone solution containing the appropriate dosage of the test compounds was used to impregnate a piece of

cardboard ($2.2 \times 3.5 \times 0.02\text{cm}$). After the acetone evaporated, a copper plate was attached to the treated cardboard and kept at 50°C . After 5 days, reactivity between the test compounds and the copper plate was evaluated. A single formulation of the test compounds and their mixed formulations were examined.

Topical toxicity tests of the test compounds (Topical application method)

$0.5 \mu\text{l}$ of an acetone solution containing the appropriate dosage of the test compound was applied to the dorsum prothorax of a housefly using a micro-syringe dispenser (a hand applicator, Burkard Co., Ltd., England).

$0.25 \mu\text{l}$ of an acetone solution containing the appropriate dosage of the test compound was applied on the case of a casemaking clothes moth larva, and the same quantity was put on the dorsum prothorax of a black carpet beetle larva and varied carpet beetle larva, using the dispenser.

LD₅₀ values were calculated by FINNEY'S graphic method.

Volatile effect tests of the test compounds (Vapor action method)

One milliliter of an acetone solution containing the appropriate dosage of the test compounds was applied to the inner base surface of an upper petri dish A (9cm in diameter and 2 cm in height) (Fig. 2). After the acetone evaporated, the applied petri dish was placed on the same size dish B with the base surface up. Between the dishes, a wire netting (12

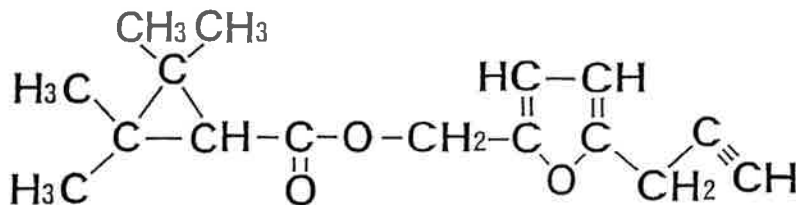


Fig. 1 Tefuramethrin (5-propargyl-2-furymethyl 2,2,3,3-tetramethyl cyclopropanecarboxylate).

mesh) was set to prevent insects from touching the applied test compounds, and the test insects were released into the lower dish B (Fig. 2).

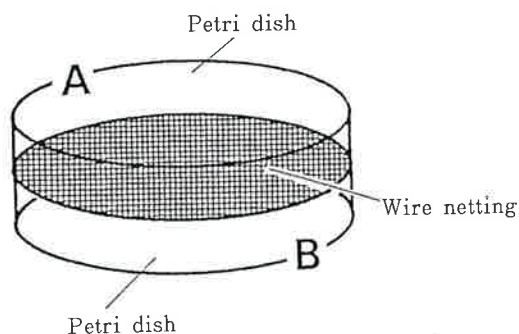


Fig. 2 Device for vapor action method. A : test compounds were applied to the inner base surface of the petri dish (9 cm ϕ \times 2 cm) ; B : test insects were released into the lower dish (9 cm ϕ \times 2 cm).

Results

Reactivity between the test compounds and copper

Reactivity of the test compounds (empenthrin, tefuramethrin and their mixed formulations) to the copper plate are presented in Table 1. A formulation of empenthrin reacted with the copper plate, and developed a shiny yellow-

greenish oil on the surface of the plate and on the applied cardboard. No reactions were observed between a formulation of tefuramethrin and the copper plate. In proportion to the gradual increase in the mixing ratio of tefuramethrin, it was found that the reaction between empenthrin and copper was more strongly inhibited. Tefuramethrin completely inhibited the reaction at the mixing ratio of 70% or more. We also found that such inhibition was an effect specific to tefuramethrin, and that inhibition was not dependent on the absolute amount of tefuramethrin, but on its mixing ratio.







Topical insecticidal activity of the test compounds

The basic efficacy of tefuramethrin on houseflies and fabric pests measured by the topical application method is shown in Table 2 and in Table 3, respectively. The LD₅₀ value of tefura-

Table 2 LD₅₀ values of tefuramethrin and empenthrin against the housefly

Compounds	LD ₅₀ (μ g/insect)
Tefuramethrin	0.21
Empenthrin	0.19

Table 1 Reactivities of the test compounds with copper plates

Compounds	Formulations (mg A. I.)					
	0	0	30	50	70	100
Tefuramethrin	0	0	30	50	70	100
Empenthrin	30	100	70	50	30	0
Reactivity						
	+	+	+	+	-	-

+ : reaction between the treated cardboard and the copper plate was observed.

- : reaction between them was not observed.

Table 3 LD₅₀ values of tefuramethrin and empenthrin against the larvae of fabric pests by the topical application method

Fabric pests	Compounds	LD ₅₀ (μ g/insect)
Casemaking clothes moth	Tefuramethrin	1.7
	Empenthrin	0.7
Black carpet beetle	Tefuramethrin	2.7
	Empenthrin	0.6
Varied carpet beetle	Tefuramethrin	0.3
	Empenthrin	0.1

methrin for the adult female housefly was 0.21 μ g/housefly, which was approximately the value for empenthrin (0.19 μ g/housefly). The LD₅₀ values for three kinds of fabric pests were slightly higher than those of empenthrin.

Volatile insecticidal activity of the test compounds

The volatile effect of tefuramethrin on houseflies measured by the vapor action method is shown in Table 4. The KT₅₀ values of tefuramethrin for the housefly were quite near to those of empenthrin. However, at low dosages, mortality ratios of tefuramethrin after 48 hours were inferior to those of empenthrin.

Table 5 and Table 6 show the volatile effect of tefuramethrin on the larvae of the casemaking clothes moth and black carpet beetle, respectively, measured by the same method. The

KT₅₀ values of tefuramethrin for the two species of the fabric pests were near those of empenthrin. However, the mortality ratios of tefuramethrin for the black carpet beetle after 72 hours were lower than those of empenthrin.

Discussion

The results indicate that tefuramethrin has vaporific insecticidal activities on houseflies and fabric pests. Tefuramethrin is a derivative of furamethrin (Fig. 3), which was developed by KATSUDA (1970). The vapor pressure of furamethrin is lower than that of empenthrin, but furamethrin has higher vapor pressure compared with allethrin, and has been used as an active ingredient in household insecticide formulations for thermal dispenser, such as mosquito coils and electric mosquito killers (mat-

Table 4 KT₅₀ values and mortality rates of the test compounds against the housefly by the vapor action method

Compounds	Content (mg/dish)	KT ₅₀ (min)	% Mortality at 48 hrs.
Tefuramethrin	1	4.8	100.0
	0.1	5.8	100.0
	0.01	7.0	40.0
Empenthrin	1	4.7	100.0
	0.1	6.0	100.0
	0.01	7.4	100.0
Untreated	—	—	3.3

Table 5 KT₅₀ values and mortality rates of the test compounds against the larvae of casemaking clothes moth by the vapor action method

Compounds	Content (mg/dish)	KT ₅₀ (min)	% Mortality at 72 hrs.
Tefuramethrin	1	30	100.0
	0.1	35	100.0
	0.01	53	100.0
Empenthrin	1	20	100.0
	0.1	23	100.0
	0.01	43	100.0
Untreated	—	—	5.0

Table 6 KT₅₀ values and mortality rates of the test compounds against the larvae of black carpet beetle by the vapor action method

Compounds	Content (mg/dish)	KT ₅₀ (min)	% Mortality at 72 hrs.
Tefuramethrin	1	57	78.1
	0.1	72	51.3
	0.01	228	0.0
Empenthrin	1	58	100.0
	0.1	75	100.0
	0.01	>240	0.0
Untreated	—	—	0.0

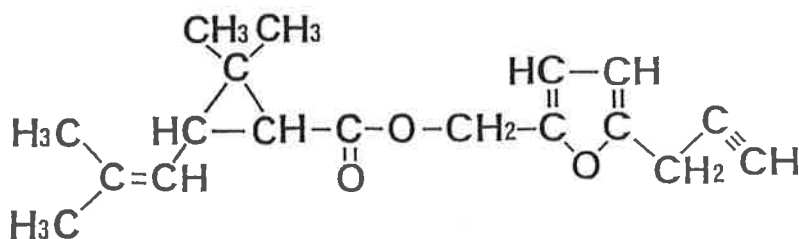


Fig. 3 Furamethrin (5-propargyl-2-furylmethyl chrysanthemate).

type and liquid-type).

Though insecticidal activities of tefuramethrin toward fabric pests were found to be slightly lower than those of empenthrin, its effectiveness to inhibit the reaction between empenthrin and copper was observed. Tefuramethrin or its mixed formulations with empenthrin at a

certain mixing ratio strongly promise to provide a novel type of vaporific mothproofing agents having no undesirable reactions with copper-containing materials as well as strong insecticidal activities against fabric pests. Adopting the mixed formulations, practical useful mixing ratios of tefuramethrin with

empenthrin are 70% or more.

Acknowledgements

We are grateful to the staff of the biological department in the laboratory for their cooperation and support.

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昆虫成長制御剤ピリプロキシフェン 水溶性粒剤のチカイエカおよびアカイエカ に対する防除効果

川田 均¹⁾・小浜 卓司²⁾・安部八洲男¹⁾

1) 住友化学工業(株)農業化学品研究所

2) 有恒薬品工業(株)

(受領：1994年5月25日；受理：1994年6月29日)

Larvicidal Activity of a Water-soluble Granular Formulation of the Insect Growth Regulator, Pyriproxyfen, against *Culex* Mosquitoes. Hitoshi KAWADA, ¹⁾Takuji KOHAMA ²⁾ and Yasuo ABE¹⁾ (¹⁾Agricultural Chemicals Research Laboratory, Sumitomo Chemical Co., Ltd., Takarazuka, Hyogo 665, Japan; ²⁾Yuko Yakuhin Kogyo Co., Ltd, Nishinomiya, Hyogo 663, Japan). *Jpn. J. Environ. Entomol. Zool.* 6 : 68-77 (1994)

Larvicidal activity of a water-soluble granular formulation (WSG) of an insect growth regulator, pyriproxyfen, was evaluated under laboratory and field conditions. IC₅₀ values (concentration required for 50% inhibition of emergence of mosquitoes) for *Culex pipiens pallens* COQUILLET (Gose strain) and *C. pipiens molestus* FORSKAL (OP resistant strain) were in the same range, between 0.01 and 0.1 ppb as an active ingredient (AI). Effective duration of pyriproxyfen WSG against *C. pipiens pallens* at the rate of 0.01 ppm as AI under semi-field conditions with sunlight was less than 10 days, which was nearly the same as that with an EC (emulsifiable concentrate) formulation. A field test was done in basement rooms of a post office in Osaka city. Treatment with pyriproxyfen WSG at the rate of 0.01 ppm as AI accompanied with an ULV (Ultra Low Volume) space spray of permethrin suppressed the population growth of larvae and adult *C. pipiens molestus* significantly more for more than 1 month compared to that of permethrin ULV spray only. Effective duration with 0.01 ppm of pyriproxyfen WSG was 2 to 3 weeks by a bioassay with samples of water from the test area.

Key Words: Mosquito, Pyriproxyfen, Insect growth regulator, Efficacy

昆虫成長制御剤ピリプロキシフェンの水溶性粒剤 (WSG) の効力を室内、準実地および実地条件で評価した。アカイエカ (御所系) およびチカイエカ (西宮市産、有機リン剤抵抗性系) 幼虫に対する50%羽化阻害濃度はいずれも0.01 ppb から0.1 ppbの範囲にあり、両者の感受性差は認められなかった。太陽光照射下の温室内での、準実地的な条件におけるピリプロキシフェン水溶性粒剤の有効期間は10日以内であり、乳剤処理によるそれとほぼ同等であった。大阪市内の郵便局地下の貯水槽においてチカイエカを対象とした野外試験を行った結果、ベルメトリン乳剤のULV (超微粒子) 散布のみでは1週間以内に個体数の回復が見られたのに対し、ピリプロキシフェン水溶性粒剤はベルメトリン乳剤のULV散布による成虫駆除と併用することにより、0.01 ppmの濃度で1カ月以上にわたってチカイエカ幼虫および成虫の密度上昇を抑えた。現地からの採取水による生物検定により、ピリプロキシフェン水溶性粒剤の幼虫に対する有効期間は0.01 ppmで2~3週間であることがわかった。

はじめに

昆虫のアラタ体より分泌される幼若ホルモン (JH) は、前胸腺より分泌される脱皮ホルモンと共に作用し、昆虫の変態を司る働きを持つ。昆虫は幼虫から蛹、あるいは若虫から成虫への劇的な変態が行われる終齢幼虫期に最もJHに対する感受性が強くなり、この時期に過剰のJHを与えると、蛹死、変態異常、過剰脱皮等の羽化阻害現象がみられる。JHは昆虫に特異的なホルモンであり、哺乳動物や魚に対する毒性が低いことから、これを殺虫剤として使用することを目的とした幼若ホルモン様物質 (JHM) の合成研究、およびその応用研究が数多くなされてきた。これらの研究によって見出された化合物の多くは、環境中での安定性の低さや、製造コスト、そして殺虫剤としての効力が不十分であったことなどの問題から実用までには至っていない。その中で既に実用に供されているJHMとしては、メトプレンやハイドロプレン (HENRICK *et al.*, 1976)、フェノキシカーブ (DORN *et al.*, 1981)、そしてピリプロキシフェン (HATAKOSHI *et al.*, 1987) が代表的なものである。メトプレンはハエ、蚊等の双翅目昆虫やノミ類に、またハイドロプレンはゴキブリに高いJH活性を示すが、これら2種のJHMがもっぱら衛生害虫防除の分野に適用されてきた最大の理由は、このような天然JHの構造的模倣物 (テルペン系) の外界での安定性が低く、農業用の分野まで適用できなかったことにある。

ピリプロキシフェンはメトプレンとは著しく構造の異なる4-フェノキシフェノキシ構造を有するピリジルエーテル化合物であり、ハエ、蚊、ユスリカ、ゴキブリ、ノ

ミ、アリなどの衛生不快害虫に対し極めて高い羽化阻害活性を示すばかりでなく (BANKS and LOFGREN, 1991; KAWADA *et al.*, 1987a, 1987b, 1989, 1993; PALMA and MEOLA, 1990; 千保ら, 1993)、その極めて高い活性と優れた安定性のために、アブラムシ、カイガラムシ、オンシツコナジラミ等の農業害虫に対しても高い防除効果を示すことが認められている (COOPER and OETTING, 1985; ISHAAYA and HOROWITZ, 1992)。著者らは、衛生害虫、特にハエ、蚊に対するピリプロキシフェンの活性を最大限に生かす製剤形態として、粒剤の開発を進めてきた。粒剤は特殊な散布機を必要とせず容易に散布可能で、有効成分の放出制御が可能であることを特徴としており、これまで多くの実地試験により、ピリプロキシフェン粒剤の優れた防除効果が実証されてきている (ALI *et al.*, 1993; KAMIMURA and ARAKAWA, 1991; KAWADA *et al.*, 1987a, 1987b; KERDPIBULE, 1989; 森川ら, 1990; MULLA *et al.*, 1986; MULLA and DAWAZEH, 1988; OKAZAWA *et al.*, 1991; SCHAEFER *et al.*, 1988; SUZUKI *et al.*, 1989)。しかし一方で粒剤は、1) 広域な散布には適さない、2) 浄化槽等に散布した場合、担体である砂粒が残留するなどの欠点を有しており、何らかの改善が必要とされてきていた。そこで著者らは、上記の問題を解決するために、従来の粒剤の特徴をある程度供え、かつ水希釈液の状態でも広域に散布可能な水溶性粒剤を開発した。本稿では、この水溶性粒剤を使用したアカイエカ、およびチカイエカに対する防除試験結果を報告し、従来の粒剤との効力的な差違について考察を行う。