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BIOLOGICAL AND PHYSICOCHEMICAL PROPERTIES OF FENITROTHION MICROCAPSULES AS A RESIDUAL SPRAYING FORMULATION FOR MOSQUITO CONTROL

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ABSTRACT. Insecticidal properties of fenitrothion microcapsules (MC[3]) as a new residual spraying formulation for mosquito control were studied. Adult female *Anopheles albimanus* were confined so as to contact several surfaces treated with fenitrothion MC or wettable powder (WP) for 10 and 30 min. Residual efficacy of MC at the rate of 1 g/m^2 was almost equivalent to that of WP at the rate of 2 g/m^2 on a plywood surface and was superior to that on an unglazed pottery surface. Greater than 50% mortality was maintained for more than 28 wk after treatment by fenitrothion MC on a mud surface with 10 min of contact at the rate of 2 g/m^2. The effective duration on a mud surface treated with WP was less than 44 wk with 30 min of contact and was less than 12 wk with 10 min of contact at the rate of 2 g/m^2. Microcapsule particles were observed to be mechanically broken (trampled) by mosquito contact, and the amount of fenitrothion released from MC increased as contact time increased. Comparison of MC with WP and emulsifiable concentrate formulations in this study indicates that the increased residual activity results from protection from evaporation and decomposition afforded by capsule walls of the MC formulation.

INTRODUCTION

Microencapsulation of insecticides is beneficial from biological, toxicological, and environmental points of view. Interfacial polymerization is one of the most convenient and suitable means of microencapsulation. Utilization of this technology means diameter and wall thickness of the microcapsule particle can be controlled to suit the manufacturer's purpose. Through a number of screenings of samples with various diameters and wall thicknesses, we have developed 3 different types of microencapsulated formulations, each of which has its own particular design, for residual spraying against pests of household and medical importance: these are 20% fenitrothion (Sumithion®) and 10% cyphenothrin (Gokilaht®) for cockroach control, and 20% fenitrothion for mosquito control.

The entry of the active ingredient contained in a microcapsule particle into cockroaches is achieved by the trampling of microcapsule particles and subsequent oral intake during grooming behavior. It was found that fenitrothion microcapsule particles for mosquito control have a fumigation effect due to the high vapor pressure of the active ingredient, a different mode of action from that seen in cockroach control (Ohtsubo et al. 1987, Tsuda et al. 1987, Kawada et al. 1990).

In this paper, we report on the short-contact efficacy of microencapsulated fenitrothion against anopheline mosquitoes and on the physicochemical properties of microencapsulated fenitrothion for mosquito control. We also discuss the similarities and differences in the modes of entry from those for cockroach control.

MATERIALS AND METHODS

Microencapsulation procedure: Microcapsules (MC) containing 20% fenitrothion (*O, O*-dimethyl *O*-4-nitro-m-tolyl phosphorothioate) were prepared by interfacial polymerization using polyurea as a wall material (Ohtsubo et al. 1987, Tsuda et al. 1987).

Bioassay—residual confined contact test: Three different kinds of panels (plywood, unglazed pottery, and mud [10 parts fine textured yellow soil, 2 parts plaster, and 10 parts water mixed and dried for more than 7 days at 25°C]) were cut into 15 x 15-cm squares. The 20% microencapsulated (20MC) and 40% wettable powder (40WP) formulations of fenitrothion were suspended in deionized water, and each suspension was sprayed on each type of panel at the rate of 50 ml/m^2. Panels were dried for 24 h at 25°C. Three-day-old adult female *Anopheles albimanus* Wied. (a susceptible strain introduced from The University of London in 1984) were used for the bioassay. The LD_{50}s of insecticides by topical application for this strain are 0.00054 μg/female

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[3] In this paper, in accordance with the authors' usage, MC is used as the abbreviation for microencapsulated product; in the USA MC is the abbreviation for miscible concentrate and ME is the abbreviation for microencapsulated product. Ed.

Table 1. Residual 10-min contact efficacy of fenitrothion microencapsulated and wettable powder formulations against *Anopheles albimanus* on various surface materials.

Formulation	Dosage (g/m^2 AI)	Material	% mortality at different weeks after treatment			
			0	4	12	20
Microencapsulated	2	Plywood	100.0	100.0	100.0	100.0
		Unglazed	100.0	100.0	100.0	100.0
		Mud	100.0	96.3	96.7	80.0
	1	Plywood	100.0	100.0	100.0	100.0
		Unglazed	100.0	100.0	100.0	100.0
Wettable powder	2	Plywood	100.0	100.0	100.0	100.0
		Unglazed	100.0	100.0	100.0	96.6
		Mud	100.0	53.6	6.7	—
	1	Plywood	100.0	100.0	100.0	100.0
		Unglazed	100.0	100.0	90.0	26.7

with permethrin, 0.0082 μg/female with DDT, 0.0075 μg/female with malathion, and 0.0050 μg/female with fenitrothion (Kawada et al. 1993a).

Ten mosquitoes were confined on each treated surface with a Petri dish (10 cm diam and 1 cm height) for 10 and 30 min. Insects were then transferred into a clean plastic cup with 5% sugar solution *ad libitum,* and mortality was recorded after 24 h. Panels were stored at 25°C and 60% RH. Residual efficacy was examined by using the same panels in the manner described above. Three replicates were made at each dose. At each test an untreated control was prepared and all data were excluded from results when more than 5% mortality was recorded in the untreated control. Correction with Abbott's formula for mortality was not done.

Bioassay—determination of effective dosage by confined contact test: The 20MC, 40WP, and 10% emulsifiable concentrate (10EC) formulations of fenitrothion were suspended in or diluted with deionized water and the suspensions sprayed on polyethylene terephtharate panels at the rate of 50 ml/m^2. Panels were dried for 24 h at 25°C. Three-day-old adult female *Culex pipiens pallens* Coq. (Gose strain) were used for the bioassay. The LD_{50}s of insecticides by topical application for this strain are 0.0042 μg/female with permethrin, 0.12 μg/female with DDT, 0.0022 μg/female with malathion, and 0.0062 μg/female with fenitrothion (Kawada, unpublished data). Ten mosquitoes were confined to contact each treated surface for 30 min and mortality was recorded in the manner described above. The LD_{50}s were calculated with Bliss's probit analysis (Bliss 1934). Three replicates were made at each dose.

Determination of evaporation rate of fenitrothion: The 20MC, 40WP, and 10EC formulations of fenitrothion were suspended in or diluted with deionized water and the suspensions sprayed on an overlaid plywood panel at the rate of 50 ml/m^2. After the panels were dried for 24 h at 25°C, they were preserved in 2 different

Table 2. Residual 30-min contact efficacy of fenitrothion microencapsulated and wettable powder formulations against *Anopheles albimanus* on various surface materials.

Formulation	Dosage (g/m^2 AI)	Material	% mortality at different weeks after treatment			
			0	4	12	20
Microencapsulated	2	Plywood	100.0	100.0	100.0	100.0
		Unglazed	100.0	100.0	100.0	100.0
		Mud	100.0	96.7	90.0	83.3
	1	Plywood	100.0	96.7	100.0	100.0
		Unglazed	100.0	100.0	100.0	100.0
Wettable powder	2	Plywood	100.0	100.0	100.0	100.0
		Unglazed	100.0	100.0	100.0	100.0
		Mud	100.0	76.9	27.6	69.0
	1	Plywood	100.0	100.0	100.0	100.0
		Unglazed	100.0	100.0	100.0	58.6

Table 1. Extended.

% mortality at different weeks after treatment			
28	36	44	52
100.0	100.0	100.0	96.6
100.0	96.7	93.3	100.0
86.7	37.9	—	—
93.3	90.0	80.0	67.9
100.0	100.0	100.0	100.0
100.0	100.0	100.0	82.8
10.0	—	—	—
—	—	—	—
93.3	83.3	90.0	6.9
—	—	—	—

conditions: 1) in an open room condition at 25°C and 60% RH, and 2) in an enclosed condition in an aluminum bag at 25°C. After storage each panel was immersed in 50 ml of acetone and sonicated for 10 min for extraction of fenitrothion. The amount of fenitrothion on the panel was analyzed by a gas chromatograph equipped with a flame photometric detector, under the following conditions: glass column, 1.1 m × 3 mmϕ, packed with 3% XE-60 on Chromosorb W (AW, DMCS) 60–80 mesh; carrier gas, nitrogen; injection temperature, 230°C; column temperature, 170°C. The percentage of fenitrothion that decomposed during storage and the percentage that evaporated were calculated as follows: % decomposed = 100 − % residue in closed condition; % evaporated = % residue in closed condition − % residue in open condition. Two replicates were made for each sample.

Determination of breakdown rate of microcapsule particle wall by contact with mosquitoes: The 20MC formulation was suspended in deionized water and was applied to a Petri dish (8.6 cm diam) at the rate of 1 g/m² active ingredient. After the Petri dish was dried for 24 h at 25°C, 50 3-day-old adult female *Cx. pipiens pallens* were confined so as to contact the treated surface in the manner described above. The amount of fenitrothion outside and inside the microcapsules on the treated surface and the amount of fenitrothion picked up by the insects were analyzed as described by Tsuda et al. (1987) after extraction with deodorized kerosene (Neochiozol®, Chuoo Kasei Co., Ltd., Japan) and acetone. Analysis was done by a gas chromatograph equipped with a flame photometric detector under conditions described above. Two replicates were made for each sample.

Table 2. Extended.

% mortality at different weeks after treatment			
28	36	44	52
100.0	100.0	100.0	100.0
100.0	100.0	100.0	100.0
80.0	53.3	80.0	24.1
100.0	100.0	100.0	56.7
100.0	100.0	100.0	100.0
100.0	100.0	100.0	100.0
96.7	93.5	60.0	0.0
60.0	—	—	—
93.3	90.0	100.0	3.4
—	—	—	—

Observation of microcapsule particles on the treated surface: Particles of the microcapsules on the treated surface were observed with a scanning electron microscope (HITACHI Ltd., S-2400) or a polarized light microscope.

RESULTS AND DISCUSSION

Residual short-time contact efficacy of fenitrothion MC against anopheline mosquitoes: Residual 10-min contact efficacy of fenitrothion MC and WP on the different surfaces is shown in Table 1. More than 80% mortality was maintained for more than 52 wk with MC and WP sprayed on plywood panels at 2 g/m² and for more than 44 wk at 1 g/m². Mortality was reduced through 20 wk and 12 wk at 2 g/m² and 1 g/m², respectively, on unglazed pottery with WP. With MC, on the other hand, no reduction in mortality was observed in 52 wk at 1 g/m². Reduction in mortality was least effective with the WP treatment on mud surface; the effective duration of WP on mud surface was less than 12 wk at 2 g/m². The MC treatment on a mud surface provided higher mortality for more than 28 wk at 2 g/m².

Residual 30-min contact efficacy of fenitrothion MC and WP on the different surfaces is shown in Table 2. High mortality was maintained for more than 52 wk with MC and WP sprayed on plywood panels at 2 g/m² and for more than 44 wk at 1 g/m². Mortality lasted through 36 wk and 12 wk at 2 g/m² and 1 g/m², respectively, on unglazed pottery with WP. However, with MC no reduction in mortality was observed in 52 wk at 1 g/m² on unglazed pottery. A lowered level of mortality with the WP treatment on a mud surface was also observed, that is, the effective duration of WP on a mud surface was less than 4 wk at 2 g/m². The MC treatment on a mud surface maintained high mortality for more than 28 wk at 2 g/m². There seemed to be little difference in the active duration between 10- and 30-min contact.

In an earlier paper we reported the effective duration of fenitrothion MC and WP against *Cx.*

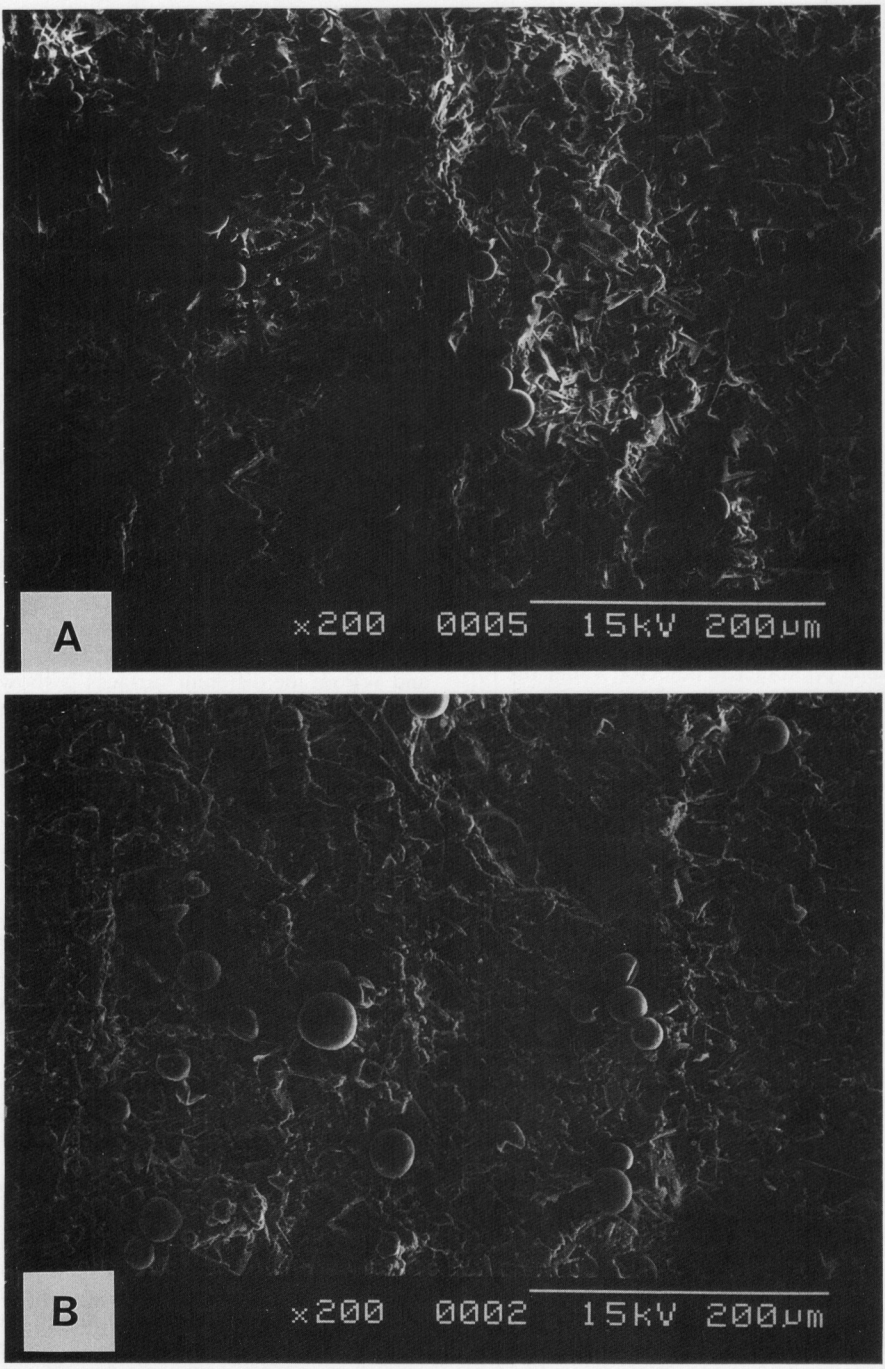

Fig. 1. Surface appearance of mud panels by SEM. A. One day after spraying; B. One month after spraying of fenitrothion MC. White bar indicates 200 µm.

Table 3. Evaporation rate of fenitrothion from overlaid plywood surface with microencapsulated, wettable powder, and emulsifiable concentrate formulations.

Formulation	% after 1 month				% after 6 months			
	Remain in open condition	Remain in closed condition	Decomposed[1]	Evaporated[2]	Remain in open condition	Remain in closed condition	Decomposed	Evaporated
Microencapsulated	97.7	100.0	0.0	2.3	66.7	100.0	0.0	33.3
Wettable powder	87.3	100.0	0.0	12.7	45.8	85.6	14.4	39.8
Emulsifiable concentrate	82.0	100.0	0.0	18.0	34.2	88.8	11.2	54.6

[1] % decomposed = 100 − % residue in closed condition.
[2] % evaporated = % residue in closed condition − % residue in open condition.

pipiens pallens by 2-h contact to be much longer than those in the present paper (Kawada et al. 1994). The susceptibilities of the 2 species to fenitrothion is thought to be in the same range: LD_{50} by topical application is 0.0050 μg/female for *An. albimanus* (Kawada et al. 1993a) and 0.0062 μg/female for *Cx. pipiens pallens* (Kawada, unpublished data). The difference in residual activities, therefore, seems simply to be due to the difference in contact time. Resting time of mosquitoes after a blood meal seems to range from a few minutes to several hours. High activity in short-term contact is necessary for obtaining adequate control. In our experiment 10-min contact with the surfaces treated with fenitrothion MC was found to be enough for controlling mosquitoes for more than half a year on absorptive surfaces such as mud walls.

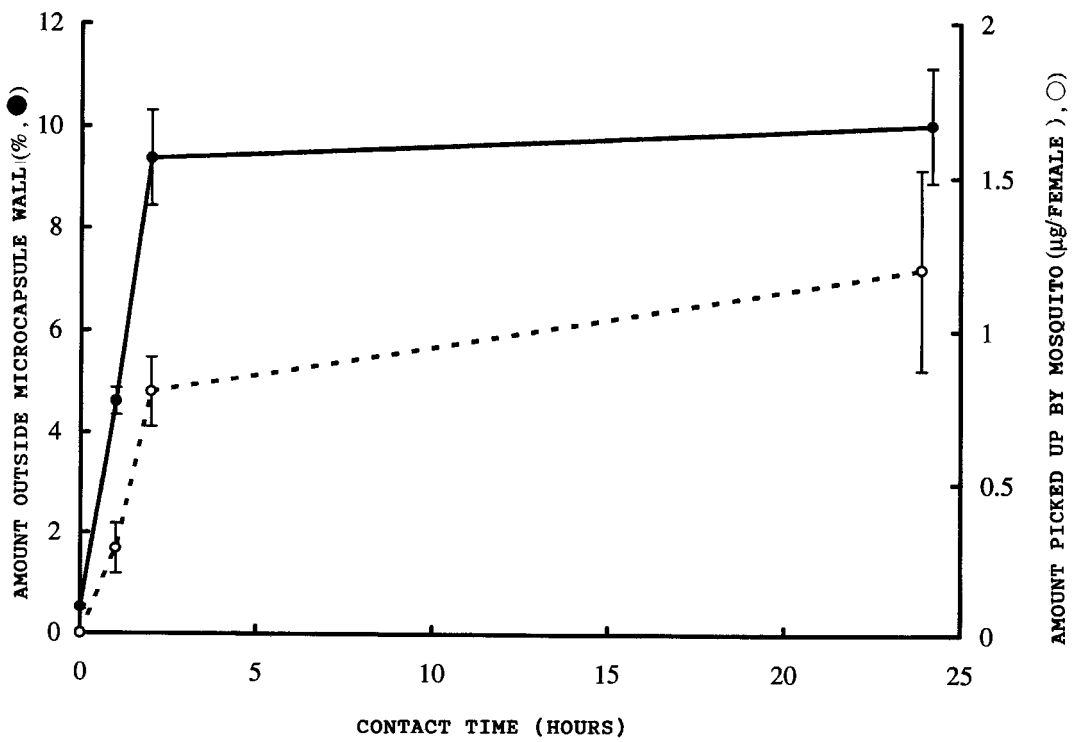

Fig. 2. Changes in the amount of fenitrothion outside the microcapsule wall (●) and that picked up by an adult female *Culex pipiens pallens* mosquito (○) after contact with a glass surface on which fenitrothion MC was applied at the rate of 1 g/m² as active ingredient. Each bar indicates 95% fiducial limit.

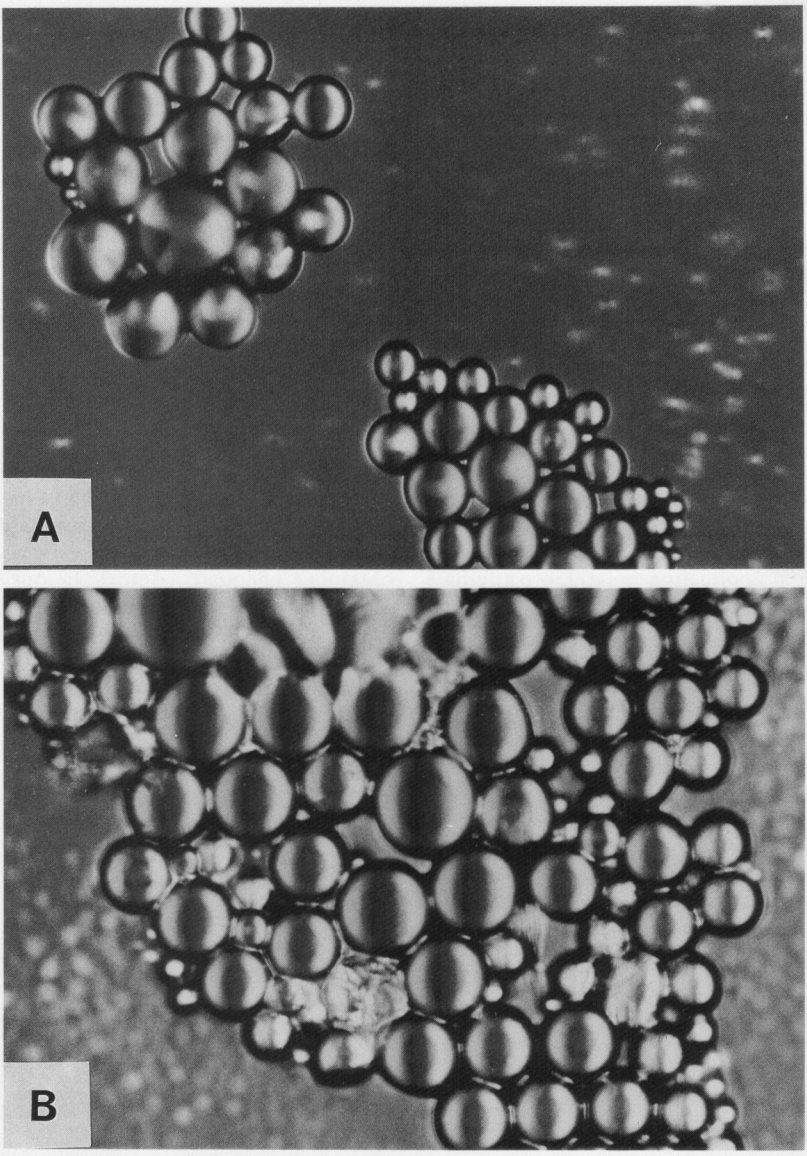

Fig. 3. Microcapsule particles observed by a polarized light microscope on a glass surface on which fenitrothion MC was applied at the rate of 1 g/m² as active ingredient. A. Before contact; B. One hour after contact of *Culex pipiens pallens.*

Effective dosage of fenitrothion for mosquitoes: The LD_{50}s of fenitrothion MC, WP, and EC against *Cx. pipiens pallens,* given as active ingredient, were 1.25 mg/m² with MC, 2.5–5.0 mg/m² with WP, and 3.54 mg/m² with EC. This result was contrary to what we had anticipated because we believed discharge of the active ingredient would be regulated by the microcapsule wall. It is suggested that the amount of fenitrothion picked up by mosquitoes will be higher in MC treatment than in the other formulations. The pick up of microcapsule particles, trampling rate, and evaporation rate of fenitrothion in MC are presented below.

Evaporation rate of fenitrothion from the treated surfaces: Results are shown in Table 3. We regarded the percentage of fenitrothion decomposed during storage as that which remains when the percentage of residue in the closed condition is subtracted from 100 and the percentage evaporated as that which remains when the percentage of residue in the open condition

is subtracted from the percentage of residue in the closed condition. Photodegradation was thought to be negligible as preservation of samples was done in relatively dark conditions. The percentage of fenitrothion remaining in the open condition was the highest in the MC formulation of the 3 formulations, and no decomposition of fenitrothion in the MC formulation was observed, whereas more than 10% decomposition was observed in the WP and EC formulations after 6 months of storage. Similarly, the percentage of fenitrothion evaporated was the least in the MC formulation. The total loss of fenitrothion was the highest in the EC formulation. Kawada et al. (1994) attributed the higher residual activity of the MC formulation to the stability of fenitrothion in the MC on the treated substratum. The present results not only support their conclusion but also indicate that stability results from protection of the active ingredient from decomposition and evaporation. The contribution of microencapsulation to extension of residual activity is thought to be 21.3% more against evaporation and 11.2% more against decomposition as compared with the EC formulation, and 6.5% more against evaporation and 14.4% more against decomposition as compared with the WP formulation. The MC formulation, therefore, provides greater protection of an active ingredient from decomposition compared to EC and WP formulations. This seems to explain the fact that the MC formulation has an evaporatively generated airborne effect against mosquitoes, as does the WP formulation (Kawada et al. 1994).

Microcapsule particles on the mud surface at 1 day after and 1 month after treatment are shown in Fig. 1. Almost all particles were found to remain on the treated surface although some particles appeared to be deflated after 1 month of storage in room conditions. A part of fenitrothion was, therefore, thought to evaporate through the capsule wall. However, most of fenitrothion was thought to remain on the surface without evaporation and decomposition.

Breakdown rate of the microcapsule wall by the contact of mosquitoes: Changes in the amount of fenitrothion outside the microcapsule wall and in that picked up by a mosquito by the contact of *Cx. pipiens pallens* adults on the MC-treated glass surface are shown in Fig. 2. Sufficient amounts of fenitrothion to kill a mosquito (more than 0.1 μg/female) was picked up by a mosquito within 1 h. Both the amount of fenitrothion picked up by a mosquito and that outside the microcapsule increased as contact time increased. The result seems to suggest trampling of capsules by the mosquito, although the test condition was rather different from the practical one.

Microcapsule particles on the glass surface without contact and those after 1-h contact with mosquitoes are shown in Fig. 3. Destruction of capsule particles was obvious on the surface after contact with mosquitoes. Kawada et al. (1993b) reported that pyrethroid (cyphenothrin) MC for cockroach control has to be "softer" than organophosphate (fenitrothion) MC for cockroach control because of the different modes of action of the 2 insecticides. The diameter/wall thickness ratio, proposed by Ohtsubo et al. (1987) as a parameter by which the strength of a MC is expressed, for fenitrothion MC for mosquito control in the present paper is greater than that of fenitrothion MC for cockroach control. The design of the MC is, therefore, similar to that of cyphenothrin MC for cockroach control, and it may be concluded that a different design is required for each MC according to the insect species. In conclusion, the breakdown of MC particles by contact was found to be one of the main modes of entry of MC fenitrothion for mosquito control, although it is still unclear what proportion of fenitrothion acts as a contact toxin and what proportion acts as a vapor toxin.

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