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Original Article

Microencapsulated Fenitrothion
Formulation and Characteristics

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Microencapsulated Sumithion was developed for vector control use by interfacial polymerization method in which polyurethane was adopted as capsule wall material. Microencapsulated Sumithion showed superior initial and residual efficacy to conventional Sumithion emulsifiable concentrate and also equivalent or superior residual efficacy to Sumithion wettable powder. Microencapsulated Sumithion does not show any dustiness and contamination on sprayed surfaces which are drawbacks of wettable powder. It was confirmed that the larger Sumithion amount in microcapsules against the wall material gives better biological efficacy.

INTRODUCTION

Microencapsulated pesticides are generally one to a hundred micron diameter particles which are composed of pesticides (or pesticide solution) and wall materials. The wall is a thin polymeric membrane which is generally semipermeable. The pesticides, as core materials, can be released from the microcapsules by diffusing through the wall. The release rate of the pesticides from the microcapsules can be controlled by the chemical structure of the capsule wall, its thickness and the particle size of the microcapsules.

Encapsulated pesticides have the following characteristics.

(1) Improvement of residual activity
(2) Stabilization of pesticides against environmental degradation
(3) Decrease in mammalian toxicity
(4) Decrease in chemical reactivity
(5) Occlusion of odor
(6) Decrease in human mucous membrane irritation
(7) Decrease in phytotoxicity
(8) Decrease in drift phenomena
(9) Solidification of liquid pesticides

Microencapsulated pesticides are generally formulated as water-based suspensions or microcapsule concentrate, which can be diluted with water prior to application. They can be applied by spraying with the conventional spraying equipment.

Some encapsulated pesticides are already on the market such as Pencap M® (methylparathion),[1] Knox out 2 FM® (diazinon)[2,3] and Sectrol TM® (pyrethrin).[4] Characteristics of these encapsulated formulations are decrease of mammalian toxicity and longer residual activity. Several works are also reported on encapsulated pesticides with starch membrane.[5]

We are now carrying out research work on encapsulation of pesticides.[6–8] In the case of encapsulated Sumithrin®, it was found that the encapsulated formulation gave consistent efficacy against german cockroaches irrespective of application surfaces.

In general, wettable powder (WP) is widely used for vector control purpose. In this case, however, there are problems of nuisance and hazards of handling pesticide dust, which takes place when WP is poured onto water for

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dilution. Contamination on the sprayed surface is also a problem. When emulsifiable concentrate (EC) is used instead of WP, these kinds of problems are solved, but biological efficacy is insufficient because of absorption into the sprayed surface. In order to develop an efficient formulation without these kinds of problems, encapsulated formulation of *O,O*-dimethyl *O*-(3-methyl-4-nitrophenyl) phosphorothioate (fenitrothion, Sumithion ®) were prepared.

In this paper, the relationship between the ratio of the active ingredient to wall materials and biological efficacy are dicussed.

MATERIALS AND METHODS

1. *Microencapsulation of Sumithion®*

According to our previous research work,[7] a polyurethan wall was found to be the most suitable for this microcapsule. Therefore the polyurethane microcapsules were used for this research.

Polyurethane microcapsules were prepared by interfacial polyaddition reaction of polyisocyanate (Adduct of toluene diisocyanate and trimethyrol propane) and ethylene glycol.

Table 1 shows the ingredients for microencapsulation of Sumithion®. Cyclohexanone was used in order to dissolve both Sumithion® and polyisocyanate. Polyisocyanate was charged at various rates to prepare microcapsules which had different ratios of the active ingredient (A.I.) to wall materials, while the amount of A.I. was constant.

The procedure for preparing the microcapsules is shown in Fig. 1. The first step of the process is dispersion of an organic mixture (containing Sumithion®, polyisocyanate and cyclohexanone) into a 2% aqueous solution of gum arabic which was added as a dispersing agent. Then the prescribed amount of ethylene glycol was added to the resulting dispersion under light agitation. The wall forming reaction was carried out for two hours at 60°C. The total weight of the resulting mixture was adjusted by addition of water according to the concentration listed in Table 1. The median diameter of the microcapsules was 40 μm to

Table 1 Components of microencapsulated Sumithion®.

Sample No.	1	2	3	4
Amount of components (g)				
Sumithion A.I.	12.0	12.0	12.0	12.0
Cyclohexanone	0.5	0.5	0.5	0.5
Polyisocyanate	0.8	0.4	0.25	0.15
Ethyleneglycol	0.35	0.35	0.35	0.35
Gum arabic	1.0	1.0	1.0	1.0
Water	Balance	Balance	Balance	Balance
Total	100.0	100.0	100.0	100.0

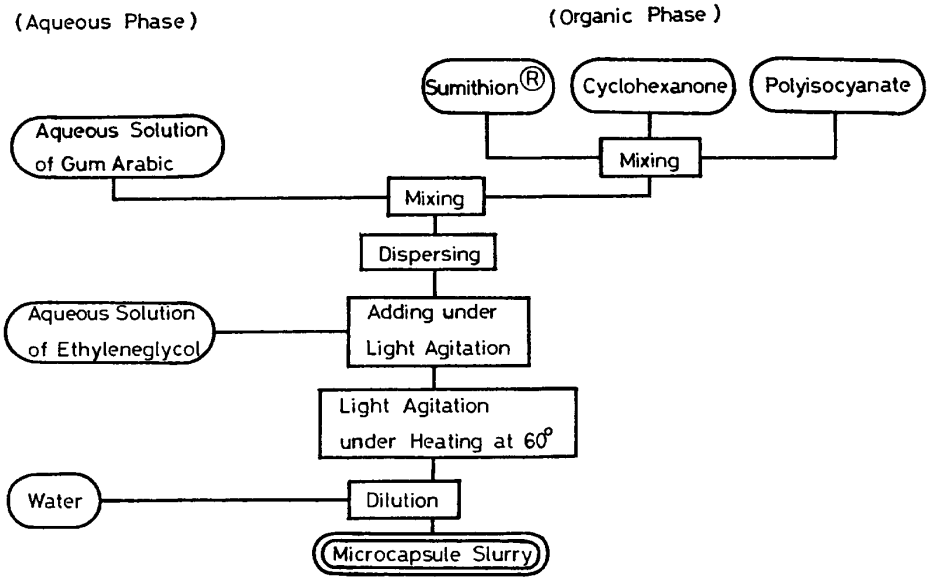

Fig. 1 Preparation process of encapsulated Sumithion® (Polyaddition reaction).

50 μm and the range of particle sizes was 5 μm to 110 μm.

2. Evaluation of Biological Efficacy against Mosquitoes (Culex pipiens pallens, female adults) by the Confined Contact Method

Each microcapsule concentrate was diluted with water to the prescribed concentration and applied uniformly at the rate of 40 ml/m² onto 3 pieces of plywood panel which were 3 cm in width and 15 cm in length. The panels were put together into a triangular shaped test holder, which had a treated inner surface. Two ends of the test holder were covered with nylon nets in order to prevent the insects from escaping.

The test holders were applied for biological efficacy measurement at two hours, two and four weeks after treatment. During the test period, the test holders were kept in the room at 25°C with 50–60% relative humidity.

Experiments were conducted in triplicate using 20 mosquitoes in each trial. After the exposure time, all the insects were transferred into a fresh container with 5% sugar solution as diet to observe mortality after 24 hr.

The efficacy of Emulsifiable Concentrate (EC) and Wettable Powder (WP) was also evaluated in the same way in order to compare with the microcapsule concentrate.

3. Evaluation of Biological Efficacy against the German Cockroach (Blattella germanica) by the Confined Contact Method

Each microcapsule concentrate was diluted with water to various concentrations and applied to the surfaces of plywood panels (15× 15 cm) with a microsyringe at the rate of 50 ml/m² (1.125 ml per panel).

The panels were applied for biological efficacy measurement at two hours, four, and eight weeks after treatment. During the test period, the panels were kept in the room at 25°C with 50–60% relative humidity.

Experiments were conducted in triplicate using 10 german cockroaches in each trial. The insects were released into a stainless steel ring 5 cm high and 10 cm in diameter, which was put on the treated panel. After two hours, they were transferred into the recovery containers for mortality counts (after 72 hr).

The efficacy of Wettable Powder (WP) was also evaluated in the same way in order to compare with the microcapsule concentrate.

4. Release Rate of Sumithion® from the Microcapsule Concentrates

Release rate of Sumithion® from the microcapsule concentrates were determined by chemical analysis according to the following procedure.

The microcapsule concentrates were diluted to 1% w/v A.I. with water. The dilution was applied uniformly into the bottom of four petri dishes (11 cm inner diameter) at the rate of 1 cc per dish (10 mg A.I. per dish).

The treated petri dishes were stored on a dark shelf at an ambient temperature (*ca.* 20°C) and were quickly rinsed with 5 ml of *n*-hexane per dish into a distilling flask at the end of prescribed periods (2 hr, 3 days, 7 days and 14 days, respectively). The four dishes corresponded to the series of storage periods.

The collected solutions were evaporated to near dryness by means of a rotary evaporator. The amount of Sumithion® recovered was equivalent to the amount of Sumithion® released during the prescribed period. These values were converted into the release rates (%) by dividing with 10 mg (*i.e.* total amount of treated Sumithion® per dish).

RESULTS AND DISCUSSION

Table 2 shows the efficacy of four kinds of microcapsules against mosquitoes in comparison with EC and WP. EC formulation shows low efficacy, especially residual effect. This could be caused by absorption of EC into the plywood panel.

Sample 4 has almost the same high biological efficacy as that of WP.

Moreover Fig. 2 shows that the release rate of A.I. through the microcapsule wall in sample 4 is the highest of the four kinds of microcapsules, especially at the initial stage.

It is apparent that the larger A.I. amount against the wall materials gives better biological efficacy. This is caused by a larger amount of diffusion of A.I. through the capsule wall.

In the case of sample 1 and sample 2, the capsule wall is too thick for the core materials

Table 2 Activity of encapsulated Sumithion® against mosquito by confined contact method on plywood panel (3 replication).

Sample	Dosage (A.I. mg/m²)	Age of deposit in weeks	% Mortality[a] at...contact time		
			5 min	15 min	30 min
No. 1	250	0	5	14	25
		2	2	4	14
		4	0	0	3
No. 2	250	0	49	57	80
		2	5	7	26
		4	0	0	12
No. 3	250	0	78	88	94
		2	30	33	59
		4	10	18	48
No. 4	250	0	86	95	100
		2	32	48	87
		4	5	27	72
WP	250	0	97	95	100
		2	43	62	87
		4	11	23	41
EC	250	0	29	78	80
		2	15	5	55
		4	4	7	18

[a] 24 hr after.

Table 3 Residual activity of encapsulated Sumithion® in comparison with WP against German cockroach by 2 hr confined contact method on plywood panel (3 replicates).

Sample	Dosage (A.I. mg/m²)	Mortality (%)		
		Initial	4 weeks	8 weeks
No. 3	0.25	100	33	0
	0.5	100	63	47
	1.0	100	100	100
No. 4	0.25	100	60	10
	0.5	100	93	80
	1.0	100	100	100
WP	0.25	100	40	3
	0.5	100	100	10
	1.0	100	100	100

to diffuse to the surface of the microcapsules.

Also, residual activity of microcapsules in sample 3 and sample 4 was evaluated in comparison with WP against German cockroaches by two hour confined contact on plywood panels. Table 3 shows that the efficacy of sample 4 is slightly better than that of WP against cockroaches.

Figure 3 is the microscopic picture of the microcapsules (sample 4) initially applied into the petri dish. Figure 4 is the one observed after 4 hr.

As water vaporised with the lapse of time, the particles became flat and the released Sumithion® was observed only around the particles.

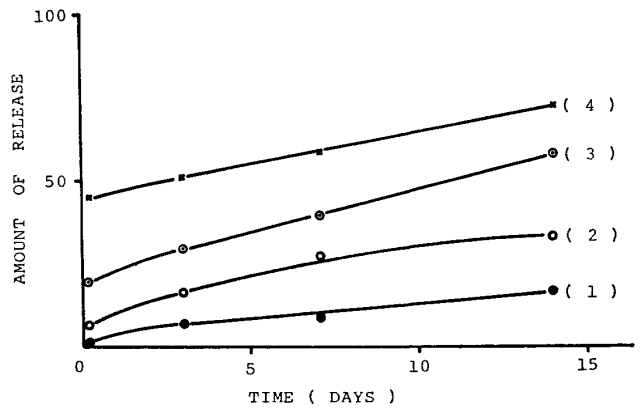

Fig. 2 Release behavior of Sumithion® from the microcapsules.

Numbers (1), (2), (3) and (4) correspond to the components in Table 1.

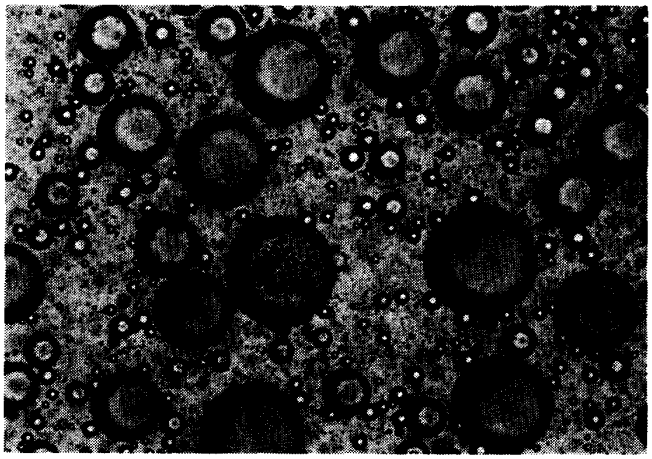

Fig. 3 Microscopic picture of the microcapsules (sample 4) initially applied into the petri dish.

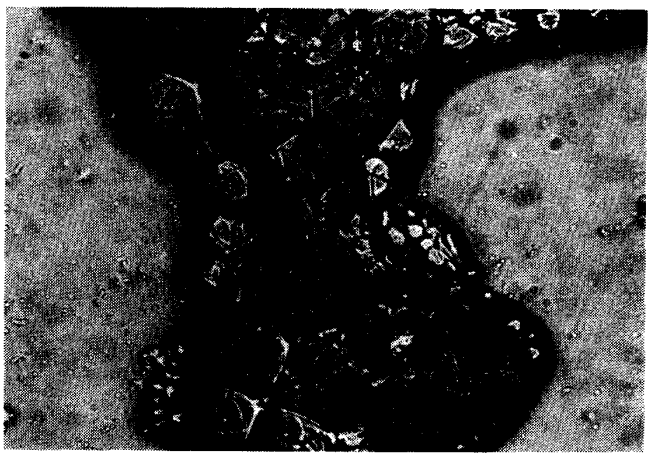

Fig. 4 Microscopic picture of the microcapsules (sample 4) after 4 hr.

Table 4 Decomposition of Sumithion® A.I. in microcapsule and WP after accelerated storage.*

Sample	Storage condition	Decomposition of A.I.
Microencapsulated Sumithion® (sample 4)	40°C after 30 days	0.25%
	50°C after 30 days	1.8 %
	60°C after 30 days	8.0 %
WP	40°C after 30 days	1.4 %
	50°C after 30 days	4.3 %
	60°C after 30 days	8.4 %

* in sealed glass-ampules.

Table 4 shows the stability of Sumithion® in the capsules after accelerated storage in sealed ampules in comparison with that of WP. Sumithion® in the capsules is as stable as that in WP.

From the results mentioned above, by microencapsulation techniques, good formulations can be developed which show equivalent or superior biological efficacy to that of WP without any of the drawbacks caused by pesticide dust.

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要　　約

マイクロカプセル化フェニトロチオン——製剤とその特性

麩山　浩，新庄五朗，辻　孝三

ポリウレタン膜を用いてスミチオンを界面重合法でマイクロカプセル化することによって衛生害虫用の新しい製剤を開発した．スミチオンマイクロカプセルは，従来の製剤である乳剤より高い初期効力および残効性を示し，水和剤に比べて同等もしくは，それ以上の残効性を示した．マイクロカプセルは，水和剤において見られるような，粉立ち，処理面の汚染等の取扱い上の問題がない．マイクロカプセル化されるスミチオンの量とマイクロカプセルの膜物質の量との関係はスミチオンの割合が増えるほど効力的に優れていることがわかった．