Microencapsulated animal repellents for agricultural applications

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Introduction

The development of animal repellent microcapsules is a relatively new scientific and industrial field. The vast majority of publications on microencapsulated repellents are patents (Figure 1), indicating the importance of industrial property rights, deriving from scientific research and development of marketable products. The literature typically describes microencapsulated insect repellents against mosquitoes, ticks, lice, mites, cockroaches and termites, and rodent repellents against rats, mice and voles. Publications on microencapsulated herbivore game repellents (deer, rabbits, hares) are scarce, as indicated in Figure 2.

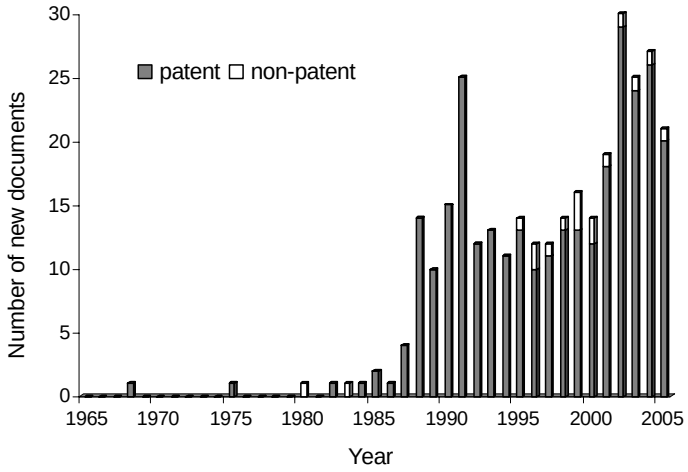


Figure 1: Bibliometric analyses of new publications on microencapsulated repellents (CAPlus database).

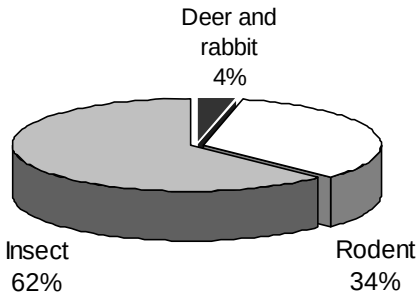


Figure 2: Ratio of publications on insect, rodent, and herbivore game repellents (CAPlus database).

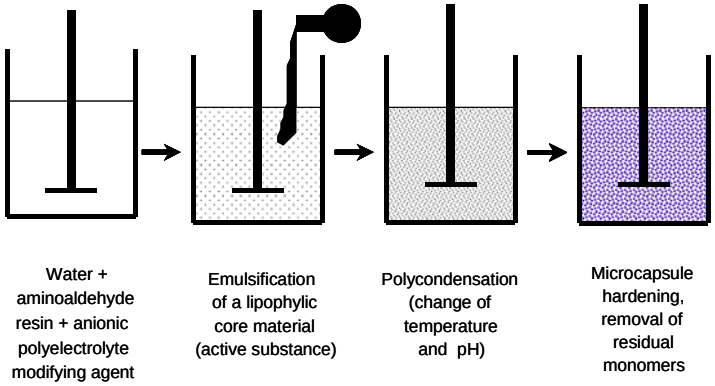
Some publications on the microencapsulation of repellents focus primarily on achieving controlled / prolonged release of active substances. Complex coacervation, interfacial polymerisation, *in situ* polymerisation, multilamellar liposome formation and impregnation of porous microparticles are the methods most often employed for this purpose. The majority of patents describe repellent microcapsules and formulations for specific applications, such as (1) protection of electric cables, wires and gas hoses; (2) protection of building and packaging materials (3) rodent and cockroach control; (4) skin applications against mosquitoes, ticks, and lice; (5) specialised repellent textiles; and (6) agricultural applications against deer, rabbits, hares, birds, rodents, insects and slugs.

Deer and rabbits are an important source of damage for agricultural and ornamental plants, especially in winter, when the quantity of natural food supplies becomes limited. To reduce the damage, several synthetic and natural animal repellent substances have been developed. Most of them are effective only for a short period of time, primarily due to high volatility and/or washing off by rain, thus requiring frequent re-application. In our work, two deer and rabbit repellents were microencapsulated in order to reduce the volatility and prolong their activity.

Materials and methods

Two active compounds were selected for microencapsulation: (1) Daphne (Symrise GmBH), a repellent based on a mixture of essential oils and other volatile compounds. Its main components are vanillin, heliotropin (3,4-methylenedioxybenzaldehyde), cyclamaldehyde [3-(4-isopropyl-phenyl)-2-methylpropanal], citronellol (3,7-dimethyl-6-octen-1-ol) and dimethylphthalate as a solvent; (2) *Psiadia punctulata* diterpenoid extract, which was obtained by ethyl acetate extraction of the resinous surface leaf exudate (Midiwo et al., 1979). *Psiadia punctulata* Vatke (synonym *P. arabica* Jaub & Spach) is a plant species from Eastern Africa, which is known to be avoided by browsing herbivores even during severe drought.

A modified *in situ* polymerisation method by Knez (1995) and Kukovič & Knez (1997) was used for the preparation of microcapsules (Figure 3). Partly methylated trimethylolmelamine and a hexamethoxymethylolmelamine resin (both procured from Melamin, Slovenia) were used as prepolymers for microcapsule walls, and a styrene-maleic acid anhydride copolymer with average mol. weight 350,000 (Hercules) as a modifying agent and emulsifier for *in situ* polymerisation. Analytical grade sodium hydroxide (Kemika, Croatia) and sodium metabisulphite $Na_2S_2O_5$ (BASF) were used for the termination of polymerisation reaction and removal of free formaldehyde from the suspension of microcapsules. Water-soluble polyvinyl alcohol Mowiol (Clariant) and acrylic latex (BASF) were used in formulations as binders. Four formulations of microencapsulated repellents were prepared for testing (Table 1).



Main process parameters:

- Melamine-formaldehyde prepolymer: 11 g/100g of core material
- Modifying agent/microcapsule core: 6.5 g/100g of core material
- Diameter of dissolver plate: 80 cm
- Mixing speed, rpm: 1500 min^{-1}
- Emulsification time: 20 min
- Share of dispersed phase in emulsion: 35 vol%
- Polymerisation time: 90 min
- Polymerisation temperature: 75 ^oC

Figure 3: Main steps and process parameters of microencapsulation by *in situ* polymerisation of amino-aldehyde resins in a 10 L reactor

Table 1: Main characteristics of microencapsulated repellent formulations

Formulation	***Microcapsule core***	***Binder (g/100g dry microcapsules)***
D1	Daphne (90%), isopropyl myristate (10%)	acrylic latex (14,2)
D2	Daphne (90%), isopropyl myristate (10%)	polyvinyl alcohol /acrylic latex 1:1 (13,2)
PP	*Psiadia punctulata* extract (20%), dibutyl phthalate (80%)	no binder
PPA	*Psiadia punctulata* extract (20%), dibutyl phthalate (80%)	PPA acrylic latex (10,0)

Repellent effects on roe deer (*Capreolus capreolus* Linne) were studied during the winter season in two consecutive field experiments with baits, set up on a field near a forest (Table 2). The first experiment consisted of 6 trials, and the second of 7 trials, all in 4 replicates. Each replicate consisted of a bait with 10 one-year-old apple branches (*Malus communis* - Jonathan), approx. 1 m

long, with 15 cm distance between branches. The distance between the baits was 20 m. A hydraulic knapsack sprayer Solo was used for spraying the baits. The quantity of spray used was 0.25 L per bait. The climatic conditions (average day temperature, precipitation) were monitored by an automatic microclimatic station. The damage caused by deer was evaluated as the number of damaged / eaten off branches. For each trial, the maximum possible number of damaged branches was 40 (10 branches in 4 replicates).

Table 2: Trials in experiments 1 and 2

Trials in experiment 1				***Trials in experiment 2***			
Trial number	*Formulation*	*Concentra-tion in water (%)*	*Active compound (%)*	*Trial number*	*Formulation*	*Concentra-tion in water (%)*	*Active compound (%)*
1	PP	5	0.30	1	PP	10%	0.60
2	PP	10	0.60	2	PPA	10%	0.60
3	Daphne*	1	1.00	3	Daphne*	1%	1.00
4	D2	1	0.24	4	D2	1%	0.24
5	D1	1	0.24	5	D2 + PP	1% + 10%	0.24 + 0.60
6	control**	-	-	6	D1	1%	0.24
				7	control**	-	-

* non-encapsulated oil (all other formulations contained microcapsules)
** control trial: baits were not sprayed

Results and discussion

Experiment 1 (Figures 4 and 5) was set up in January in severe winter conditions (average day temperature was -13°C, 5 to 10 cm of snow). The pressure of animals on apple branches in baits was strong. First damage in the non-sprayed control was observed already the first day after setting up the experiment. For the whole evaluation period of experiment 1, the largest number of damaged branches was recorded in the non-sprayed control (trial 6). The highest repellent efficacy was recorded in trial 2 with microencapsulated *Psiadia puctulata* extract.

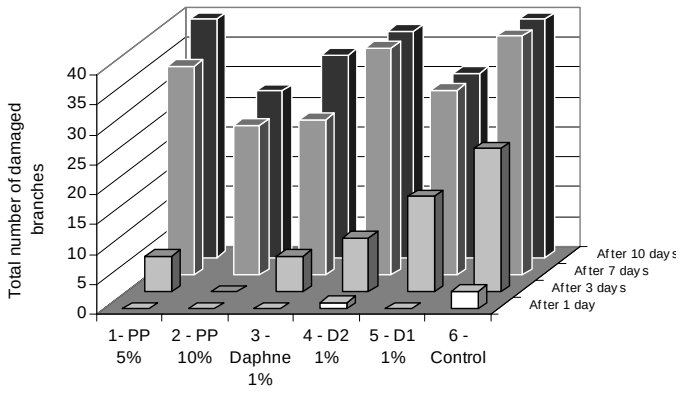


Figure 4: Total number of damaged branches in trials as shown in four evaluations of experiment 1

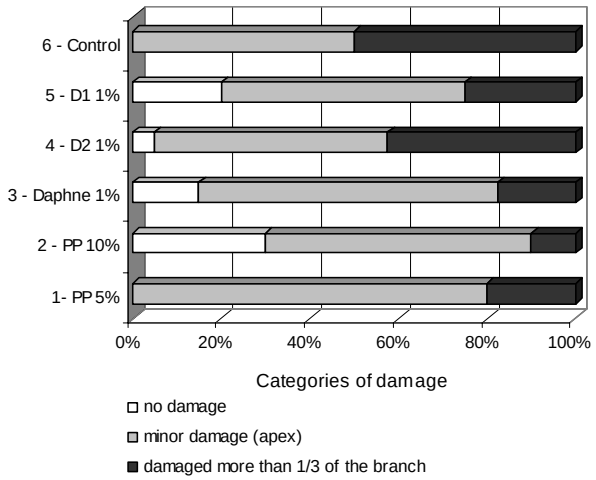


Figure 5: Ratio of evaluated categories of damaged branches in trials of experiment 1 at the last evaluation (10 days after spraying)

Experiment 2 (Figures 6 and 7) was set up under milder climatic conditions in March (average day temperature +2°C, no snow; rain precipitation 10 mm on day 12, 52 mm on day 25). The results show longer repellent activity in milder winter conditions. In all trials with microencapsulated

repellents, the damage was less intensive, compared to the non-sprayed control (trial 7) and non-encapsulated Daphne oil (trial 3), which indicates higher efficacy and prolonged activity of all microencapsulated repellents. The strongest repelling effect was recorded in trial 2 with microencapsulated *Psiadia puctulata* extract and acrylic latex binder. The importance of a binder in case of heavy rains can be seen in a comparison of trial 2 with trial 1 (*Psiadia puctulata* extract with no binder). In trial 1 the repelling effect was strong during dry days, and was lost after rains. Formulations with microencapsulated Daphne were all more effective than non-encapsulated oil, but their repelling effect seemed to be weaker than that of *Psiadia punctulata* extract. The possible explanation lies in the mode of activity. Daphne is primarily a smell-based repellent, while *Psiadia* extract possesses a combined smell- and taste-based animal repellent effect.

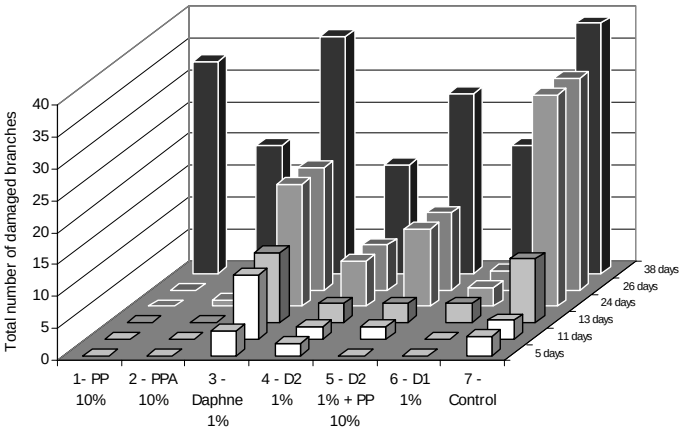


Figure 6: Total number of damaged branches in trials as shown in six evaluations of experiment 2

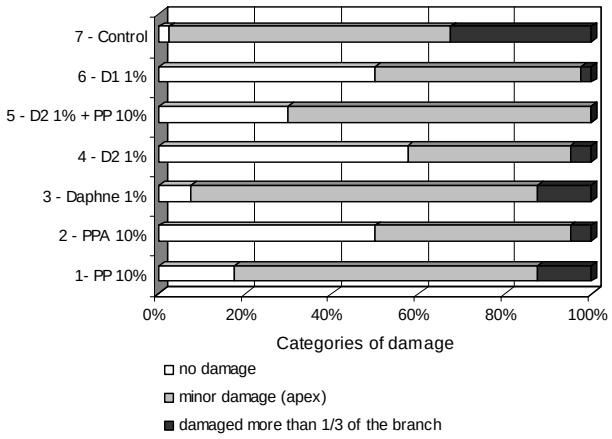


Figure 7: Ratio of evaluated categories of damaged branches in trials of experiment 2 at the last evaluation (38 days after spraying)

Conclusions

Two animal repellents - Daphne oil and *Psiadia punctulata* extract - were microencapsulated by *in situ polymerisation* method. Aqueous suspensions of microcapsules were mixed with polyvinyl alcohol and acrylic binders, and were tested with baits in two winter experiments against roe deer (*Capreolus capreolus* Linne). The results showed a prolonged activity and improved efficacy of microencapsulated repellent formulations in comparison with the standard non-encapsulated Daphne repellent and non-sprayed control. *Psiadia punctulata* leaf exudate exhibited a stronger repelling effect than Daphne. The activity of both microencapsulated repellents was stronger in milder winter conditions, when the pressure of animals was not extreme. In addition, at higher temperatures microcapsules prevented the premature evaporation of volatile compounds, and reduced the washing off by rains.

Acknowledgement

Psiadia punctulata diterpenoid extract was donated by Professor Jacob O. Midiwo, University of Nairobi, Kenya

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