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Controlled-Release Systems for the Insect Growth Regulator Pyriproxyfen

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A simple technique was developed for the production of controlled-release systems (CRSs) for pyriproxyfen, an insect growth regulator active against the larvae of *Culex pipiens*, the most common species of mosquito found in developed countries. The CRSs consisted of a spongy core material encapsulated in a coating of a polyurethane or polyurea hydrogel, into which the active ingredient had been incorporated. The coating also included a surfactant to improve the low solubility in water of pyriproxyfen. The light core material enabled the CRS to float on the water surface, where the mosquitoe larvae are found. The type and amount of the polymeric coating and the amount of surfactant influenced the release profiles into water of the active ingredient. The release profiles of the CRSs were adjusted to the life cycle of the *C. pipiens* mosquito in order to obtain their optimal activity on the eighth day, which corresponds to the time of larval maturity.

KEYWORDS: Controlled-release systems; insect growth regulators; pyriproxyfen; coating; encapsulation; life cycle of mosquitoes

INTRODUCTION

Synthetic insecticides used for the control of mosquitoes vary greatly in their structure, toxicity, persistence, and environmental impact (*1*). These chemicals include organochlorines, organophosphates, pyrethroids, and carbamates, many of which have high mammalian toxicity. Ongoing use of these chemicals and their consequent buildup in the environment have led to pest resurgence, secondary pest outbreaks, and development of resistance (*2*, *3*). As a result of these environmental issues, the current tendency is to use preparations having apparent minimal adverse impact on the environment, such as insect growth regulators (IGRs) and biological insecticides.

IGRs act by disrupting the normal development of insects by mimicking juvenile hormone (JH) or molting hormone or by interfering with chitin synthesis, typically resulting in larval or pupal mortality (*4*, *5*). The main advantage of IGRs over conventional pesticides is their relative nontoxicity to mammals, because they target only insects. The matrix in which the pesticides are included is biodegradable. The most commonly used IGRs that are active against larvae, including those of species that have developed resistance to conventional insecticides, are methoprene, cyromazine, buprofezin, fenoxycarb, pymetrozine, and kinoprene and pyriproxyfen (*6*). The latter compound, a relatively new JH mimetic of synthetic origin with high efficacy and minimal impact on natural enemies (*7*, *8*), has been used successfully in the management of the white fly (*9*), the German cockroach *Blatella germanica* (*10*, *11*), and certain species of mosquito (*12*).

There are, however, a number of drawbacks associated with the use of IGRs. These materials are stage specific, which means that they are effective only if applied at the correct growth stage in the insect life cycle. In addition, IGR larvicides act slowly, and they may take days or weeks to accomplish what organophosphates achieve within hours. Finally, IGRs are more expensive than conventional insecticides. This latter disadvantage may be offset by the lower application rates required, particularly if the active ingredient is formulated into a controlled-release system (CRS), making it cost-effective (*13*).

CRSs confer the advantage that the active ingredient is maintained in the encapsulated CRS or in the environment in the appropriate dosage for a specified period of time, which may be months or even years (*14*). They may thus increase the effectiveness of the active agent while decreasing the potential of hazard to the environment (*15*, *16*). Other advantages presented by CRSs include reducing losses from volatility of liquid formulations, reducing phytotoxicity, protecting larvicides from environmental degradation, reducing leaching of the active material into soil and aquifers, facilitating the formulation of

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Table 1. Composition of Pyriproxyfen CRS Formulations with Polyurethane or Polyurea Coatings[a]

					coating components						
formulation	pyriproxyfen	perlite	Induce	coating type	PEG 2000	PEG 6000	PEG 10000	Bayflex	polyisocyanate	ETA	TEPA
PC-1	3	65	20	polyurethane	10.6				1.4		
PC-2	3	65	20	polyurethane		11.5			0.5		
PC-3	3	65	20	polyurethane			11.7	-	0.3		
PC-4	3	65	20	polyurethane				7.2	4.8		
PC-5	3	71	20	polyurea					4.2	0.9	0.9
PC-6	3	71	20	polyurea					4.2	0.9	0.9
PC-7	3	71	20	polyurethane	5.3				0.7		
PC-8	3	65	20	polyurethane	10.7				1.3		
PC-9	3	61	30	polyurethane			5.8		0.2		
PC-10	3	55	30	polyurethane			11.7		0.3		
PC-11	3	61	30	polyurea					4.2	0.9	0.9
PC-12	3	55	30	polyurea					10.0	2.0	2.0
PC-13	3	85	0	polyurethane			11.7		0.3		
PC-14	10	48	30	polyurethane			11.7		0.3		

[a] ETA, ethylenediamine; TEPA, tetraethylene pentaamine; values in the table are % (w/w).

liquids into solid granules or flowable powders, separating reactive components, and providing for ease of handling (*17*, *18*).

Progress in polymer science over the past few years has provided a range of materials suitable for CRS applications. These materials must be environmentally degradable (*19*, *20*), cheap, and suitable for application over large-scale areas (*21*). Synthetic biodegradable polymers that are commonly used include polyethylene, polyvinyl alcohol, ϵ-caprolactone polyester, polyethers, and polyurethanes (*19*, *20*, *22*, *23*). The U.S. Environmental Protection Agency (EPA) has proposed that polymers of the future be both photodegradable and biodegradable to ensure their complete removal from the environment (*19*).

Other types of polymers that have been developed for encapsulating a variety of biocides include Intelimer polymers for microencapsulating pesticides such as diazinon or trifluralin (*24*), the Culigel system of acrylate polymers for bioactive agents used in the management of pests such as *Aedes taeniorhynchus* and *Culex quinquefasciatus* mosquitoes (*22*), cellulose granules (Biodac) and corn cob granules for encapsulating biological agents or IGRs (B.t.i., B. Sphaericus, methoprene, or pyriproxyfen) against *A. taeniorhynchus*, *Anopheles albimanus*, and *C. quinquefasciatus* mosquito larvae and *B. germanica* nymphs (*23*), a sugar/flour matrix for coating the insecticide dimethoate suspended in latex paint (*25*), starch matrices for sprayable formulations (*26*), and a starch matrix containing a gel promoter for entrapping nonvolatile pesticides (*27*).

Our group recently developed a series of CRSs for formulating cyromazine, which proved to be an effective treatment against the larvae of the *C. pipiens* mosquito (*13*), the most common species of mosquito found in developed countries and the primary vector in the transmission of West Nile virus (*28*−*30*). We extended the work on the formulations to the controlled release of plant growth factors by means of hydrogel coatings (*31*). The latter polymeric materials exhibit the ability to swell in water and to retain a significant portion of water without dissolving (*32*). In the current study, we further developed the hydrogel CRS formulations for the IGR pyriproxfen, which is also an effective larvicide in the control of *C. pipiens*. The IGR was incorporated into a polymeric envelope coating a spongy core material, which enabled the CRS to float on the surface of the water. The influence of the nature and amount of coating on the release of the active ingredient was studied, and preliminary in vivo testing of the larvicidal activity of the formulations was conducted.

MATERIALS AND METHODS

Materials. Pyriproxyfen [2-[1-methyl-2-(4-phenoxyphenoxy)ethoxy]-pyridine], the active ingredient of technical grade 99%, produced by Sumitomo Chemical Corp., is a pale, stable yellow solid, melting point = 47 °C, flash point = 119 °C, and solubility in water of 0.37 mg/L at 25 °C. The polyisocyanates Voranate 580 and Isonate M342 (diphenyl methane-4,4′-diisocyanate) were purchased from Dow Benelux, N.V.; the polyester Bayflex (Desmophen 2200) was from Bayer; Induce surfactant is a mixture of several surfactants manufactured by Helena Chemical Co.; and polyethylene glycol (PEG), ethylenediamine, and tetraethylenepentamine were from Fluka.

Polyurethane and Polyurea Coatings. Three types of polyurethane coating were prepared by conventional interfacial condensation of Voranate 580, as the hydrophobic component, with PEG 2000, PEG 6000, or PEG 10,000 as the hydrophilic component (formulations PC-1, PC-2, PC-3, PC-7, PC-8, PC-9, PC-10, PC-13, and PC-14, **Table 1**). A fourth polyurethane coating was prepared from Voranate 580 and Bayflex (formulation PC-4, **Table 1**). The polyurea coating was produced by interfacial polymerization between a polyisocyanate, either Voranate 580 or Isonate M342, and a polyfunctional amine obtained by reacting ethylenediamine with tetraethylenepentamine (formulations PC-5, PC-6, PC-11, and PC-12, **Table 1**).

Encapsulation Technology. The choice of perlite as the inert core material was made on the basis of its low density, which enables it to float on water, and its low cost. The physical and chemical properties of pyriproxyfen, particularly its low solubility in water and its flash point, dictated the coating method for encapsulation, which was performed as follows. Pyriproxyfen was dissolved in acetone (as the organic solvent used for the polymerization process) and then homogenized with Voranate 580. The mixture was sprayed over a known weight of dried and filtered perlite in a coating pan (Apex Engineering Industries) rotating at 40 rpm. As the acetone evaporated and a relatively homogeneous distribution of the mixture on perlite was obtained, a second mixture, comprising molten PEG and a surfactant (Induce) dissolved in acetone, was added. The interfacial polymerization condensation process was allowed to proceed for 1 h in the rotating coating pan at room temperature.

Dissolution Tests. These tests were performed to determine the chemical release of pyriproxyfen from the CRSs. The tests were carried out in a dissolution test system (model 2100B, Distek, North Brunswick, NJ), comprising six glasses (11.5 cm high × 10.2 cm i.d.), each filled with 800 mL of distilled water. Each glass was fitted with a basket, which held 1.0 g of a formulation containing 0.03 g of pyriproxyfen (**Table 1**). The grains of the chosen formulation had approximately the same size and shape. The dissolution system was held at 25 °C and operated at a basket rotation of 50 rpm. Due to the very low

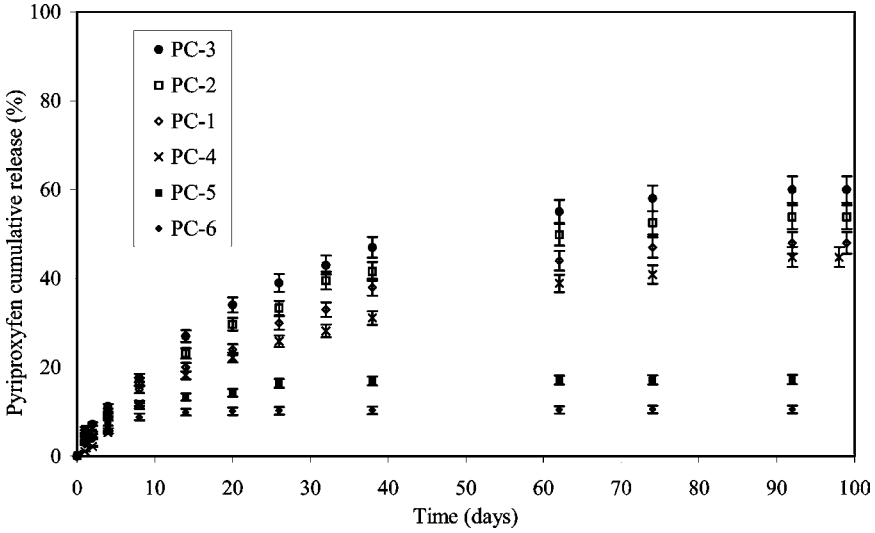

Figure 1. Release of pyriproxyfen into water from formulations with different types of polymeric coating (PC-1, PC-2, PC-3, PC-4, PC-5, and PC-6) as a function of time. Bars represent confidence intervals (for 95% confidence coefficient for *t* distribution).

solubility of pyriproxyfen in water, the water in each glass was replaced every 24 h at the beginning, a step that prevented saturation and thus facilitated analysis of the entire amount of active ingredient released into the water. The interval between the changing of water was gradually increased during the course of each experiment as in the case of taking of the samples for HPLC analysis. This way the dissolution system was kept below the saturation level.

At irregular time intervals, liquid samples were withdrawn from the upper layers of each glass (because the aqueous solution was stirred, the concentration of pyriproxyfen was the same in any part of the solution), and the amount of pyriproxyfen released into the water was determined by HPLC. Because the solubility of pyriproxifen is very low, at the beginning the samples were taken each time after 1 day, and later the interval was increased up to 100 days. The HPLC system (Shimadzu, Tokyo, Japan) was equipped with a Hypersil ODS 5-μm column, 250 mm $\times$ 4.6 mm i.d. (Runcorn, U.K.). The eluents used were water/methanol, 20:80 v/v, with a flow rate set at 1 mL/min; the UV detector wavelength was set at 226 nm during the 10-min analysis. The weight percentage of the pyriproxyfen released versus saturation time for each CRS was determined.

For each experimental run, a sample was taken from two different batches (each one consisting of three glasses) of the same formulation prepared under the same conditions. This sampling technique was used to exclude random factors, such as technological conditions (temperature), nonhomogeneous distribution of the IGR in the coating, and variations in coating thickness.

RESULTS AND DISCUSSION

The encapsulation technology was designed to overcome the three major problems associated with the use of pyriproxfen as an efficient larvicide against *C. pipiens* mosquitoes, that is, its low solubility, high cost, and tendency to sink to the bottom of water bodies, where large amounts of material can be lost by adsorption on organic materials and sand. Problems of solubility and material loss were addressed by encapsulating the active ingredient into a hydrogel, that is, polyurethane or polyurea. The requirement for the active material to be released at the surface of the water where the mosquito larvae float was fulfilled by coating the embedded active ingredient onto a very light inert material—perlite—that enables the final formulation to float on the surface. Studies of the cumulative release of pyriproxyfen into water were designed to determine the influence of the following factors on the release rate: type and amount of polymeric coating, presence of the surfactant, and amount of pyriproxyfen embedded in the polymeric coating.

Table 2. Cumulative Pyriproxyfen Release from the Studied Formulations versus Saturation Time

formulation	pyriproxyfen released[a] (%)	saturation time (days)
PC-1	47.5 ± 4.8	~75
PC-2	52.0 ± 3.9	~75
PC-3	58.1 ± 4.9	~92
PC-4	44.8 ± 6.5	~92
PC-5	16.7 ± 7.1	~26
PC-6	10.3 ± 4.1	~32
PC-7	43.1 ± 4.5	~74
PC-8	58.1 ± 5.1	~92
PC-9	53.5 ± 3.9	~95
PC-10	72.2 ± 5.5	~75
PC-11	17.1 ± 2.8	~62
PC-12	11.9 ± 3.7	~38
PC-13	3.2 ± 0.9	~78
PC-14	68.5 ± 4.4	~92

[a] Values are means ± confidence intervals (at 95% confidence level for *t* distribution).

Effect of the Type of Polymeric Coating. It has recently been shown that the nature of the hydrogel coating of a formulation has a marked influence on the release rate of the active ingredient into water (*31*). In the first series of formulations, PC-1 to PC-4 (**Table 1**), it was found that the cumulative release of pyriproxyfen from formulation PC-4, having the polyester-based coating, was slower than that from the other three formulations, PC-1, PC-2, and PC-3, the coatings of which contained the more hydrophilic PEG (**Table 2** and **Figure 1**). The release of pyriproxyfen from the latter three formulations was, in turn, dependent on the molecular weights of the starting PEGs: the higher the molecular weight of the PEG, the better the hydrogel properties of the polyurethane and, hence, the higher the release of active ingredient.

The finding that the entire amount of pyriproxyfen is not released into water may be explained in terms of the encapsulation of this very hydrophobic material in a polymeric envelope. The release of some of the pyriproxyfen into the bulk water takes place in a two-step process, that is, dissolution of the pyriproxyfen into the water taken up into the hydrogel, followed by diffusion from the hydrogel. The rest of the pyriproxyfen remains trapped in the polymeric envelope. In preliminary experiments analytical tests on the formulation granules were performed (data not presented), which clearly indicated that the rest of pyriproxyfen remained entrapped in the polymeric

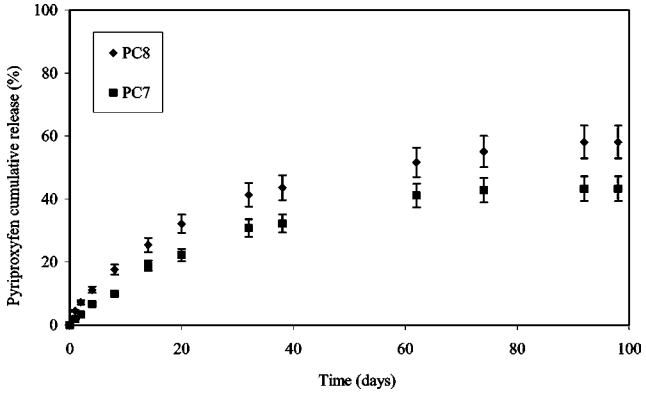

Figure 2. Influence of the amount of the polymeric coating on the release of pyriproxyfen into water from formulations with polyurethane coatings (PC-7 and PC-8) as a function of time. Bars represent confidence intervals (for 95% confidence coefficient for *t* distribution).

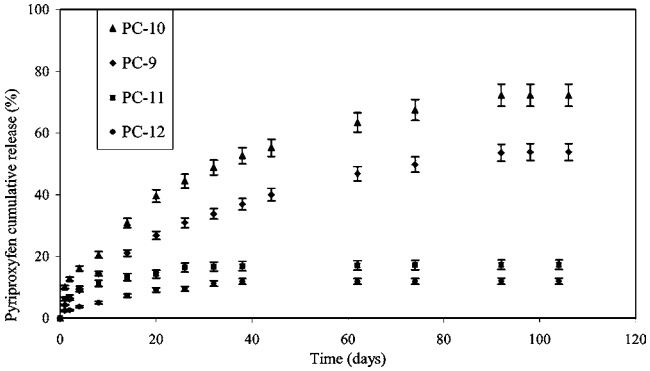

Figure 3. Influence of the amount of the polymeric coating on the release of pyriproxyfen into water from the formulations PC-9, PC-10, PC-11, and PC-12 as a function of time. Bars represent confidence intervals (for 95% confidence coefficient for *t* distribution).

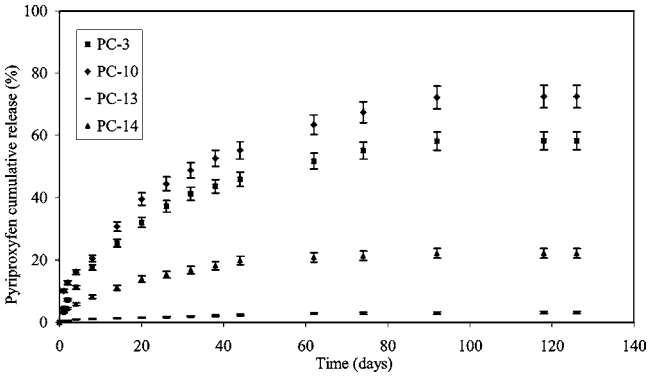

Figure 4. Influence of the amount of surfactant (Induce) on the release of pyriproxyfen into water from the formulations PC-3, PC-10, PC-13, and PC-14 as a function of time. Bars represent confidence intervals (for 95% confidence coefficient for *t* distribution).

envelop, thus excluding the assumption that it may have degraded in any way. The time after which there is no further release of the active ingredient into the water is defined in this case as the saturation time.

When polyurea was used as the polymer in the envelope coating, that is, in formulations PC-5 and PC-6 (**Table 1**), there was, as expected from its very low hydrophilicity, a very low cumulative release of pyriproxyfen up to saturation time, 16.7 $\pm$ 7.1% after ~26 days and 10.3 $\pm$ 4.1% after ~32 days (**Figure 1**).

Effect of the Amount and Composition of Polymeric Coating. The higher the amount of the polyurethane coating, the higher the release of pyriproxyfen into water. Of the PEG 2000 formulations, PC-8, containing 10.7% of coating, released pyriproxyfen faster than PC-7, containing 5.3% of coating (**Table 2**). These results were to be expected, because the higher amount of hydrogel will absorb a higher amount of water and thus facilitate the dissolution and subsequent diffusion of relatively increased amounts of pyriproxyfen. Similarly, the type of hydrogel will influence the uptake of water, as is reflected in the superior release of the active ingredient from the 11.7% high molecular mass PEG 10,000 coating (PC-10) versus that from the 10.7% PEG 2000 coating (PC-8) (**Table 2**). The same trend was observed for the 5.8% coating, PC-9, versus the 5.3% coating, PC-7 (**Table 2**). For the formulations coated with polyurea, PC-11 and PC-12, the greater the coating thickness, the slower the release of pyriproxyfen (**Table 2**; **Figure 3**).

Effect of Surfactant. The influence of the surfactant Induce on pyriproxyfen release was tested in three formulations with different surfactant contents, PC-10 (30%) and PC-3 (20%) versus PC-13 (0%). Induce is a nonionic surfactant that is used in agriculture as a spreader and an activator. It is designed for quick wetting, good penetration, and uniform droplet distribution. It was found that the formulation with the highest surfactant concentration gave markedly better release (**Table 2**; **Figure 4**). The role of the surfactant was to increase the solubility in water of the active ingredient entrapped in the polymer.

Effect of Amount of Pyriproxyfen. A comparison of the release of active ingredient from a formulation containing 3% (w/w), PC-10, with that containing 10% (w/w), PC-14, showed that the solubility of pyriproxyfen in water is so low that the amount of the active ingredient in the formulation had no effect on the release into water, with approximately the same proportion of pyriproxyfen being released from the two formulations (**Table 2**).

Conclusion. In this study we developed a series of CRSs for the controlled release of pyriproxyfen, an IGR mimicking the action of JH, as a larvicide against *C. pipiens*. The choice of the core material type of the formulations enabled the CRSs to float on water—and thus come in contact with the larvae—and the controlled release of the active material was obtained by incorporating it into a coating of various polymers and a surfactant. The latter served to improve the release of the active ingredient from ~20 to ~50−70%. It is extremely important to consider biological parameters and to take into consideration the environmental conditions (temperature, humidity, etc.) when chemical formulations are developed. Therefore, by constructing different types of CRSs with different rates of release, we can afterward, depending on the climate conditions or the developmental stage of larvae, combine different systems to obtain the optimal result. In other words, a "programming" of these CRSs can be possible by taking into consideration different parameters. Our results showed a good potential of the CRSs for field application, and in light of the in vitro preliminary experimental findings, more comprehensive in vitro and in vivo studies on *C. pipiens* mosquito larvae are under way and testing of the CRSs under field conditions is being planned. The next steps in the continuation of this research will comprise the application of statistical tools and mathematical models to the design of optimal CRS formulations on the basis of in vitro results.

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