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SLOW-RELEASE POLYMERIC COMPOSITIONS FOR MOSQUITO LARVICIDING[1]

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Considerable interest has developed in the use of slow-release polymeric formulations of larvicides since the concept was introduced a few years ago. The authors have been cooperating with a number of interested agencies in the evaluation of these materials for public health applications.

Slow-release polymers are solutions of pesticide in solid polymer formulations of both rubber and plastics. Examples of materials which have been used are SBR rubber and polyvinylchloride plastic. When the slow-release formulation is placed in water, the larvicide is slowly dissolved from the surface. As the concentration of larvicide in the surface is depleted, additional larvicide diffuses from within the polymer to the surface so that release of active agent continues. By appropriate research slow-release polymer formulations can be produced for almost all organic larvicides.

Slow-release polymers are a new tool for mosquito control workers. Although they are not yet commercially available, it is desirable that the directors of mosquito abatement districts understand the potential for these new products as an aid to their future planning.

By using slow-release polymers, it may be possible under some conditions to obtain mosquito larval control for an entire season with a single application. Indeed, it might even be possible to obtain two or more seasons' control by a sufficiently heavy dosage. In the use of slow-release polymers to prevent marine fouling an active lifetime of five years has been indicated. However, mosquito abatement districts are normally budgeted on a one-year basis, and from a practical point of view, single-season control is probably most desirable.

Advantages to the use of larvicidal slow-release polymers include: lower labor costs for application since less frequent applications are required; better localization of application; greater safety in handling and application; and the possibility of preflood application. The dry pellets are inactive. If they are scattered over a dry pasture, they will not release larvicide until the pasture is irrigated.

There are, of course, some disadvantages as well. These include: a higher cost for the material; and larvicidal pellets which remain in the environment for a long time.

In discussing slow-release polymers

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with a number of workers and researchers in mosquito control, we found that it was not well understood how these materials should be used. Possibly, therefore a few observations on both the theory and practice relating to their use may be helpful.

Slow-release polymers, like larviciding sprays must be broadly distributed over the surface of the water in which larvae are to be controlled. In general, it is expected that these materials will be scattered over the water surface in granular form. Figure 1 shows some possible granule sizes and shapes. With most of the polymer compositions made to date, it is necessary that a relatively large polymer surface be exposed to the water. Thus the granules will be relatively small. It is not, for example, possible to control mosquitos in a 20 x 20 pond by placing one chunk of polymer in the pond even though the polymer may contain several times the amount of larvicide which would be lethal to mosquito larvae if it were uniformly dispersed in the water. In order to obtain good dispersion of the larvicide in the body of the water, it is desirable that these polymer granules be scattered rather uniformly over the surface so that after they sink to the bottom there is not too great a distance between granules. Incidentally, slow-release polymers which float can also be made.

The larvicide while within the polymer is not "available" for killing larvae, or for that matter, for killing or damaging anything else in the environment, unless, of course, the granule itself is ingested. It must leave the polymer and enter the water before it is active. This brings us to another important characteristic of slow-release polymers. The rate of release is not constant with time. The initial release rate decreases as time goes on. The rate of release of active agent is a function of the concentration of active agent remaining in the polymer particles. Figure 2 shows experimental data on release rate obtained with an organotin in neo-

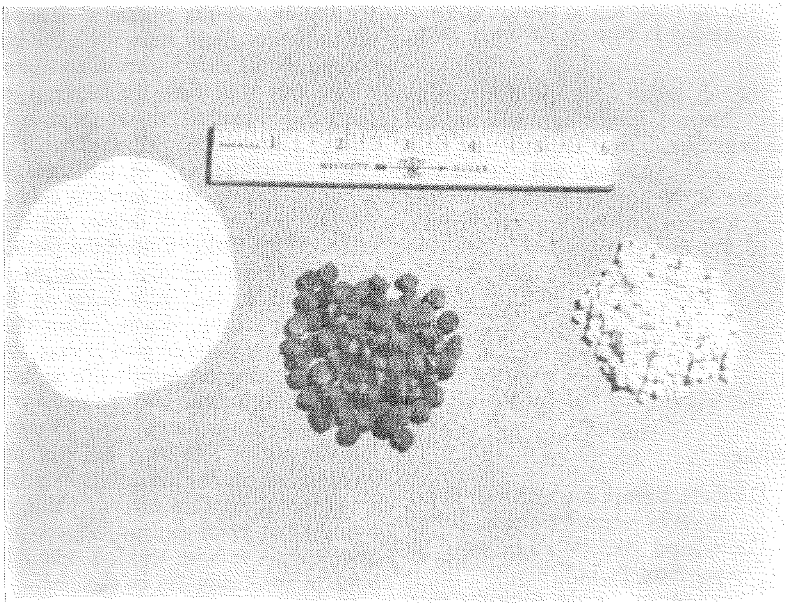

FIG. 1.—Typical Range of Particle Sizes of Slow-Release Polymeric Larvicides.

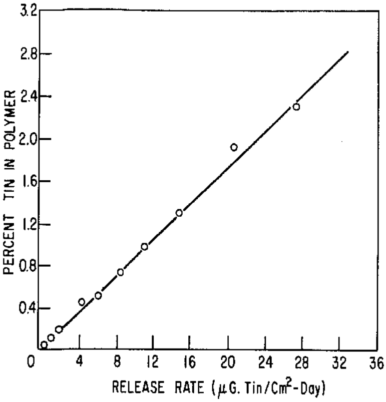

FIG. 2.—Release Rate as Function of Tin Concentration in Neoprene Containing TBTO

prene. It shows a straight line relationship between release rate and residual concentration. The concentration of pesticide in the polymer and the rate of release of the pesticide from the polymer as a function of time are shown in equations (1) and (2):

$$C = C_0 e^{\frac{-KAt}{V}} \qquad (1)$$

$$R = KC_0 e^{\frac{-KAt}{V}} \qquad (2)$$

where:

C =bulk average concentration of pesticide in polymer at time t, lb./in.^{3}
C_0=original pesticide concentration in polymers, lb./in.^{3}
K =a constant for the particular polymer/pesticide system, $\frac{\text{in.}}{\text{day}}$
A =surface area of the particle, in.^{2}
V=volume of the particle, in.^{3}
t =time, days
e =base of natural logarithms
R =rate of release of pesticide from polymer, $\frac{\text{lb.}}{\text{in.}^2 \text{ day}}$

These equations, and those that follow, apply strictly only to the most simple case, that where the release rate of the pesticide is controlled by the rate of dissolution from the surface of the polymer. They serve, also, as a useful approximation for the mathematically more complex case where diffusion within the polymer is the controlling mechanism.

These equations result in a release rate which decreases logarithmically with time, as shown in Figure 3, where the same data are plotted on rectangular coordinates (the lower curve) and on semilogarithmic coordinates (the upper curve). In the latter case, a straight line results.

Note that the term A/V appears in the exponent in the equations. This is the surface-to-volume ratio. Both the initial release rate and the rate of change of release rate with time are functions of this ratio. The surface-to-volume ratio in turn is a function of particle size. The effect of particle size on this ratio is shown in Table 1. It is seen that cutting the particle size in half doubles the surface-to-volume ratio. Cutting the particle size to one-tenth multiplies the surface-to-volume ratio by ten. Also shown on the table is the number of particles per unit volume for the different particle sizes. We see that cutting the particle size in half increases the number of particles by a factor of eight, the cube of two. Decreasing the particle size by a factor of ten increases the number of particles by a factor of 1,000, the cube of ten. Utilizing a larger number of particles obviously increases the ease with which a relatively uniform application can be obtained.

Equation 3 is a rearrangement of the previous equation.

$$t = \frac{V}{KA} \ln \frac{C_0}{C} \qquad (3)$$

In this form it gives the time required to release any desired fraction of the contained pesticide.

For the application of a fixed number of pounds per acre, the volume applied can be considered constant. It is evident that the time to release a fixed fraction

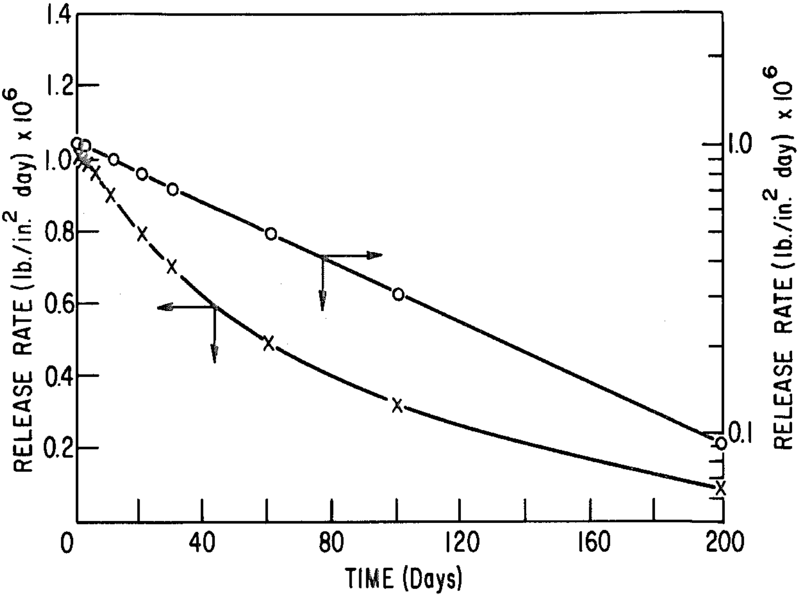

FIG. 3.—Release Rate as Function of Time TBTO in Neoprene, 0.05 inch Cubes Computed from Data on Sheets

of the pesticide from the polymer varies inversely with the area through which release is occurring or inversely with the surface-to-volume ratio. To be more specific, the time required to release one-half of the pesticide from the polymer will be cut in half if the particle size is cut in half.

The dosage to be applied to a given volume of water can be calculated from Equation (4).

$$W = wy\,(1 - e^{\frac{-KAt}{V}}). \qquad (4)$$

where:

W = weight of active agent released in time period, t, lb.

w = weight of bioactive polymer applied, lb.

y = weight fraction of active agent in the polymer

It gives the weight of active agent released for a given time period from a known amount of bioactive polymer containing a known weight fraction of active agent. In Equation (4), the term "wy" is the total amount of active agent applied. In practice, the supplier of the bioactive polymer can be expected to determine release rate data in the laboratory and provide them to the user in the form of easy-to-use tables or nomographs.

Environmental factors play an important part in determining the actual concentration of pesticide in the water. Figure 3 showed a typical release rate curve for a polymer which contains a larvicide. As discussed before, if surface-to-volume ratio of this particular polymer were increased the time scale would be shortened. If the dosage were increased, either by increasing the concentration within the particle, or increasing the total weight of the polymer, the curve would be pushed upward. Water temperature plays a small part. Increasing the water temperature will increase the rate of release slightly. In Figure 4, the upper curve describes the

TABLE 1.—Surface to volume ratio for cubes.

Edge Size, in.	Volume, in.^{3}	Surface in.^{2}	Surface, in.$^{-1}$ / Volume	Particles per in.^{3} of polymer
1	1	6	6	1
0.5	0.125	1.50	12	8
0.1	0.001	0.060	60	1,000
0.05	0.000125	0.0150	120	8,000
0.01	0.000001	0.0006	600	1,000,000

increase in concentration of the larvicide within a pool if all the larvicide remained in the water.

There are, however, competing reactions occurring in the pond which result in removal of the larvicide from the water at the same time that larvicide is coming out of the polymer. These reactions are decomposition and adsorption. The rate of decomposition is dependent upon the concentration of larvicide at any time, the temperature of the water, the pH of the water, and the amount of light, to name the more important variables.

In the case of the adsorption phenomena the important variables are again, concentration, and temperature, and to some extent pH. Of greatest importance is the nature and amount of adsorbents. These include the debris present in the pond, such as leaves and branches, and the bottom and sides of the pond itself. The amount of the adsorption on the pond bottom depends upon the nature of the material on the bottom and ratio of the volume of the water to the area of the bottom. The rate of adsorption will increase with concentration and will be more important in a shallow pond than in a deep pool, where the ratio of bottom surface area to water volume is the smallest. Sketched in Figure 4 is a curve approximating the amount of pesticide removed from the pond. The difference between the amount of larvicide released from the polymer and the larvicide adsorbed or hydrolyzed is the effective concentration in the pond for mosquito control. It is the difference between the two curves. You can see that, as time goes by, the concentration in the pond decreases and eventually reaches some threshold value beyond which no further larvicidal action would be expected.

In order to get a better feel for what this threshold value is, it is necessary to look at the time-to-kill for various concentrations of larvicide. Figure 5 shows a typical curve for this effect. The curve

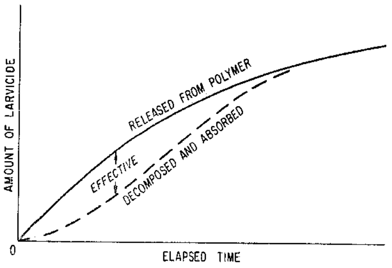

FIG. 4.—Typical Cumulation Curves for Larvicide Release and Loss

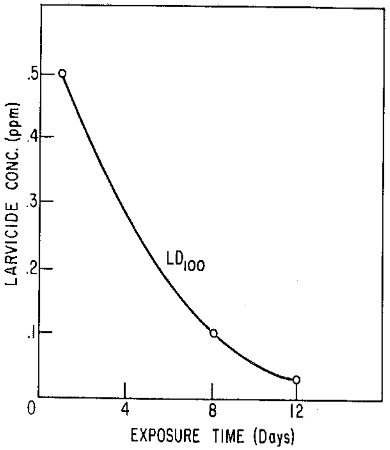

FIG. 5.—TBTO Concentration vs. Exposure Time for LD$_{100}$ (CULEX PIPIENS)

illustrates that at high concentrations only a relatively small time is needed to give 100 percent control, but that 100 percent control can also be obtained if the concentration is maintained at a low level over a prolonged period. Consequently, this effect should be taken into account when considering the dosage required for control in ponds. The position of this curve is dependent upon some of the environmental factors, principally those related to the rate of metabolism in the larvae. Increasing the temperature, increasing the food supply, increasing the oxygen supply should effectively raise this curve. Both Figures 4 and 5 illustrate that the environmental factors play an important role in determining the effectiveness of a larvicidal application. The removal curve shown is a hypothetical example. It is not based on any experimental data. There will be a considerable variation in actual removal curves, depending on the environment.

DROPLET SIZE, DENSITY, DISTRIBUTION AND EFFECTIVENESS IN ULTRA-LOW VOLUME AERIAL SPRAYS DISPERSED WITH TEEJET® NOZZLES[1]

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TeeJet® flat fan and hollow cone hydraulic nozzles are now the most commonly used devices for dispersing ultra-low volume (ULV) aerial sprays and aerosols of concentrated insecticides. The efficacy of applications made with these nozzles was described by Lofgren (1970) in his review of ULV sprays for use in mosquito control; however, only limited research has been reported on the physical characteristics of these sprays, especially in relation to mosquito control. Also, in a subsequent paper, Mount *et al.* (1970) suggested that increased efficiency might be achieved by improvements in the physical characteristics of the ULV sprays. However, before improvements can be made, we must have precise information about the physical characteristics of the sprays presently being used. The research reported here was designed to provide additional data on the droplet size, density, distribution and effectiveness of ULV aerial sprays dispersed with TeeJet flat fan and hollow cone nozzles produced by Spraying Systems, Inc.

Methods and Materials. The ULV sprays were applied with two Stearman and one Air Force UC-123 aircraft. One Stearman aircraft was equipped with a battery-powered ULV propulsion system for most of the tests. This system consisted of a 12-volt DC motor, a 2000R bronze oberdorfer gear pump, and a B & G stainless steel insecticide tank. The insecticide flowed from the tank to the nozzles through polyethylene tubing. A breaker switch with cables to the battery positioned in the pilot's compartment activated the system. In tests against natu-

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[1] Mention of a pesticide or a proprietary product does not constitute a recommendation or an endorsement of this product by the USDA or the Department of Defense.

[2] Major, Biomedical Science Corps, U. S. Air Force, Armed Forces Pest Control Board, Liaison Officer, Entomology Research Division, ARS, USDA, Gainesville, Florida 32601.