Controlled Release of Propham from Plastic Granules

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Synopsis

Urea-formaldehyde granules containing propham, a volatile herbicide, have been tested in the laboratory as well as in the field as a suitable controlled release system. The advantages of such a system compared to the present commonly used formulas are being demonstrated.

INTRODUCTION

Weed growth in cultivated fields is controlled by using various herbicides, which are usually marketed as emulsifiable concentrates or wettable powders. Since the active ingredients in such formulations are volatile materials, their effective period is rather limited. In order to extend the period, it became common to spray the field with a herbicide solution and subsequently to plow under and/or irrigate; the effective period is still much too short. In the next stage, strong adsorbents such as clay particles have been examined as carriers for the active ingredients. In such a system, the herbicide is adsorbed only on the particle surface (which is relatively large); therefore, the volatilization process is not sufficiently retarded and the system is still not satisfactory.

To extend the activity of the herbicide, controlled-release systems have been sought. Recently, various granular formulations have been developed.¹⁻⁴ Formulations were prepared with several polymers such as cellulose acetate, polyamide, polyester, urea-formaldehyde, polyurethane, and poly(vinyl chloride). The active ingredient is dissolved, dispersed, or encapsulated in the polymeric matrix, and under field conditions it is released owing to diffusion through the polymer to the surrounding environment. Such systems have demonstrated the possibility of designing plastic formulations with varying rates of release. Controlled-release granular formulations have economical and ecological⁵ advantages over liquid or powder versions.

In the present work, plastic granules containing volatile herbicide have been studied. Propham was chosen as the representative of a large group of volatile herbicides to examine the effectiveness of plastic granules as a suitable system for its controlled release. The volatility of nonencapsulated propham was found to be relatively high and sensitive to temperature, wind, and soil moisture.⁶ For ecological reasons, urea-formaldehyde was selected as the plastic component of the granules since it may act as a fertilizer and does not pollute the fields. The potential of such a system has been tested in the laboratory as well as in the field.

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Propham is the common name for isopropyl N-phenylcarbamate (IPC). Its solubility in water at 20°C is 100 ppm, and it is soluble in most organic solvents. Technical propham manufactured by Bayer (W. Germany) is 98% pure and melts at 87°C. Propham is mainly used for the control of annual grass weeds in peas, beet, alfalfa, onion, etc. This herbicide is absorbed by the weeds through the roots.⁷

Propham-containing urea-formaldehyde granules, about 1 mm in diameter, were prepared on a laboratory scale.⁸ The concentration of the active ingredient in the formulation was determined by GLC. Approximately a 2-g granular sample was weighed into a flask. After adding chloroform (50 ml), the capped flask was shaken for 8 hr at room temperature and aliquots (4 μ l) were analyzed by GLC.⁹

TEST PROCEDURES AND RESULTS

Laboratory Tests

Laboratory-scale release tests were conducted in three media: air, water, and organic solvent. In the first test, approximately 1-g samples of granules containing 50 mg propham were spread in a crystallization dish and heated in an air-ventilated oven at the desired temperature. After specified times, the samples were cooled to room temperature, and the remaining herbicide concentration in the granules was determined as described above. For comparison, a 50-mg sample of pure herbicide was exposed to the same conditions, and its weight was followed as a function of time. The results of tests conducted at 60°C are shown in Figure 1. As can be seen in this figure, the evaporation rate at 60°C of propham incorporated in a polymeric matrix is lower than that of the pure herbicide.

To test the release rate of propham into an organic solvent, chloroform (50 ml) was added to a granular sample (2 g) in a capped flask and was shaken at room temperature. Chloroform was selected as a very good solvent for propham,

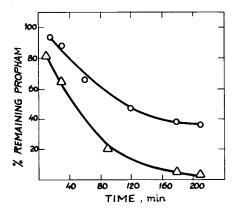


Fig. 1. Release of propham into air at 60° C: (Δ) pure propham; (O) controlled-release formulation.

having the ability to extract the whole amount of propham from the granules in 8 hr. At desired time intervals, aliquots of the solution were analyzed by GLC to determine the herbicide concentration. The percentage of remaining herbicide in the granules having an initial concentration of 10.35% is shown in Figure 2 as a function of time. This figure shows an initial rapid drop in the percentage of herbicide remaining in the granules followed by a gradual decline for the remainder of the test period.

The release rate into water was tested by immersion of granules (1 g) containing 4% propham in distilled water (500 ml) at 45°C. The capped flask was held under constant shaking. After the desired time, the propham was extracted from the water solution by chloroform in a separating funnel. Excess chloroform was evaporated to about 20 ml solution which was transferred into a 25-ml volumetric flask, followed by dilution to volume with chloroform. A 4- μ l aliquot of the solution was analyzed by GLC. This procedure was followed for every run with different immersion times in water. The percentage of remaining herbicide in the granules as a function of time is shown in Figure 3. The release of propham into water is much slower than into chloroform. This is due to the better solu-

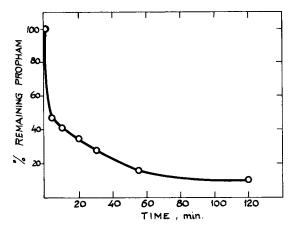


Fig. 2. Release of propham from plastic granules into chloroform at 25°C.

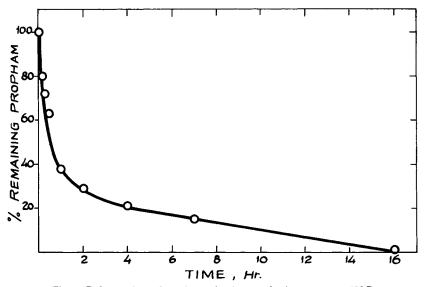


Fig. 3. Release of propham from plastic granules into water at 45°C.

bility of propham in the organic solvent and probably also due to the solvent effect on the polymeric matrix.

Field Tests

A series of field tests with the plastic granular formulation were conducted in 12 beds 1 m² each. In each bed, six rows were seeded, including *Beta vulgaris* L. and *Lactuca satival* L. and grass such as *Avena satival* L., *Lolium perenne* L., *Triticum aestivum* L. and *Hordeum volgare* L. One day after seeding, the beds were treated with various propham formulations followed by irrigation. The germination percentage was counted ten days after seeding and was compared with the control. The effectiveness of the herbicide was tested by seeding grass at various time intervals after the treatment and counting the germinated seeds (tests were repeated twice). An increase in germination indicates a decrease in propham concentration available to the plant in the soil.

For demonstration purposes, the results of two field tests are described. One test was conducted in the winter (average temperature during test 8°-18°C, few rainy days with about 16 mm rain per day) and the other, in the summer (average temperature during test 17°-25°C, no rain). Each test included three kinds of beds. One kind was seeded followed by no herbicide treatment; the second kind was seeded and subsequently treated with standard propham formulation (wettable powder containing 50% active ingredient), $600 \text{ g}/1000 \text{ m}^2$; and the third kind was treated with the controlled-release formulation (containing 8% pure propham), $4 \text{ kg}/1000 \text{ m}^2$. In both treatments, the propham level was about the same. As can be seen in Figures 4 and 5, there is a dramatic difference between the three kinds. In the winter test, in those cases where herbicide was applied at the seeding day, no difference between the effectiveness of the two formulations was found. However, when beds were seeded a few days after treatment, the controlled-release formulation was advantageous. The difference in effectiveness was found to increase as the seeding date after treatment was postponed. In the summer test, it was found that full germination of grass look place already four days after treating with standard herbicide, whereas in the controlled-release formulation-treated beds, practically no grass germinated in 12 days.

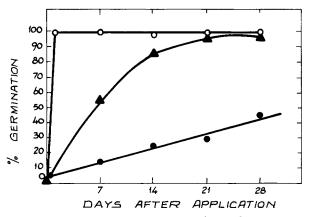


Fig. 4. Grass germination: (\blacktriangle) after application of 600 g/1000 m² propham (w.p. 50%); (\blacklozenge) after application of 4 kg/1000 m² controlled-release formulation propham; (O) after no application (winter time).

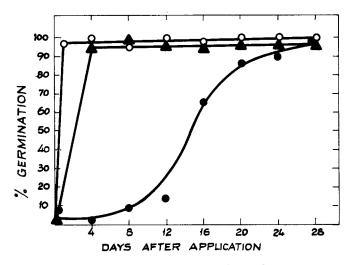


Fig. 5. Grass germination: (**A**) after application of 600 g/1000 m² propham (w.p. 50%); (**O**) after application of 4 kg/1000 m² controlled-release formulation propham; (**O**) after no application (summer time).

Granular samples were collected from the field and their propham concentration was determined. As expected, the herbicide content decreased gradually with exposure period in the field (see Fig. 6).

DISCUSSION

A volatile herbicide included in plastic granules exhibits longer effective periods than standard formulations. The difference between the two is more pronounced as the ambient temperature increases. As known, the effectiveness of volatile ingredients is dramatized at elevated temperatures. In the summer, the standard formulation was not yet active after three days, whereas the controlled-release formulation was still highly active even after 12 days. Negitt et al.¹⁰ came to similar conclusions when they evaluated granular applications including DNBP and EPIC as herbicides for weed control in potatoes. A quali-

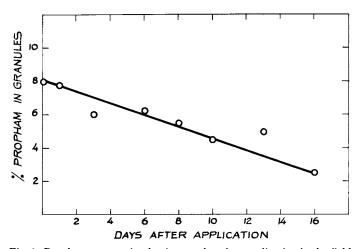


Fig. 6. Propham content in plastic granules after application in the field.

tative correlation has been found between laboratory and field tests. The water immersion test was found to be the most reliable one among the laboratory tests. At this stage, laboratory release tests are only indicative tests for comparison purposes.

In conclusion, granular formulations in which each granule acts as a slowrelease source of active ingredients protect agricultural plants for long periods of time. A granular formulation has advantages over a standard formulation. The standard formulation kills only those weeds which germinate shortly after the application, while the granular slow-release formulation affects also those which germinate later.

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