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Evaluation of release time and efficiency of a botanical insecticide pellet under laboratory conditions

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Abstract
Application of controlled release formulations in agricultural pest control has sharpened recently. Evaluation of controlled release time and its permeation is one of the most important factors of pellet formulation efficiency against stored products pests. The aim of present study was evaluation of controlled release time and efficiency of 1 g insecticide pellets, made based on Eucalyptol, botanic constituent, and Poly(vinyl alcohol), the biodegradable polymer, in laboratory condition. Pellets containing active ingredient (250 µl of 1,8-Cineole) was located in n-hexane medium (120 ml) for slow release, and sampling was carried out in 12 h intervals for 72 h with three replications. Spectrometry of samples was performed by UV-visible spectrometer. Obtained data were used for concentration-maximum absorption counting, and for drawing of concentration-time graphs. Obtained results were indicated that release rate was highest in first 12 h (33% of total release) and reduced by time gradually, and release in 60-72 h interval was inappreciable (0.65% of total release). Overall, pellets release efficiency (released amount of total 1,8-Cineole from pellets) was estimated about 83%.

Keywords: 1,8-Cineole, controlled release, pellet formulation, poly(vinyl) alcohol, UV-visible

1. Introduction
Controlled release methods were applied according to blend of biological active ingredient with polymer materials used both physical blending, for providing a release rate control mixture, and chemical binding, as a carrier for active ingredient. Selection of the best method for active ingredient release in perfect quantity for exert of its optimum biological and ecological characteristics and minimum side effects is related to active ingredient physicochemical properties in controlled release system [1].
Two attitudes were reported about active ingredients and polymer materials physical mixing. First, biological ingredient could be encapsulated in and released from polymer micropores. Second, active ingredient could be heterogenically blended with a polymer matrix. This polymer matrix could be biodegradable or not, and its release controlled with diffusion in matrix, chemical-biological erosion or both of them [2].
Poly (vinyl alcohol) (PVA) is a water soluble polymer based on petroleum resources with unique properties such as good transparency, luster, antielectrostatic properties, chemical resistance and toughness [3]. It has also good gas barrier properties and good printability. Its water solubility, reactivity, and biodegradability make it a potentially useful material in biomedical, agricultural, and water treatment areas [4]. Because of biodegradable characteristic and its low cost, Poly(vinyl alcohol) has been used as a matrix for encapsulation of the pharmacological reactive agents too [5].
Poly(vinyl) alcohol was used several times in agriculture as release matrix for agrochemicals. Loaded agrochemicals on PVA are consisting of fertilizers and chemical pesticides (for example fungicides, herbicides and insecticides) [1]. Also botanical pesticides as Azadirachtin-A were physically loaded on both PVA and Poly(vinyl) acetate and used in experiments [6]. It is believed that application of essential oil and their constituents in formulations such as granule and pellets in crop mass, enhance their ability to use in higher levels and additionally resolved their shortages as low penetration power and vapor pressure. Measurement of release time of produced formulation for evaluation of product efficiency in crop mass is very important.

Different methods are used to estimate the amount of released material in case of agrochemicals. UV-visible spectrometry is a widely used method to evaluate the released amount of agrochemicals in recent years [7, 8, 9, 10]. In this method, selection of appropriate solvent that had not any UV absorption in the same wavelength as the released material is important. The second important point is lack of chemical bonding within the solvent and solute that could alter the absorption peak.

The aim of current study was to determine the 1,8-Cineole release time and efficiency of an insecticide pellet formulation in n-Hexan solvent using UV-visible spectrometry.

2. Material and Methods:

2.1. Chemicals

n-Hexan (86.18 g/mol), 1,8-Cineole with 154.25 g/mol molar mass (Assay (GC, area%) ≥ 98%) and Poly(vinyl alcohol) (Molar Weight: 72000 g/mol; hydrolysis mole: 98) were purchased from Merck Co. (Darmstadt, Germany). All experiments were done in 2011 (June-September).

2.2. Pellets preparation and related measurements

PVA and 1,8-Cineole mixture was prepared by Dry Mixing Method (DMM) in ice temperature on stirrer with 500 rpm for 4 h. Preparation of mixture was done in tight 100 ml balloons and mixing power provided with magnetic stirrer. Then prepared mixtures were transferred to FTIR pellet maker apparatus (Thermo Nicolet part No. 0016-035, USA) and pellets were produced by hydraulic pressure under 100 Kg. Production of a pellet from prepared mixture was done in about 1 min. Average of ten 1 g pellets diameter and thickness were measured by calliper that were 1.3±0.1 and 0.93±0.01 cm, respectively [11].

2.3. Release estimation

For measuring the release time of pellets, n-hexane was used as the basis release medium. Reasons of this choice were lack of absorption in the ultraviolet light by n-hexane in 1,8-Cineole absorption wave length, no polarity, lack of hydrogen bonding with the desired component, 1,8-Cineole solubility in n-hexane, and no solution of poly(vinyl alcohol) in n-hexane. 1,8-Cineole maximum absorption and maximum wavelength (λ_M) were obtained During preliminary spectroscopies. According to obtained data, Concentration-Maximum absorption line was drawn and the line equation, for different concentrations of 1,8-Cineole was obtained by Excel 2007 software. One gram pellets with 2:5 (1,8-Cineole:PVA) were used for release time evaluation.

2.4. Wave length and maximum absorption of 1,8-Cineole

In this step n-hexane absorption by UV-visible spectrometry apparatus (PG Instruments Ltd. model: T 80) was considered as base line. Then absorption of 1,8-Cineole in n-hexane solution was obtained by mentioned apparatus and obtained spectroscopy was considered as 1,8-Cineole absorption.

2.5. Concentration-Maximum absorption of 1,8-Cineole

First solutions of 1,8-Cineole in n-hexane were prepared in different concentrations (5, 10, 15, 20 and 25 ppm) and their maximum absorption were obtained with three replications. Ten ml glass vials with robber cap (that prevented 1,8-Cineole evaporation) were used as solutions container. Using Concentration-Maximum absorption line for different concentration of 1,8-Cineole help us to predict the probable extract concentrations of pellets by analyze of their maximum absorption. Using obtained line equation in previous step, maximum absorption data were changed to released concentration (ppm) and their diagram was drawn in different sampling times.

Thereafter, the pellets with dose of 2:5 were located in n-hexane for release. Sampling of the contents for spectrometry (volume 120 ml) was performed by conventional syringe (1 ml), at successive times with constant intervals (12 h). Samples were so concentrated for spectrometry and they diluted (10 times) for successful spectrometry, and the recorded maximum absorption was multiplied by 10, ultimately. N-hexane solution was considered as base line in the spectrometer, then samples were transferred to the apparatus cells and their maximum absorption was recorded.

Glass vials were located in room temperature, without moving (in order to elimination of 1,8-Cineole solution increase error due to solvent moving) and in shadow during the experiments. Three replications were performed for each time and their mean point was considered as maximum absorption for that time.

3. Results

3.1. Concentration-maximum absorption line equation

After fitting the obtained maximum absorbance points to a trend line, resulted line equation was:

$$Y = 0.014X - 0.014$$

Where Y is maximum absorption and X is 1,8-Cineole concentration. Correlation coefficient (R^2) for obtained points with drawn line was 0.88 that indicate test precision. Because concentrations of 1,8-Cineole were diluted 10 times, evaluated concentrations were multiplied to ten after the spectrometry. Maximum absorbance for 1,8-Cineole in different concentrations is shown in figure 1.

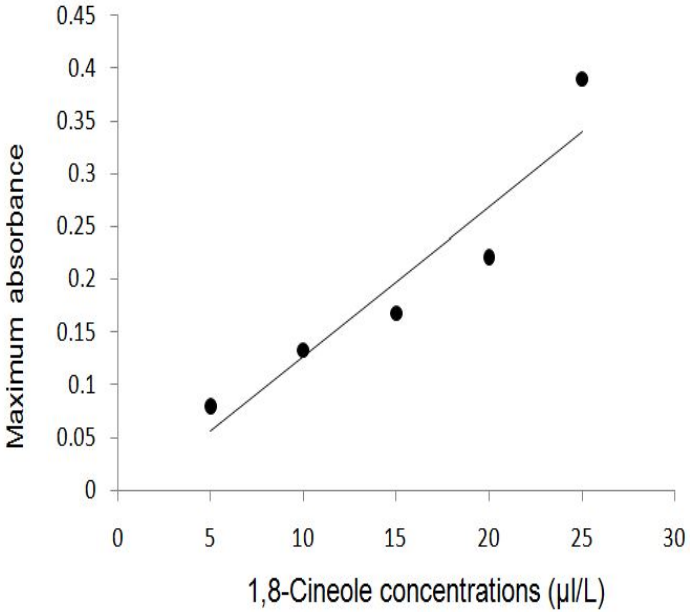

Fig 1: Maximum absorbance for 1,8-Cineole in different concentrations

3.2. Pellets release/time diagram

At first, diagram of maximum absorption-release in different times was drawn using obtained data from spectrometry of released 1,8-Cineole in sampling times. Maximum absorbance of 1,8-Cineole in n-hexan solvent at different times is shown in figure 2.

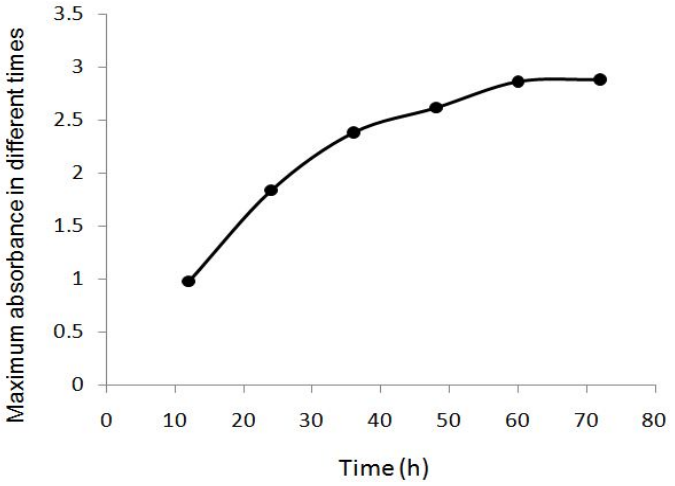

Fig 2: Maximum absorbance of 1,8-Cineole in n-hexan solvent at different times

1,8-Cineole released concentrations (ppm) at different times are shown in figure 3.

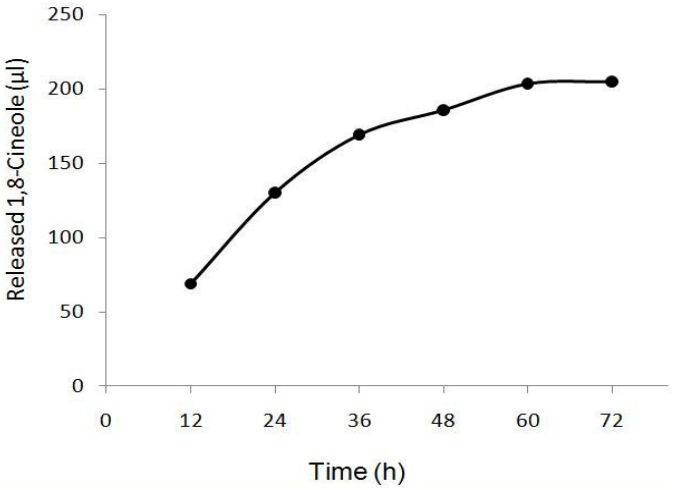

Fig 3: 1,8-Cineole released concentrations (ppm) at different times

According to release curve slope, released amount could be observed. Release amount was maximum in first 12 h (about 33%) and release rate decreased gradually by time until 60-72 h interval, that in which release amount was negligible.

Table 1: 1,8-Cineole released amount and release efficency of insecticide pellet formulation in different times

Release time (h)	Release percent*	Release efficency (%) †	1,8-Cineole captured amount (µl)
12	33.65	27.6	181
24	29.92	25.53	119.67
36	19.02	15.6	80.67
48	8.13	6.67	64
60	8.45	7.06	46.34
72	0.65	0.54	45
Total	100	83	---

* (Released amount on 12 h period of time/total released amount (205 µl) of 1,8-cineole) × 100

† (Released amount on 12 h period of time/total amount (250 µl) of 1,8-cineole) × 100

As showed in table 1, release rate was highest in first 12 h (33.65% of total release) and reduced by time gradually, and release in 60-72 h interval was inappreciable (0.65% of total release) and 45 µl of 1,8-Cineole is remained captured in polymer structure, finally. Overall, pellets release efficiency (released amount of total 1,8-Cineole from pellets) was estimated about 83%.

4. Discussion

Size and dimensions of produced formulations have been different in similar researches. Evidently, Size and dimensions of produced formulations have distinct effects on efficiency and rate of release. Release time and pest exposure to pesticide increased by enhancement in granule and pellets diameter [7, 10]. Hu *et al.* [7] used 8 cm diameter spheres as feeding stimulant and Singh *et al.* [10] used 1.07-1.34 mm diameter beads in their release rate study. Average pellets diameter and thickness in the current study were 1.3±0.1 and 0.93±0.01 cm, respectively. It would be predictable that reduction in pellets size can enhance the release efficiency.

Release media in different works were various, according to produced formulations. Some of them used media suitable for active ingredient release and solution, and the others used media similar to formulation action site. Hu, *et al.* [7] used water as release medium that was compatible with application site of formulation. While Singh, *et al.* [10] used Acetonitrile for their formulation release medium that was not compatible with fungicides usual application environment and their obtained results had purely experimental aspects. Riyajan and Sakdapipanich [6] used Methanol as release medium that was not natural environment of Neem application. In current study, we used n-hexane for release medium of produced pellets because of its suitable characteristics for 1,8-Cineole and despite of its non similarity to pellets use area. Because of the media that pellets should be use, it will be better that air be considered as pellets release media. However evaluation of release amount in air is difficult and needs gas chromatography with Solid Phase Micro Extraction (SPME) method.

In current study we used UV-visible spectrometry for evaluation of pellets released efficency that is similar to some other studies [7, 8, 9, 10]. Cao, *et al.* [12] and Riyajan & Sakdapipanich [6] used HPLC for their experimental formulation release time evaluation. Release time of controlled release formulations have been found to vary among different works due to their dimensions, their active ingredient nature and release medium. This time had ranged between 15-20 h [6] to 7 days [8].

We suggest to investigate the effect of barrier polymer on efficiency of pellets using different polymers such as poly(vinyl) acetate and starch. Also, other solvents such as acetonitrile and ethyl acetate could be used to determine the effect of solvent on pellets release efficiency. Overall, the experimented insecticide pellet formulation showed potential efficiency enough to release the botanical active ingredient, 1,8-Cineole and could be used against stored product insects in storage.

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