

Efficacy of Some Nanoparticles against the Adults of Red Flour Beetle *Tribolium castaneum* (Herbst) under Laboratory Conditions

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Abstract

Stored grain insect pests cause high deterioration to grains and seeds in storage, such as weight loss, less germination and reduced nutrition values of grains. In the present study, the efficacy of three nano-particles nano-malathion, organic nano-silica and inorganic nano-silica were evaluated by mixing with grains under laboratory conditions against the adults of *Tribolium castaneum* (Herbst) (Coleoptera: Tenebrionidae). The results showed that nano-malathion had the highest adverse effect on all parameters after 14 days of treatment. The median lethal concentrations (LC_{50s}) of organic nano-silica and inorganic nano-silica on adults of *T. castaneum* were 2.76 and 0.90% after 10 days of treatment, respectively. The lethal concentration (LC₅₀) of nano-malathion was 34×10^{-5} % while the (LC₅₀) of conventional malathion was 0.10% after 3 days of treatment. Inorganic nano silica showed significant inhibitor on the number of *T. castaneum* adult progeny at the lower concentration.

Key words: Malathion, Nano-malathion, Nano-silica, *Tribolium castaneum*, The median lethal concentration.

Introduction

The red flour beetle, *Tribolium castaneum* (Herbst, 1797) (Coleoptera: Tenebrionidae) is an important worldwide pest of stored products, it's major secondary pest of stored grain based products, including maize, rice and wheat, particularly in the tropic regions (Howe 1965; Gonzalez *et al.* 2013; Sabbour, 2014 and Waganet *et al.* 2016). This pest can cause considerable damage to stored grains and completely destroy kernels. Moreover, the product quality is affected by presence of eggs and dead insects, and holes on the grains (Lu and He 2010). The efficient control of stored grain pests has long been the aim of entomologists throughout the world. Synthetic chemical pesticides have been used for many years to control stored grain pests (Salem *et al.* 2007). Malathion is the most commonly used, and is a choice in the stored grain in different countries because of its high potency to a wide range of pests and its relatively low toxicity in mammalian (Flehi-Slim *et al.* 2015; Rezget *et al.* 2010; Wang *et al.* 2014). Nanotechnology has been considered one of the main technologies of the 21st century. This technology involves design, synthesis, characterization and application of particles or systems with dimensions less than 100 nanometer (Hoyt and Mason 2008). Also, nanoparticles a new generation of environmental remediation technologies that could provide cost-effective solution to some of the most challenging environmental clean-up problems (Chinnamuthu and Murugesu Boop-Palhi 2009) and help us to produce new pesticides, insecticides and insect repellent (Owolade *et al.* 2008). Also, researchers believe that nanotechnology will revolutionize agriculture including pest management in the near future (Bhattacharyya *et al.* 2010). Silica

nanoparticles (SNPs) have been evaluated against the cotton leafworm *Spodoptera littoralis* (El-bendary and El-Helaly 2013).

Yang *et al.* (2009) expressed that nano-particles loaded with garlic essential oils is efficacious against *Tribolium castaneum* (Herbst). Vani and Brindhaa (2013) used Silica nanoparticles (SNPs) to control stored grain pests *Corcyra cephalonica*. El-Samahy *et al.* (2014) studied (SNPs) against the lesser grain borer beetle *Rhyzopertha dominica* and the red flour beetle *Tribolium castaneum*. Gamal (2018) tested two nano-particles, silica oxide (SiO₂) and aluminium oxide (Al₂O₃) were used as stored product insect protectants in comparison with malathion as standard reference, by mixing with grains against *Sitophilus oryzae*. The present work was carried out to evaluate the efficacy of three nanoparticles nano-malathion, organic nano-silica and inorganic nano-silica insecticide against the red flour beetle *Tribolium castaneum* under laboratory conditions.

Materials and methods

Insect used:

The red flour beetle *T. castaneum* (Herbst) (Tenebrionidae, Coleoptera) was used in this study. Tests were performed in the stored product pests Laboratory at the Plant Protection Department, Faculty of Agriculture, Benha University. The insects were reared in glass jars (approx. 500 ml) containing semi-artificial diet (crushed wheat mixed with yeast (5% w/w). All cultures and experiments were held at 28 ± 2 °C and 65 ± 5 RH with 16 hours light and 8 hours dark. The glass jars were covered with muslin.

Tested Materials:

1-Malathion

Common name: Malathion

Chemical name: O, O dimethyl 1-5 (1, 2dicarboxyl) ethylphosphorodithioate.

Formula: C₁₀H₁₉O₆PS₂

The applied formulation: 57 % EC

Source: Brought from El-Nasr Chemical Industry Co., Egypt

2-Nano-malathion

The applied formulation: 10 % EC

Source: purchased from Nano Way C., Elmaadi, Egypt

3-Silica (SiO₂): and organic nano-silica (nano-SiO₂):

Appearance color: white

Appearance form: powder

Solubility: Dispersion into water or ethanol

Source: purchased from Nano Way C., Elmaadi, Egypt

Avg. Size (TEM): 100 nm

Toxicity test:

Different concentrations of tested materials were mixed with crushed wheat grains to determine their effect on adults of *T. castaneum*. These concentrations were 0.1, 0.05, 0.025, 0.0125, 0.006 and 0.003% (w/w) in the case of malathion and 5×10^{-4} , 2.5×10^{-4} , 1.6×10^{-4} , 1.25×10^{-4} and 1×10^{-4} (w/w) for nano-malathion. Also, different concentrations of organic nano-silica and inorganic nano-silica were 3, 2.5, 2, 1.5, 1 and 0.5% (w/w) each prepared concentration was added to 10g of treatment crushed wheat grains infested with 30 newly emerged adults (1-2 weeks old) of *T. castaneum*. Experiments were applied in jars (500 ml) with three replicates for each treatment and the untreated control. All replicates were kept at 28±1°C and 65-70 RH for all treatment and control. Mortality percentages were recorded at 1, 2, 3, 5, 7, 10 and 14 days after treatment. Adults of *Tribolium castaneum* were removed from the jars after 14 days holding periods. The insect progeny was inspected after 60 days post-treatment and the

reduction in the progeny was calculated according to the following equation:

$Reduction \% = (No. \text{ of adults in control} - No. \text{ of adults in treatment} / No. \text{ of adults in control}) \times 100$

Results and discussions

Toxic effect of nano-malathion at different concentrations on the mortality percentage and the reduction in progeny of *T. castaneum* adults is summarized in **Table (1)**. The mortality percentage increased with increasing concentration and exposure time. The results showed that nano-malathion at the highest concentration of 5% and exhibited highest mortality of 100%. Reduction percentages of progeny were increased with increasing concentration. The high reduction was observed with concentration of 5% for nano-malathion 100%.

Data set up in **Table (2)** indicated that the conventional malathion caused high mortality and reduced the number of progeny when the concentration increased compared to control. These data are in agreement with **Ali et al. (2007)** and **Abd-El-Salam et al. (2015)**, who found that malathion had the highest activity against the all parameters studied of *T. castaneum* adults mortality, offspring and weight loss percentage.

The LC₅₀ values for malathion and nano-malathion on adults of *T. castaneum* are shown in **Table (3)**. The calculated lethal concentrations were 34×10^{-5} and 0.10% for nano-malathion and malathion, respectively. According to these values, nano-malathion was more potent c.a. 300 fold than that of malathion against the adults of *T. castaneum*. These results indicated that nano-malathion particles has a promising agent could be used against stored insect in the future.

Table 1. Toxicity of nano-malathion mortality and reduction in progeny of *Tribolium castaneum* adults at indicated days

Concentration %(w/w)	Accumulative Adult mortality (%) after indicated days							Mean No. of the progeny after 60 days	Reduction in progeny%
	1	2	3	5	7	10	14		
5×10^{-4}	46.7	67.8	82.2	91.1	100	-	-	-	100
2.5×10^{-4}	42.2	54.4	65.6	78.9	85.6	94.4	97.8	13	97
1.6×10^{-4}	35.6	46.7	55.6	70	75.6	81.1	88.9	41	90.6
1.25×10^{-4}	32.2	43.3	51.1	64.4	72.2	76.7	82.2	89	79.5
1×10^{-4}	28.9	37.8	45.6	61.1	66.7	67.8	75.6	124	71.5
Control	0	0	0	1.1	2.2	2.2	3.3	435	-

Table 2. Toxicity of malathion and reduction in progeny of *Triboliumcastaneum* adults at indicated days

Concentration %(w/w)	Accumulative Adult mortality (%) after indicated days							Mean No. of the progeny after 60 days	Reduction in progeny%
	1	2	3	5	7	10	14		
0.1	66.7	81.1	92.2	100	-	-	-	-	100
0.05	38.9	61.1	71.1	81.1	92.2	100	-	9	97.9
0.025	35.6	55.6	60.0	65.6	74.4	82.2	87.8	51	88.3
0.0125	27.8	47.8	51.1	58.9	65.6	75.6	80.0	93	78.6
0.006	17.8	32.2	42.2	50.0	58.9	70.0	71.1	115	73.6
0.003	12.2	26.7	36.7	42.2	52.2	63.3	65.6	194	55.4
Control	0	0	0	1.1	2.2	2.2	3.3	435	-

Table 3. Comparison of the LC₅₀(w/w) % between nano-malathion and malathion on *Triboliumcastaneum* adults after three days exposure time.

Pesticides	Lethal concentration % (w/w)			Slope ± SE
	LC ₅₀	LC ₉₀	LC ₉₅	
nano-malathion	34×10 ⁻⁵ (6×10 ⁻⁶ – 175×10 ⁻⁴)	0.0145 (0.0028 – 0.074)	0.080 (0.016 – 0.416)	0.50 ± 0.36
Malathion	0.010 (0.003 – 0.031)	0.575 (0.189 – 1.751)	1.805 (0.593 – 5.495)	0.732 ± 0.24

The efficiency of both organic nano-silica and inorganic nano-silica were assessed against *T.castaneum* adults under laboratory conditions of 28 ±1°C and 65±5% RH. The results of the effect of organic nano-silica on adult mortality and reduction in progeny *Triboliumcastaneum* are presented in **Table (4)**. Mortality percentage was increased with increase both concentration and prolonged exposure time at all treated treatment. As mortality increased by increasing concentrations and time of exposure to reach 62.2, 60, 58.9, 53.3, 43.3

and 23.3% at concentrations 3, 2.5, 2, 1.5, 1 and 0.5% after 14 days, respectively for *T. castaneum*. Reduction in progeny was varied between 92.9 to 65.01% at various tested concentrations of organic nano-silica. These data were in agreement with **El-Samahyet et al. (2015)** who estimated the efficiency of silica nano particles as mortality percent on stored wheat insects *S. oryzae* and *T. castaneum* under laboratory conditions. The SiNPs mixture showed high protective effect for wheat grains against tested insects by using high concentrations.

Table 4. Efficiency of organic nano-silica against *T. castaneum* adults under laboratory conditions

Concentration %(w/w)	Accumulative Adult mortality (%) after indicated days							Mean No. of the progeny after 60 days	Reduction in progeny %
	1	2	3	5	7	10	14		
3	15.6	27.8	35.6	41.1	52.2	57.8	62.2	20	92.9
2.5	11.1	25.6	28.9	33.3	40	50	60	31	89.04
2	8.9	23.3	27.8	34.4	38.9	46.7	58.9	43	84.8
1.5	7.8	10	12.2	23.3	30	36.7	53.3	54	80.9
1	5.6	8.9	11.1	16.6	23.3	28.9	43.3	68	75.9
0.5	4.4	6.7	7.8	11.1	12.2	18.9	23.3	99	65.01
Control	1.1	1.1	2.2	2.2	2.2	3.3	3.3	283	-

Table 5. Efficiency of inorganic nano-silica against *T. castaneum* adults under laboratory conditions

Concentration (w/w)%	Accumulative Adult mortality (%) after indicated days							Mean No. of the progeny after 60 days	Reduction in progeny %
	1	2	3	5	7	10	14		
3	24.4	55.6	66.7	78.9	85.6	88.9	100	0	100
2.5	18.9	34.4	45.6	54.4	68.9	80	87.8	0	100
2	15.6	32.2	37.8	48.9	61.1	65.6	77.8	18	93.6
1.5	12.2	30	35.6	53.3	56.7	61.1	65.6	29	89.7
1	10	28.3	22.2	43.3	51.7	55	61.7	53	81.3
0.5	7.8	14.4	21.1	26.7	36.7	43.3	48.9	61	78.4
Control	1.1	1.1	2.2	2.2	2.2	3.3	3.3	283	-

The results in **Tables (4 and 5)** indicated that the efficiency of both organic nano-silica and inorganic nano-silica against *T. castaneum* adults under laboratory conditions of $28 \pm 1^\circ\text{C}$ and $65 \pm 5\%$ RH accumulative mortality percentages of *T. castaneum* increased gradually by increasing the exposure time. And the mortality percentage after treatment with organic nano-silica particles at the concentration 3% was higher as it reached 15.6 to 62.2% after one day and 14 days, respectively. Also, results showed that the organic nano-silica particles inhibited the number of progeny against *T. castaneum*. The highly reduction in F1 progeny was

observed with concentration of 3% for organic nano-silica particles (92.9%). The data in **Table (5)** showed that the differences in the mortality percentages of *T. castaneum* among treatments, as recorded one day and 14 days, increased gradually by increasing the concentration and exposure time of inorganic nano-silica particles. The number of mortality was higher as it reached 24.4 to 100% after one day and 14 days at concentration of 3%, respectively. The results revealed that the inorganic nano-silica particles inhibited the number of progeny against *T. castaneum*. Completely reduction in progeny was recorded with concentration of 3%.

Table 6. Comparison of the LC_{50} (w/w) % between organic nano-silica and inorganic nano-silica on *Tribolium castaneum* adults after ten days exposure time.

Materials	Lethal concentration% (w/w)			Slope \pm SE
	LC_{50}	LC_{90}	LC_{95}	
Organic nano silica	2.76 (1.58 – 4.81)	23.69 (13.60 – 41.29)	43.58 (25.01 – 75.94)	1.37 \pm 0.12
Inorganic nano silica	0.90 (0.55 – 1.482)	6.26 (3.81 – 10.27)	10.84 (6.61 – 17.79)	1.55 \pm 0.11

Data in **Table (6)** revealed that the values of the calculated lethal concentrations were less in inorganic nano-silica than that of organic nano-silica which LC_{50} and LC_{90} values were 0.90, 6.26 and 2.76, 23.69% for inorganic nano-silica and organic nano-silica, respectively.

These data were similar to **Rouhani et al. (2012)** who found that the presence of nano-silica and nano-silver resulted 100 and 75% mortality for adults of *Callosobruchus maculatus* at the highest concentration of 2.5 g kg⁻¹ on day 14, respectively. Also, **Ali et al. (2017)** found that the effect of nano-silica on the mortality percentages of *C. Chinese* when the unsexed adults were exposed to broad bean seeds treated with different concentrations of both tested silica types (0.5, 1.0 and 2.0 g/100 g seeds) for different periods (48, 96, 144 and 192 hrs.), it was found that NPS pronounced more insecticidal activity against the bruchid beetle *C. chinensis*.

The present study is clarified that nano-practical's especially nano-SiO₂ can be effectively used to control insect pests. Also, **Abd-El-Salam et al. (2015)** found that malathion achieved the highest effect on mortality of progeny and weight loss against *T. castaneum* as compared with Al₂O₃ and ZnO nano-particles. In addition, they indicated that Al₂O₃ had higher effect than ZnO against *T. castaneum*. **Ebeling and Wagner (1959)** proposed that insecticidal efficacy of the dust became enhanced if the particles are finally divided. Damage occurs to the insect protective wax coat on the cuticle, by sorption and abrasion. **Debnath et al. (2011)** demonstrated that the insect began to lose water due to damage of the water barrier. **Eveling (1971)** found that the insects die due to desiccation. This hypothesis for the physical mode of action makes the case in the use of nanocides stronger and the nanocides can be removed by conventional milling process unlike sprayable formulations of conventional pesticide leaving

residues on the stored grain. Therefore, silica and aluminum oxides nano-particles have a good potential to be used as grainprotecting agent and alternatives to conventionalchemical insecticides.

Conclusion

The insecticidal activity of the two types of nano-silica and nano-malathionagainst *T. castaneum*indicated the potential use of these nanoparticles as a new technology for insecticidal materials. Insecticidal activity was confirmed in nano-particles, although the results showed that the two types of nano-silica and nano-malathion varied in their effectiveness against *T. castaneum*nano-malathion had the highest effect, followed by inorganic nano-SiO₂ andorganic nano-SiO₂. The ability of using inorganic nano-SiO₂ and organic nano-SiO₂as alternatives to the chemical control of *T. castaneum*(stored grain insect pests) is possible. This approach can help reduce the amount of insecticides applied and subsequently minimize its hazards to human health and environment. Nanoparticles are promising and require some improvement in their physical properties. Further research is needed to identify its mode of action, its non-target toxicity and to determine the potential of other nano-structured materials as pest control options for insects.

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فاعليه بعض جزيئات النانو ضد خنفساء الدقيق الصدئيه تحت الظروف المعملية

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تسبب حشرات الحبوب المخزونه الكثير من المشكلات للحبوب و البذور المخزونه, مثل خساره الوزن, قلة انبات البذور وكذلك قلة القيمه الغذائيه للحبوب. أجرى هذا البحث بهدف تقييم ثلاثه من جزيئات النانو (نانوملائثيون , نانو سيليكيا عضويه و نانو سيليكيا غير عضويه) تحت الظروف المعملية كمبيدات لحشرات الحبوب المخزونه مقارنة بالملائثيون التقليدى , وذلك عن طريق خلطها بالحبوب ضد الحشرة الكاملة لخنفساء الدقيق الصدئيه. وقد اوضحت النتائج ان النانو ملائثيون كان اعلى تأثيرا فى كل المعاملات بعد ١٤ يوم من المعامله. وكان التركيز اللازم لقتل ٥٠% من التعداد للسيليكيا العضويه و الغير عضويه ٢,٧٦ و ٠,٩٠ % بعد ١٠ ايام على التوالي. وكان التركيز اللازم لقتل ٥٠% من التعداد فى حاله الملائثيوم التقليدى ١٠,١٠ % بعد ٣ ايام , بينما كانت ٣٤×١٠^{-٥} % فى حاله النانو ملائثيون. وكان للسيليكيا غير العضويه تاثير هام فى تقليل عدد الحشرات فى النسل الناتج عند التركيزات المنخفضه منها.