ANSF APPLIED AND NATURAL SCIENCE FOUNDATION

Journal of Applied and Natural Science
17(1), 285 - 292 (2025)
ISSN : 0974-9411 (Print), 2231-5209 (Online)
journals.ansfoundation.org

Research Article

Formulating hexamine, mineral oil, mixture and evaluation of its insecticidal activity against Papaya Mealybug *Paracoccus marginatus* (Hemiptera: Pseudococcidae) under laboratory conditions

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Article Info
https://doi.org/10.31018/jans.v17i1.6181
Received: September 15, 2024
Revised: February 24, 2025
Accepted: March 01, 2025

How to Cite
Eskander, M. A. *et al.* (2025). Formulating hexamine, mineral oil, mixture and evaluation of its insecticidal activity against Papaya Mealybug *Paracoccus marginatus* (Hemiptera: Pseudococcidae) under laboratory conditions. *Journal of Applied and Natural Science*, 17(1), 285 - 292. https://doi.org/10.31018/jans.v17i1.6181

Abstract
Hexamine is a heterocyclic organic chemical that has a wide range of applications. It is primarily employed as an anti-infective agent to treat urinary tract infections. Mineral oils are refined petroleum-based hydrocarbon blends of aromatic oils, paraffins, and naphthenes. Cutting and grinding fluids are made with mineral oils with a wide range of compositions. This research paper's primary goal was to formulate hexamine, mineral oil, and their combination in appropriate formulation types and assess their biological activity against papaya mealybug *Paracoccus marginatus.* The mineral oil was formulated as an emulsifiable concentrate (EC), hexamine as a soluble powder (SP), and its mixture as oil in water emulsion (EW). All developed formulations passed all tests that the WHO and FAO had prescribed for these types of formulations. Biological tests were conducted in the laboratory using serial concentrations (0.125, 0.25, 0.50, and 1%) on *P. marginatus* nymphs and adults. Mineral oil and hexamine mixture (EW) formulation demonstrated the highest activity against *P. marginatus*, nymphs and adults, followed by hexamine (SP) formulation and then the mineral oil (EC) formulation. Their LC_{50} values were, respectively, 0.0509, 0.0653, and 0.1015 for nymphs and 0.0513, 0.0767, and 0.1224 % for adults. Also, the efficacy of these formulations displayed a higher effect on nymphs than adults. After completing the necessary research, these new formulations may be applied in the field after trial to control the papaya mealybug, *P. marginatus*.

Keywords: Emulsifiable concentrate, Emulsion, Hexamine, Mineral oils, *Paracoccus marginatus,* Soluble powder

INTRODUCTION

Oils are among the safest and most efficient substitutes for synthetic insecticides and fungicides, having been used for centuries as pesticides. Mineral oils are thought to be a potential pest management solution all across the world. The benefit of mineral oils remains their ability to combat strain resistance effectively. It was also claimed that resistance to mineral oils had not been documented (Mohammad, 2016)

Hexamine is a white powder in crystal form that has a faintly sweet taste and no smell. Its formula is $(CH_2)_6N_4$. Figure 1 illustrates its chemical structure (Shurooq *et al.,* 2022).

Hexamine is mostly used in organic synthesis; for example, it is used as a vulcanizing agent, a component of pesticides, and in manufacturing explosives and plastics (Zdeka *et al.,* 2011). The invasive mealybug, *P.marginatus*, is a pest insect native to Central America that poses a significant risk to over 60 plant species from over 22 families worldwide. It is one of many invasive mealybug species that have destroyed most globally significant agricultural and horticultural crops, such as papaya, guava, cotton jackfruit, jatropha eggplant, and mango. The papaya mealybug feeds by sucking plant sap (Laneesha *et al.,* 2020).

The papaya mealybugs infest the underside of the leaves of the papaya plant along their veins, eventually moving on to the immature fruits and making them unfit for human eating and unsalable (Ahmed *et al.,* 2011). Depending on the type of the host plant, *P. marginatus* can complete its life cycle in 15 - 32 days on average. Depending on the temperature and relative humidity, it can produce 11 - 13 generations annually (Laneesha *et al.,* 2020).

Mealybugs are currently controlled using various pesticides, but their cryptic behaviour, waxy body cover, and clumped distribution pattern make many ineffective. This raises the risk of pesticide resistance. Multiple applications are needed to control mealybugs, and the active ingredients in different insecticide classes may reduce their efficacy. As pesticide resistance grows, the effectiveness of these treatments may decrease (Venkatesan *et al.,* 2016).

To create a product that is safe, easy to use, has a good shelf life, and doesn't have any unfavourable side effects, the majority of technical pesticides are prepared in advance by combining active components with inert, diluents, preservatives, adjuvants, etc (Hazra and Purkait, 2019). In Egypt in 2023, *P. marginatus* was discovered, damaging 14 host plants (Fatma and Asmaa, 2023). The present research paper aimed to find new active ingredients, test them on *P. marginatus* and formulate them in a local commercial formulation to use them to control *P. marginatus* after laboratory and field studies are completed.

MATERIALS AND METHODS

Tested chemical

Fine chemicals: Hexamethylenetetramine (methenamine, hexamine, molar mass 140.186 $g.mol^{-1}$), supplied by El- Gomhoria Co., Cairo, Egypt. Mineral oil: supplied by Cairo Oil Refining Co., Tanta Branch, Egypt. Surface active agents: Toximol-H, Tween 80 and Tween 20 were supplied by EL- Gomhoria Co. Cairo Egypt; Sodium lauryl sulfate (SLS) and poly ethylene glycol 600 di laurate (P. E. G. 600 DL) were supplied by the Egyptian Starch, Yeast and detergents Co., Alexandria, Egypt; Solvents: Acetone, xylene, ethanol and dimethylformamide (DMF) were supplied by EL-Gomhoria Co., Cairo, Egypt.

Physico-chemical properties of all formulation components

Active ingredient

The physico-chemical properties of all used substances as active ingredients were determined as follows.

Solubility: It was determined according to (Nelson and Fiero, 1954). Free acidity or alkalinity: This was determined using the same methods outlined in the WHO recommendation (1979).

Surface active agents

Surface tension: According to ASTMD-1331 (2001), it was ascertained by employing a Du-Nouy tensiometer for solutions containing 0.5% (W/V) surfactant.

Hydrophilic-lipophilic balance (HLB): A surfactant's water solubility is thought to be a rough indicator of its hydrophilic-lipophilic balance (Lynch and Griffin, 1974).

Critical micelle concentration (CMC): The method outlined by Osipow (1964) was used to determine the CMC of the tested surfactants at which the surface tension of the solution doesn't decrease with a further increase in surfactant concentration.

Free acidity/ alkalinity: It was determined as mentioned before.

Local prepared mineral oil 90 % emulsifiable concentrate (EC) formulation

Several trials were conducted to prepare mineral oil as an emulsifiable concentrate. These trials involved adding varying weights of emulsifier or blending emulsifier with different weights of oil and stirring until homogeneity was achieved. The developed formulations were subjected to an emulsion stability test using the methodology outlined in CIPAC MT 36.1(2002) to ascertain which would pass and be appropriate for use.

Locally prepared hexamine as 90 % soluble powder (SP) formulation

Soluble powder formulations were developed after determining the physical characteristics of the active ingredient and surface-active agents. Several trials were conducted by varying the weights of the active ingredients and surface-active agents, and the solubility, surface tension, foam, pH, acidity, and free alkalinity or acidity were measured to ascertain which of the resultant formulas passed the reported tests and were appropriate for use.

Preparation of hexamine and mineral oil mixture as 45 % oil in water emulsion (EW) formulation

After a series of trials, hexamine and mineral oil mixture was prepared as 45% (EW) formulations following Salvica *et al.* (2012). The emulsion stability, foam, and free acidity or alkalinity were measured both before and after storage at 54 ± 2 degrees Celsius for 14 days to determine the best formula for application following CIPAC (2002)

Physico-chemical properties of locally prepared mineral oil 90 % emulsifiable concentrate (EC) formulation and hexamine, mineral oil mixture 45% oil in water emulsion (EW) formulation

Viscosity: Using a Brookfield viscometer model DVII+Pro, the measurement was made using centipoises, in accordance with ASTM D-2196 (2005).

Surface tension: It was determined as shown before.

Emulsion stability test: It was done according to FAO/ WHO MT 36.3. (2010).
Ph it was determined by using Cole-Parmer pH conductivity meter 1484-44 according to Dobrat and Martijn, (1995).
Foam: It was measured according to the method described by (CIPAC 2002).
Free acidity or alkalinity: It was performed as mentioned earlier.

Physico-chemical properties of locally prepared hexamine as 90 % soluble powder (SP) formulation

Solubility, Foam, PH, Free acidity/ alkalinity were determined as mentioned before.

Physico-chemical properties of local, newly prepared formulations spray solution at a field dilution rate (0.5 %)

Surface tension, Viscosity, and pH, were determined as mentioned before.
Electrical Conductivity and Salinity: The Cole-Parmer pH/Conductivity meter 1484-44 was utilized to determine it, with µmhos serving as the unit of measurement for electrical conductivity, as per Dobrat and Martijn (1995).

Bioassay

With a few changes, El-Hefny *et al. 's* (2011) approach was used to assess the toxicity of the tested formulations. Eight leaves per treatment were randomly selected from infested shrubs to provide samples of infested papaya leaves, which were then placed in paper bags and brought to the laboratory. Spraying leaves involved employing concentrations of 1, 0.5, 0.25, and 0.125 % for each of the three locally developed formulations employed in this study. In a petri dish, the treated leaves were maintained in four replicates for each treatment, with four replicates receiving water treatment as a control. Using a Stereomicroscope, the inspection was carried out 1, 3, 5, and 7 days after treatment, counting the dead and living in various phases of *P. marginatus.* Additionally, a pre-count was performed as an index for each treatment.

Statistical analysis

Utilizing the Abbott formula (1925), the corrected mortality % was calculated. To estimate the LC_{50} values, the corrected mortality percentages were obtained according to Finney (1971).

RESULTS AND DISCUSSION

Formulation part

The physico-chemical characteristics of hexamine and mineral oil as active components are mentioned in Table 1. The mineral oil fraction that was utilized demonstrated total miscibility in all organic solvents was immiscible with water. Additionally, it displayed an acidic characteristic, as shown by the sulphuric acid percentage taken (0.07802). Conversely, hexamine showed total solubility in water and total insolubility in organic solvents and an alkaline property as sodium hydroxide percentage (0.2452). According to the data acquired, depending on the solubility values of each of them, the mineral oil might be formulated as an emulsifiable concentrate and the hexamine as a soluble powder (El-Sharkawy *et al.,* 2020).

The physical and chemical characteristics of the examined surface-active compounds are mentioned in Table 2. When compared to water, the surface tension values of all the surfactants that were utilized were lower; the lowest surface tension was shown by Sodium lauryl

Table 1. Physical properties of mineral oil and hexamine used as active ingredients

Compound	% Solubility (W/V)					Free alkalinity as % NaOH	Free acidity as % H_2SO_4
	Water	Acetone	DMF	Ethanol	Xylene		
Mineral oil	-*	miscible	miscible	miscible	miscible	-	0.07802
Hexamine	soluble	-*	-*	-*	-*	0.2452	-

-*: means insoluble.

Table 2. Phsico-chemical properties of the suggested surface-active agents

Surface active agent	Surface tension Dyne/ cm	CMC* %	HLB*	Free acidity as H_2SO_4	Free alkalinity as NaOH
Toximol-H	35.3	0.3	10- 12	0.2	-
Tween 80	38.4	0.5	>13	0.6	-
P.E.G. 600 Do. *	39.1	0.9	10-12	-	0.31
SLS*	31.7	0.8	>13	-	0.024
Tween 20	38.7	0.2	>13	0.37	-

P. E. G. 600 DO. *: poly ethylene glycol 600 dioleate; CMC*: Critical micelle concentration;SLS*: Sodium lauryl sulphate; HLB*: Hydrophilic-lipophilic balance sulphate (SLS), which was followed by Toximol-H, Tween 80, Tween 20, and P. E. G. 600 DO. that displayed the greatest value; their values were 31.7, 35.3, 38.4, 38.7, and 39.1 dyne/cm respectively. The CMC % of tween 20 was the lowest at 0.2, followed by Toximol-H at 0.3, Tween 80 at 0.5, SLS at 0.8 and P. E. G. 600 DO displayed the maximum value of 0.9. While all other surfactants used showed higher hydrophilic-lipophilic balance (>13), Toximol-H and P. E. G. 600 DO displayed the same hydrophilic-lipophilic balance (10 –12). P. E. G. 600 DO. and SLS demonstrated an alkaline property evaluated as free alkalinity, with values of 0.31 and 0.024 %, respectively, whereas Tween 80, Tximol-H, and Tween 20 revealed an acidic property measured as free acidity, with respective values of 0.6, 0.37, and 0.2. Because of the compatibility of the active ingredients' and surfactants' physical and chemical properties, some surfactants may be used to formulate the used mineral oil as an emulsifiable concentrate, while others may be used to prepare hexamine as soluble powder, depending on the characteristics of the tested surfactants and active ingredients as stated earlier (Eskander *et al.,* 2020)

The physical and chemical characteristics of the prepared EC and EW formulations are given in Table 3 before accelerated storage. Both formulations demonstrated full emulsion stability in both types of water and an acidic pH value for the EC formulation and an alkaline pH value for the EW formulation; respectively, these values were 5.2 and 7.4. The EC formulation displayed free acidity, calculated as sulfuric acid (0.1373), while the EW formulation displayed free alkalinity, calculated as sodium hydroxide (0.140). Neither formulation type produced any foam for either type of water used. In comparison to the EC formulation, the EW formulation had greater viscosity and surface tension values: 23.9 and 17.94 centipoise for viscosity and 33.42 and 28.95 dyne/cm for surface tension, respectively.

After accelerated storage, the physical and chemical characteristics of the prepared EC and EW formulations are shown in Table 4. The emulsion stability test revealed that neither the developed formulation experienced precipitation nor oily cream separation in hard or soft water. The results that seemed evident from free acidity or alkalinity values were that the EC formulation showed a free acidity value of 0.1128 while the EW formulation showed a free alkalinity value of (0.140). The EC formulation revealed an acidic pH value of 5.33, while the EW formulation revealed an alkaline pH value (7.55). Neither formulation produced a little foam in both soft and hard water. Similar to the situation before accelerated storage, the EW formulation's viscosity and surface tension values were higher than those of the EC formulation; the former displayed values of 23.84 centipoise and 33.6 dyne/cm, while the latter displayed values of 16.81 centipoise and 28.71 dyne/cm, respectively.

The storage outcomes under both circumstances showed that the characteristics of the newly developed formulations were unchanged, as evidenced by the values of emulsion stability, PH, and free acidity or alkalinity.

Before and after accelerated storage, the newly developed SP formulation's physical and chemical character-

Table 3. Physico - chemical properties of the prepared (EC) and (EW) formulations before accelerated storage

Formulation	Viscosity centipoise	Surface tension Dyne/cm	Emulsion stability ml cream separation		Foam		PH	Free acidity as H_2SO_4	Free alkalinity as NaOH
			H. W	S. W	H. W	S. W			
Mineral oil (EC)	17.94	28.95	0	0	0	0	5.2	0.1373	-
Mineral oil + hexamine (EW	23.9	33.42	0	0	0	0	7.4	-	0.140

P. marginatus before treatment by EW formulation

P. marginatus after treatment by EW formulations (at 0.5 %)

Fig. 1. *Showing the insect died , swelled, and the cell walls decomposed, and their contents came out*

istics were displayed in Table 5. Before being stored, the new formulation demonstrated full solubility in soft and hard water, with minimal foam values in both 5, 3 ml. Sodium hydroxide (0.2) measurements of free alkalinity confirmed the alkaline PH value of 7.9. The newly developed soluble powder formulation also demonstrated an alkaline pH value (7.7) with free alkalinity (0.22) and total solubility with minimal foam values (4, 2 ml) in soft and hard water, respectively, after being stored under accelerated conditions. The results obtained before and after accelerated storage made it abundantly evident that the newly developed soluble powder formulation can maintain its chemical and physical characteristics before and after accelerated storage (Hamouda *et al.,* 2022).

The physical and chemical characteristics of the spray solution for the developed SP, EC, and EW formulations at a field dilution rate of 0.5% are displayed in Table 6. In comparison to water (72 dyne/cm), the surface tension values of all freshly produced formulations were lower. The EC formulation recorded the lowest surface tension value, followed by the SP and the EW formulation, with respective values of 29.41, 33.55, and 35.75 dyne/cm. In the same way, the electrical conductivity and PH values that the EC formulation displayed were the lowest, followed by those of the EW formulation and the SP formulation, which showed the highest values. The EC formulation revealed an acidic pH value, while the EW and SP formulations declared an alkaline PH value. The corresponding electrical conductivity values for the EC, EW and SP formulations were 5.1, 45.6, and 432 μ mhos, while the corresponding pH values were 5.92, 7.76, and 7.99. While the other two formulations displayed no salinity (0.00 g/l), the SP formulation displayed a minor salinity value (0.2 g/l). According to the relative viscosity values of the recently developed formulations, the SP formulation had the highest viscosity value (1.44 centipoise), followed by the EC formulation (1.1 centipoise) and the (EW) formulation (0.704 centipoise). The spray solution's physical and chemical characteristics immediately impact the pesticide formulation's behaviour in the field and the target pest's biological efficacy. Because wettability arises when the attraction between the leaf surface and water is stronger than the surface tension of the liquid, the lower the surface tension of the spray solution, the higher the predicted biological activity of the pesticide. Accordingly, the agrochemical's surface tension shouldn't be extremely high (Pereira *et al.,* 2016). A needed other characteristic is the decrease in pH value with an increase in electrical conductivity, since the former would lead to a rise in the attraction between the spray solution and the treated plant, increasing deposition and penetration on the tested surface and, ultimately, increasing effectiveness (Molin and Hirase, 2004). According to (Zhang *et al.,* 2017) the second property will cause the pesticide formulation to get ionized, improving its deposit and penetration through the plant's surface. As a result, the pesticide effectiveness of these formulations will rise. A higher viscosity is also necessary since it can lead to a higher biological efficacy since it reduces drift and retains spray solution sticking to plant leaf surfaces (Carvalho *et al*., 2018).

Biological activity

The evaluated formulations' insecticidal activity against *P. marginatus* nymphs and adults in the lab are shown in Table 7 at serial concentrations of 0.125, 0.25, 0.5, and 1%. After three days of treatment, the highest concentration used 1% revealed the highest percentage of mortality, 79 and 75, while the lowest concentration used revealed the lowest percentage of mortality, 52

Table 4. Physico-chemical properties of the prepared EC and EW formulations after accelerated storage

Formulation	Viscosity centipoise	Surface tension Dyne/cm	Emulsion stability ml cream separation		Foam		PH	Free acidity as H_2SO_4	Free alkalinity as NaOH
			H. W.	S. W.	H. W.	S. W.			
Mineral oil (EC)	16.81	28.71	0	0	2	1	5.33	0.1128	-
Mineral oil + hexamine (EW)	23.84	33.66	0	0	3	2	7.55	-	0.14

Table 5. Physico-chemical properties of the newly prepared SP formulation before and after accelerated storage

Formulation	Before storage						After storage					
	Solubility		Foam		pH	Free alkalinity as NaOH	Solubility		Foam		pH	Free alkalinity as NaOH
	S. W	H. W	S. W	H. W			S. W	H. W	S.W	H. W		
Hexamine (SP)	100	100	5	3	7.9	0.2	100	100	4	2	7.7	0.22

Table 6. Physico-chemical properties of spray solution of the prepared (SP), (EC), and (EW) formulations at field dilution rate (0.5 %)

Formulation	Viscosity centipoise	Surface tension Dyne/cm	Conductivity (μ mhos)	Salinity %	PH
Hexamine (SP)	1.44	33.553	432	0.2	7.99
Mineral oil (EC)	1.1	29.241	5.1	0	5.92
Mineral oil + hexamine (EW)	0.704	35.753	45.6	0	7.76

Table 7. the insecticidal efficacy of the tested formulations against nymphs and adults of *Paracoccus marginatus* under laboratory conditions

Tested formulations	Conc. %	% Mortality of developmental stages of P. marginatus after							
		1 Day		3 Days		5 Days		7 Days	
		N	A	N	A	N	A	N	A
Mineral oil (EC)	1	55	45	79	75	95	93	100	100
	0.5	47	40	73	69	93	91	100	98
	0.25	40	35	64	61	84	82	97	94
	0.125	35	30	52	49	78	75	88	87
Hexamine (SP)	1	75	65	86	81	100	98	100	100
	0.5	67	57	81	76	97	96	100	100
	0.25	57	52	73	69	92	90	100	100
	0.125	43	40	59	55	82	81	100	100
Mineral oil + hexamine (EW)	1	84	81	99	100	100	100	100	100
	0.5	78	74	98	95	100	100	100	100
	0.25	66	61	93	88	100	100	100	100
	0.125	56	51	77	77	100	100	100	100

N= nymph A= adult

and 49% for nymphs and adults, respectively. Throughout all treatment periods, the mineral oil (EC) new formulation demonstrated a direct relationship between the percentage of mortality and the increase in concentration. As the concentration increased, the percentage of mortality increased for both stages under study. For all concentrations used, a further direct relationship was found between the percentage of mortality and treatment duration; the longer the treatment duration, the higher the percentage of mortality. For nymphs, the concentration of 1% resulted in 55, 79, 95, and 100% mortality percentage; for adults, it produced 45, 75, 93, and 100% mortality percentage after 1, 3, 5, and 7 days of treatment, respectively. For the two stages under investigation, the same relationships were also noted for the other two formulations, hexamine soluble powder and mineral oil, hexamine mixture oil in water emulsion formulation. Increasing concentration led to a rise in the percentage of mortality, and extending the treatment period also increased the percentage of mortality for each formulation. After three days of treatment, the EW formulation gave the highest mortality value (99 and 100%) followed by hexamine SP formulation (86 and 81%) followed by the mineral oil EC formulation (79 and 75%) for nymphs and adults, respectively. Hexamine and hexamine mixture oil in water emulsion formulation showed the highest activity with the highest concentration compared to mineral oil formulation on both stages under study for all concentrations and all treatment periods.

The LDP line parameters of the formulations evaluated against *P. marginatus* nymphs and adults in laboratory are shown in Table 8. After 3 days of treatment, the mineral oil (EC) formulation had the greatest LC_{50} values for both nymphs and adults, at 0.1015 and 0.1224, % respectively. Similarly, at three days after treatment, the LC_{50} value of hexamine mixture oil in water emulsion formulation was the lowest compared to the LC_{50} values of the other two formulations; their LC_{50} values were 0.0509, 0.0653, and 0.1015 % for nymphs and 0.0513, 0.0767, and 0.1224 % for adults for (EW) formulation, (SP) formulation, and (EC) formulation, respectively. After three days from treatment, the slope values for the three formulations under study were 1.9972± 0.3901, 0.9542± 0.2111, and 0.8434± 0.1985 for nymphs and 1.7921± 0.3351, 0.8312± 0.2939, and 0.7735± 0.1946 for adults, for the three previously arranged formulations. These results were corroborated by the calculated slope values, which showed an increase for each formulation with the increase in the period of treatment. For all formulations under study hexamine mixture (EW) demonstrated the highest ac-

Table 8. LC_{50}, LC_{90} and Slope of the tested formulations against nymphs and adults of *P. marginatus* under laboratory conditions.

Tested formulations	Parameter	Developmental stages of P. marginatus after			
		1 Day		3 Days	
		N	A	N	A
Mineral oil (EC)	LC_{50}	0.6413	1.9004	0.1015	0.1224
	LC_{90}	114.6922	1537.8402	3.3577	5.5526
	Slope	0.5690 ± 0.1890	0.4407± 0.1911	0.8434± 0.1985	0.7735± 0.1946
Hexamine (SP)	LC_{50}	0.1782	0.2604	0.0653	0.0767
	LC_{90}	4.1285	20.2258	1.4384	2.6704
	Slope	0.9391 ± 0.1948	0.6780± 0.1894	0.9542± 0.2111	0.8312± 0.2939
Mineral oil + hexamine (EW)	LC_{50}	0.0879	0.1202	0.0509	0.0513
	LC90	1.8821	2.4927	0.2331	0.2661
	Slope	0.9633 ± 0.2051	0.9730 ± 0.2001	1.9972± 0.3901	1.7921± 0.3351

N= nymph A= adult

tivity (the lowest LC_{50} and highest slope values), while the mineral oil (EC) formulation demonstrated the lowest activity (the highest LC_{50} and lowest slope values). These findings were based on the LC_{50} and slope values of the tested formulations.

The obtained results may be attributed to the mode of action of the tested active ingredients either in single formulations (the Mineral oil 90 % (EC) and hexamine 90 % (SP)) or in mixed formula (the mixture 45% (EW)). Mineral oils dissolve the insect body's waxy layer causing dehydration (Helmy *et al.,* 2012). Hexamine has some fumigant properties and acts as a stomach and contact poison (Chemotechnique, 2017). Therefore, the mixture of both tested materials in one formula (EW) increased the activity against *P. marginatus* more than mineral oil or hexamine alone due to the two ways of toxicity that the mixed formula may show. These results also agreed with the results reported by Eskander *et al.* (2020), who reported that diesel oil mixture with thymol or camphor showed higher mortality than diesel oil alone against mealybug Ferrisia virgate, whereas the mixture of diesel oil with thymol showed higher efficacy than diesel oil with camphor. LC_{50} values were 4.02, 4.51, 4.86 and 5.33 ml / l for diesel oil alone, while it was: 0.17, 0.17, 0.25 and 0.25 ml / l for diesel oil mixed with thymol and 0.88, 0.99, 1.12 and 1.24 ml / l for diesel oil camphor mixture for 1^{th} , 2^{nd}, 3^{rd} and adult of *F. virgate* respectively.

Conclusion

Hexamine was formulated as a soluble powder (SP), mineral oil as an emulsifiable concentrate (EC), and the mixture of the two as oil in water emulsion (EW), depending on the physic-chemical properties of hexamine, mineral oil, and their mixture. All prepared formulations passed all tests that were required. The prepared materials tested in the laboratory against papaya mealybug, *P. marginatus* adults and nymphs showed that. The maximum activity against *P. marginatus* nymphs and adults was shown by the mineral oil (EC) and hexamine (SP) mixture oil in water emulsion (EW), followed by hexamine soluble powder (SP) formulation and the mineral oil emulsifiable concentrate formulation and their effect on nymphs were higher than on adults. From the above results, the newly formulated hexamine oil mixture as 45 % EW could be used for controlling mealybug, *P. marginatus,* after conducting other required field experiments.

Conflict of interest

The authors declare that they have no conflict of interest.

REFERENCES

1. Abbot, W. S. A. (1925). A method of computing the effectiveness of an insecticide; *J. Econ.Ent.*, 18(2), 265-267. https://doi.org/10.1093/jee/18.2.265a
2. Ahmed K. N.; Al-Helal M. A.; Khanom N-E-P. & Bulbul S. (2011). Control strategies of papaya mealybug, *Paracoccus marginatus* Williams & Willink infesting vegetable crops in Bangladesh, *The Journal of Plant Protection Sciences*, **3**(1), 44 – 47
3. American Society of Testing Materials ASTM (2001). Standard Test Method for Surface and Interfacial Tension Solution D-1331.
4. American Society of Testing Materials ASTM (2005). Standard Test Method for Rheological Properties of Non – Newtonian Materials by Rotational (Brookfield type) Viscometer, D-2196 copyright ASTM, Bar Harbor Drive, West Conshohocken, PA 19248-2959, United States.
5. Carvalho, F. K., Antuniassf, U. R., Chechetto, R. G. & Mota, A. A. B. (2018). Viscosity, surface tension and droplet Size of spray liquids containing formulations of insecticides and fungicides with oil- based adjuvants. *International Advances in Pesticide Application. Aspects of Applied Biology* **137**, 183- 190.

6. Chemotechnique, (2017). MB Diagnostics AB Modemgatan 9, (SE- 235 39) Vellinge- Sweden.
7. CIPAC (2002). Collaborative International Pesticides Analytical Council Limits Hand book Vol. F. Physico- chemical Methods for technical and formulated pesticides.
8. Dobrat, W. &Martijn, A. (1995). Collaborative International Pesticides Analytical Council CIPAC hand book, F, Physicochemical Methods for Technical and Formulated Pesticides.
9. El-Hefny, S. Amany; ELsahn, N. M. & Yacoub, S. SH. (2011). Effect of some plant extracts on citrus mealy bug *Planococcus citri* (Risso), *Egyptian Journal of Agricultural Research*, 89(2), 511-519. DOI: 10.21608/ejar.2011.175651
10. El-Sharkawy R. A., Hamouda S. E. S. & Elmasry S. N., Formulation of the newly synthesized arylidene derivative as 10 % flowable and evaluation of their insecticidal efficacy on cotton leafworm *Spodoptera littoralis* (Lepidoptera: Noctuidae). *Egyptian Journal of Plant Protection Research Institute*, 3 (1), 433 - 443 (2020).
11. Eskander, M. A., Fatma, A. Moharum & Sannaa, A. M. Abd El- Mageed (2020) Toxicity of the locally formulated diesel oil alone and mixed with botanical synthetic materials against mealybug *Ferrisia virigate* (Cocherell). *Journal of plant protection and pathology*, Mansoura Univ., 11 (4), 211- 214. DOI: 10.21608/jppp.2020.96006
12. FAO/WHO (2010). Manual on Development and Use of FAO and WHO Specifications for Pesticides, 1st Ed. 3rd Rev. FAO *Plant Production and Protection, FAO*, Rome, MT 36.3
13. Fatma, A. M. & Asmaa, A. T. (2023). First record of papaya mealybug, *Paracoccus marginatus* Williams and Granara de Willink (Hemiptera: Coccomopha: Pseudococcidae) in Egypt. *Future Journal of Agriculture Science*. (4), 59-66.
14. Finney, D. J. (1971). Probit Analysis, A statistical Treatment of the Sigmoid Response Curve, 7th Ed., Cambridge Univ. Press, England.
15. Hamouda, S. E. S., Abd-Alla, H. I. & Abd Elattif, Nosa S. (2022). Schiff base synthesis, formulation as emulsifiable concentrate and study its nematicidal efficiency on root-knot nematode Meloidogyne spp. under greenhouse conditions, *International Journal of Agricultural Technology* .18 (5),.12-1
16. Hazra, D. K. & Purkait, K. (2019). Role of pesticide formulations for sustainable crop protection and environment management: A review. *Journal of Pharmacognosy and Phytochemistry*, 8(2), 686-693
17. Helmy, E. I.; Kwaiz, F. A. & El-Sahn, O. M. N. (2012). The usage of mineral oils to control insects, *Egyptian Academic Journal of Biological Sciences,* 5(3), 167 -174. Doi.10.21608/eajbsa.2012.14277
18. Laneesha M.; Sachin S. S.; Babasaheb B. F. & Shankarganesh K. (2020). Papaya mealybug (*Paracoccus marginatus*) (Hemiptera: Pseudococcidae): A new threat to agri-horticulture ecosystem. *Indian Journal of Agricultural Sciences* .90 (3), 455–62, Review Article https://doi.org/10.56093/ijas.v90i3.101448.
19. Lynch M. I. & Griffin W. C. (1974). Food Emulsions in: Emulsion Technology, by Lissant K. J., Marcell Decker, Inc., New York. Mukerjee P. & K. J. Mysels (1971) Critical Micelle Concentration of Aqueous Surfactant Systems. National Bureau of Standards Washington DC, PP. 1-21.
20. Mohammad R. D., (2016). Comparison of mineral spray with current synthetic pesticides to control important pests in citrus orchards and their side effects. *Arthropodos*, 5 (2), 56- 64.
21. Molin W. T. & Hirase K. (2004). Comparison of Commercial Glyphosate Formulations for Control of Prickly Side, Purple Nutsedge, Morning Glory and Sicklepood. *Weed Biology and Management*, 4(3), 136-141. DOI: 10.1111/j.1445-6664.2004. 00130.x
22. Nelson, F. C., &Fiero, G. W. (1954). Pesticide formulations, a selected aromatic fraction naturally occurring in petroleum as a pesticide solvent. *Journal of Agricultural and Food Chemistry, 2*(14), 735-737. https://doi.org/10.1021/jf60034a005.
23. Osipow I. L. (1964). Theory and Industrial Applications, Reinhold Publishing Crep., New York, 473PP.
24. Pereira V. J., Da Cunha J. P. A. R., De Morais T. P., de M., Ribeiro-Oliveira J. P. & De Morais J. B. (2016). Physical-chemical Properties of Pesticides: Concepts, Applications, And Interactions with the Environment *Bioscience Journal*, 32(3), 627-641, DOI: https://doi.org/10.14393/BJ-v32n3a2016-31533
25. Salvica G., Dragica B., Ljiljana R., & Andelka T. (2012). Development of water-based pesticides system. *Journal Pesticides and Phytomedicine* (Belgrade). 27 (1), 77-81. DOI: 10.2298/PIF1201077G
26. Shurooq T. A.; Riyadh S. A.; Nasma B. A.; Mohammad F. A.; Ali H. & Hashim M. (2022). Statistical investigation of study the effects of different parameters on hexamine production using response surface methodology (RSM), University of Thi -Qar, *Journal for Engineering Sciences*, 12 (1), 146-152. DOI: 10.31663/tqujes.12.1.429.
27. Venkatesan, T. V.; Jalali, S.K.; Ramya, S.L. & Prathibha M. (2016). Insecticide Resistance and Its Management in Mealybugs. © Springer India 2016 223 M. Mani, C. Shivaraju (eds.), Mealybugs and their Management in Agricultural and Horticultural crops, DOI 10.1007/978-81-322-2677-2_17
28. World Health Organization, WHO (1979) Specification of Pesticides Used in Public Health, 5th Ed. Geneva.
29. Zdeka M.; Martin B.; Tomáš M. & Ivan Š. (2011). Determination of Hexamine in the Dithane DG Fungicide Using Capillary Isotachophoresis, *Sensing in Electrolysis*. 6, 368 – 379.
30. Zhang H., Lu J., Wei D., *et al*. (2017). Wetting state and dynamic spreading behaviour of alkyl polyglucoside drop on cucumber leaf surface. *Transaction of the Chinese Society of Agricultural Engineering,* 33 (3): 81-87, doi 10.11975/j.issn. 1002-6819.2017.03.011.