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Primary Screening of German Chamomile Oil as Insecticides, Baits, and Fumigants in Nanoformulations against the Health Pest *Periplaneta americana* (L.) (Dictyoptera: Blattidae)

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Cockroaches are household pests with a health risk to humans. Natural products may have insecticidal activities against pest insects and thus inspire the development of novel insecticides as "biopesticides". In this study, we report a strategy to prepare nanoparticles of German chamomile (*Matricaria chamomilla* L.) oils (= essential oils or EO) to control the American cockroach in the two forms: one is coated with polyethylene glycol (PEG) as nanocapsiolation (NC) and the other is loaded with oil as a nanoemulsion (NE). First, the nanoformulations were prepared and characterized by evaluating droplet size, zeta potential, polyspersion index and morphology. The compounds of EO were also analyzed by Gas Chromatography–Mass Spectrometry (GC–MS). A complete FTIR characterization was made to confirm the formation of EO–PEG composite. In a fumigant bioassay, EO and NE caused 100% and 44.44% mortality of cockroaches at 10 mg/liter after 1 h and 72 h of application, respectively. In a dusting toxicity bioassay, NC caused 100% mortality at 1.5 mg/500 ml after 24 h. Overall, the obtained results suggested that the oil–based nanoformulations could serve as an alternative method for producing biopesticides to overcome the disadvantage of the natural and semisynthetic pesticides and to increase their efficacy to control cockroaches. In addition, the PEG improved the stability of oil products and increased their dispersion in the aqueous phase. Although we found the essential oils of German chamomile could be useful to control the cockroaches, the techniques should be improved the nanoformulations to obtain practical effectiveness.

Key words: Pest control, botanical pesticides, insecticidal activity, toxicity, nanotechnology

INTRODUCTION

The American cockroach, *Periplaneta americana* (L.) (Dictyoptera: Blattidae) is a large and robust pest insect and prefers warm and wet urban environments such as heating rooms and sewage systems (Wannigama *et al.*, 2014; Zou *et al.*, 2016). The American cockroach is active at night and come out of its hideouts for feeding. The adults are long–lived (one year or more) and can produce a large number of ootheca (*i.e.*, egg capsules) depending on the availability of food (Strong *et al.*, 2000; Baggio–Deibler *et al.*, 2018). Current control methods targeting adult cockroaches include indoor residual spraying and/or the use of several synthetic insecticides classified into nine chemical groups, *i.e.*, sulfonamide, arylpyrazole, pyrethroids, organophosphates, neonicotinoids, insect growth regulators, inorganic, hydrazine, and carbamates (WHO, 2006). However, the overuse of chemically synthesized pesticides has resulted in a number of problems such as acute or/and chronic toxicity to humans and vertebrates (Govindarajan *et al.*, 2011; Muthukumaran *et al.*, 2015) and development of pesticide resistance through behavioral adaptation or metabolic activities (Sogorb and Vilanova, 2002; Khan *et al.*, 2014; Nkya *et al.*, 2014; Tahir *et al.*, 2017). These issues are prompting the search for safe and effective alternatives such as eco–friendly natural products or organic pesticides (Suzuki and Yamato, 2018).

Natural products have long been used as botanical insecticides because they contain a variety of plant secondary chemicals that have physiological activities or biological reactions against a wide range of organisms, some of which can be used in pest management programs (Bacci *et al.*, 2015; Campos *et al.*, 2015; Tran *et al.*, 2016, 2017; Benelli and Pavela, 2018). Also, botanical insecticides are recognized as safer alternatives with lower toxicity to human and animals, and, hence, can be used in environmentally friendly pest control (Isman, 2015; Laosinwattana *et al.*, 2018). One of the most important botanical insecticides that has traditionally been used is essential oils (Suzuki and Yamato, 2018). Essential oils (EOs) are extracted from natural plants by distillation methods and are hydrophobic liquids containing complex mixtures of low molecular weight volatiles and other compounds. These oils are rich in phenylpropanoids and terpenoids (*e.g.*, monoterpenes and sesquiterpenes) (Bhavya *et al.*, 2018), and have proved to have a toxic effect against several pests: They can be used in pest control as growth inhibitory (Isman, 2006), contact pesticide (Lee and Lee, 2016), fumigant (Liang *et al.*, 2017), repellant (Rajendran and Sriranjini, 2008), and antifeedant (Kanda *et al.*, 2017). Nevertheless, these compounds may have characteristics that represent obstacles to their use such as water insolubility, limited physical stability, chemical instability, quick degradation,

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and short residual activity (Pavela *et al.*, 2018).

A significant tool to overcome the aforementioned drawbacks can be applications of nanotechnology (Campolo *et al.*, 2017; Oliveira *et al.*, 2017). Nanotechnology is currently considered a novel approach in diverse fields of research and also has potential for the development of improved insecticide formulations with small particle sizes ranging from 50 to 200 nm (Tadros *et al.*, 2004). Thus, by applying nanotechnology tools, it is possible to develop pesticides with nanoemulsions and nanopowder (Athanassiou *et al.*, 2018; Cui *et al.*, 2018). Nanoemulsions are oil–in–water dispersions composed of small lipid droplets in the range of 50–300 nm, achieving protection of active ingredients while enhancing stability or reducing isolation of components and decreasing volatility (Huang *et al.*, 2018). Also, nanopowders synthesized with polyethylene glycol (PEG–400) loaded with essential oils may show increasing solubility in water, increasing stability and efficiency at a lower dose, reducing the economic cost for each application, protecting active ingredients from losses from degradation and evaporations, and also decreasing toxicity to beneficial insects (Choudhury *et al.*, 2012). Thus, nanoformulations of essential oils can be used in the production of effective bio–pesticides that have advantages over conventional pesticides, including ease of availability, low mammalian toxicity (not true in all cases) and rapid degradation (Rajendran and Sriranjini, 2008; Hashem *et al.*, 2018). Essential oils having biopesticide potential are known from many plant families such as Lauraceae, Rutaceae, Lamiaceae, Piperaceae, Apiaceae, Cupressaceae, Poaceae, Zingiberaceae, and Asteraceae (Boulogne *et al.*, 2012; Hashem *et al.*, 2018)

German chamomile (*Matricaria chamomilla* L.), an annual plant belonging to Asteraceae, is widely used as a food supplement and is considered a healthy food (Singh *et al.*, 2011; Raal *et al.*, 2012). Its essential oils and their compounds exhibit antimicrobial, antifungal, antiaflatoxigenic activities as well as antioxidant, pesticide, or repellent activities against different organisms (Awadalla *et al.*, 2017). The main active constituents are flavonoids, *i.e.*, apigenin and its derivatives, chamazulene, farnesene, α-bisabolol, dicycloethers, and coumarins (Petrul'ová–Poracká *et al.*, 2013). Numerous compounds from the essential oils can exhibit insecticidal activity through neurotoxic effects mainly on octopamine synapses and GABA, or by inhibiting acetylcholinesterase (Pavela and Benelli, 2016).

In the present study, we highlighted the usefulness and efficacy of German chamomile oils modified by physical and chemical nanotechnology as an organic pesticide against cockroach pests. For this purpose, we created some formulations to apply and test the application methods such as surface contact, topical application, fumigation, dust, and toxic baits. To the best of our knowledge, this paper is the first report using the essential oil of *M. chamomilla* and their nanoformulations against *P. americana*.

In this study, two novel functionalized *M. chamomilla* essential oils, *i.e.*, nanoemulsion and nanopowder, were prepared based on colloidal delivery systems to improve their chemical and physical properties. Then, the chemical components of the essential oils and their nanoemulsion were analyzed with a Gas Chromatography–Mass Spectrometry (GC–MS). The nanoformulation was characterized with regard to their particle size distribution, zeta potential (ZP), entrapment efficiency (EE), and morphology. The particle size, interfacial charge, crystallinity, suspensibility, wettability, stability and bioavailability of nanopowder were also characterized to evaluate the formulation performance. Afterwards, the toxicity of essential oils and their nanoformulation against American cockroach were evaluated. Our study provides a promising strategy to construct effective nanoformulations for sensitive pesticides. This solid nanodispersion could substantially reduce the surfactant dosage and decrease the frequency of administration relative to conventional formulations, and have a significant prospect in preparing biopesticides for crop and environmental protections.

MATERIALS AND METHODS

Plant essential oils and chemicals

Polysorbate 80 (Tween 80), polyethylene glycol 6000 (PEG) and ethanol were obtained from El–Gomhouria for Trading Chemicals & Medical Appliances Co., Egypt. The water used throughout this study was distilled water (18 0; Sartorius–arium@611VF). Essential oils (EO) of German chamomile (*Matricaria chamomilla*) were kindly provided by Hashem Brothers Company for Essential Oils and Aromatic Products (Kafr–Elsohby, Kalyoubeya, Egypt).

Preparation of nanoformulations

Nanoemulsions (NE), *i.e.*, oil–in–water nanoemulsions of the two oils (14%), were prepared according to Hamouda *et al.* (1999), which were further detailed by Joe *et al.* (2012). Briefly, Tween 80 was used as a non–ionic surfactant, and the oil phase of the nanoemulsion consisted of the essential oil (14% of the total emulsion); ethanol (3%) and biosurfactant (Surfactin, Tween 80; final concentration 3%) representing 20% (v/v) of the emulsion (Hashem *et al.*, 2018). The oil phase was mixed and kept for 1 h at 86°C, subsequently mixed with distilled water (80% v/v), kept for 3 min and finally centrifuged at 10,000 rpm for 15 min to produce nanoemulsion formula.

Nanoencapsulation (NC) of *M. chamomilla* oils was prepared by a melt dispersion technique according to Yang *et al.* (2009). Different parts of PEG 6000 were heated at 65°C. The mixture was stirred with glass rod to ensure even distribution and after being cooled at 25°C, grounded in a mortar pestel, sieved (using a sieve mesh 200), and stored in an air tight container till further experiments. The process parameters during preparation of NC were precisely controlled to avoid any significant influence on nanoparticles characteristics.

Characterization of essential oils and their nano-formulations

A TRACE™ 1310 gas chromatography equipped with Fisher Trace ISQ mass spectrometer (GC–MS) column (SLB™–5ms 30 m × 0.25 mm × 0.25 μm, Sigma–Aldrich, St. Louis, MO, USA) was used for the separation of the EO and NE compounds. A helium (purity 6.0, Westfalen AG, Münster, Germany) carrier gas with a flow rate of 1.5 mL min^{-1} was used with a sample injection volume of 1 μL and a split of 1:5 at an injection temperature of 250°C. Regarding to GC settings, the oven program was started at 40°C for 1 min, and increased at 3 K min^{-1} to 60°C and then at 30 K min^{-1} until 280°C was reached; the final temperature was kept constant for 8 min (Stenzel *et al*., 2018). The MS operating parameters were conducted through injector and MS transfer line temperatures were 250°C; ion source temperature: 250°C; ionization energy: 70 eV; total ion scan mode with mass scan range of 50–500 amu (scan time: 0.2 s) (Li *et al*., 2016). Compounds of both samples were identified by comparing linear retention indices (RI), retention times (RT), and their mass spectra with those obtained from authentic samples (purchased from the Sigma–Aldrich Group) , the NIST/NBS in Wiley libraries, and the literature, and then individual compounds were determined based on GC peak area (Adams, 2007). All of the steps of sample preparation, extraction and analysis procedures were carried out in the Laboratory of the Hashem Brothers Company (Abdel Moneim Riad St., Giza, Egypt).

The isotropy and morphology of each sample were examined using Olympus BX–51 microscope equipped with a Color Evolution LC digital camera (PL–A662) and PixelLINK image analyzer software. One drop of each sample was transferred onto a glass slide, covered by a coverslip and then analyzed under polarized light microscopy. The average droplet size (d.nm), viscosity (cP), and poly dispersivity (PDI) were determined by a dynamic laser scattering method (Zetasizer Nano ZS90), and also the zeta potential (mV), Zeta Deviation (mV) and Conductivity (mS/cm) were measured by photon correlation spectroscopy using the kit ZetaPlus (Zhermack, Badia Polesine, Italy) (Arancibia *et al*., 2016). Fourier transform infrared spectrophometer (FT–IR) was used to investigate the functional groups and the presence of vibration/stretching bands to characterize the chemical compatibility of the NC and its compounds (EO and PEG); the spectrum was scanned over the frequency range of 4000–500 cm^{-1}.

Insects

A colony of *P. americana* was established on bread with molasses and provided with distilled water at 27 ± 2°C and 70 ± 5% RH. Newly emerged adults (2–4 days old) of both sexes were used in the following bioassays; test insects were kept in 500 ml jars and the jars were placed in a room conditioned at 27 ± 2°C in semi–darkness. All bioassays were replicated three times with a control group to compare.

Bioassays

Fumigant toxicity. Appropriate concentrations of the EO dissolved in acetone (2.5, 5.0, 7.5 and 10 mg/liter) and NE dissolved in distilled water (2.5, 5.0, 7.5 and 10 mg/liter) were applied individually onto filter paper circles (Whatman No. 1, 2.5 cm in diameter). After solvent evaporation for 30 min, each of the filter papers was attached to inner surface of the cap of glass containers (500 mL), in which nine cockroach adults of mixed sex each had been placed. Filter papers that were impregnated with distilled water or acetone were used as control for comparison with NE and EO treatments, respectively. The caps were secured tight, and three replicates were prepared for each treatment and control. When knockdowns were observed, insects were considered intoxication or dead. Mortality was evaluated at 1, 2, 3, 6, 12, 24, 48 and 72 h after admission.

Topical toxicity. Aforementioned doses of the EO and NE were topically applied with a micro–applicator (Burkard, UK) to the abdomen of adult cockroaches that had been anesthetized using CO_2. The controls received only acetone and distilled water (2 μm) as an alternative of EO and NE, respectively. Batches of three treated adults were put into glass containers (250 mL), and mortality was determined 1, 2, 3, 6, 12, 24, 48 and 72 h after application. Each assay was performed 3 times.

Spray and dusting toxicity. EO dissolved in acetone (2.5, 5.0, 7.5 and 10 v/v) and NE dissolved in distilled water (2.5, 5.0, 7.5 and 10 v/v) were tested for spray toxicity to cockroaches. Also, dry dust applications were assessed with NC (0.25, 0.5, 1.0 and 1.5 gm/500 ml glass) to examine dusting toxicity. A PEG dust only was further included as a positive control. The dust deposit of NC and the solutions of NE and EO were distributed on the whole cockroach parts. Three replications each containing nine cockroach adults per dose were conducted. Mortality was assessed at 1, 2, 3, 6, 12, 24, 48 and 72 h after the applications.

Feeding toxicity. Cockroach diets were mixed with one milliliter of EO or NE at different concentrations (2.5, 5.0, 7.5 and 10 ml/litter for EO and 0.25, 0.5, 1.0 and 1.5 gm/5 gm diet for NC) and were then given as food to adult cockroaches. Acetone, distilled water and PEG 6000 each was mixed with diets as a negative control for EO, NE and NC, respectively. Three replications containing nine cockroach adults per dose were conducted. Mortality was assessed at 1, 2, 3, 6, 12, 24, 48 and 72 h after the applications.

Statistics Analyses. Raw data of *P. americana* mortality were corrected by Abbott's formula (Abbott, 1925) when they exceeded 5% with regard to mortality in untreated control variants, and the corrected data expressed as percentages were reported.

RESULTS AND DISCUSSION

Chemical constituents of EO and NE

The GC–MS detected 41 different components, which formed 99.97% of the total components of the tested samples (Table1). Overall, EO and NE shared 17

Table 1. Chemical compositions of the essential oil from *Matricaria chamomilla* as EO and after preparation as nanoformulation (NE)

	Compounds	Percentage (%)	
		EO	NE
1	**Artemisia ketone**	0.17	0.34
2	Berkheyaradulene	0.11	
3	Cyclohexane,1–ethenyl–1–methyl–2,4–bis (1–methylethenyl)	0.13	
4	Isocaryophillene	0.13	
5	Aromandendrene	0.12	
6	**cis–á–Farnesene**	22.34	10.21
7	cis–Z–à–Bisabolene epoxide		0.16
8	α–Bisabolol	0.18	
9	γ–Muurolene	0.22	
10	**α–Cubebene**	2.00	0.83
11	**Eudesma–4 (14),11–diene**	0.47	0.17
12	**γ-Elemene**	1.00	0.54
13	α–Muurolene	0.12	
14	**α–Farnesene**	0.45	0.46
15	**Cadina–3,9–diene**	0.96	0.23
16	Naphthalene,1,2–dihydro–2,5,8–trimethyl		0.41
17	γ–Muurolene	0.13	
18	Caryophyllene oxide		0.22
19	1–Heptatriacotanol	0.11	
20	trans–Geranylgeraniol		0.13
21	**Spathulenol**	0.70	0.76
22	**(–)–Globulol**	0.16	0.28
23	Bicyclo[3.1.1]hept–3–ene,2–formylmethyl–4,6,6–trimethyl		0.30
24	Bicyclo[3,3,1]non–2–ene,7–oxa–2,8,9–trimethyl–5–acetoxymethyl	0.19	
25	(–)–Spathulenol		0.14
26	Limonen–6–ol, pivalate		0.60
27	α–Bisabolol oxide B	0.36	
28	**tau.–Cadinol**	1.03	1.07
29	**α–Bisabolol oxide B**	9.30	10.34
30	**Ledene oxide–(II)**	0.59	0.54
31	α–Santalol		0.27
32	**Cyclohexane,1–methyl–2,4–bis (1–methylethenyl)**	7.41	7.95
33	**α–Bisabolol**	1.11	1.52
34	cis–Z–à–Bisabolene epoxide		0.15
35	**Benzene, (4,5,5–trimethyl–1,3–cyclopentadien–1–yl)**	1.27	1.92
36	**Bisabolol oxide A**	48.61	56.65
37	**2–Pentadecanone, 6,10,14–trimethyl**	0.14	0.17
38	**2–Vinyl–6–methoxy–8–aminoquinoline**	0.39	3.17
39	2–Vinyl–6–methoxy–8–aminoquinoline		0.24
40	**Oxacycloheptadec–8–en–2–one, (8Z)**	0.12	0.13
41	Tetradecanamide		0.13

compounds detected (Table 1), and the compatibility ratio of the components was 41.48% between EO and NE. The most abundant component was bisabolol oxide A (48.61% vs 56.65%), followed by cis–á–farnesene (22.34% vs 10.21%), and à–bisabolol oxide B (9.30% vs 10.34%) of the EO and NE, respectively. Eleven components (*e.g.*, α–bisabolol oxide B, γ–muurolene, aromandendrene, etc.) were present only in the EO and they made up 1.8% of the total composition correspondingly. Meanwhile, 11 components were detected in the NE formula (*e.g.*, limonen–6–ol, pivalate, etc.) that formed 2.75% of the NE composition, which were not found in the EO formula.

In the present study, bisabolol oxide A and cis–á–farnesene were the major compounds present in the EO and NE. The concentrations of bisabolol oxide A and cis–á–farnesene reported in the previous literatures vary between (13 to 56%) and (4 to 21%), respectively, even in chamomile (Tolouee *et al.*, 2010; del Carmen Romero *et al.*, 2012; Raal *et al.*, 2012; Formisano *et al.*, 2015; Göger *et al.*, 2018; Piri *et al.*, 2019). The wide variation in the amount of bisaboloids (bisabolol oxide and bisabolol oxide B) could be attributed to growth conditions of the plant (Rafieiolhossaini *et al.*, 2010) and/or the number of sunny hours (Gosztola *et al.*, 2010). Also, the amount of bisapolol has been shown to increase with temperature (Seidler–Lozykowska, 2010). Late planting dates and soil characteristics (Cd, Cu, nitrogen, and amino acids) seem to affect the amount of cis–á–farnesene (Rafieiolhossaini *et al.*, 2010). Thus, the variations in chemical components of chamomile oils may be related to biotic and abiotic factors such as climate, geographic origin, stage of maturity, fertilization and cropping systems (Roby *et al.*, 2013; Luz *et al.*, 2014; Santos *et al.*, 2016), which can influence the plant growth and, consequently, the plant physiology.

Beside, the present study showed that the EO of *M. chamomilla* differed in its chemical compositions between pre– and post–formulations (Table 1). This difference should be related to reactions of the compounds during the manufacturing process of NE. An increase in the bisaboloid concentration and the appearance of new components in NE may be due to the high temperature condition during preparation. This indicates that the NE preparation process can potentiate the concentration of some components whereas it may cause a decrease or exclusion of the others. Thus, the consequences of such preparation process on bio–pesticide activity should be focused in future studies.

Physical characterization

The microscopic examination revealed that diameters of spherical particles that had been formed differed markedly among the EO, NE and NC (Fig. 1). Several studies have examined the optimal conditions for the formation of stable emulsions, and various concentrations of *M. chamomilla* have been formulated. For example, Binks *et al.* (2009) have studied the effect of temperature on the stability of emulsions, showing that an increase in temperature decreased the emulsion sta-

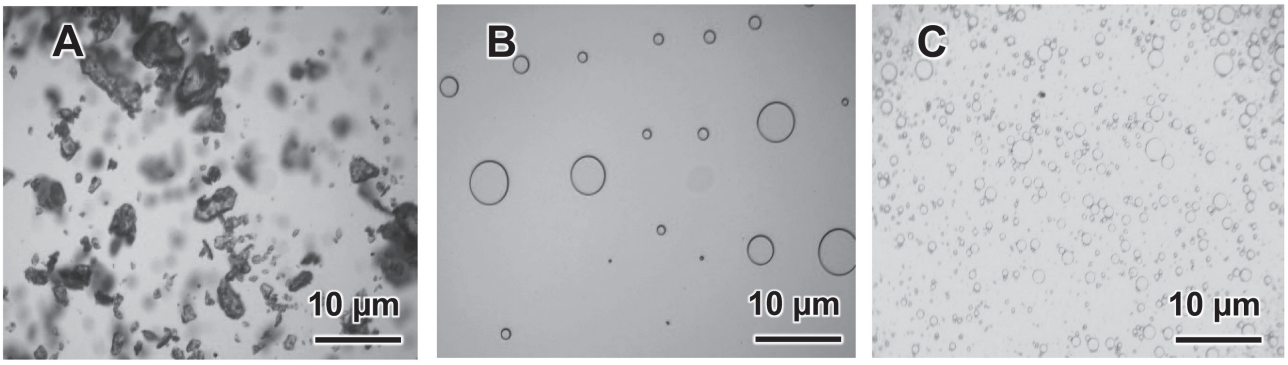

Fig. 1. Light microscope images of: (A) Nanoencapsulation NC; (B) Essential oils EO; and (C) Nanoemulsion NE.

Table 2. Characterization of the essential oil and their nanoformulations

Sample	Particle Size (nm)	Zeta Potential (mV)	Polydispersity (PDI)	Viscosity	Conductivity
Essential oils (EO)	3143	– 10.2	0.308	0.8872	0.033
Nanoemulsion (NE)	221.4	– 15.4	0.722	0.8872	0.019
Nanoencapsulation (NC)	403.4	– 14.3	0.412	0.8872	0.035

bility. Nanoemulsions containing pulegone at a starting concentration of 10% were found to have the highest release capability for prolonged periods at all temperatures (Binks *et al.*, 2009). In some previous studies, nanoemulsions are shown to have superior results in terms of the quantity of the released pulegone and allowed significantly prolonged release of this agent as compared with the coarse emulsions (Golden *et al.*, 2018). Nanoemulsions have a higher surface area in comparison to coarse emulsions (McClements and Rao, 2011) and can release a higher amount of pulegone. In addition, nanoemulsions are more stable than coarse emulsions (Arnon–Rips and Poverenov, 2016).

We found that NE and NC treatments in the present study greatly reduced the particle size, demonstrating that nanoformulations of the essential oils were successfully achieved (Table 2). Based on previous studies (Nakajima, 1997; Sonneville Aubrun *et al.*, 2004; Lett 2016), we regarded that the emulsion with average droplet diameter (Z) lower than 300 nm as a nanoemulsion. In our study, the *M. chamomilla* essential oil showed Z–average size of 3143 nm and conductivity of 0.033 mS/cm, and the zeta potential was highly negative (–10.2) (Table 2). On the other hand, the poly–dispersity index (PDI) value was slightly higher (0.308) than the values previously reported in Hashem *et al.* (2018), with low viscosity of 0.8872 cP, which might be due to the low oil content in our study. The decrease of PDI value by increasing the aniseed oil concentration can be due to the higher effective concentration of the thickener in the aqueous phase. Indeed, it has been reported that PDI values lower than 0.25 indicate a narrow particle size distribution, proving good physical stability of the nanoemulsion, due to the reduced Ostwald ripening (Hoeller *et al.*, 2009). Thus, with our experimental procedure, some measurements suggested improvements in nanoformulations whereas the others were not.

On the other hand, it is known that an increase in viscosity of the continuous phase reduces oil droplet mobility, which delays instability phenomena, resulting in oil droplets with a more homogeneous particle size (Arancibia *et al.*, 2016). However, we did not detect any change in the viscosity values after nanoformulations (Table 2). Also it is demonstrated that zeta potential values greater than + 25 mV or lower than – 25 mV typically point out high degrees of stability (Shi *et al.*, 2017). In the present study, again, we did not find enough improvements on zeta potential after nanoformulations though the values became lower (Table 2). Z–average sizes below 20 nm indicate a high mobility in the solution, due to the applied field and Brownian motion (Berne and Pecora, 2000). Moreover, conductivity of 5 mS/cm or more indicates the presence of highly conductive ions in nanoemulsion, which can lead to electrode polarization and degradation (Patakangas, 2014). Therefore, the conductivity was not improved in our study.

The infrared spectra measurements of EO, PEG 6000 and fragrant nanoparticle by the FT–IR analysis are shown in Fig. 2. In case of pure PEG, the absorption peak at 3447 cm^{-1} corresponded to the O–H group. The peaks at 2886 cm^{-1}, 1470 cm^{-1}, 1346 cm^{-1} and 842 cm^{-1} belonged to the C–H stretching vibration while 1109 cm^{-1} peak was due to C–O asymmetric stretching vibration. Furthermore, the obtained band at 1652 cm^{-1} represented the C = O stretching vibration. In case the *M. chamomilla* oil, the strong wide band at 3447 cm^{-1} indicated the stretching vibration of O–H group, typical not only for carbohydrates. Aliphatic fragments may be observed with maximum 2966 cm^{-1} and 2928 cm^{-1} which attribute to narrow bands of C–H stretching vibrations of aromatic rings, and polyphenolic compounds were

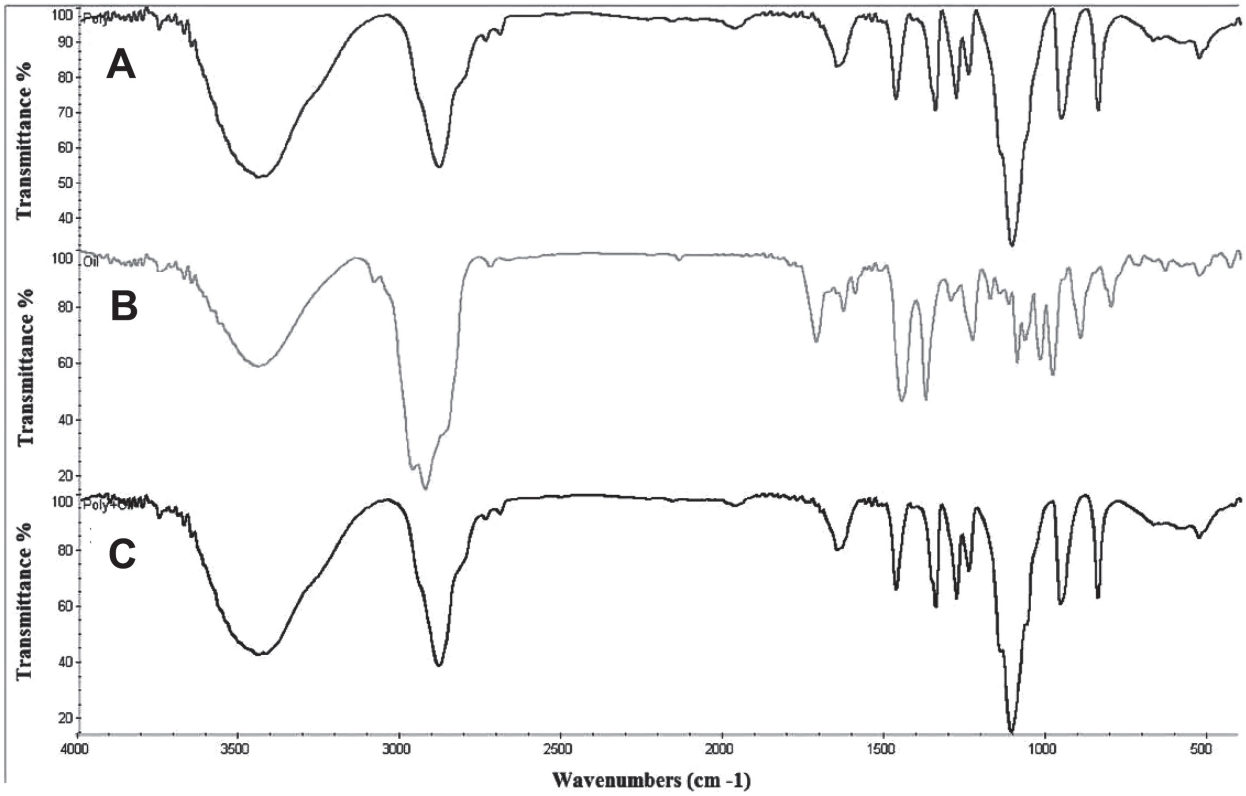

Fig. 2. Fourier–transform infrared (FTIR) spectra for: (A) PEG; (B) EO; (C) PEG + EO compositions.

related to C = O and C = C bonds at 1716 cm^{-1} and 1632 cm^{-1}, respectively. The spectral shape of the dominant group of intensive bands at 1452 cm^{-1} and 1376 cm^{-1} regions (Fig. 2A), typical for responses of symmetric stretching vibrations from non–esterified carboxylic groups (COO–) of hexuronic acids like glucuronic acid or galacturonic one. In addition the lack of strong signals at characteristic ranges 1231 and 1177 cm^{-1} in tandem with 1119–1093 cm^{-1} indicates the absence of sulfated esters in the *M. chamomilla* oil preparation. Aforementioned evidence indicated the EO and PEG are closely matching at the majority of characteristic peaks confined between 3752 – 2696 cm^{-1} ranged, 1452 – 1109 cm^{-1} ranged and 983– 528 cm^{-1} ranged, thus the incorporation of both compounds is good. In case of PEG + EO composition, the resulting spectra of the PEG encapsulated nanoparticles illustrated no significant interaction between the oils as active ingredient and PEG is a nanoparticle shell material. In addition to , infrared spectroscopy was done to confirm the formation of PEG nanoparticles. The spectrum confirmed PEG 6000 polymer is a good nanoparticle shell material.

Bioassays

The mortalities of adult cockroaches largely depended on the type of formulas, *i.e.*, EO, NE, and NC, and also on the form of treatments, *i.e.*, fumigant, topical, spray, dust or feeding treatments. We summarized in Table 3 the relationships between the type or form of the treatments and the effectiveness measured on the basis of cockroach mortality, in which we described the results as E (= effective in killing cockroaches), NE (not effective because mortality rate was less than 5%) and NA (not applied because the material formula did not correspond to the method of use). The three types of application, that is, topical, spray and feeding treatments, mostly did not result in knockdown of test cockroaches; the percentages of mortality were less than 5%. Therefore, we regarded these three types of treatments with EO and NE as not effective or practical (Table 3). Feeding application with NC was also not effective. However, knockdown of the adult cockroaches due to fumigant toxicity was frequently observed for EO or NC (Table 3). Dusting toxicity also was found effective when NC formula was applied though knockdown of test cockroaches seldom occurred when this formula was used as feeding treatments (Table 3).

Cockroach mortality differed markedly among the three formulas (EO, NE, and NC). The results for fumigant toxicity of EO and NE are shown in Fig. 3. EO showed the highest fumigant toxicity in comparison to the NE at all tested concentrations and all exposure periods (Fig. 3). For both control samples (distilled water for NE and acetone for EO), percentages of mortality were extremely low (mortality < 5%), except when the acetone concentration was 10 ml/l. However, at higher concentration (10 mg/liter) mortality of adult cockroaches notable increased, reaching 100% after 1 h. until 72 h. for the EO, and reaching 44.44% after 72 h. for the NE (Fig. 3d).

For NC treatment as dust formulation (Fig. 4), the result showed that the treatment carried out with the

NC from 1 h to 72 h, and the knockdown or mortality of tested cockroaches increased with time while it remained 0 in the negative control (PEG only) throughout the experimental periods. The NC powder caused 100% of adult mortality at 1.5 mg/500 ml after 24 h or after 48 h at 1.00 and 0.5 mg/500 ml (Fig. 4).

Among the members of the genus *Matricaria* grown all over the world, *M. chamomilla* has been of particular interest because of a wide range of useful biological activities (Berry, 1995). The main constituents of *M. chamomilla* are α–bisabolol oxide B, α–bisabolol and bisabolol oxide A, and these have been used as anti–

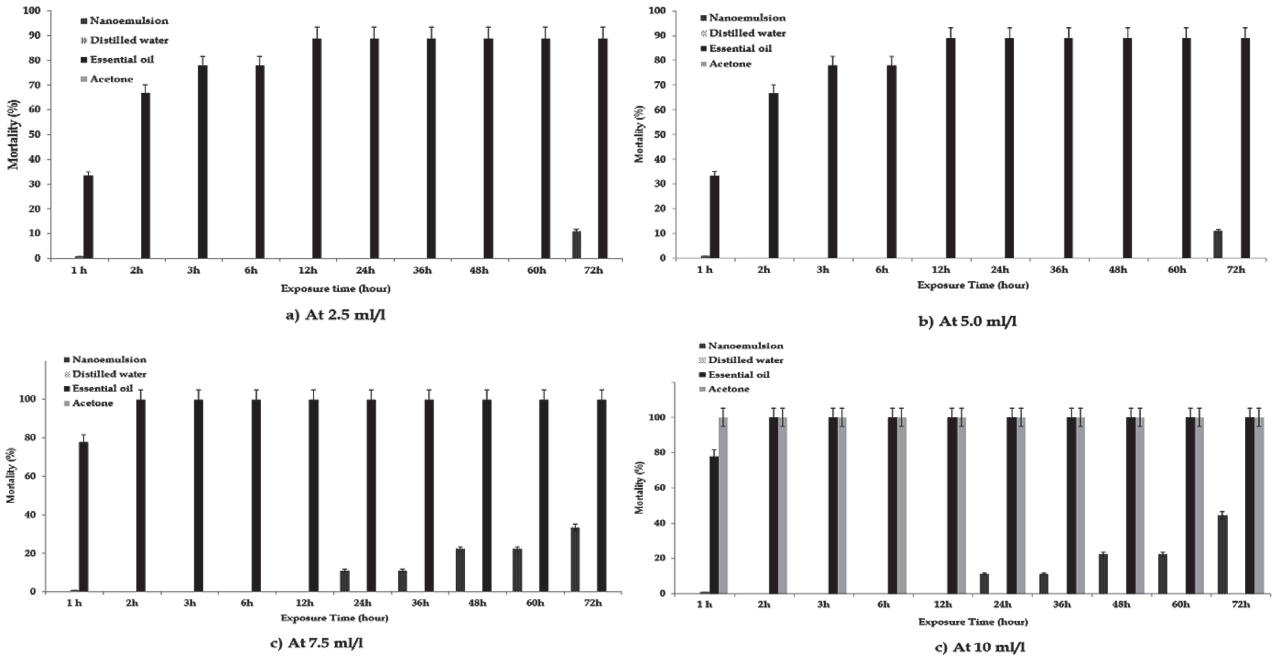

Fig. 3. Percentages of mortality (mean ± SE) of *Periplaneta americana* adults when exposed to nanoemulsions (with distilled water as a negative control) and essential oil (with acetone as a negative control) for 1, 2, 3, 6, 12, 24, 48 and 72 h at different concentrations (a. 2.5 mg/liter; b. 5.0 mg/liter; c. 7.5 mg/liter and d. 10 mg/liter).

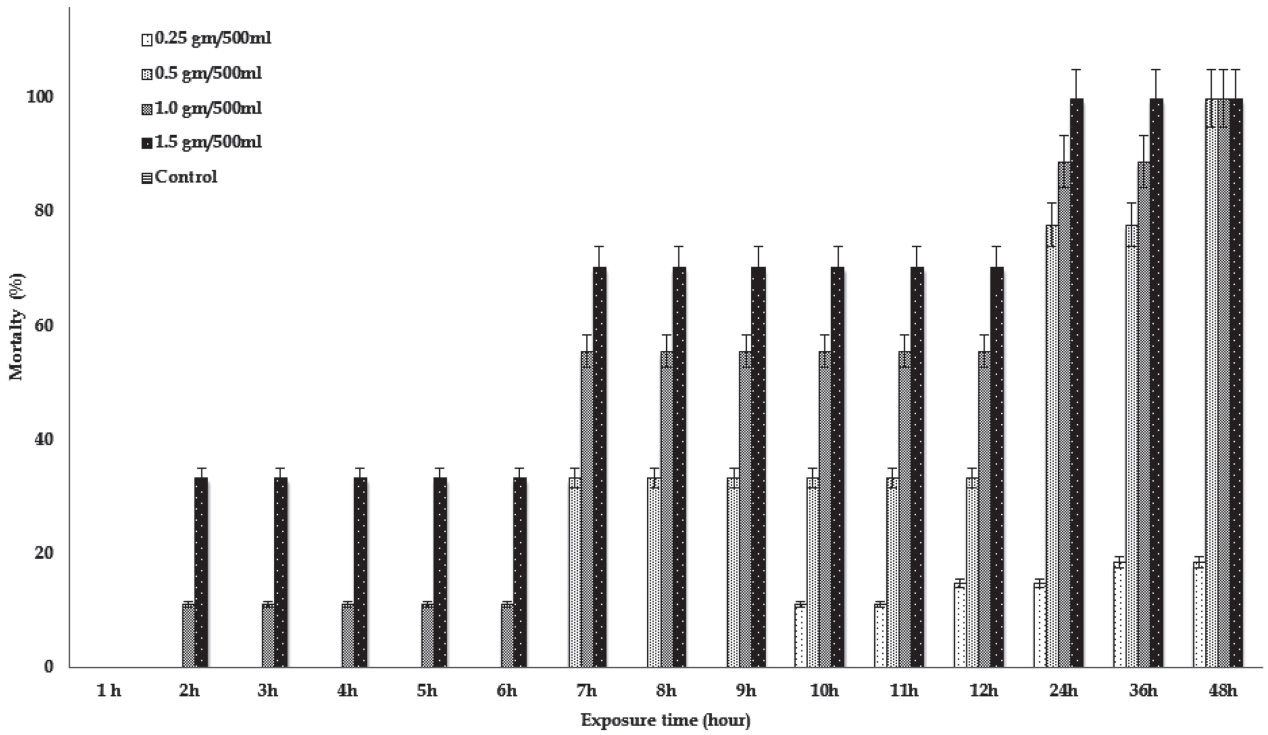

Fig. 4. Efficacy of nanocapsulation dusts (NC) on *Periplaneta americana* adults after 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 24, 36 and 48 h at different concentrations (0.25; 0.5; 1.0 and 1.5 gm/500 ml glass) and PEG only as a negative control.

inflammatory and antispasmodic (Szöke *et al*., 2004). To the best of our knowledge for the first time, no report on the effect of *M. chamomilla* oil against cockroaches was reported. Some researchers reported antifungal activity of the plant oil against different phytopathogenic and medically important fungi (Soliman and Badeaa, 2002; Magro *et al*., 2006), while the others showed weak or no growth inhibition in this regard (Rauha *et al*., 2000; Lee *et al*., 2007; Bluma *et al*., 2008). Philips *et al*. (2010) and Phillips and Appel (2010) have already reported that the fumigant and contact toxicities of 12 essential oil components against German cockroaches: carvacrol, 1,8–cineole, trans–cinnamaldehyde, citornellic acid, eugenol, geraniol, limonene, linalool, menthone, α–pinene, β–pinene, and thymol. They reported also that 1,8–cineole was the most toxic to male and female adult German cockroaches in a fumigant test. The fumigant toxicity of carvacrol and thymol was less than that of α–pinene, β– pinene, and limonene. However, thymol and carvacrol showed strong contact toxicity against adult male and female German cockroaches.

Finally, it can be concluded that *M. chamomill*a flower in EO, NE and NC formula is a potent inhibitor of *P. americana* growth (I do not know why you can say so because there are no data about cockroach growth or development). With the proven safety and high stability together with the present data, essential oils of *M. chamomilla* and their nano–forms may be useful as control agents against American cockroaches. As novel cockroach–control agents, chamomile essential oils and their nano–forms can provide us with various treatment methods to control cockroaches. The future studies will focus on their safety to humans, animals and nontarget organisms as well as their mode of action.

AUTHORS' CONTRIBUTIONS

A. S. El–Khodary, N. F. Ghanem designed the study and revised the final manuscript. O. M. Rakha helped during the experiment. N. W. Shoghy prepared the nanoformulations and analyzed the data, and wrote the first draft of the manuscript. T. Ueno discussed the results, polished up the research concept and the manuscript.

CONFLICT OF INTEREST

The authors declare no conflicts of interest.

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