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Silica-Microencapsulated Orange Oil for Sustainable Pest Control

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An ultralow amount of sub-micron spherical SiO_2 particles encapsulating 7 wt% crude orange oil (SiliOrange) suspended in water shows surprisingly high insecticidal activity against the cotton leafworm *Spodoptera littoralis*, and significantly reduces the progeny of cotton aphid *Aphis gossypii* under laboratory testing conditions. Considering the ease of reproducible preparation of the material and the biocompatible nature of both silica and orange essential oil, these results may open the route to sustainable pest control using new biopesticide water-based formulations based on sol–gel microencapsulated orange oil.

1. Introduction

Mechanically extracted from the orange peel cells via a water jet stream before fruit squeezing for juice production, orange oil is a valued essential oil (EO) mostly composed of *d*-limonene.[1] Edible and with an exquisite scent, orange oil finds its major applications in the food, beverage, cosmetic, perfumery and oil industries.[1]

Its current value is so high (exceeding \$10 kg^{-1}) that today's orange juice makers earn a large and increasing share of their revenues from its sale.[2] The 15 year (2003–2017) average global supply of orange oil has been around 57 000 tons, with a 9000 tons shortage in 2016/2017 due to constant decline of Florida's production hit by citrus greening disease. The price of orange oil has gone from then record high \$9.45 kg^{-1} in the summer of 2017 for bulk samples,[2] to over \$11.50 kg^{-1} in the summer of 2019.[3]

The amount of aldehydes contained in orange oil, both terpenes (neral and geranial) and aliphatic aldehydes (decanal and octanal), crucially affects the olfactory notes of orange oil. The concentration of carbonyl compounds, which substantially influences the orange oil value and price, exceeds 1% in EOs from organically grown oranges, whereas is less than 0.4% in oils containing pesticides.[4]

Citrus essential oils are a promising tool for the sustainable control of economically relevant agricultural pests, but their field application is limited by several constraints such as the high instability and degradability patterns. However, the encapsulation of essential oils into nanostructures can overcome such limitations, ensuring the preservation of the insecticidal properties.[5,6]

Among the many and varied microencapsulation technologies, the encapsulation of organic liquids in porous SiO_2 microspheres stands out for multiple benefits, including the vastly enhanced chemical and thermal stability of the microencapsulated organic species, high mechanical strength, ease of handling (since silica does not carry a static charge and is always free flowing), and dispersion of the microspheres in different liquid media.[7] Silica, furthermore, is a biocompatible and environmentally benign (biodegradable) material ideally suited for biological, medical and pharmaceutical applications.[8]

Co-formulated with other substances such as urea and borax, orange oil is highly effective in killing or repelling several noxious insects, fungi, bacteria and even viruses.[9] At least two broad-spectrum biopesticides based on orange oil have been recently commercialized.[9] Orange oil terpenes, however, are easily oxidized when exposed to high temperature, oxygen and humidity, namely, the typical open field conditions.

From complex coacervation with biopolymers[10] through spray-drying with gelatin or lignin[11] several techniques have been described to microencapsulate orange oil. To the best of our knowledge, encapsulation in spherical SiO_2 microcapsules has not been reported. In this study we describe the outcomes of using sol–gel microencapsuled crude orange oil, herein dubbed SiliOrange, formulated in water as insecticide against the cotton aphid *Aphis gossypii* Glover (Hemiptera: Aphididae) and the cotton leafworm *Spodoptera littoralis* (Boisduval)

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(Lepidoptera: Noctuidae) under laboratory conditions. Both insect pests are extremely polyphagous and can cause serious economic losses to a multitude of agricultural and ornamental crops.[12,13]

2. Experimental Section

2.1. Chemicals

Tetraethylorthosilicate (TEOS, >99%), cetyltrimethylammonium bromide (CTAB, >99%), glycerol (>99.5%), methyltrimethoxysilane (MTES, >99%), and ammonium hydroxide solution (puriss. p.a., 25 wt% NH_3 in H_2O) were purchased from Sigma-Aldrich. All chemicals were of high purity and were used without any further purification. Crude orange oil was kindly donated by Agrumi-Gel (Barcellona Pozzo di Gotto, Italy).

2.2. Material Synthesis

Glycerol (25 mL), CTAB (360 mg), and ammonium hydroxide (2.5 mL) were added to deionized water (250 mL) in a glass balloon kept under stirring at room temperature for 10 min after which the solution was added with crude orange oil (6 mL). The mixture kept in an ice-bath was sonicated by an ultrasonic processor (Bandeline, Sonoplus HD 4100) for 10 min at 25% amplitude. The resulting microemulsion kept at 40 °C under mechanic stirring (IKA Eurostar digital Werke) at 400 rpm was added dropwise with TEOS (16 mL), and left under stirring for 48 h. The white precipitate was filtered, washed extensively with deionized water and mildly dried in an oven (30 °C) overnight.

A blank sample was prepared with the same procedure substituting the orange oil with an equivalent volume of *n*-hexane. The synthesis of the methyl-modified silica microcapsules started from a similar solution of deionized water (250 mL), CTAB (3 g) and NH_4OH 25% (2.5 mL) kept under stirring at room temperature for 10 min and thus added with a solution of orange oil (6 mL) in cyclohexane (20 mL). The resulting emulsion was put in an ice-bath and sonicated by ultrasonic processor (Bandeline, Sonoplus HD 4100) for 10 min at 25% amplitude. The microemulsion was then transferred to a flask and kept under mechanical stirring at 400 rpm at 40 °C while a solution of TEOS (38 mL) and MTES (1.8 mL) was added dropwise. The white precipitate readily obtained was filtered, washed extensively with deionized water and dried in an oven (30 °C) overnight.

2.3. Characterization

The SiliaOrange essential oil loading was assessed by GC-FID analysis referring to the amount of limonene. A small amount (100 mg) of SiliOrange was added to a test tube along with 2 mL of 1 M NaOH aqueous solution in order to ensure complete dissolution of the silica shell. After stirring the mixture for 24 h, the non-polar fraction was extracted with 2 mL dichloromethane (DCM) and filtered through a syringe filter (Whatman PTFE, pore size 0,2 μm). The amount of limonene in the organic phase was measured via GC-FID analysis using undecane as internal standard. The analyses were carried out on a Shimadzu GC-FID 17A equipped with a Supelco SPB-1701 capillary column. TGA/DSC analysis was performed under nitrogen on a Mettler Toledo TGA/DSC1.

The orange oil maximum additive concentration (MAC) in CTAB was determined measuring the absorbance at 630 nm of the CTAB solution upon the addition of orange oil (0.025% to 1%) using a UV–VIS spectrophotometer (Shimadzu UV-1800) at 25 °C. The wavelength of 630 nm was chosen to avoid interference with the absorbance of orange oil.

Dynamic light scattering (DLS) and Zeta potential measurements were performed using a Malvern Zetasizer Nano ZS equipped with a He–Ne laser at a power $P = 4.0$ mW operating at 633 nm wavelength. The average diameter and size distribution (polydispersity index, PDI) of SiliOrange aqueous suspensions were determined by Photon Correlation Spectroscopy (PCS). Each solid sample was dispersed in filtered (0.2 μm) bi-distilled water. The instrument setting conditions were equal to those described above for the particle size measurements. Each sample was analyzed in triplicate.

The scanning electron microscope (SEM) pictures were obtained using a Zeiss EVO MA10 microscope equipped with an SE-Everhart–Thornley secondary electron detector. A lanthanum hexaboride cathode was used as the source of electrons. The accelerating voltage was 20 keV and probe current 10 pA. All imagines were acquired in ultra vacuum condition (10^{-7} mbar). To increase the conductivity of the sample, each material sample was deposited with an ultrathin layer (2 nm) of gold nanoparticles using an AGAR Sputter Coater for gold.

2.4. Toxicological Bioassays

The biopesticide formulation used throughout this work was comprised of silica (SiO_2) SiliOrange microcapsules (0.5 g) dispersed in distilled water (150 mL).

Larvae of *S. littoralis* used for the bioassays were collected from the laboratory insect colony maintained in a climatic chamber (Refrigerated incubator model IRE-475, Raypa R. Espinar, S.L., Spain) at 24 ± 1 °C, 50 ± 10% R.H. and L12:D12 h photoperiod. *S. littoralis* larvae were reared on artificial diet according to a published method[14] inside plastic boxes (10 × 15 × 5 cm) that were covered with fine mesh to provide ventilation.

The rearing of *A. gossypii* was initiated from aphid infested organic greenhouse solanaceous crops inside ventilated propylene boxes (56 × 39 × 42 cm) offering zucchini plants (*Cucurbita pepo* L., cv "Bianca di Trieste") as hosts. Host plants were replaced twice a week and kept at the aforementioned controlled laboratory condition. For the insect rearing and the bioassays, zucchini plants were used at the phenological stage of 3rd true leaf grown in 2 L pots in greenhouse conditions. For the experiment, newly molted adults (24 ± 12 h old) were obtained by isolating 3rd instar nymphs on clean zucchini pots one week before the bioassay started. Nymphs were maintained at the same environmental laboratory conditions. The biological experiments were carried out at the Department of Agriculture, Food and Environment of the University of Catania (Italy).

2.5. Lethal and Sublethal Toxicity on *Spodoptera littoralis*

The contact toxicity and the consequent sublethal effect of the aqueous suspension of SiliOrange on *S. littoralis* larvae were evaluated by spraying same age larvae. For each replicate five to ten 2nd instar larvae of *S. littoralis* were topically sprayed with SiliOrange on an absorbent paper sheet and were then transferred to a plastic ventilated arena (4 × 5 × 2 cm) containing the artificial diet, provided *ad libitum*. Every 48 h the following biological parameters of insects were measured: i) larval survival (%), ii) development time (i.e., time elapsed, expressed in hours, from egg hatching to pupa formation), and iii) pupal weight using a 10 mg sensitive Mettler–Toledo microbalance.

An untreated control (distilled water), the blank of SiliOrange, and a treated control with the pyrethroid lambda-cyhalothrin (Karate Zeon, Syngenta Italia, a.i. 9.48%) at the highest label dose recommended for the control of noctuid moths in tomato protected crops (i.e., 0.125 L ha^{-1}), were used as controls. The boxes containing sprayed *S. littoralis* larvae were maintained in a climatic cabinet at the controlled climatic conditions reported above. Larval frass was removed every two days from the experimental arena, were cleaned up by and the factitious food was also renewed. Six experimental replicates were performed with the microencapsulated formulations and the controls.

Figure 1. Zucchini plant sample hosting *Aphis gossypii* insects prior to spraying the SiliOrange suspension that is in the spraying device on the right side.

2.6. Lethal and Sublethal Effect on *Aphis gossypii*

The contact toxicity of SiliOrange was evaluated on the survival and the progeny of *A. gossypii* by spraying young, same age adult aphids feeding on a zucchini plant. For this bioassay, twenty same age young adults feeding on a fresh 2-week old zucchini plant (that represented a single replicate) were sprayed from a distance of 0.3 m with a 0.1 L hand sprayer until runoff. A total amount of 1 g SiliOrange suspension was sprayed. Such methodology was chosen because it mimics a potential field scenario in which the chemical control of aphid pests is routinely pursued through insecticide foliar spraying devices (**Figure 1**).[15]

Sprayed aphid infested plants were left to dry under laboratory conditions and each plant was fixed with modelling-clay in a two-superposed transparent plastic covered with a net[16] and kept inside a climatic cabinet at the previously described condition.

The aphid survival was assessed under a stereomicroscope 24 h after the beginning of the treatment. The aphid progeny (i.e., number of produced nymphs by females) was counted daily for ten consequent days. To compare the toxicity with a control and an established insecticide, an untreated control was used (i.e., distilled water), the blank of SiliOrange and the neonicotinoid Afidane 200 SL (Chimiberg, Italy, a.i. imidacloprid 17.7%) at the highest recommended concentration for controlling aphid pests in greenhouse crops (i.e., 0.75 L ha^{-1}). This bioassay was replicated five times per treatment group.

2.7. Data Analyses

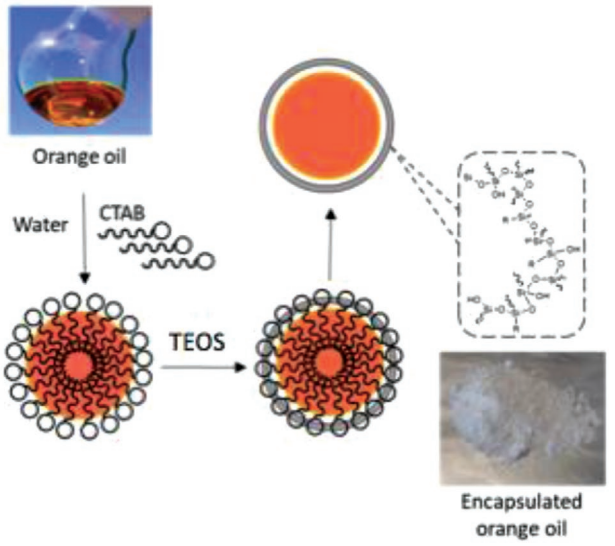
The homogeneity and normality of the dependent variables (i.e., measured biological parameters) were checked through Levene and Shapiro-Wilk tests and the dataset was log-transformed whenever needed. The effect of the independent variables (i.e., chemicals) on the biological parameters (i.e., larval survival, the developmental time and the pupal weight for *S. littoralis* and the adult survival and the progeny for *A. gossypii*) were tested by carrying out a single factorial ANOVA. Least Significant Difference (LSD) post hoc test was used for multiple means comparisons among the chemical treatments at $p \leq 0.05$. This statistical analysis was performed in RStudio (Version 4.0.0 – 2020).[17]

3. Results and Discussion

The template-driven sol–gel microencapsulation of orange oil displayed in Scheme 1 was successful, affording SiO_2 spherical microcapsules with a 7 wt% orange oil load, consistently assessed by means of GC. Though spherical and homogeneous, the methylated microcapsules had a modest 0.5 wt% content of essential oil (EO). This shows that the presence of the methyl-modified silane during the sol–gel polycondensation favored the exit of the hydrophobic molecular components of the EO from the micellar structures, which is required for the entrapment of an hydrophobic substance in water (Scheme 1).

We briefly remind that in the two-step sol–gel microencapsulation process of organic molecules in silica-based microcapsules from oil in water (O/W) emulsions, pioneered by Avnir and co-workers,[18] the emulsion droplets act as a microreactor environment for the hydrolysis and condensation reactions of Si alkoxides.

In the present case of SiliOrange synthesis, glycerol added to the precursor sol–gel mixture decreases the dielectric constant of the continuous phase thereby reducing self-aggregation



Scheme 1. Synthesis of SiliOrange microcapsules: i) sonication of the essential oil in the presence of aqueous CTAB forms a stable microemulsion, ii) addition of TEOS forms the silica shell encapsulating the nanoemulsion core, iii) precipitation of the microcapsules followed by filtration and washing.

of the cationic surfactant CTAB molecules via enhanced electrostatic interactions in an elegant way in which glycerol and CTAB molecules do not directly interact.[19]

We thus measured the maximum additive concentration (MAC), namely the highest concentration of a lipophilic compound that can be incorporated into a micellar surfactant solution at a given surfactant concentration, of orange oil in CTAB to maximize the efficiency of sol–gel microencapsulation by assessing the amount of orange oil that was effectively solubilized by the surfactant (CTAB).

Figure 2 shows that CTAB micelle saturation occurs between 0.10% and 0.35% orange oil concentration, which leads to the rapid increase in the absorbance at 630 nm due to formation of un-dissolved orange oil droplets remaining dispersed in the aqueous phase. At this concentration micelles were no longer able to accumulate the oil in the core. This absorbance increase

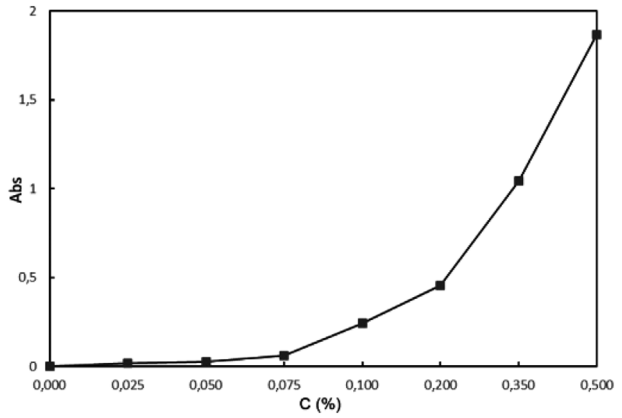


Figure 2. Absorbance of CTAB micelles at increasing concentrations of orange oil.

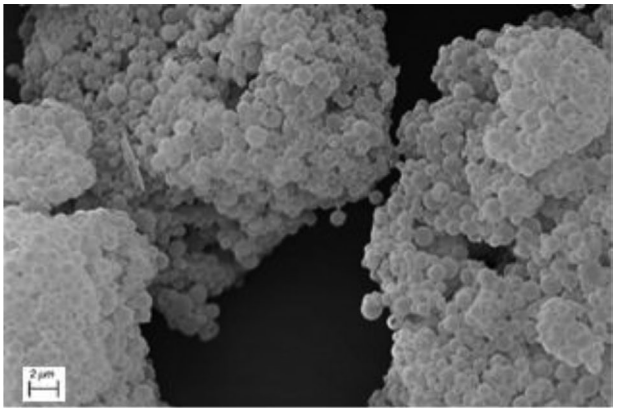


Figure 3. SEM photograph of SiliOrange SiO_2 microcapsules having 7 wt% orange oil load.

allowed us to determine 0.05 wt% as the ideal concentration of orange oil that ensures full microencapsulation of the EO volume (6 mL) added to the sol–gel precursor mixture (see the Experimental Section).

The SEM photograph (**Figure 3**) clearly shows that SiliOrange is comprised of sub-micron spherical SiO_2 capsules of narrowly distributed size.

The particle size distribution from dynamic light scattering (**Figure 4**, top), derived from a deconvolution of the measured intensity autocorrelation function of the sample using a non-negatively constrained least squares fitting algorithm, pointed to a predominant population constituted of rather monodispersed microparticles of 0.153 µm diameter with a polydispersity index (PDI) of 0.249.

As an indirect measure of the net charge of ceramic particles in suspension, the zeta potential of silica microparticles measured in a dispersant indicates the stability of a suspension. Hence, the ζ (zeta) potential distribution (Figure 4, bottom) measured at the pH (6.3) determined by the simple addition of the microparticles to deionized water using the principles of

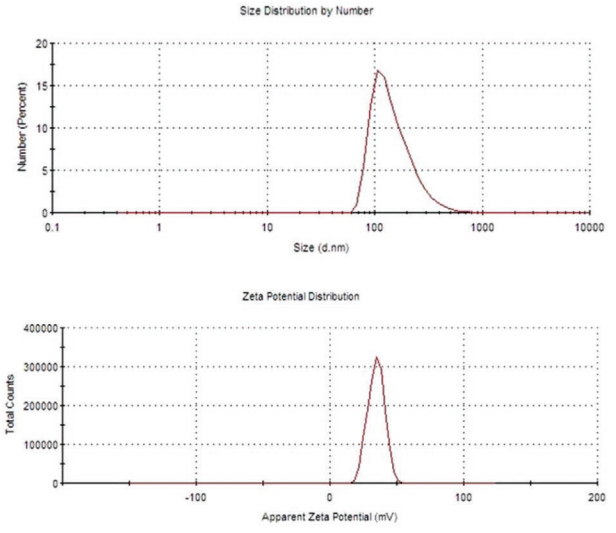


Figure 4. Particle size distribution for SiliOrange SiO_2 microcapsules doped with 7 wt% orange oil (top); ζ potential distribution (bottom).

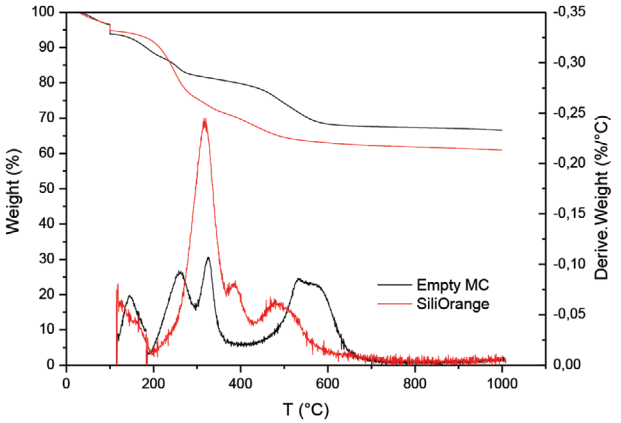


Figure 5. Top: TGA curves of empty (black curve) and SiliOrange microcapsules (red curve). Bottom: derivative thermogravimetry (DTG) plot.

laser Doppler velocitometry and phase analysis light scattering (M3-PALS technique) indicated good stability of aqueous suspensions of SiliOrange microspheres (34.5 mV).

The larger the absolute zeta potential value, the more stable the colloidal dispersion. A rule of thumb based on the Derjaguin, Landau, Verwey, and Overbeek theory suggests that values of ζ equal to or larger than ±30 mV are enough to promote stable water suspensions.[20] In light of practical applications, it is also relevant herein to notice that for amorphous silica, namely for glassy particles such as those comprising the SiliOrange material, the ζ potential rapidly decreases toward more negative values when increasing pH,[21,20] thus enabling the use of these microcapsules in aqueous systems of widely different pH.

The thermogravimetric analysis (TGA) describes the material's thermal stability and its fraction of volatile components. The plot shown in **Figure 5** indicates successful encapsulation of orange oil. In the blank sample, the first weight loss in the TGA was due to water evaporation (–6.18%) followed by a more significant weight loss peak at 300 °C likely due to water formed by condensation of silanol groups at the SiO_2 surface. Eventually, at 550 °C also the entrapped surfactant molecules were released and thermally decomposed.

The SiliOrange TGA profile is simpler. The significant weight loss (–32.15%) at around 300 °C following the same first weight loss due to water evaporation (–5.90%) was caused by concomitant orange oil and silanol groups condensation, similarly to what happened with core–shell silica microcapsules encapsulating glycerol (**Table 1**).[22]

A sample of SiliOrange (0.5 g) dispersed in water (150 mL) was tested as potential sustainable biopesticide against two

Table 1. Weight loss in SiliOrange and empty SiO_2 microcapsules during the thermogravimetric analysis.

T (°C)	SiliOrange weight loss (%)	Blank SiO_2 weight loss (%)
100	5.91%	6.19%
300	32.15%	13.25%
590	–	13.15%
	Total loss: 38.06%	Total loss: 32.59%

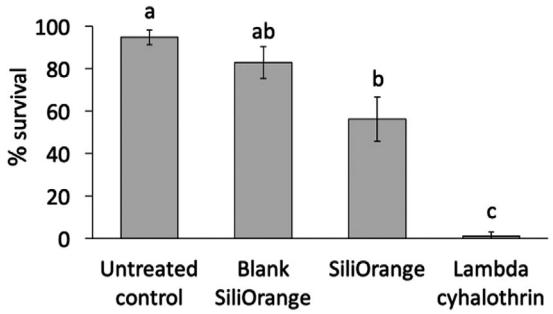


Figure 6. Mean survival (±SE) of *S. littoralis* larvae in the contact toxicity bioassay. Bars with different letters indicate significant differences at $p < 0.05$ (ANOVA, LSD test).

insect herbivores having different feeding strategies. We chose the cotton leafworm *S. littoralis* as chewing pest and the cotton aphid *A. gossypii* as sap-feeding pest. In our standardized experimental setup, we chose larvae of similar age to exclude any potential effect of age on the observed biological parameters (i.e., mortality), as well as for ensuring the replicability of the results.

In the contact toxicity bioassay, the SiliOrange aqueous formulation used in this work reduced the survival of larvae by ≈40% in comparison to the untreated control ($F_{3, 22} = 45{,}17$; $p = 0.0001$). Nevertheless, *S. littoralis* larvae sprayed with SiliOrange survived significantly longer than larvae treated with the pyretroid-based insecticide that caused ≈100% mortality (**Figure 6**).

Conversely, no statistical differences were found between the survival of larvae sprayed with distilled water (i.e., untreated control) and the blank of SiliOrange. Similarly, treatment with SiliOrange significantly decreased the development time of *S. littoralis* larvae by 1.6 times in comparison to the untreated control ($F_{3,22} = 45{,}59$; $p = 0.0001$) (**Figure 7**).

As a consequence, also the pupal weight of *S. littoralis* was negatively affected by SiliOrange ($F_{2, 17} = 6.86$; $p = 0.007$) (**Figure 8**, 192.11 ± 36.62 mg) compared to larvae sprayed with distilled water (578.07 ± 16.16 mg). Pupal weight was not determined in the treated control (i.e., lambda-cyhalothrin), because none of the sprayed larvae survived this bioassay.

The entomotoxicity of silica-based nanoparticles per se has been proven against different lepidopteran pests[23,24,25] including *S. littoralis*.[26,27] Similarly, the insecticidal activity of

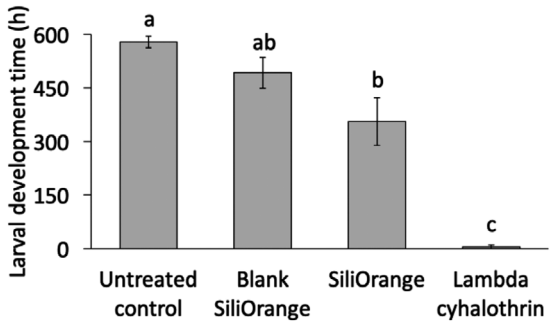


Figure 7. Mean development time (±SE) of *S. littoralis* larvae in the contact toxicity bioassay. Bars with different letters indicate significant differences at $p < 0.05$ (ANOVA, LSD test).

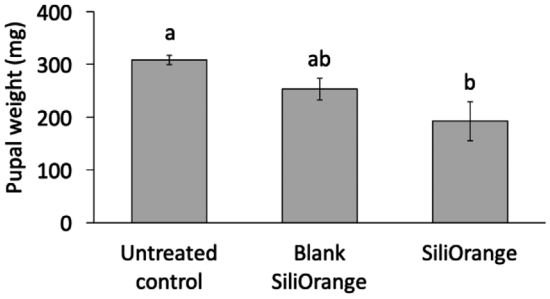


Figure 8. Mean weight (mg) (±SE) of *S. littoralis* pupae formed from larvae sprayed in the contact toxicity bioassay. Bars with different letters indicate significant differences at $p < 0.05$ (ANOVA, LSD test).

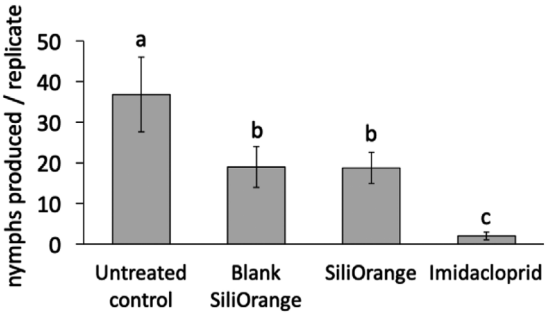


Figure 10. Mean number (±SE) of *Aphis gossypii* progeny produced by sprayed female adults in the contact toxicity bioassay. Bars with different letters indicate significant differences at $p < 0.05$ (ANOVA, LSD test).

citrus essential oils has been demonstrated in several lepidopterans including the cotton leafworm.[5,28,29]

Although a limited number of studies proved the toxicity of silica nanostructures prepared by the sol–gel method on *Spodoptera* spp.,[23,30] no information is currently available on the toxicity of silica nanoparticles encapsulating *Citrus sinensis* essential oil against *S. littoralis*. Silica nanoparticles can be physisorbed by the insect cuticular lipids causing mostly mechanical damages that lead to death.[31] Furthermore, the terpenes contained in *Citrus* essential oil can cause insect mortality through multiple modes of action.[32] SiliOrange at the tested ultralow amount caused no acute toxicity in *A. gossypii* adults, whereas the imidacloprid-based treated control caused the highest mortality ($F_{3,20} = 233.84$; $p = 0.001$) (**Figure 9**).

This absent toxicity of SiliOrange microcapsules in *A. gossypii* bioassay using the ultralow amount of encapsulated crude orange oil might be explained by the different susceptibility of the pest to the citrus essential oil encapsulated in the nanoparticles.[33] Moreover, considering that *Citrus sinensis* is a host plant of *A. gossypii*, a possible insect adaptation to the secondary metabolites of the host plant may have reduced the susceptibility of this species to citrus essential oil constituents as a result of a co-evolutionary process.[34]

By contrast, SiliOrange significantly decreased the offspring by ≈48% in *A. gossypii* in comparison with the untreated control ($F_{3,20} = 4.08$; $p = 0.025$). However, the same depletion in the progeny production was found in the SiliOrange group and its blank control (**Figure 10**). This reduction of aphid fertility in both silica-based treatments may be caused by the action of amorphous silica gels through contact leading to desiccation as in different agricultural pests, such as the black bean aphid *A. fabae* Scopoli (Hemiptera: Aphidiidae).[35]

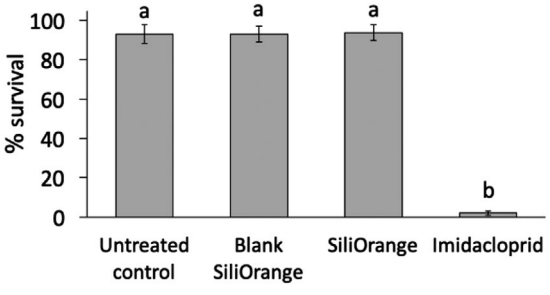


Figure 9. Mean survival (±SE) of *A. gossypii* adults in the contact toxicity bioassay. Bars with different letters indicate significant differences at $p < 0.05$ (ANOVA, LSD test).

4. Outlook and Conclusions

This work reports a reproducible synthetic sol–gel route to relatively monodispersed (PDI = 0.249) spherical SiO_2 submicron particles (153 nm in size) loaded with 7 wt% crude orange oil (SiliOrange), and its first application as a potential formulation for the control of economically relevant agricultural pests with different feeding strategies.

We have demonstrated for the first time that an ultralow amount of SiliOrange capsules dispersed in water show surprisingly high insecticidal activity against the cotton leafworm *Spodoptera littoralis*, and significantly reduce the number of offspring of cotton aphid *Aphis gossypii* under laboratory testing conditions. The tested water-based formulation contained 500 mg of SiliOrange microspheres in 150 mL water. This translates into 35 mg of crude orange oil only, which equates to a nominal concentration of 0.23 ppm of orange oil in water.

The activity of this highly diluted dispersion was compared to that of commercially available insecticides with well-known active ingredients.

From the viewpoint of pesticide safety, the entomotoxicity of SiliOrange toward *S. littoralis* as well as its environmental impact at different levels should be assessed in the field.[36] Our research also revealed that SiliOrange sub-micron silica capsules obtained through the sol–gel process[37] can impair the fertility of *A. gossypii*. Therefore, further studies on the sublethal impact of the chemical at transgenerational level should be investigated by the estimation of population demographic indexes.[38–40]

Although preliminary studies on the toxicity of nanomaterials against insect pests have been reported,[41] current knowledge on their mechanisms of action and physicochemical characteristics in open field conditions is still limited, thus new investigations are urgently needed.[40]

Regardless of the commercial importance of orange essential oil,[1] including its use as active ingredient in new biobased insecticides,[9] successful sol–gel microencapsulation in spherical SiO_2 particles has not been reported so far. Previous work synthesized silica microparticles functionalized with limonene,[42] or silica and organosilica microcapsules doped

with bergamot oil.[43] Though similar, the aforementioned preparative route cannot be applied to the microencapsulation of orange oil, which required the crucial presence of glycerol in the sol–gel precursor mixture, likely to reduce the aggregation of the surfactant CTAB molecules in the micellar nanoreactors (Scheme 1).[19,37]

For the purpose of pest control using SiliOrange microcapsules, two most important aspects are the safety for human health[44] and the environmentally friendly nature[8] of amorphous mesoporous SiO_2. Both aspects are promising for the approval of the SiliOrange material as solid pest control agent.

Considering the ease of the preparation of the material, the biocompatibility of both silica and orange essential oil, and the ultralow amount of SiliOrange microcapsules used, these results may open the route to sustainable pest control based on sol–gel microencapsulated orange oil.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

All raw data concerning the biological parameters represented in the graphs of this article are available upon contacting one of the corresponding authors (L.Z.). All data concerning the characterization of the SiliOrange materials are available upon contacting one of the corresponding authors (R.C.).

Keywords

orange oil, microencapsulation, silica, sol–gel, pest control, cotton aphid, cotton leafworm

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