Chinese Chemical Letters 31 (2020) 2137–2141

Contents lists available at ScienceDirect

Chinese Chemical Letters

journal homepage: www.elsevier.com/locate/cclet

Communication

Fabrication linalool-functionalized hollow mesoporous silica spheres nanoparticles for efficiently enhance bactericidal activity

Li Jin[a], Xun Liu[a], Changhao Bian[a], Jie Sheng[a,b], Yishan Song[a,b,*], Yongheng Zhu[a,b,*]

[a] College of Food Science and Technology, Shanghai Ocean University, Shanghai 201306, China
[b] College of Food Science and Technology, Laboratory of Quality & Safety Risk Assessment for Aquatic Products on Storage and Preservation (Shanghai), Ministry of Agriculture and Shanghai Engineering Research Center of Aquatic-Product Processing & Preservation, Shanghai Ocean University, Shanghai 201306, China

ARTICLE INFO

Article history:
Received 8 November 2019
Received in revised form 3 December 2019
Accepted 12 December 2019
Available online 13 December 2019

Keywords:
Linalool
Hollow mesoporous silica spheres
Encapsulation
Bactericidal activity
Antimicrobial mechanism

ABSTRACT

To develop a novel food preservation technology for efficiently enhance bactericidal activity in a long term, hollow mesoporous silica spheres (HMSS) with regular nanostructures were applied to encapsulate natural organic antimicrobial agents. The chemical structures, morphologies and thermal stabilities of linalool, HMSS and linalool-functionalized hollow mesoporous silica spheres (L-HMSS) nanoparticles were evaluated by polarimeter, field emission scanning electron microscope (FE-SEM), transmission electron microscope (TEM), fourier transform infrared (FT-IR), thermal gravimetric analyzer (TGA), nitrogen adsorption-desorption, zeta potential and small angle X-ray diffraction (SXRD). The results show that the linalool was successfully introduced into the cavities of HMSS, and the inorganic host exhibited a high loading capacity of about 1500 mg/g. In addition, after 48 h of incubation, the minimum bactericidal concentrations (MBC) of L-HMSS against *Escherichia coli* (*E. coli*), *Salmonella enterica* (*S. enterica*) and *Staphylococcus aureus* (*S. aureus*), *Listeria monocytogenes* (*L. monocytogenes*) were decreased to be 4 (< 5) mg/mL and 8 (< 10) mg/mL, respectively. These results revealed linalool-functionalized hollow mesoporous spheres could efficiently improve the bactericidal activities of the organic component. Furthermore, SEM images clearly showed that L-HMSS indeed had an extremely inhibitory effect against gram-negative (*E. coli*) and gram-positive (*S. aureus*) by breaking the structure of the cell membrane. This research is of great significance in the application of linalool in nano-delivery system as well as food industry.

Hollow mesoporous silica spheres nanoparticles and its non-metallic nanostructures feature as nontoxic, tunable pore diameter, large surface area, high thermal stability and great biocompatibility [1–3]. With those benefits and advantages, it is no doubt that hollow spheres have been applied in various fields, including biochemistry, medicines, agricultures, foods and other industries [4–6]. However, there are few researches about the combination of hollow mesoporous silica spheres nanoparticles with natural products to maintain/enhance its bioactivities, especially against food-borne bacteria [7,8]. The mechanism of efficiently improve the bactericidal activity of functionalized nanoparticles may be due to the strong electrostatic attraction between negatively charged cell surface and positively charged nanoparticles [9]. This binding between bacterial cell walls and functionalized hollow mesoporous silica spheres could cause locally high concentrations of agents to attack cell walls, easily damaging cell membrane and causing cell death [10–12].

Interestingly, linalool (3,7-dimethyl-1,6-octadien-3-ol), an active substance, is a major composition of certain essential oils including coriander, basil, lavender and so on [13]. It shows antimicrobial, anti-inflammatory, anticancer, anti-hyperlipidemic and analgesic properties [14,15]. However, linalool often presents an intense smell, continuous volatility, poor water solubility and unstability, which extremely restrict its applied prospects and academic values [16]. In order to overcome these disadvantages, it is necessary to adopt some novel strategies, such as nano-encaplusation, nanoemulsion and liposome [17–19], to perfect the biochemical properties of linalool and meantime still efficiently enhance antimicrobial activity [20]. In view of this, mesoporous silica materials as carriers come into being to meet the needs of long-term and sustained-release properties in recent

* Corresponding authors at: College of Food Science and Technology, Shanghai Ocean University, Shanghai 201306, China.
E-mail addresses: yssong@shou.edu.cn (Y. Song), yh-zhu@shou.edu.cn (Y. Zhu).

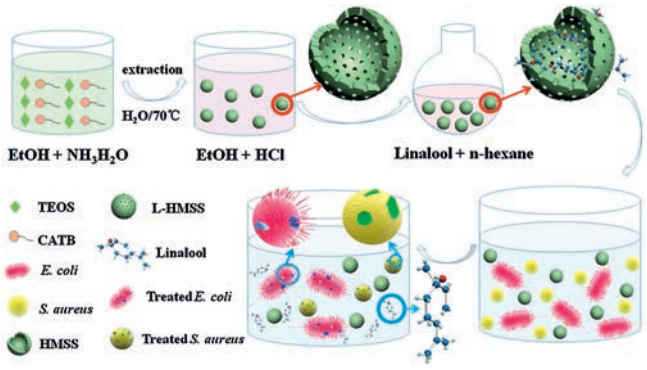
https://doi.org/10.1016/j.cclet.2019.12.020
1001-8417/

years [21–24]. As the most promising nanoloading system for smart delivery, active agents entrapped in mesoporous silica nanoparticles have attracted considerable prospects due to their well-defined morphology, high loading capacity, on the nanoscale with pore channels penetrating from the outside to the inner hollow core and favorable for the target molecular adsorption/release [25–27], which can improve the antibacterial efficacy [20,28].

In our research, hence, natural antimicrobial agents encapsulated hollow mesoporous silica spheres were firstly prepared successfully to inhibit the growth of food-borne bacteria, intending to form a long-term and stable molecule delivery system (linalool functionalized nanoparticles). In contrast to linalool, L-HMSS showed the total inhibitory concentrations against *E. coli*, *S. enteric* (< 5 mg/mL) and *S. aureus*, *L. monocytogenes* (< 10 mg/mL) after 48 h of incubation. At the same time, the chemical characterizations of the HMSS and L-HMSS were conducted and the antimicrobial mechanism was also studied by disrupting cellular membrance structure. This study would have an important significance in the application of natural antimicrobial products in nano-delivery system and bactericidal activity enhancement. The experimental procedures are described in detail in Supporting information.

Linalool is so-called 3,7-dimethyl-1,6-octadien-3-ol with the molecular formula $C_{10}H_{18}O$. The structural formula of monomer might be (*R*)-(−)-, (*S*)-(+)- or (*RS*)-(±)- forms. From the specific rotation analysis with the three authentic form, the commercial specimen was recognized as (*R*)-(+)-linalool with $[\alpha]_D$ = 17.4°. The structural formula of (*R*)-(+)-linalool was presented in Fig. S1 (Supporting information). To be exact, the rotatory direction and optical rotation are determined by the molecular stereostructure [29]. Moreover, the deviations of the structure and function of linalool may vary link with species, harvest time, geographic regions, local climate, extraction methods and some abiotic factors [30]. This would have a predominant impact on physico-chemical properties of linalool.

SEM and TEM images determined the size and morphology of HMSS and L-HMSS. The synthetic procedure of HMSS/L-HMSS was illustrated in Scheme 1. As shown in Figs. 1a and c, the synthesized nanoparticles size was *ca.* 600 nm. The difference of size distribution was due to the influence of ammonia concentration. Under otherwise constant experimental conditions, the particle sizes became larger with the increase of ammonia hydroxide concentration (up to 8 mmol/mL) [31]. Furthermore, the unique and uniform spherical morphology were formed by hydrothermal and carbonization treatment. The reason for that is the modified spherical silica templates polymerized onto the surface with high monodispersity [32]. Thus, there were traces of molecular



Scheme 1. Schematic illustration of the preparation of HMSS/L-HMSS and the bactericidal process against *Escherichia coli* (*E. coli*) and *Staphylococcus aureus* (*S. aureus*) of L-HMSS.

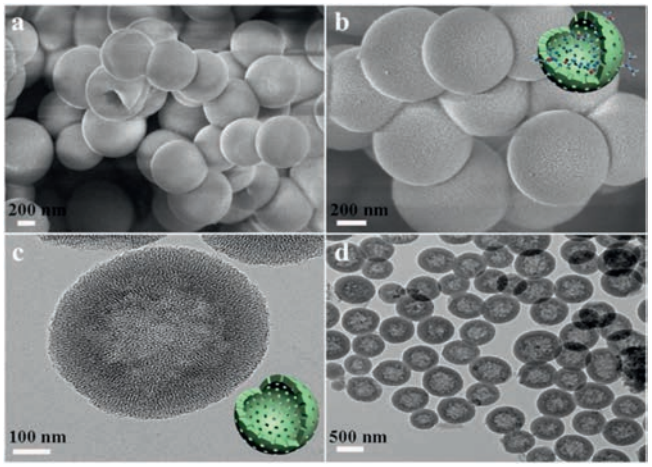


Fig. 1. SEM images of (a) HMSS and (b) L-HMSS. TEM images of (c) HMSS and (d) L-HMSS.

condensation properly on the silica surfaces. It can be also seen in Figs. 1b and d that the L-HMSS remained unchanged in loading process. That is the fact that the high stability of hollow mesoporous silica spheres [33].

Fig. S2 (Supporting information) presented the FT-IR spectra of HMSS, L-HMSS and linalool, and Table S1 (Supporting information) collected the respective peak assignments. The strong peak of hollow spheres at 1046 cm^{-1} is attributed to Si-O-Si group. The weak peaks at ~2920 and ~2853 cm^{-1} are due to asymmetric stretching of CH_2 group of CTAB and C–H in the hollow spheres structure. As for linalool, the chemical structure was found to be remarkable similar with the previous report [34]. The L-HMSS showed similar infrared absorption bands except for the shoulders peaks at 3378, 3084, 2970, 1637, 1452 and 1375 cm^{-1} which are the absorption bands of linalool compared with IR, indicating that the ligand was embedded onto the wall of hollow spheres. In addition, the absorption band of C—C should appear in the 1112 cm^{-1} but it is overlapped by the absorption band of the Si-O-Si bonds in the 1046 cm^{-1} [35]. Interestingly, the relative absorbance intensity between the antisymmetric stretching vibration of C–H bond of L-HMSS is bigger than that of the hollow spheres and linalool, which means that the C–H number is increased, because the superposition of the C–H group between HMSS and linalool. Meanwhile, those imply linalool functionalized hollow mesorporous silica spheres nanoparticles successfully.

Fig. S3 (Supporting information) shows the TGA curves of HMSS, L-HMSS and linalool. It is a very intuitive tendency that showing a slight and steady weight loss between 30 °C and 180 °C attributed to the water and solvent (~5%) removal from the surface of hollow spheres. The organic substance (~20%) is thermally degraded at 180−330 °C. Likewise, TGA curve of linalool illustrates a two-step weight loss. The initial weight loss occurred at the temperature range of 30−100 °C due to the solvent evaporation. Moreover, with the temperature goes up to 180 °C, linalool is pyrolyzed completely in the second stage. Most notably, TGA results demonstrate the thermal degradation of L-HMSS including three steps. The first degradation step begins at 30 °C due to the loss of water with a weight loss of about 4%. At the same time, the second step is accompanied with the thermal degradation of linalool which is about 60% between 30 °C and 150 °C. At temperatures above 150 °C, the remained aromatic carbon components turn to ash with the value of about 10%. More importantly, TGA analysis was also performed to quantify the amount of linalool encapsulated into HMSS. The curves exhibit the remain weight mass of linalool from L-HMSS. It exhibited the

loading capacity of about 1500 mg/g. The results showed a high loading capacity of L-HMSS.

To explore investigate the special structure of the hollow spheres, the nitrogen adsorption-desorption isotherm is shown in Fig. S4 (Supporting information), where it could be defined as classical type-IV curve (Fig. S4a) with a hysteresis loop in the P/P_0 range from 0.4 to 1.0. The result implied the existence of a highly ordered mesoporous network. The pore size distribution of HMSS is evaluated from Fig. S4b (Supporting information). The BET total pore volumes (1.52 cm^3/g) and specific surface areas (887.8 m^2/g) of HMSS are summarized. The mesoporous material presented a relatively narrow pore size distribution, suggesting that the pore channels could be filled with the targeted molecules [36]. Therefore, the non-metallic nanostructures are expected to be related with the loading capacity enhancement. It also further confirmed that L-HMSS exhibiting a high loading capacity.

Table S2 (Supporting information) showed that the average size of HMSS were 616 ± 9.93 nm while that of L-HMSS were 634 ± 18.46 nm. Besides, the zeta potential of pure matrix and L-HMSS were -51.95 ± 3.37 and -40.91 ± 1.41 mV, respectively, the change of values suggested that linalool had been introduced to cavities of hollow spheres.

The SAXRD patterns of preparing host HMSS and L-HMSS were presented in Fig. S5 (Supporting information). For preparing host, the pattern clearly showed the order of the hexagonal array of the hollow spheres structure and exhibited distinct Bragg peaks in the 2θ range of 0.6°-6°, which can be indexed as (100) reflections. The L-HMSS showed similar curve with that of pure matrix, only the diffraction intensity was lower, which was due to the presence of linalool moieties inside the pore channels of the HMSS, resulting in the decrease of the mesoporous order. The results further verified the preparation of the linalool functionalized hollow mesoporous silica spheres successfully.

By detecting the growth reduction of four typical food-borne microbial bacteria, gram-negative (*E. coli*, *S. enterica*) and gram-positive (*S. aureus*, *L. monocytogenes*), to investigate the antimicrobial and bactericidal activities of pure linalool. The bactericidal activity of pure linalool was referred as the MBC. The results meant that the growth rate of the initial microbial amounts decreased by 99.9%.

As can be seen from Fig. 2, the growth reduction rates of the four tested bacteria (linalool concentration ranging from 1 mg/mL to 12 mg/mL) during 24 h of incubation were observed. Most importantly, there is no remarkable difference between 24 h and 48 h incubation with respect to the MBC on both gram-negative and positive bacteria. In addition, the MBC of linalool for *E. coil* and *S. enterica* were both 5 mg/mL as suggested in Figs. 2a and b. In different antimicrobial concentrations of linalool including 1, 2, 3, 4 mg/mL, the microbial growth inhibition rate of *E. coil* were 0.006 ± 0.006, 0.239 ± 0.024, 0.583 ± 0.039 and 0.865 ± 0.033, while those of *S. enterica* were 0.055 ± 0.045, 0.363 ± 0.013, 0.614 ± 0.034 and 0.800 ± 0.020, respectively. The result demonstrated that linalool has a significant inhibitory effect against *E. coil* (gram-negative). However, the presented MBC for *E. coil*, in the presence of linalool, in the test was lower than the concentration established by another report [37]. These differences may be caused by the chosen bacterial strain, tested condition and matrix effect [38]. In that case, higher amounts of linalool were required to attain the same effect on inhibiting the growth of microbial.

For *S. aureus* and *L. monocytogenes* in Figs. 2c and d, the total inhibition concentration (MBC) were both 10 mg/mL. In a series of antibacterial concentrations of linalool including 2, 4, 6 and 8 mg/mL, the microbial growth inhibition rate of *S. aureus* were 0.004 ± 0.004, 0.517 ± 0.049, 0.718 ± 0.049 and 0.827 ± 0.027, while those of *L. monocytogenes* were 0.071 ± 0.019, 0.317 ± 0.034, 0.653 ± 0.028 and 0.815 ± 0.015, respectively. These data implied that the linalool inhibited the growth of the gram-positive bacteria in the concentration between 2 mg/mL and 10 mg/mL after 24 h and 48 h of incubation.

Compared the bactericidal activities between gram-negative and positive bacteria, it was obvious that the MBC for gram-negative bacterium were lower than that of negative bacterium, which manifested that the effect of linalool on membrane permeability to gram-negative bacterium may be much more remarkable. Likewise, linalool showed greater antimicrobial activities to gram-negative than positive bacterium at the same inhibition concentration.

Most interestingly, a few researches reported that gram-positive bacteria are inclined to be more sensitive than negative bacteria when treated with certain natural antimicrobial products. On the contrary, Nakatani found out that the MBC of linalool to gram-negative bacteria was lower than that to gram-positive [39], which was in line with our study. Different functional groups of linalool were crucial for antimicrobial activity of bacteria, and different structures of cell biofilm led to different sensitivities. More concretely, linalool was more effective in inhibiting gram-negative bacteria, which might be due to the fact that the gram-negative bacteria with lipid bilayer had better lipophilic–hydrophobic properties than gram-positive ones with thick layer of peptidoglycan [40]. Besides, we would have a further explore and discuss the morphological changes of the cells.

To analysis the antibacterial activity of the mesoporous silica matrix, the hollow mesoporous silica spheres were tested at concentrations ranged from 1 mg/mL to 5 mg/mL in TSB. The hollow spheres are a mesoporous material with large specific surface area and high stability. When compared the results with positive controls, the two kinds of particles did not reduce the growth of any bacteria. These phenomena demonstrated that the different size particles have no effect on bacterial viability. The conclusion is in accordance with the previous report [41].

As seen in Figs. 3a and b, the MBC of the L-HMSS against *E. coil* and *S. enterica* reduced to 4 mg/mL after 24 and 48 h incubation. Apparently, the functionalized nanoparticles showed significantly

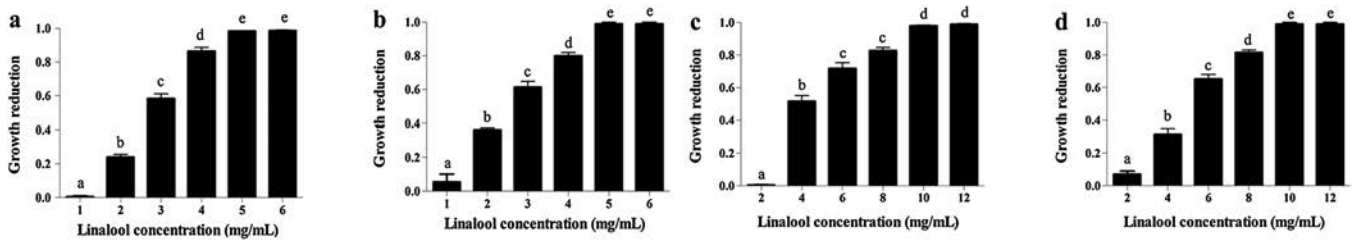


Fig. 2. The growth reduction of (a) *E. coli*, (b) *S. enterica*, (c) *S. aureus* and (d) *L. monocytogenes* treated with different linalool concentrations at 24 h. The same letters in the bars show homogeneous group membership ($P < 0.05$) (n = 3).

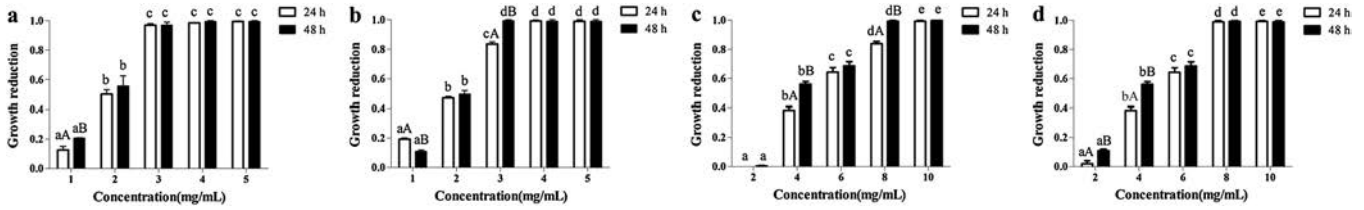


Fig. 3. The growth reduction of (a) *E. coli*, (b) *S. enterica*, (c) *S. aureus* and (d) *L. monocytogenes* treated with different L-HMSS concentrations at 24 h (white) and 48 h (black). Different letters in the bars indicate statistically significant differences ($P < 0.05$) from levels of concentration (small letters) and exposure time (capital letters) ($n = 3$).

greater bactericidal activity at low concentrations. But after 48 h incubation, the growth reductions were more significant at the same concentrations. Similarly, the total inhibition concentration value (10 mg/mL) for L-HMSS against *S. aureus* and *L. monocytogenes* were the same with that of linalool in 24 h incubation, while the MBC decreased to 8 mg/mL after culturing for another 24 h in Figs. 3c and d. At 24 h, the treatment with 4, 6 and 8 mg/mL linalool loaded into hollow spheres also had a bacterial growth reduction. However, after 48 h incubation, the growth reductions had a better antimicrobial activity at the same concentrations.

These results indicated that loading process did not affect the bactericidal activity of linalool. Besides, the L-HMSS indeed had a better inhibitory effect against the bacteria for a long time. In other words, the encapsulated nanoparticles not only maintain the antibacterial activity, but also showed a long-term and sustained-release property in *vitro*. As a consequence, nanoencapsulation would have a great promising for both controllable release and bactericidal activity enhancement in organic delivery system.

When comparing the antimicrobial activity of monomer linalool and L-HMSS (Figs. 2 and 3), it was shown that the growth reduction of the linalool and L-HMSS against *S. aureus* and *L. monocytogenes* still provided with the same range (2−10 mg/mL). However, linalool more effectively inhibited the growth of *E. coil* and *S. enterica*. The efficiently improve the bactericidal activity of functionalized nanoparticles may be due to the strong electrostatic attraction between negatively charged cell surface and positively charged nanoparticles, which destroy cell membrane easily and induce the cell death. Besides it may be caused by the controllable release of linalool from L-HMSS. These results also implied that the hollow spheres nanoparticles are applicable matrix for linalool functional modification and slow release delivery for antibacterial system.

As stated above, Fig. 4 depicts the SEM images of the changes of *E. coil* and *S. aureus* cells morphology to further probe the mechanism of L-HMSS inhibiting bacterial growth. As a representative of food-borne pathogens, the SEM images show the classic morphology of *E. coil* cells with an intact membrane and inerratic structure (Fig. 4a). Most noticeably, the *E. coil* cells were treated with 4 mg/mL of L-HMSS, showing an irregular cell surface and notable ruptures or pores in cell membrane (Fig. 4b). Likewise, as shown in Figs. 4c and d, the morphological of treated *S. aureus* cells with 4 mg/mL of L-HMSS underwent obvious changes compared with that of untreated controls. It is even more obvious that the cell surface was broken and the collapsed cells even fused with each other.

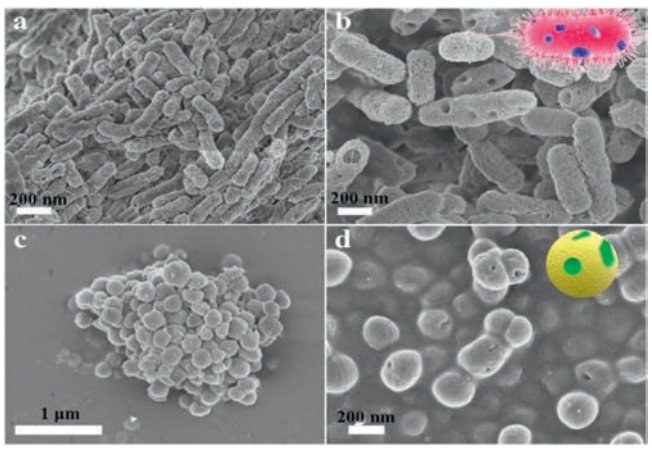


Fig. 4. SEM images of (a,b) *E. coil* and (c, d) *S. aureus* with 4 mg/mL of L-HMSS for 3 h.

The SEM images also demonstrated the structure deeper damage degree of *E. coil* at the same concentration of L-HMSS compared with *S. aureus* cells, it implied the *E. coil* (gram-negative) is more sensitive than *S. aureus* (gram-positive) when the same L-HMSS concentrations are used. In addition, the process of centrifugation may separate the hollow spheres from the samples, which caused the HMSS to go undetected by SEM.

In the light of several studies, the mechanism of action of linalool on bacteria cells is based on the alteration of the different essential processes occurring at the cell envelope, which inhibits the growth of microorganism [42]. The cellular division and death may be due to change its permeability and destroy the integrity of the cell membrane by active groups of linalool [43].

In conclusion, a novel hollow sphere with stable structure was synthesized by silica templating method for linalool protection applications. We encapsulated linalool into hollow spheres to form a remarkable bactericidal formula. The regular morphology of hollow spheres remained stable and unchanged during linalool embedding process analyzed by TEM and SEM, and results from FT-IR and TGA demonstrated that the L-HMSS was successfully formed with good stability and high loading capacity. Most importantly, the new formulation showed an effective bactericidal effect against food-borne bacterium especially *E. coli* and *S. enterica* (gram-negative bacteria). The long-term and continuous release of L-HMSS achieved by nanoloading gave them greater bactericidal effect than linalool after 48 h incubation. Furthermore, SEM images clearly showed that L-HMSS indeed had a remarkable inhibitory effect against *E. coli* and *S. aureus* by breaking the structure of the cell membrane. As we expected, linalool functionalized into hollow spheres are a novel nanomaterial for the controllable release of natural agents as well as for enhancing antimicrobial activity.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

The National Natural Science Foundation of China (No. 31701678), and the Shanghai Key Research Projects of Promoting

Agriculture by Science and Technology (No. 2019-02-08-00-15-F01147) supported this work.

Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:https://doi.org/10.1016/j.cclet.2019.12.020.

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