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SHORT COMMUNICATION

Chitosan-based nanofragrance with antibacterial function applied to wallpaper

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Abstract

Adding fragrances to the wallpaper can optimize our living environment and office environment. However, the poor adhesion and rapid release of fragrances on wallpapers have limited their application. In this study, vanillin was encapsulated in particles based on chitosan and poly(lactic-co-glycolic acid), thereby achieving a slow release of the fragrance. In addition, due to the addition of chitosan, the adhesion of the fragrance on the wallpaper was enhanced, and the wallpaper was given antibacterial properties.

KEYWORDS

antibacterial function, chitosan, fragrance, slow release, wallpaper

1 | INTRODUCTION

Wallpapers are widely used in our lives [1–4]. Adding fragrances to the wallpaper can make our working and living environment full of aroma, so that we can work and live more comfortably. However, the aromatic wallpaper prepared by directly adding fragrances to the wallpaper has great defects. First, there is no interaction between most fragrance molecules and wallpaper. Therefore, the adhesion of fragrances on the wallpaper is poor. Second, the release of fragrances from most aromatic products is rapid [5–8]. Excessive release of fragrance molecules leads to a strong

Abbreviations: CS, chitosan; PDI, polymer dispersibility index; PLGA, poly(lactic-co-glycolic acid).

aroma in the air, which can cause us discomfort. In addition, the rapid loss of fragrances shortens the life of aromatic products. Finally, wallpapers easily absorb microorganisms such as bacteria, which affects people's health. Therefore, the key to preparing aromatic wallpaper lies in the following three aspects: (1) enhance the interaction between fragrance and wallpaper; (2) slow down the release rate of fragrance; (3) give wallpaper antibacterial function.

Although the interaction between the fragrances and the wallpaper is poor, the fragrance can be encapsulated in particles that can better adhere the wallpaper, thereby improving the adherence of the fragrances to the wallpaper. Besides, encapsulating fragrances in nanoparticles can also slow the release rate of fragrances. In addition, by adding an antibacterial component to the surface of the nanoparticles, wallpaper could have antibacterial effect. Many kinds of nanofragrances have been reported. For example, Zhou et al. reported two kinds of metal-organic frameworks for encapsulation of fragrance and long-term sustained release of fragrances.[9,10]

Poly (lactic-co-glycolic acid) (PLGA) is a polymer made by random polymerization of lactic acid and glycolic acid. It is an organic polymer compound with good biocompatibility [11,12], degradable function [13,14], nontoxicity [15,16], good encapsulation [17,18], and film-forming properties [19,20]. PLGA is widely used in pharmaceutical, medical engineering materials, and modern industrial fields. Therefore, PLGA could be chosen to encapsulate fragrance. The main ingredient of wallpaper is cellulose. Chitosan (CS) has been reported to have a strong interaction with cellulose, which can improve the adhesion of perfumes to wallpaper [21,22]. Besides, CS has a good antibacterial effect [23-25]. Therefore, the use of CS can not only enhance the adhesion of the fragrance on the wallpaper but also can impart an antibacterial effect on the wallpaper.

In this study, Vanillin@CS-PLGA nanocapsules were prepared by double emulsion-solvent evaporation method(W/O/W) to achieve a better sustained release effect and a significant antibacterial properties of the films for *Escherichia coli* and *Staphylococcus aureus*. The nanocapsules were then added in wall to paper Vanillin@CS-PLGA-W. The Vanillin@CS-PLGA can improve the adhesion of vanillin to wallpaper.

PRACTICAL APPLICATION

Fragrances and wallpaper are the most commonly used daily necessities. Scented wallpaper can provide us with a comfortable living and working environment. However, the fragrances on traditional aromatic wallpaper have poor adhesion and the fragrances are released too quickly. In this study, CS-PLGA@Vanillin was designed and prepared to overcome the shortcomings of traditional aromatic wallpaper. It not only improved the adhesion of the fragrance to wallpaper, but also slowed down the release rate of the fragrance. In addition, it also gave the wallpaper antibacterial function. More importantly, it had low production cost, simple preparation method, and could be industrially produced. Therefore, CS-PLGA@Vanillin has a great application prospect.

2 | MATERIALS AND METHODS

2.1 | Materials

PVA (polymerization degree, 1700, hydrolysis degree, 99%) was purchased from Innochem. PLGA was bought from Sigma. Vanillin and CS were obtained from J&K Chemical. *Staphylococcus aureus* (ATCC25923) and *E. coli* (ATCC25922) were purchased from Rishui Biotech Co. Ltd (Qingdao, China). All other chemicals such as ethanol and tetrahydrofuran were analytical grade reagents and obtained locally.

2.2 | Preparation of CS-PLGA loaded with vanillin (Vanillin@CS-PLGA) and wallpaper loaded with vanillin (Vanillin@CS-PLGA-W)

Note that 500 mg of CS was dissolved in 500 mL of dilute acetic acid solution (1% v/v) to prepare 1 mg/mL of CS acetic acid solution. PLGA and vanillin solution with different concentrations were prepared. PLGA solution was dissolved in vanillin solution to form a primary emulsion, which was further emulsified in 1 mg/mL of CS acetic acid solution (PVA solution 1% v/v). Then the emulsion was purified by ultracentrifugation at 50 602 × *g* under cooling (4°C) for 20 min, followed by washing thrice with double-distilled water to obtain Vanillin@CS-PLGA.

Finally, wallpaper (50 cm^2) was immersed in Vanillin@CS-PLGA aqueous solutions (1 mg/mL). After 12 h, the wallpaper was then dried at 40°C for 1 h in an oven to obtain Vanillin@CS-PLGA-W.

2.3 | Characterization of Vanillin@CS-PLGA

The morphology of Vanillin@CS-PLGA was observed using a scanning electron microscope (SEM, JEOL, Japan).

2.4 | Particle size

The particle size of the above sample in three replicates was determined by a dynamic light scattering using a Malvern Zetasizer nano ZS apparatus (Malvern Instruments, Malvern, United Kingdom). Each sample was measured by a solid state

TABLE 1 The hydrodynamic diameters and PDIs of the Vanillin@CS-PLGA with different mass ratios of CS–PLGA–vanillin

CS–PLGA–vanillin	Average size (nm)	PDI
4:3:3	271.7	0.407
4:3:5	342.8	0.38
4:4:3	213.2	0.268
4:4:5	320.2	0.381
4:5:3	269.7	0.310
4:5:5	273.7	0.310

He–Ne laser of 633.0 nm at 25°C with an angle detection of 90°. The size distribution was evaluated for all samples according to the polymer dispersibility index (PDI).

2.5 | The morphology of the wallpaper

The surface morphologies of the untreated wallpaper and the wallpaper finished by Vanillin@CS-PLGA were studied via scanning electron microscope (SEM, JEOL, Japan).

2.6 | Determination of loading efficiency

The content of vanillin in the product was determined by a UV-Vis spectrophotometer at 231 nm wavelength, in which the standard curve was determined by vanillin of known concentration. The loading efficiency of vanillin in Vanillin@CS-PLGA was calculated by the following formula:

$$\text{Loading efficiency } (\%) = W_1/W_2 \times 100\%,$$

where W_1 is the content of vanillin and W_2 is content of Vanillin@CS-PLGA.

2.7 | The content and release of vanillin

The content and release of vanillin was measured by HPLC. For HPLC analysis, we used a mobile phase with water/acetonitrile (v/v = 40%/60%) at a flow rate of 1.0 mL/min and a C18 reversed-phase column (250 × 4.6 × 5 μm). The concentrations of linalool based on the peak area at the retention time of 5 min at 231 nm. The release rate is calculated via the following formula:

$$\text{Release rate } (\%) = W_1/W_2 \times 100\%$$

where W_1 is the weight of released vanillin and W_2 is the total weight of vanillin.

2.8 | Tests for the antibacterial properties

For the inhibition zone, *S. aureus* (ATCC strain 29523) and *E. coli* (ATCC strain 29522) were used to evaluate the antibacterial activity of the Vanillin@CS-PLGA. The bacterial suspension (50 μL) was taken out by pipette, poured into an aureus agar plate, and coated uniformly by a glass coating rod. Then, Vanillin@CS-PLGA, Vanillin@PLGA, and water were loaded onto the surface of the agar. After this, the aureus agar plate was incubated at 37°C for 24 h. The inhibition zone was determined by a digital camera.

2.9 | Statistical analysis

All the data were analyzed in triplicate and presented as a mean value with standard deviation (mean ± SD).

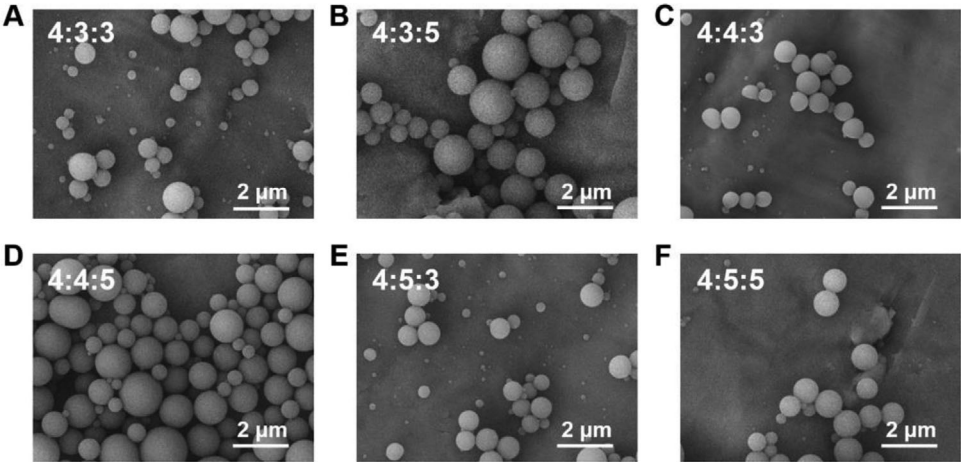

FIGURE 1 (A–F) The SEM images of Vanillin@CS-PLGA with different mass ratio of CS–PLGA–vanillin. Scale bars: 2 μm

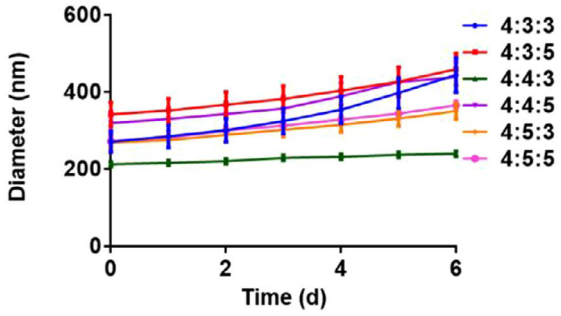

FIGURE 2 The stability of Vanillin@CS-PLGA with mass ratio of CS–PLGA–vanillin

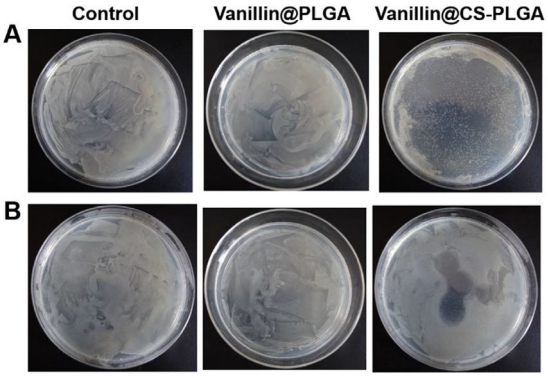

FIGURE 4 (A) The antibacterial effect against *E. coli*. (B) The antibacterial effect against *S. aureus*

3 | RESULTS AND DISCUSSION

3.1 | The preparation and characterization of Vanillin@CS-PLGA

The ratio of CS, PLGA, and vanillin is critical to the performance of Vanillin@CS-PLGA. As shown in Table 1, 6 ratios of chitosan, PLGA, and vanillin were used to prepare Vanillin@CS-PLGA. The results showed that when the mass ratio of CS–PLGA–vanillin was 4:4:3, the particle size and PDI of Vanillin@CS-PLGA were the smallest, which indicated that when the mass ratio of CS–PLGA–vanillin was 4:4:3, Vanillin@CS-PLGA was the most homogeneous. Therefore, this mass ratio was chosen to prepare Vanillin@CS-PLGA.

The morphologies of Vanillin@CS-PLGA with different mass ratios of CS–PLGA–vanillin were characterized by SEM. As shown in Figure 1A–D, all the Vanillin@CS-PLGAs were spherical. In addition, the size of Vanillin@CS-PLGA was smallest and the Vanillin@CS-PLGA was the most homogeneous when the mass ratio of CS–PLGA–vanillin was 4:4:3. These were consistent with the hydrodynamic diameters and PDIs of the Vanillin@CS-PLGA with different mass ratios of CS–PLGA–vanillin in Table 1.

Stability is also one of the important indicators of Vanillin@CS-PLGA performance. The hydrodynamic diameters of Vanillin@CS-PLGA with different mass ratio at different times were measured to evaluate the stability of these nanofragrances. As shown in Figure 2, the average diameter of the Vanillin@CS-PLGA only increased by 12.85% after 6 days when the mass ratio was 4:4:3. In contrast, the average diameters of Vanillin@CS-PLGA with other mass ratios increased by more than 30%. These results indicated that the nanofragrance was the most stable when the mass ratio of CS–PLGA–vanillin was 4:4:3. Therefore, Vanillin@CS-PLGA with this mass ratio was selected.

3.2 | The loading efficiency and release of vanillin

Loading efficiency is an important indicator of Vanillin@CS-PLGA. The loading efficiency of vanillin was measured by UV-Vis. First, the standard curve of vanillin was measured (Figure 3A). Subsequently, the concentration of vanillin in Vanillin@CS-PLGA was calculated by measuring the absorbance of Vanillin@CS-PLGA, and the loading efficiency of vanillin was finally calculated to be 21.5%.

Fragrance release performance is another important indicator of Vanillin@CS-PLGA. The amount of vanillin released by HPLC was detected. As shown in Figure 3B, free vanillin was released rapidly. In contrast, the release rate of Vanillin@CS-PLGA was very slow. After 24 h, less than 14% of the fragrance was released from Vanillin@CS-PLGA. In contrast, more than 96% of the free fragrance was released. In addition, only 35.46% of the vanillin was released

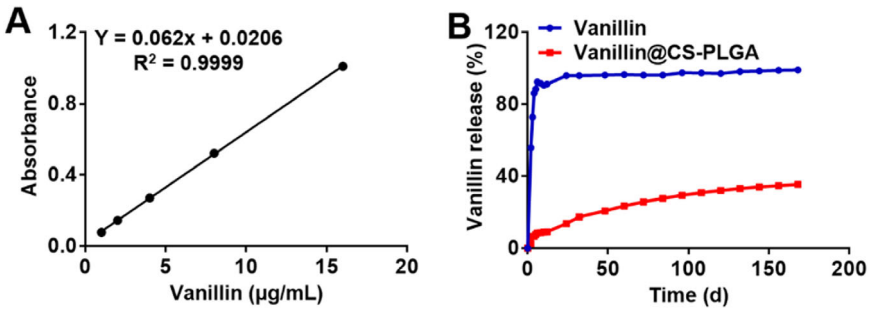

FIGURE 3 (A) The standard curve of vanillin. (B) The cumulative release profiles of vanillin in different samples

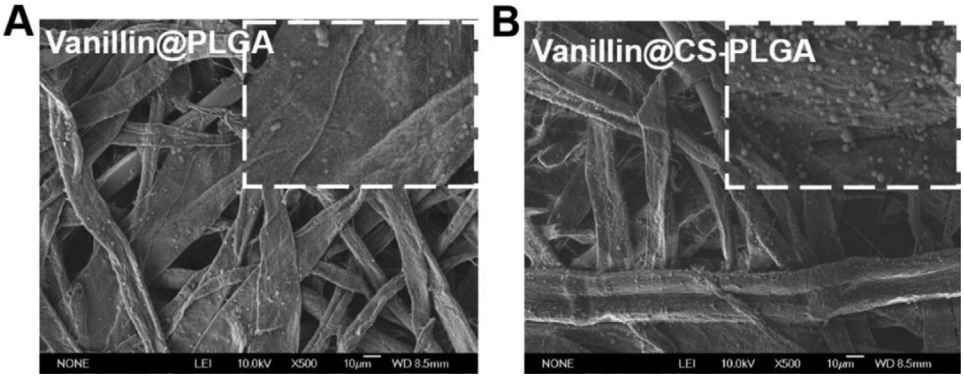

FIGURE 5 The SEM images of (A) Vanillin@PLGA-treated wallpaper and Vanillin@CS-PLGA-treated wallpaper

from Vanillin@CS-PLGA after 7 days. This result indicated that Vanillin@CS-PLGA had excellent sustained release properties.

3.3 | The antibacterial property of Vanillin@CS-PLGA

Escherichia coli is the most representative Gram-negative bacterium. *Escherichia coli* is a conditional pathogen that can cause gastrointestinal infections or urinary tract infections in humans and a variety of animals under certain conditions. *Staphylococcus aureus* is the most representative Gram-positive bacterium. Staphylococcus is the most common pyogenic coccus and an important source of hospital cross infection. Therefore, *E. coli* and *S. aureus* were selected to evaluate the antibacterial effect of Vanillin@CS-PLGA. In order to explore the antibacterial function of CS in Vanillin@CS-PLGA, nanofragrance without CS (Vanillin@PLGA) was prepared. The average diameter of the Vanillin@PLGA was 286.2 nm, which was approximately the same as the average diameter of the Vanillin@CS-PLGA.

As shown in Figure 4A, Vanillin@PLGA had no antibacterial effect against *E. coli*, while Vanillin@CS-PLGA had a very obvious antibacterial effect. Figure 4B showed the antibacterial effect of different fragrances against *S. aureus*. The results showed that Vanillin@PLGA also had no antibacterial effect against *S. aureus*. In contrast, Vanillin@CS-PLGA has a better antibacterial effect on *S. aureus*. Therefore, the addition of CS gave Vanillin@CS-PLGA an excellent antibacterial effect.

3.4 | The application of Vanillin@CS-PLGA to wallpaper

Free fragrance has poor adhesion on wallpaper. Therefore, adding free fragrance to the wallpaper is not effective. And the adhesion of fragrance on the wallpaper determines whether they can achieve industrial applications. In this study, CS was added to improve the adhesion of fragrance to wallpaper. The adhesive ability of the fragrance on the wallpaper was evaluated by the number and density of the nanoparticles on the aromatic wallpaper. As shown in Figure 5A, only a small amount of Vanillin@PLGA adhered to the wallpaper. And a large amount of Vanillin@CS-PLGA adhered to the wallpaper. Therefore, CS significantly improved the adhesion of fragrance on the wallpaper. This also meant that Vanillin@CS-PLGA had a good application prospect.

4 | CONCLUDING REMARKS

In this study, Vanillin@CS-PLGA nanocapsules were prepared by double emulsion-solvent evaporation method (W/O/W). The loading efficiency of vanillin was up to 21.5%. Vanillin@CS-PLGA had an excellent sustained release fragrance effect. In addition, Vanillin@CS-PLGA had a good antibacterial effect against both Gram-positive bacteria and Gram-negative bacteria due to the addition of CS. Moreover, the addition of CS also enhanced the adhesion of Vanillin@CS-PLGA on the wallpaper. Therefore, Vanillin@CS-PLGA was expected to have a good application prospect.

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CONFLICT OF INTEREST

The autors have declared no conflict of interest.

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