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Heat-Resistant Sustained-Release Fragrance Microcapsules

Yan Li,[1,2] Yi-Qing Huang,[1,2] Heng-Feng Fan,[1,2] Qiang Xia[1,2,3]

[1]School of Biological Science and Medical Engineering, State Key Laboratory of Bioelectronics, Southeast University, Nanjing 210096, People's Republic of China

[2]Suzhou Key Laboratory of Biomedical Materials and Technology, Suzhou 215123, People's Republic of China

[3]Suzhou Nanohealth Biotech Limited Corporation, Suzhou 215123, People's Republic of China

Correspondence to: Q. Xia (E-mail: xiaq@seu.edu.cn)

ABSTRACT: In this study, fragrance microcapsules were prepared by a spray-drying method, in which the osmanthus flower fragrance acted as the core material and gum arabic and maltodextrin acted as shell materials. Scanning electron microscopy images showed that the microcapsules were approximately spherical in shape with a concave surface. Fourier transform infrared spectroscopy was used to prove the formations of the microcapsules. The fragrance retention rate at high temperatures (80–120°C) after a short heating time (30 min) reached 85.20 ± 2.72% and the retention rate after a long heating time (a week) at 60°C reached 95.40 ± 2.88%. The retention rate after 100 days exceeded 90%, and the transdermal release experiments showed that on the surface of the skin, the fragrance in the microcapsules stayed longer than in the pure fragrance oil. These results indicate that the fragrance microcapsules had an excellent aroma-reserving ability. The results of the release test proved that the transport mechanism of the fragrance microcapsules conformed to the Weibull equation. © 2013 Wiley Periodicals, Inc. J. Appl. Polym. Sci. **2014**, *131*, 40053.

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INTRODUCTION

Spices and perfume compounds are important in everyday life; they are used worldwide not only in the food industry, textiles, cosmetics and medicine but also in many scented household items. However, their disadvantages include little solubility, high volatilization, oxidation, instability under light or heat, and easy loading that distorts the scent, and these limit their use.[1,2] So, researchers are eager to find a practical way to protect fragrances and to improve their stability and processability.

It is known that controlling the volatilization rate and degradation is the key to prolonging the sensory characteristics and improving the stability of fragrance materials. Effective encapsulation is one way to deal with this. Microcapsules have been used in a variety of fields, including carbonless copying papers, adhesives, cosmetics, insecticides, drug carriers, and pharmaceutical applications.[3–5] They are used to encapsulate a selected functional material in a shell of polymer ranging in size from several nanometers to micrometers. Microcapsules perform a variety of functions, including the controlled release of functional substances and the protection of unstable materials within a particular environment.[6–8] Consequently, microencapsulation has become a mainstream technology of fragrance encapsulation to protect fragrant materials, improve their heat-resistance ability, extend their aroma-reserving time, and increase their applicability for various products.

Existing fragrance microencapsulation technologies include the molecular inclusion method, solid lipid nanoparticles method, spray-drying method, complex coacervation, interfacial polymerization, and *in situ* polymerization.[9–15] However, complex coacervated products may be inappropriate for many applications, and polymerization reactions with side reactions between the monomers could lead to an alteration of the encapsulation products. The spray-drying process is a popular one for the formation of powder products for many applications for good liquidity and instant products. Moreover, to ensure high stability and satisfying long-lasting controlled release properties, biocompatible materials are adopted to encapsulate fragrance molecules effectively without crosslinking agents.

Therefore, in this study, microcapsules were prepared by a spray-drying method with natural, nontoxic, and biocompatible shell materials. The loading capacity (% loading) and microencapsulated efficiency (% ME) were determined, and Fourier transform infrared (FTIR) spectroscopy was used to analyze the chemical structure of the fragrance microcapsules. The objective of this study was to investigate the thermal stability, aroma-reserving ability, morphology, and release characteristics of the

fragrance microcapsules. When we fit the experimental data to kinetic equations, the most appropriate one to describe the fragrance-release process was discovered. In this study, osmanthus flower fragrance was used as a core material, and blends of gum arabic and maltodextrin were applied as wall materials.

EXPERIMENTAL

Materials

The osmanthus flower fragrance was obtained from Kedao Flavors and Fragrances Co., Ltd. (Shanghai, China). Gum arabic was purchased from TICGUMS, and maltodextrin (dextrose equivalent of 20) (DE = 20) was purchased from Baolingbao Biology Co., Ltd. (Shandong, China). Anhydrous ethanol and NaCl were purchased from Sinopharm Chemical Reagent Co. (Shanghai, China). The water used for all experiments was ultrapurified water with a conductivity of 18.2 MΩ and was obtained from a MilliQ-Plus purification system (Millipore, Germany).

Preparation of the Fragrance Microcapsules

The osmanthus flower fragrance was encapsulated into the gum arabic and maltodextrin at core material weight ratios of 5, 15, and 25%. At first, a shell material solution was prepared. Gum arabic and maltodextrin, with a weight ratio of 1:1, were dissolved in water at room temperature. The solid content of the solution was 35%. Then, the desired amount of osmanthus flower fragrance was added and stirred to form an emulsified mixture at room temperature. The last step was to turn the emulsion into a microencapsulated powder by a spray-drying method. The inlet and outlet air temperatures were 180 and 90°C.[16]

% Loading and % ME

Because the microcapsules were water-soluble and the fragrance was soluble in ethanol, 50% v/v aqueous ethanol was used to dissolve the shell materials of the microcapsules and extract the fragrance. The microcapsule samples were weighed accurately and dissolved in 10 mL of 50% v/v aqueous ethanol. After 30 minutes of ultrasonic treatment, the sample suspension was filtered by syringe filters with a pore size of 0.45 μm. The filtrate was measured at an optical absorbance wavelength of 275.5 nm by ultraviolet spectrophotometry according to calibration curve spectrophotometry, and then, the weight of the fragrance in the microcapsules was obtained by calculation. Because the microcapsules could dissolve in the ethanol and the fragrance could not, the ethanol acted as the medium to separate the fragrance and the microcapsules. Some other accurately weighed microcapsule samples were extracted by ethanol for 30 min and then similarly filtered by syringe filters. The filtrate was measured at an optical absorbance wavelength of 275.5 nm, and then the weight of the fragrance at the surface of microcapsules was obtained. The % loading and % ME were obtained by the following formulas:[17]

$$\%\text{Loading} = \frac{\text{Weight of fragrance in the microapsules}}{\text{Weight of the fragrance microapsules}} \times 100 \quad (1)$$

$$\%\text{ME} = 1 - \frac{\text{Weight of fragrance at the suface of the microapsules}}{\text{Weight of fragrance in the microapsules}} \times 100 \quad (2)$$

Morphological Observation

The microcapsule samples were coated by a layer of gold with a thickness of 20 nm. Observation of its appearance was obtained by a Hitachi S-4800 scanning electron microscope (Hitachi, Japan). The particle size of the fragrance microcapsules was measured with the SmileView analysis program. More than 300 microcapsules were recorded.

FTIR Spectrum

To investigate the chemical structure of the osmanthus flower fragrance microcapsules, the pure fragrance oil, a mixture of the shell materials, the osmanthus flower fragrance microcapsules, and a physical mixture of the fragrance oil and shell materials were tested by a Nicolet 6700 FTIR spectrometer (Thermo Scientific), respectively. The fragrance microcapsules were extracted by ethanol to remove the fragrance adhered to the surface of the microcapsules at first.

Thermal Stability

Four sets of microcapsule samples were weighed accurately. Three of the samples were heated for 30 min at 80, 100, and 120°C, respectively, and one sample was not heated. The samples were dissolved in 10 mL of ultrapure water and measured for optical absorbance at a wavelength of 275.5 nm by ultraviolet spectrophotometry on the basis of the calibration curve spectrophotometry. Another set of samples were treated at 60°C for 7 days to investigate the thermal stability after a long heating time. The same quality of osmanthus flower fragrance was weighed and treated in the same way as a control group. The osmanthus flower fragrance was dissolved in 10 mL of ethanol and measured at 275.5 nm. The retention rate of the fragrance was obtained with the following formula:

$$\text{Retention rate } (\%) = \frac{\text{Absorption of the sample after heating}}{\text{Absorption of the sample before heating}} \times 100 \quad (3)$$

Aroma-Reserving Ability

The fragrance microcapsules were exposed to the air at 20 and 40°C and the % loading was measured every 15 days. The final % loading was the % loading after 15 days, the initial % loading was the % loading before 15 days, and so on. The % ME was also determined. The % loading and % ME were calculated by eqs. (1) and (2). The fragrance retention rate was calculated by the following formula:

$$\text{Fragrance retention rate}(\%) = \frac{\text{Final\% loading}}{\text{Initial\% loading}} \times 100 \quad (4)$$

Aroma-Reserving Ability against Skin

It is well known that fragrance is widely used in skin products, such as cosmetics. The purpose of this experiment was to investigate whether the microcapsules could extend the aroma-reserving time at the surface of the skin. To compare the aroma-reserving ability against the skin of the pure fragrance oil with that of the fragrance microcapsules, a transdermal release method was used as a reference. Skin of male hairless rabbits were obtained from the epidermis region. The experiment was performed with Franz diffusion cells with an effective area of 2.80 cm^2. The microcapsule solution and pure fragrance oil (1.0 mL) with a concentration of 1% were applied to the excised

Table I. Release Kinetics Models

Release kinetics model	Fitting equation
Zero-order equation	$M_t/M_\infty = at + b$
First-order equation	$\ln(1 - M_t/M_\infty) = at + b$
Higuchi equation	$M_t/M_\infty = at^{1/2} + b$
Ritger–Peppas equation	$\ln(M_t/M_\infty) = a \ln t + b$
Weibull equation	$\log[-\ln(1 - M_t/M_\infty)] = a \log t + b$

a, *b* was variables in each equation and $t^{1/2}$ is the square root of *t*.

skin samples. The reservoir chamber was filled with 6.5 mL of 0.9% w/w normal saline. The cells were maintained at 37.0 ± 0.5°C to ensure a surface skin temperature of 32°C and stirred at 300 rpm throughout the experiment. A volume of 200 μL of medium was extracted for specified time intervals, and then, the same volume of the pure medium was immediately added to the reservoir chamber. The extracted solutions were diluted and analyzed at an optical absorbance wavelength of 275.5 nm by ultraviolet spectrophotometry after filtering. The residual fragrance on the skin surface was removed for testing, and the fragrance concentrations of the inner skins were determined by the extraction of anhydrous ethanol after 24 h.

Release Behavior

The dynamic dialysis technique was used to determine the releasing characteristic of the osmanthus flower fragrance microcapsules.[18,19] Before the experiment, the normal saline was prepared as a release medium in a 37 ± 1°C water bath. A certain amount of samples of different % loadings were dissolved in 5.0 mL of normal saline and then placed into a dialysis bag dialyzed against normal saline (500 mL) at 37 ± 1°C. The control group was pure fragrance oil that was dissolved in the mixture solution of the shell materials. The suspension was put into the dialysis bag immediately to make sure the fragrance oil was still dissociated (not encapsulated into the shell materials). The magnetic stirring speed was set to 100 rpm. Dialysate (5.0 mL) was regularly extracted by a pipette, and we immediately added 5.0 mL of normal saline. The absorbance was determined at 275.5 nm by a UV spectrophotometer. The transport mechanism was studied by the fitting of the experimental data to five different kinetic equations and the calculation of the corresponding parameters, as shown in Table I.[20–22] In Table I, M_t is the amount of fragrance released at time *t*, and M_∞ refers to the amount of fragrance released at infinite time, so M_t/M_∞ stands for the cumulative release at time *t*.

RESULTS AND DISCUSSION

Morphological Analysis

The scanning electron microscopy (SEM) images of the osmanthus flower fragrance microcapsules magnified 1000 and 3000 times are shown in Figure 1(A,B). By observation, the microcapsules were approximate spheres; the irregular surface morphology was concave. The concave was probably caused by the high temperature during spray drying. Smooth-faced microcapsules provide better protection for fragrance materials than concave-faced ones.[23] This suggested that the appropriate reduction of the temperature may have been favorable for the formation of smooth-faced microcapsules. In addition, it could not be said that the fragrance was totally encapsulated by the shell materials. The fragrance molecules might have adhered to the surface of the shell materials or been inlaid in them. We could not determine from the figure whether all of the fragrance was encapsulated in the microcapsules.

The particle size of the fragrance microcapsules varied from 3.6 to 13.4 μm. The average size of the microcapusles was 7.34 ± 2.41 μm. It is necessary to point out that there were small openings in the shell materials, and the fragrance might have permeated through those small openings and then contacted the air and volatilized. The thickness of the shell materials at some point was approximately 629 nm, as shown in Figure 1(C).

FTIR Spectral Analysis

Generally, FTIR spectroscopy is used to characterize microcapsules structurally because it is possible to prove the existence of materials in microcapsules.[24] The FTIR spectra of the pure fragrance oil, mixture of the shell materials, osmanthus flower fragrance microcapsules, and physical mixture of the fragrance oil and shell materials from this study are given in Figure 2. If molecule groups, such as —COOH, —COOR, and C=O, exist in a polymer, their absorption peaks will appear at near 1700 cm^{-1} because of the stretching vibrations of chemical bonds, and some variations on the spectrum shape, position, and intensity will provide some evidence on whether a foreign molecule group enters into a vacancy or their interactions.

The main composition of the osmanthus flower fragrance includes linalool, citral, benzyl acetate, and some other organic

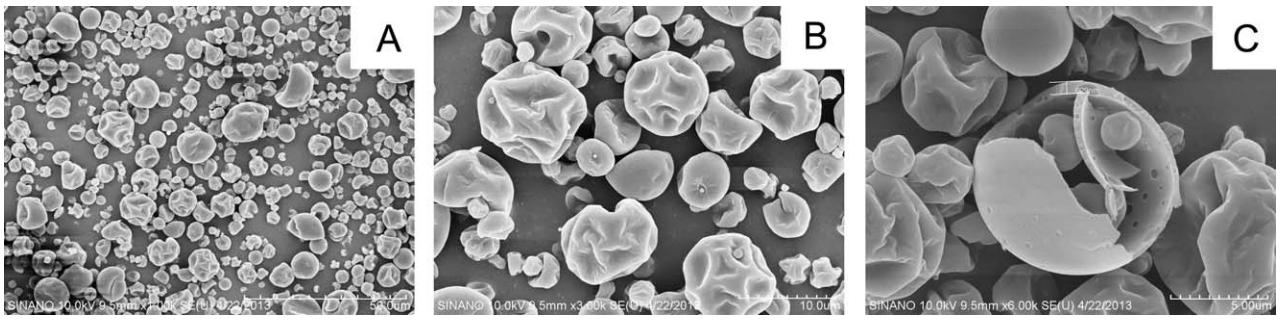

Figure 1. SEM images of the fragrance microcapsules coated by a layer of gold with a thickness of 20 nm: (A) 1000, (B) 3000, and (C) 6000×.

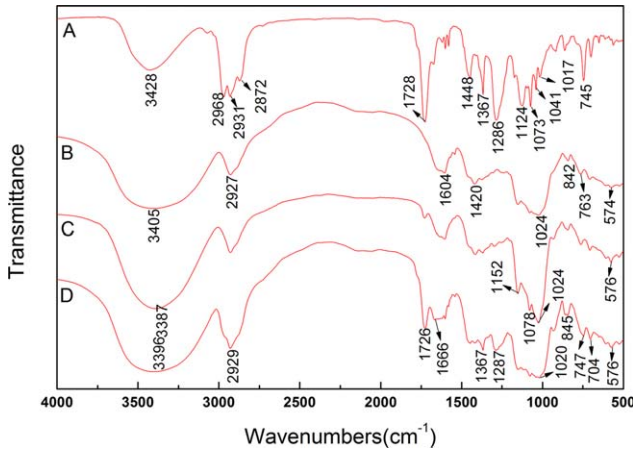

Figure 2. FTIR spectra of the (A) osmanthus flower fragrance, (B) mixture of shell materials, (C) osmanthus flower fragrance microcapsules, and (D) physical mixture of osmanthus flower fragrance and shell materials. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

molecules, which consist of molecule groups of —COOH, —COOR, and C═O. Their peaks due to strong stretching vibrations could be identified at 1728 cm^{-1}, as shown in Figure 2(A). In the infrared spectra of the fragrance microcapsules, the peak at 1726 cm^{-1} decreased significantly, and it was also available in the spectra of physical mixture. We deduced that the shell materials did successfully packet the fragrance oil and the fragrance microcapsules were formed. The peaks at 1024 and 574 cm^{-1} were present in the spectra of both the shell materials and the microcapsules. This revealed the presence of the shell materials in the structure of the microcapsules. The peaks at 1448 and 1367 cm^{-1} represented the deformation vibration of the —CH_2 and —CH_3 groups. These peaks showed a variation on the spectrum shape in the spectra of the fragrance microcapsules; this also proved the formation of the microcapsules.

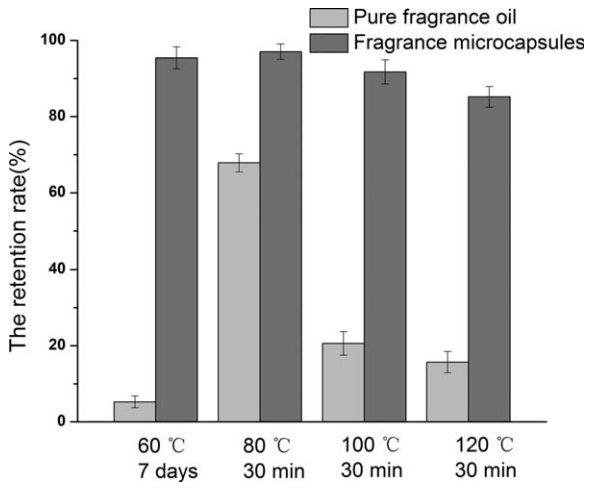

Figure 3. Retention rates of the osmanthus flower fragrance and fragrance microcapsules after they were heated at different temperatures for different times ($n = 3$; n is the test times).

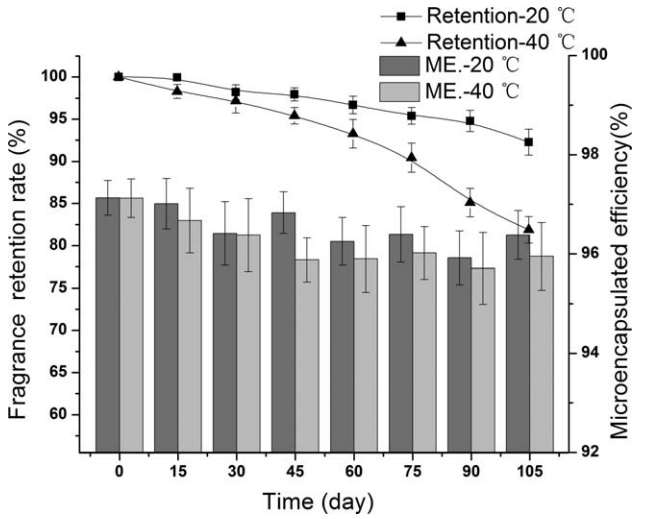

Figure 4. Plot of the fragrance retention rate and % ME versus time for 105 days at 20 and 40°C ($n = 3$).

Thermal Stability

As shown in Figure 3, the retention rate of the osmanthus flower fragrance decreased when the temperature increased. The retention rates of the microcapsules after 30 min were 97.04 ± 1.01, 91.75 ± 1.15, and 85.20 ± 2.72% at 80, 100, and 120°C, respectively, and greatly increased compared with that of the pure fragrance oil. The retention rate of the microcapsules after 7 days at 60°C was 95.40 ± 2.88% and was almost 18 times that of the pure fragrance oil; thus, the fragrance microcapsules had better thermal stability. This indicated that the encapsulation enhanced the heat resistance and provided good protection for the fragrance materials. This advantage would also guarantee easy storage for certain products.

Aroma-Reserving Ability

The fragrance retention rate of the microcapsules after 105 days decreased to 92.28 ± 1.53% at 20°C; this was obviously higher than the value of 81.88 ± 1.56% at 40°C, as shown in Figure 4. The % ME reduced within 1.5% when the time was changed at either 40 or 20°C. In addition, the % ME at 20°C was higher than that at 40°C. Bangs and Reineccius[25] pointed out that the encapsulated efficiency depended on the DE value of

Table II. Cumulative Permeation Qualities at Different Times at 37.0 ± 0.5°C

	Cumulative permeation quality ($\mu g/cm^2$)[a]	
Time (h)	Pure fragrance	Microcapsules
1	48.813 ± 6.982	312.965 ± 24.467
2	84.638 ± 18.356	328.322 ± 17.835
4	94.875 ± 13.336	430.698 ± 20.346
6	245.888 ± 19.569	466.530 ± 18.898
8	599.088 ± 31.857	476.768 ± 32.766
12	1098.163 ± 29.963	1055.196 ± 23.351
24	1514.075 ± 40.845	2299.071 ± 38.365

[a] $n = 3$, mean plus or minus the standard deviation.

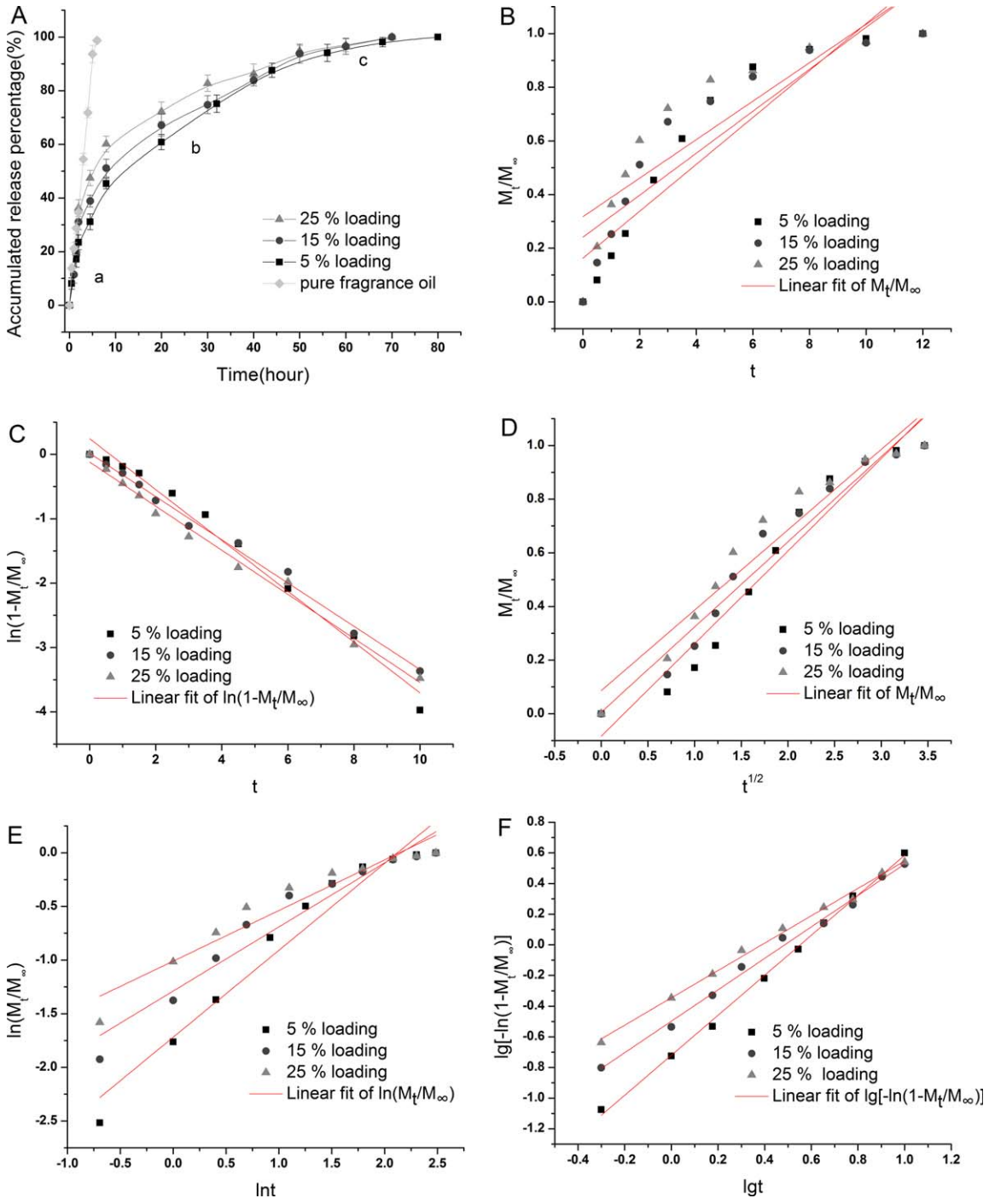

Figure 5. (A) Release curves of the osmanthus flower fragrance microcapsules with different % loadings and the pure fragrance oil ($n = 3$) and plot of the release dynamics of the osmanthus flower fragrance microcapsules in 0.9% normal saline at 37°C ± 1°C: (B) M_t/M_∞ versus time, (C) $\ln(1 - M_t/M_\infty)$ versus time, (D) M_t/M_∞ versus $t^{1/2}$, (E) $\ln(M_t/M_\infty)$ versus ln t, and (F) $\log[-\ln(1 - M_t/M_\infty)]$ versus lg t; $t^{1/2}$ is the square root of t. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

maltodextrin. The % ME decreased when the DE value increased. With a maltodextrin DE value of 10, the % ME was the highest. However, the retention rate of the fragrance rose as the DE value increased. The DE value of the maltodextrin used in this study was 20; thus, the retention rate of the fragrance maintained a high value during the process of storage.

The fragrance retention rate of the microcapsules after 6 months was still 71.53 ± 3.26% at room temperature, whereas the % ME did not change much. We speculated that the fragrance in the microcapsules gradually permeated through the shell materials and the fragrance in the surface of the microcapsules was volatilized sequentially so that the % ME did not

Table III. Relation Coefficient of the Cumulative Release Fitting Equations of the Osmanthus Flower Fragrance Microcapsules in 0.9% Normal Saline at 37°C ± 1°C

	R^2 [a]		
Release kinetics model	5% loading	15% loading	25% loading
Zero-order equation	0.84460 ± 0.01782	0.80753 ± 0.02447	0.73490 ± 0.03030
First-order equation	0.95820 ± 0.04867	0.97699 ± 0.02883	0.98349 ± 0.00561
Higuchi equation	0.94584 ± 0.00280	0.95295 ± 0.00791	0.92855 ± 0.01410
Ritger–Peppas equation	0.94649 ± 0.00385	0.933298 ± 0.00389	0.91751 ± 0.00096
Weibull equation	0.99300 ± 0.00851	0.98774 ± 0.00760	0.99189 ± 0.00275

[a] $n = 3$, mean plus or minus the standard deviation.

change significantly according to the formula of % ME. The results reveal that the fragrance microcapsules had a good aroma-reserving ability.

Aroma-Reserving Ability against Skin

The cumulative permeation quality at different times of the microcapsules and control samples (pure fragrance oil) are given in Table II. The total permeation quality of the microcapsules after 24 h was 4.5982 mg, and that of the pure fragrance oil was 3.0282 mg. The qualities of fragrance of the inner skins were 2.9179 and 6.3195 mg. The qualities of the residual fragrance on the skin surface of both samples were 1.8278 and 0.3431 mg. The fragrance (microcapsule group) concentration at the surface of the skins was 19.56 ± 0.17%, and for the pure fragrance oil group, the value was 3.54 ± 0.12%. This indicated that the fragrance in the microcapsules could remain at the surface of the skins more easily than that of the pure fragrance oil. More fragrance stayed on the inner skins than in the microcapsules, and the reason might attribute to the lipophilicity of the fragrance oil. The results of this experiment exhibited that fragrance microcapsules had a good ability to reserve aroma against skin. This may provide the possibility of application in skin care products.

Release Behavior

As shown in Figure 5(A), the osmanthus flower fragrance in the microcapsules released quickly in 5 h (section a) and then began to slow down during the 5th to 50th h (section b). The release was almost at equilibrium after 50 h (section c). This shown an initially rapid release phase followed by a slow release phase. A comparison of the three samples of different % loadings showed that the higher the % loading was, the faster the fragrance was released over 40 h, and the release rates of three samples were basically the same after 50 h. The pure fragrance oil was released very quickly, and the release was in equilibrium at the 6th h. The release rate of the fragrance in the products was dependent on its volatility and the resistance during the transmission to the air. The shell materials increased the resistance of this process, so the fragrance in the microcapsules was released more slowly than the pure fragrance oil. The release test also proved that the fragrance microcapsules had a good sustained release ability.

Pena et al.[26] showed that perfume materials can release continuously for more than 144 h (6 days). Although in this test, the fragrance could release for a week, the cumulative release rate at 60 h was set as 100% to conveniently investigate the transport mechanism. The data of the cumulative release rate of the samples at different times were fitted by a zero-order equation, first-order equation, Higuchi equation, Ritger–Peppas equation, and Weibull equation; these are given in Table I. The results are shown in Figure 5(B–F).

The linear fitting equations are shown in Table III, which shows the relation coefficient (R^2) of five kinds of release curves. The release of the microcapsules was found to fit well with a linear relation between $\log[-\ln(1 - M_t/M_\infty)]$ and $\log t$. Thus, the best fitting equation was the Weibull equation. Kosmidis et al.[27] showed that the Weibull function was the most appropriate one to describe the entire duration of the drug-release process.[27] The result of this test was consistent with this theory.

CONCLUSIONS

We successfully demonstrated that a blend of gum arabic and maltodextrin could be used to the encapsulate osmanthus flower fragrance by a spray-drying method. The fragrance microcapsules were approximately spherical in shape with a concave surface. FTIR spectroscopy was used to prove the formation of the microcapsules. By analysis of % loading and % ME, the fragrance retention rate at a high temperature (80–120°C) after a short heating time (30 min) reached 85%. Furthermore, the retention rate after a long heating time (a week) at 60°C reached 95%; this was almost 18 times greater than that of the pure fragrance oil. This indicated that the encapsulation could enhance heat resistance and provide good protection for fragrance materials. The fragrance retention rate after 100 days exceeded 90% and after 6 months was still 71% at room temperature; this exhibited a good ability for extending the time of aroma reservation. The results of the transdermal release experiments also proved that the fragrance in the microcapsules could stay longer at the surface of the skin than in the pure fragrance oil. This indicated that the fragrance microcapsules could be added to certain cosmetics for its excellent aroma-reserving ability against skin. The results of the release test showed that the transport mechanism of the fragrance microcapsules followed the Weibull equation, which was the most appropriate equation for describing the entire release process.

This study proved that the natural, nontoxic, and biocompatible fragrance microcapsules with gum arabic and maltodextrin as

shell materials improved the heat-resistance ability and showed satisfying long-lasting controlled release properties. The fragrance microcapsules introduced in this study may be appropriate for the food industry, textiles, cosmetics, medicine, and other applications. They could provide a solid foundation for the industrial applications of fragrance microcapsules.

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