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Preparation and Properties of Sandalwood Essential Oil Microcapsules in Detergents

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Sandalwood essential oil can be used to flavour and perfume detergents and cleansers, but it is volatile and unstable. In this experiment, the molecular inclusion-complex method using the β-cyclodextrin as the wall material, was adopted to encapsulate the sandalwood essential oil, and prepare the sandalwood essential oil microcapsules. Then, it studies the preparation process of sandalwood essential oil microcapsules, and explores the factors such as core-wall ratio, stirring speed, and stirring time etc. affecting the morphology of the fragrant microcapsules. The experimental results show that when the core-wall ratio is 1:16 (mass ratio), the stirring speed is 250 r/min, and the stirring time is 4 h, the sandalwood essential oil microcapsule with spherical structure and ratio of core material 10.02 % can be prepared. With good sustained release property and thermal stability, sandalwood essential oil microcapsule has an important value for additives of detergents and cleansers.

1. Introduction

Santalum album is a valuable Chinese herbal medicine belonging to the sandalwood evergreen tree. The sandalwood essential oil (SEO) is prepared from the sandalwood heartwood by distillation and other extraction methods. The essential oil of sandalwood is liquid, and the main components are α-santalol and β-santalol (Wedding et al., 2006). Thanks to the properties of anti-oxidation (Scartezzini and Speroni, 2000; Optasanu et al., 2016), anti-inflammatory (Feng et al., 2012), anti-skin tumour (Dwivedi and Zhang, 1999), and prevention of skin UV damage (Bommareddy et al., 2007), it is widely used in daily detergents such as soap, hand soap, shower gel and shampoo etc (Jahns et al., 2004)., taking a good health care effect and rich flavor. SEO is highly volatile, and easily oxidized and deteriorated. It is often easy to lose effect in detergents. Micro-capsulated technology is one of the most effective methods to maintain the stability and aromatic persistence of SEO. Common methods include polyurethane (PU) interfacial polymerization method which can achieve the microencapsulation of oily substances such as olive oil (Dong et al., 2009; Noor Amirah Azelan et al., 2018) and vitamins (Bouchemal et al., 2004) with PU wall materials. However, the microcapsules prepared by the above system have a lower content, and the reagents such as concentrated hydrochloric acid are used in the production process, which brings problems for subsequent processing. In this study, using β-cyclodextrin (Piletti et al., 2017) as the wall material, micron-sized SEO/β-cyclodextrin were prepared by molecular inclusion-complex method were prepared. Then, it explores the morphological structure, sustained release effect and thermal stability of the microcapsules prepared. It is intended to contribute to the functional addition and fragrance enhancement of SEO microcapsules in detergents and cleansers.

2. Experiment

2.1 Materials

Sandalwood essential oil (industrial grade, Guangzhou Dongfang Daily Chemical Co., Ltd.); β-cyclodextrin (RG, Adamas Reagent Co), absolute ethanol (AR, Tianjin Beichen Founder Reagent Factory), and ultrapure water.

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2.2 Preparation of sandalwood essential oil microcapsules

This method was determined to preparate of essential oil microcapsules detect had the modified published (Ayala-Zavala et al., 2008). The core material solution was prepared: Take 1g of SEO and add a certain amount of ethanol aqueous solution (10 %), prepare the essential oil solution and ensure that the mass-to-volume ratio of SEO and ethanol aqueous solution is 1:10. The wall material solution was prepared: weigh the quantitative β-cyclodextrin and dissolve it in an aqueous ethanol solution (50 %) under stirring at 55 °C, with the mass-to-volume ratio of the β-cyclodextrin to the aqueous ethanol solution 1: 10; then, slowly add the core material solution dropwise to the wall material solution, and adjust the mass ratio of the core material solution to the wall material solution (4/1, 1/1, 1/2, 1/4, 1/6, 1/8, 1/10, 1/12, 1/14, 1/16, 1/18, 1/20), at the temperature of 55 °C during the addition, maintaining a certain stirring speed (200 r/min, 250 r/min, 300 r/min); after the completion of the dropwise addition, turn off the heating and stir the mixture is stirred for a certain period of time (2 h, 3 h, 4 h, 5 h, 6 h), and then allow it to stand at 4 °C overnight. Finally, the precipitate is collected by filtration, and the precipitate is dried in a vacuum oven at 50 °C for 24 h. In this way, the SEO microcapsules are prepared.

2.3 Properties of sandalwood essential oil microcapsules

2.3.1 Surface morphology of sandalwood essential oil microcapsules

The morphology of the microcapsules was observed using FEI Quanta FEG 250.

2.3.2 Determination for the encapsulation rate of sandalwood essential oil microcapsules

1.00g of SEO microcapsules were placed into a round bottom bottle. They're heated on the electric heating sleeve for 15min to weigh and record the quality until the mass remained constant. Then, the encapsulation rate of SEO microcapsule was calculated (Fernandes et al., 2008).

Total oil retention (%)=(1-M_s'/M_s)%

M_s: SEO microcapsules initial mass (g)

M_s': 100 °C SEO microcapsules mass (g)

2.3.3 Sustained release property of sandalwood essential oil microcapsules

The sustained release properties (Partanen et al., 2002) of SEO / β-cyclodextrin microcapsules at different temperatures were mainly studied in this paper. The β-cyclodextrin, SEO, SEO / β-cyclodextrin microcapsules were divided into three groups and placed in different temperature environments (4 °C, 28 °C and 37 °C) for a certain time (every one hour), the mass change was weighed after the measurement, to analyse the change rule of the mass and investigate whether the microcapsules have sustained release properties.

2.3.4 Thermal stability of sandalwood essential oil microcapsules

Three groups were divided: β-cyclodextrin (experimental conditions: experimental loading amount about 10 mg, nitrogen flow rate 30 mL/min, heating rate 10 °C/min, room temperature for sampling starting temperature, final temperature controlled at 400 °C), SEO (experimental conditions: experimental sample loading about 10 mg, nitrogen flow rate 30 mL/min, heating rate 10 °C/min, room temperature for sampling starting temperature, final temperature controlled at 400 °C), SEO/β-cyclodextrin microcapsules (experimental conditions: experimental loading amount about 8 mg, nitrogen flow rate 30 mL/min, heating rate for 5 °C/min, 10 °C/min, 15 °C/min, 20 °C / min respectively, room temperature for the sampling temperature, and the final temperature as controlled at 400 °C). The thermogravimetry (TG, Netzsch, STA 449 F3) was applied to study the heat stability of the SEO microcapsules.

3. Results and discussion

3.1 Surface morphology characterization of sandalwood essential oil microcapsules

Through the adjustment of the composition and process, when the stirring temperature is 55 °C, the ratio of the core material solution to the wall material solution is 1:16 (mass ratio), the stirring speed is 250 r/min, and the stirring time is 4 h, a series of micron-sized SEO/β-cyclodextrin microcapsules can be prepared. It's found that the mass ratio of core solution to wall solution has the most obvious effect on the morphology of SEO / β-cyclodextrin microcapsules, and the SEM photograph is shown in Fig.1. It can be seen from the figure, most of the microcapsules are full round micron-sized spherical particles. However, a large number of spherical ruptures and surface depressions can be observed at the points indicated by the arrows in the figure. In particular, in Figure 1(c), the collapse of the shell layer characteristic in the microcapsule structure can be observed. This also explains from the reverse that the product produced does have a typical microcapsule structure. In addition, Figure 1 also shows that the SEO / β-cyclodextrin microcapsules are around 1-2 μm. The reason why SEO / β-cyclodextrin can form microcapsules may be related to the molecular structure of β-cyclodextrin. The molecular shape of β-cyclodextrin is a slightly conical hollow cylindrical structure (Villalonga et al., 2007) (Figure 2), and β-cyclodextrin has a hydroxyl group extending toward the outer end of the molecule, ensuring the cyclodextrin to have a hydrophilic appearance, whereas, inside the cavity, due to the existence of H atom and glycoside O atom etc. (Saenger, 1980), the interior of the cavity exhibits a non-polar and hydrophobic character, so the β-cyclodextrin can be complexed with sandalwood oil to form microcapsules. Because of the limited size of the β-cyclodextrin cavity, when the SEO is too much, part of the cavity that cannot enter the cavity will adhere to the surface of the β-cyclodextrin and be the agglomeration (Figure 3), so, the proportion of core solution and wall material solution should be adjusted. When the amount of SEO is appropriate, with the appropriate stirring speed and stirring time, the core material is encapsulated into the wall material, and the microcapsule molecules are dispersed and formed, thus preparing the desired SEO / β-cyclodextrin microcapsules.

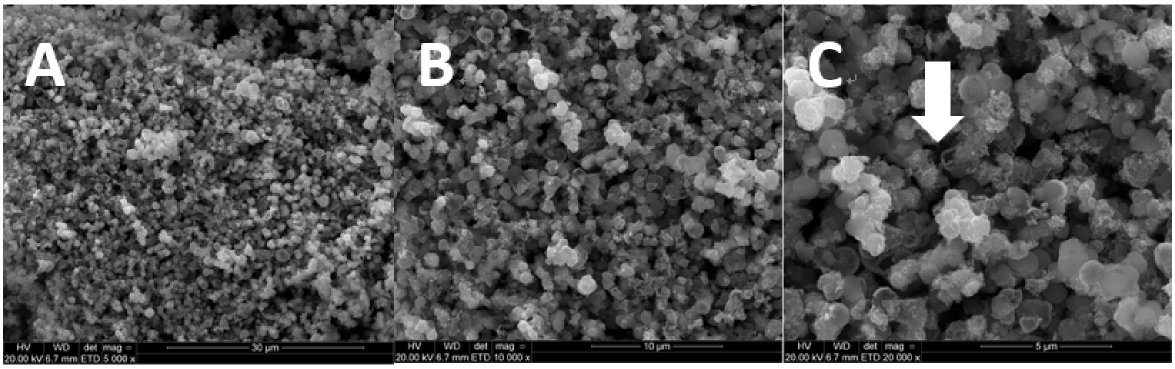

Figure 1: Typical scanning electron micrograph (SEM) of SEO / β-cyclodextrin microcapsules

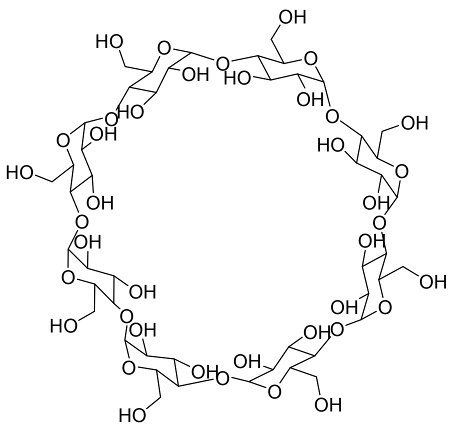

Figure 2: Schematic diagram of the β-cyclodextrin molecular structure

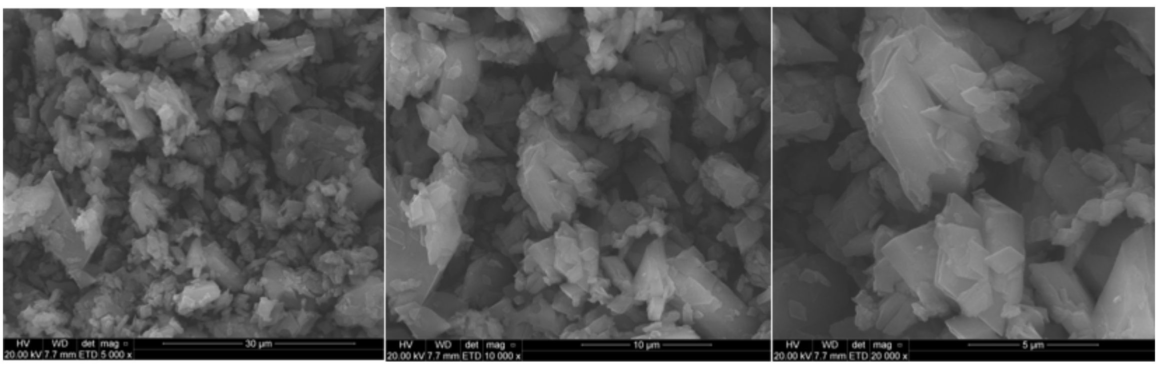

Figure 3: Scanning electron micrograph (SEM) of β-cyclodextrin agglomeration

3.2 Determination for the encapsulation rate of sandalwood essential oil microcapsules

When the stirring temperature is 55 °C, the ratio of the core material solution to the wall material solution is 1:16 (mass ratio), the stirring speed is 250 r/min, and the stirring time is 4 h, the experiment was conducted according to the determination method for the encapsulation rate of SEO microcapsules. It has been found through experiments that the mass of β-cyclodextrin changed at 100 °C. This is the weight loss of the blank wall material before 100 °C, which is negligible. Finally, the total weight loss rate of β-cyclodextrin was 85.17 % at 220°C, and the weight loss rate of SEO microcapsules was 95.19 %. The difference between the two was the encapsulation rate of SEO, which was 10.02 %.

3.2 Sustained release property of sandalwood essential oil microcapsules

It can be seen from the Figure 4 that the mass changes of SEO and SEO / β-cyclodextrin microcapsules are basically the same at the ambient temperature of 4 °C, probably because the temperature is relatively low and the essential oil itself is not volatile. When the ambient temperature reaches 28 °C, the mass change of sandalwood oil is significantly higher than that of SEO / β-cyclodextrin microcapsules, indicating that the temperature will affect the evaporation of essential oils, while the products coated with microcapsules also volatilize, but at lower rate. When the temperature reaches 37 °C, the mass change of SEO is larger than that of SEO / β-cyclodextrin microcapsules. In short, the higher the temperature, the higher the volatile oil content of sandalwood oil or microcapsules, but the sustained release of microcapsules is stronger, while β-cyclodextrin is not sensitive to temperature.

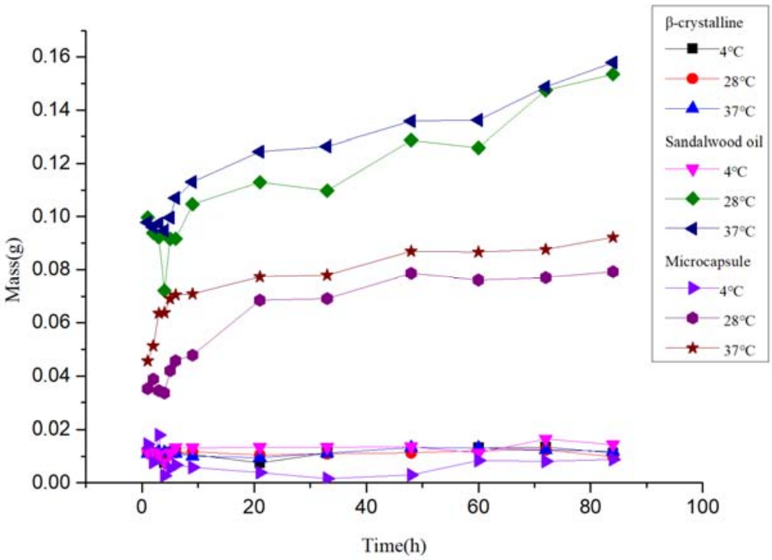

Figure 4: Slow release performance curve of SEO microcapsules at different temperatures

3.4 Thermal stability of sandalwood essential oil microcapsules

It can be seen from Figure5 that the essential oil of sandalwood begins to decompose at 77 ℃, and the second platform peak appears at 177 ℃. After that, the decomposition rate of SEO increases, which is considered to be sensitive to heat by sandalwood oil, resulting in complete decomposition. Compared with sandalwood oil, the SEO / β-cyclodextrin microcapsules have similar curves, and the first decomposition of the essential oil in the microcapsules is also at 77 ℃, but due to the encapsulation of β-cyclodextrin wall material, the decomposition process continued to 277 ℃, and there emerges the second platform, indicating that the sandalwood oil in the microcapsules continues to decompose before 277 ℃, until the β-cyclodextrin begins to decompose at 277 ℃, and the sandalwood oil is completely decomposed. The above results indicate that the thermal stability of the micro-encapsulated essential oil is much greater than that of the un-encapsulated oil. However, in the thermal decomposition process of SEO / β-cyclodextrin microcapsules, their thermal stability is also different with the change of heating rate; from 5 °C/min to 20 °C/min, as the heating rate per minute increases, the thermal stability of the microcapsules decreases, and when the heating rate decreases, the inside of the microcapsules is less affected by the temperature, and the thermal stability of the microcapsules becomes better.

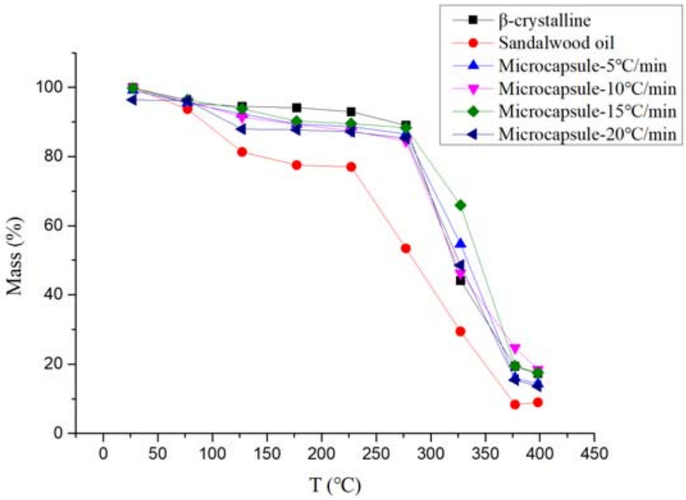

Figure 5: Thermal stability of SEO microcapsules (TG)

4. Conclusions

In this paper, using β-cyclodextrin as the wall material and SEO as the core material, the molecular inclusion-complex method was applied to obtain the SEO / β-cyclodextrin microcapsules under certain preparation conditions, and their morphology was characterized by SEM. When the stirring temperature is 55 °C, the core-to-wall ratio is 1:16 (mass ratio), the stirring speed is 250 r/min, and the stirring time is 4hr, the micro-sized spherical SEO microcapsules can be prepared, with the encapsulation rate of 10.02 %. Then, through the analysis of the experimental results, it's found that the ratio of SEO to β-cyclodextrin is a key factor in the formation of spherical microcapsules, which may be related to the structure of wall-wood sandalwood oil and β-cyclodextrin. Finally, the SEO / β-cyclodextrin microcapsules were analysed in terms of sustained release property and thermogravimetry. The test results show that the SEO capsule has excellent sustained release performance and thermal stability, which is presumably due to the wall material β-cyclodextrin acting as a barrier to encapsulate the core material and improve the sustained release property (Saenger, 2010). With good sustained release property and thermal stability, SEO microcapsule can be used to increase the flavour of detergents and cleansers.

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