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Research Progress and Prospects of Microcapsules in the Sustained-release and Lingering Fragrance Technology of Spices

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Abstract. Spice microcapsules, which encapsulate volatile compounds within protective wall materials through core–shell structures, provide enhanced stability, extended shelf life, and controlled release of active components. This review systematically explores three main preparation strategies: physical methods, such as spray-drying and freeze-drying; chemical approaches, like in-situ and interfacial polymerization; and physicochemical techniques, including complex coacervation and molecular embedding. The paper further investigates the release mechanisms of encapsulated spices, emphasizing diffusion-driven release, stimulus-responsive behavior, and wall-material degradation as the primary pathways. Applications in textiles, food, and tobacco illustrate the potential of spice microcapsules to enhance product performance. Current challenges include limited process scalability, environmental sensitivity, and cost constraints. Future development focuses on designing environmentally friendly wall materials, optimizing low-energy consumption processes, and integrating multifunctional properties into microcapsules. These efforts aim to improve encapsulation efficiency, functional stability, and controlled release, providing a practical framework for industrial-scale implementation and high-value applications.

Keywords: Spice microcapsules, Sustained-release technology, Long-lasting fragrance, Smart response, Green siding, Multi-functional integration

1. Introduction

Spices are essential functional components in everyday life and are widely used in food, cosmetics, textiles, and household products. They provide unique flavors and aromas and enhance consumer experience. However, traditional spices have some problems. They have low molecular weight, high volatility, limited polarity, and heat sensitivity. These properties cause rapid loss and low utilization efficiency. They also make the spices vulnerable to environmental factors. This limits their functional longevity and performance in practical applications, especially in textiles, food, and tobacco. To solve these problems, spice microencapsulation technology has been developed. This technology embeds spices within polymeric wall materials to create a core–shell microcapsule

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structure. This protects the active compounds from adverse external conditions such as light, heat, oxygen, and humidity.

In addition, microcapsule technology allows controlled release and stimulus-responsive behavior of the encapsulated spices. The preparation of these microcapsules can be done using physical methods (e.g., spray-drying, freeze-drying), chemical approaches (e.g., in-situ and interfacial polymerization), or physicochemical techniques (e.g., complex coacervation, molecular embedding). This makes it possible to design them precisely for specific application needs.

In recent years, microcapsule technology for fragrances has made significant progress in traditional fields such as textiles, food, and tobacco. Lavender microcapsules constructed by Zhang with multi-layer wall materials have shown excellent washability and sustained-release performance on cotton fabrics [1]. In the food industry, Liu et al. Effectively slowed down the release rate of flavorings by using Pickering emulsion microcapsules with high Encapsulation Efficiency (EE%) [2]. In the tobacco industry, the microcapsule technology studied by Zhang et al. [3] not only enhances the persistence of aroma, but also has the dual functions of antibacterial and improving the taste quality and meets the tobacco additive limit requirements [3-7]. In addition, the technology also shows broad application prospects in emerging fields such as leather, paper, and medicine.

This article mainly reviews the structural principles, preparation methods of flavor microcapsules and their application progress in textile, food, tobacco and other fields, and discusses future development directions.

2. Basic structural principles and preparation methods of flavor microcapsules

2.1. Basic structure and principles of microcapsules

Among the many ways spices are preserved, microcapsule technology is a core-shell structure consisting of a wall material, the shell material, which covers the outside and a core material which encloses the inside. Spice microcapsules are composed of a spice core material and a polymer wall material. The wall material forms a film at the interface to form a core-shell structure, which enhances the stability of the spice under light, heat, oxygen and moisture conditions, and enables the controlled release of the spice through the permeability of the wall material or external stimuli [8-11].

The way micro-nano capsules are made can be divided into three main types. This is based on how the wall materials form a film. The three types are physical methods, chemical methods, and physicochemical methods [11].

The main release mechanisms of flavor microcapsules can be grouped into several types. The first type is diffusion-driven sustained release. In this type, the encapsulated compounds move through the micropores of the wall material due to concentration differences. The second type is stimulus-responsive controlled release. Here, environmental factors such as temperature, pH, light, or enzymatic activity cause changes in wall permeability or phase. This triggers the release of the core material. The third mechanism involves release mediated by degradation or dissolution. In this case, the encapsulated substance is released due to chemical breakdown of the wall material or mechanical disruption. Additionally, multi-mechanism coupled release combines diffusion and degradation processes. This achieves synergistic control over the release profile.

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2.2. Main preparation methods

2.2.1. Physical methods

Physical preparation methods mainly include spray drying, freeze-drying and vacuum drying, etc. Spray drying is one of the most commonly used microencapsulation techniques, which works by atomizing the precursor into droplets and drying them instantaneously into powder in a hot air field, suitable for heat-stable or moderately heat-sensitive formulations [6]. Tina J et al.(2021) by atomizing a suspension containing insoluble calcium salt $CaHPo_4$ with sodium alginate at pH 5.6-8 and completing instantaneous drying ina Buchi B-290 spray dryer at an inlet temperature of 150 °C, a gas flow of 35 m^3/h, and a residence time of seconds The volatile base was released, local pH dropped, calcium salts dissolved to release Ca^{2+}, and water was rapidly evaporated to produce dried microcapsules with a cross-linking degree of 33-100 % [7]. Spray drying is often used for large-scale packaging due to the advantages of mature equipment, low cost and continuous production. The advantages lie in continuous operation and cost control, but the limitations are the adverse effects of high temperatures on the activity of the spices.

For highly heat-sensitive or resilient core materials, extrusion-ion crosslinking can be used to form gel droplets, and chitosan coatings can be applied to enhance mechanical strength and gastrointestinal tolerance; The method has a high encapsulation rate, but capacity and scalability still need to be improved.

Yue et al. Using the extrusion drop method, a mixed solution of sangin gum (SG) containing Lactobacillus planteriae and sodium alginate (SA) was dropped into a calcium chloride solution to form microcapsules through ionic cross-linking, resulting in microcapsules with EE% as high as 97.66±0.79% and viable bacteria count as 9.97±0.03 log CFU/g after freeze-drying. The microcapsules were significantly superior to those of sodium alginate alone.

The microcapsules were subsequently modified via chitosan coating. The initially prepared microcapsules were immersed in a chitosan solution, where electrostatic interactions facilitated the formation of an outer protective layer. This modification increased the encapsulation efficiency (EE%) to 99.14 ± 1.46% and significantly improved resistance to heat, bile salts, and gastric acid.

In addition, Yue et al. explored a co-crosslinking strategy, wherein chitosan was directly added to the crosslinking agent solution, allowing it to participate in the crosslinking process during microcapsule formation. This approach produced microcapsules with markedly enhanced mechanical strength, with hardness reaching 587.26 ± 36.52 gf. However, the resulting microcapsules exhibited larger internal pores, providing less effective protection for encapsulated probiotics compared to the chitosan coating method [7].

2.2.2. Chemical method

Chemical preparation methods mainly rely on monomer polymerization reactions to form polymer wall materials on the surface of the fragrance droplets, including in-situ polymerization, interfacial polymerization, emulsion polymerization, etc. In-situ polymerization is the most widely used technique in chemical methods, with a typical example being the preparation process of urea-formaldehyde resin as the wall material [8].

in situ polymerization, xu et al. used methyl methacrylate (MMA) as monomer, trimethylolpropane triacrylate (TMPTA) as copolymer monomer, and cetyltrimethylammonium bromide (CTAB) as emulsifier. Poly (MMA-co-TMPTA)/ fragrance microcapsules with an EE% of

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up to 91.6%, garnet morphology, significantly improved thermal stability, and sustained fragrance release for more than 90 days were successfully prepared [9].

The method involves mixing an oil phase containing lavender essence with an aqueous phase containing emulsifier to form an O/W emulsion, which is then polymerized at 78°C for 8 hours under nitrogen protection. Compared with pure PMMA microcapsules with an EE% of only 17.4%, the introduction of TMPTA copolymer monomers can effectively increase shell density, raise the initial evaporation temperature of the fragrance from 62°C to over 116°C, and significantly improve its long-lasting fragrance retention on fabrics [9].

The compound coagulation method separates phases through electrostatic and hydrogen bonding interactions between polymers and deposits films at the interface under mild conditions, suitable for heat-sensitive fragrances, but sensitive to pH and ionic strength. Molecular embedding is often achieved through the host-guest interaction of cyclodextrin, combined with chitosan or alginate to construct multilayer wall materials, which can significantly reduce the release rate and increase the retention rate of fragrances.

Xu et al. employed a very sophisticated interfacial coordination self-assembly technique, first emulsifying citral to form an oil/water (O/W) emulsion as a template, and then coordinating with tannic acid (TA) as an organic ligand and Fe^{3+} inorganic crosslinker at the oil-water interface of the emulsion droplets. The rapid formation of stable Metal-Phenolic Network (MPN) shells leads to the construction of core-shell microcapsules (citral@TA-FeIII). Microcapsules with uniform particle size of approximately 528 nm, EE% up to 12.79%, and significant PH-responsive release such as neutral sustained-release and acidic rapided-release were successfully obtained. The 14-day retention rate of the obtained microcapsules was 53.3%, much higher than the 7.2% of the emulsion, which not only greatly enhanced the chemical stability and antioxidant properties of citral, but also demonstrated excellent antibacterial performance due to the antibacterial properties of the shell and the controlled-release characteristics of the core material. The antibacterial effect is further enhanced by the rapid release of citral in acidic conditions such as when bacterial proliferation causes a decrease in environmental pH, demonstrating the characteristics of intelligent response [10].

In-situ polymerization and interfacial polymerization can make a dense shell layer at the spice droplet's interface. This helps a lot with barrier and heat resistance. But systems with formaldehyde or isocyanate monomers are not allowed in food and medical fields. We need to watch out for residues and do safety checks. Adding multi-functional copolymer monomers can boost crosslinking. This improves encapsulation efficiency and makes the shell more compact. Interfacial coordination self-assembly methods, like metal-phenol networks, can quickly form films at room temperature. They can also release in response to pH, which is good for applications that are sensitive to the environment.

2.2.3. Physical and chemical methods

Physicochemical methods mainly include composite coagulation methods. Dorra et al. Prepared microcapsules using coacervation, a method that successfully encapsulated essential oil mixtures through steps such as emulsification, coagulation, and curing with chitosan as the wall material.

Dorra et al. The fragrance microcapsules obtained by this method have high EE%, significant sustained-release properties and excellent stability. The microcapsules are regular spherical in shape, with particle sizes ranging from 12 to 48 μm and an average diameter of about 22 μm, which can effectively reduce the evaporation of essential oils and extend the action time. The microcapsules prepared by this method still have good retention rate and aroma release effect after multiple washes

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on fabrics, which can significantly improve the durability and thermal stability of the fragrance on cotton fabrics [11].

The compound coagulation method has mild operating conditions and is broadly applicable. It offers an effective approach for the controlled release of fragrances in textiles, food, and related applications.

Zhang used a typical molecular encapsulation method. They built multi-layered microcapsules with β-cyclodextrin as the main wall material, chitosan as the second wall material, and sodium alginate as the third wall material to encapsulate lavender essential oil. This method made sustained-release aromatic microcapsules. The particle sizes were between 400 and 900 nm.

Microcapsules prepared using this method demonstrated a high encapsulation efficiency, achieving a fragrance utilization rate of 71.56%, thereby substantially improving fragrance loading. Additionally, they exhibited excellent retention performance, maintaining approximately 80% of the encapsulated fragrance after 30 days at room temperature, indicating minimal aroma loss [1].

3. Applications of micro-nano capsules of spices

3.1. Textile industry

Fragrance microcapsule technology is most widely used in the textile industry. Long-lasting aromatic textiles such as aromatic underwear, aromatic towels, and aromatic home textiles can be prepared by fixing the microcapsules of fragrances to fabrics through finishing processes.

Under Zhang's research, multilayer wall microcapsules with lavender fragrance as the core material were cationic modified and finished onto cotton fabric. After 30 days of storage at 25 ° C in the dark, the total peak area retention rate of headspace GC was approximately 80%. After 10 water wash cycles, it still retained about 20% of its effective fragrance carrier, showing excellent water wash resistance and fragrance retention. It has broad application prospects in the field of aromatic finishing of textiles [1].

The aromatic microcapsules prepared by Sheng with sodium lignosulfonate as emulsifier, isopolketone diisocyanate as wall material and mugwort essential oil as core material, when used in laundry liquid finishing fabrics, not only have good sustained-release performance, but also have a sensory score of 3.52 72 hours after washing and drying, and still reach 2.56 after 120 hours, with long-lasting fragrance, The aromatic microcapsules have an instantaneous release property after rubbing the fabric and are washable, which can replace traditional SMA emulsifiers. These aromatic microcapsules not only have mosquito repellent effects but also achieve environmentally friendly aromatherapy [12].

3.2. Food industry

In the food industry, spice microcapsule technology is mainly used to protect sensitive flavor components, mask unpleasant odors, control flavor release, etc. Microencapsulation can prevent the volatilization, oxidation and reaction loss of flavorings during food processing and storage, and improve utilization and stability.

Flavors are highly volatile in food if the temperature rises or the EE% is low, Liu et al. Pickering microcapsules with alginate-gelatin as the wall material and SiO_2 nanoparticles as the emulsifier had a cumulative release rate of about 32.5% when stored in the dark at 25 ° C for 49 days; At the same time, the lipid oxidation rate in rapeseed oil matrix was significantly lower than that in the control group, indicating that the method contributed to extending the flavor stability period of the food [2].

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Microcapsule technology is crucial in the food industry, which significantly enhances the functionality and quality of food additives in various foods by protecting active ingredients, controlling release and improving stability, and is a key supporting technology for the development of new foods [13].

3.3. The tobacco industry

In the tobacco industry, flavoring microcapsule technology is mainly used to improve cigarette smoking quality, maintain flavoring stability and develop characteristics. Before the microcapsules of flavoring are added to new cigarettes, the flavoring is volatile and oxidized, the aroma is short and the tobacco shreds are prone to mold. The cumulative release rate of the clove phenolic flavoring microcapsules prepared with chitosan/alginate wall material was lower than that of the traditional formula when stored at a constant temperature for 70 days; Higher inhibition of common moldy strains at an addition of 0.5%. Sensory evaluation showed a more delicate and harmonious taste absorption quality, but the health effects still need to be verified by independent toxicological studies [3].

3.4. Other emerging application fields

In addition to the traditional application areas mentioned above, spice microcapsules are constantly expanding into new application scenarios. In leather and paper, spice microcapsules can give products a lasting aroma [14] and increase added value [15].

In addition, in aromatic care products, sustained-release essential oil microcapsules help achieve controlled release, but applications involving health effects require evidence-based assessment and compliance review.

4. Conclusion

Spice microcapsules provide technical benefits in enhancing fragrance persistence and environmental stability. However, they face challenges such as process scale-up, compatibility of wall materials, and cost. Physical methods can produce on a large scale but are not suitable for heat-sensitive core substances. Chemical encapsulation creates dense shells. Yet, the use of monomers and organic solvents raises safety concerns. Physicochemical approaches work under mild conditions. But they often have poor batch-to-batch consistency and limited controllability.

Future research should focus on developing environmentally friendly wall materials, optimizing low-energy processes, and designing systems that respond to multiple stimuli. Xu et al. showed “on-demand release” of fragrances by making wall materials that react to specific triggers, like pH, temperature, enzymatic activity, or light. This greatly increases the possible uses in smart packaging, targeted delivery, and responsive textiles. Developing green wall materials is important. It should use renewable, biodegradable natural polymers like cyclodextrins, chitosan, alginates, and proteins, along with food-grade emulsifiers and crosslinkers, to meet sustainability and safety standards.

At the same time, setting up standard evaluation criteria is important. These criteria include encapsulation efficiency, release kinetics, and resistance to washing, heat, and light. Together with life cycle assessment and cost analysis, these frameworks will help in large-scale production and multifunctional application of spice microcapsules.

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