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Research Progress of Fragrance Retention Technology: From Physical Encapsulation to Intelligent System

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ABSTRACT
Fragrance is the core ingredient that drives the sensory experience of consumers. It is widely used in food, cosmetics, textiles and home care. However, the inherent high volatility, chemical instability and complex release behavior of fragrance molecules pose fundamental challenges to their long-term retention and performance in products. Fragrance retention technologies aim to overcome these bottlenecks by protecting aroma compounds and achieving precise and controlled release through physical or chemical means. This paper systematically reviews the complete evolution of fragrance retention technology from the first generation of physical packaging to the seventh generation of artificial intelligence driven systems. Key technological paradigms, including polymer microcapsules, stimulus response systems, dynamic covalent precursors, self-assembling nanostructures, biotechnology methods, and systems-level engineering, are discussed in depth. Based on the comprehensive analysis of representative research papers, this paper summarizes the core mechanisms, advantages and limitations of each generation of technologies, identifies current research gaps, and puts forward prospects for future development.

KEYWORDS
Spice delivery; Microcapsules; Controlled release; Stimulus response material; Sustainability; Review

1 Introduction

Spices is food, cosmetics, textiles and household care products in an integral part of, it not only is the product of the sense of smell identification, can arouse the emotional memory, shaping the brand image, more directly influence consumers' purchase decision and the use of satisfaction [1-2]. The "front," "middle," and "back notes" of a bottle of perfume, or the changes in the aroma of a washing powder during washing, drying, and wearing, are intuitive embodiments of the complex release dynamics of spices. However, fragrance molecules usually have low molecular weight (usually < 300 Da), high vapor pressure and contain chemically active functional groups such as aldehyde-ketone and ester, which make them highly susceptible to volatilization, oxidation, isomerization or degradation in reaction with other components during product storage and use, severely limiting their effectiveness and shelf life [3-4].

Therefore, the core goal of modern fragrance retention technology has evolved from a simple "locking fragrance" to a multi-dimensional and systematic engineering challenge, which can be summarized into three points: (1) protection -- physical isolation of fragrance from oxygen, water, light and other active ingredients in the formulation during a long product storage period to prevent its chemical degradation; (2) deposition - ensure that spices or its carrier in applications such as spraying on skin, clothing can efficiently, attached to the target surface evenly, to resist external force such as friction, rinse off; (3) controlled release - in the expected time, to expect the rate and the way of delivery spices, from the moment of fragrance "outbreak" to weeks or even months of "permanent stay joss-stick", and even the response to a particular event, such as friction, sweating "intelligent sensory experience" [5]. This paper aims to systematically review the research progress of fragrance retention technology, from the early physical barrier to the current complex system integrating materials science, chemical engineering, biotechnology and artificial intelligence, to construct a complete knowledge map, and to look forward to the future development direction.

2 Literature Review

2.1 Basic Science of Spice Delivery

The development of any advanced delivery system must be based on solid basic science.

Toxicological safety is the bottom line and primary prerequisite for fragrance application. The International Fragrances Association (IFRA) and research institutions have carried out systematic toxicological evaluation of common fragrances, which provides scientific basis for safe dosage. Scognamiglio et al. showed that the 90-day no observable adverse effect (NOAEL) level of phenylephrine, a common fragrance, was 0.5 g/kg; The EC3 value of the local lymph node test (LLNA) of acetocinene is 13.95%, indicating that it is a moderate allergen, and the concentration should be strictly controlled in use [6]. These data provide a key reference for evaluating the potential risk of percutaneous absorption and sensitization when designing novel delivery systems.

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Sensory assessment methodologies provide a bridge to quantify fragrance properties and connect

physicochemical properties with subjective experience. Traditional category scales (such as 0-10 points) have problems such as large individual differences and nonlinear perception when evaluating intensity. Cortez-Pereira et al. demonstrated that the Labeled Magnitude Scale (LMS), which allows the evaluator to mark the intensity on a logarithmic coordinate with semantic anchors (such as "none", "weak", "moderate", "strong"), is more sensitive and reliable than the traditional category scale [7]. This method is particularly suitable for assessing the skin retention of spices, a core sensory indicator that measures the decay of spices over time and determines the quality of products.

Quantitative chemical engineering models aim to predict macroscopic sensory performance at the molecular level, bridging the gap between theory and practice. Rodrigues, etc from a chemical engineering perspective, the system introduces the spice ternary phase diagram (PTD), is used to predict the thermodynamic behavior of different spices mixture in a specific substrate, identify formula may form the dominant area "smell" [5]. Meanwhile, the 1D/3D diffusion model developed by them can simulate the spatiotemporal distribution of spices from the product substrate, through the delivery system, and finally released into the environment, providing a powerful theoretical tool for optimizing the release kinetics.

2.2 First Generation: Physical Encapsulation -- the Cornerstone of Commercialization

Physical encapsulation is the earliest commercialization and most mature technology for fragrance retention. Its core principle is to use liquid fragrances as a "core material", wrapped in a continuous layer of polymer "shell", through the physical barrier to isolate the outside world.

Synthetic polymer microcapsules are representative of this field. Microcapsules with polymethyl methacrylate (PMMA), melamine resin, polyurea, etc. as shell materials are prepared by interfacial polymerization, in situ polymerization, etc. Encapsulation efficiency can reach 72-95%, and can provide good mechanical strength and thermal stability [1]. However, its wide application is facing serious challenges. First, there is often formaldehyde residue in the preparation process of melamine resin and other condensates, which is not in line with the trend of green and safe consumption. Second, most synthetic polymers are difficult to biodegrade in the environment, and the upcoming ban on microplastics in the European Union, which will take effect in 2025, will directly eliminate such non-biodegradable microcapsules. In addition, the study by Foroutani et al. reveals a key application problem: Many spice molecules themselves can act as plasticizers of polymers, which can significantly reduce the glass transition temperature (T_g) of PMMA after penetration into the shell, making the originally hard and dense shell soft at room temperature, leading to accidental early release of spice, which puts forward new requirements for the long-term stability design of microcapsules [8].

Natural polymer microcapsules (such as chitosan, casein, keratin, gum Arabic, etc.) have become a research hotspot for replacing synthetic polymers due to their excellent biocompatibility and potential biodegradability. However, the mechanical strength of microcapsules is usually weak, and the preparation process is mostly physical condensation, which limits the protection of the core material. Through nanocomposite enhancement strategies, such as doping TiO_2 nanoparticles into chitosan shells, the encapsulation efficiency and sustained release time can be significantly improved, providing an effective path to improve the properties of natural polymers.

Solid lipid nanoparticles (SLNS) represent another important type of physical encapsulation. Unlike polymer shells, SLNS achieve encapsulation by dissolving or dispersants of fragrances in lipid matrices (e.g., triglycerides, fatty acids) that are solid at room temperature. Cortial et al. found that the encapsulation efficiency of SLN for different spices was significantly different (66.5 to 94.9%), and it was positively correlated with the oil-water partition coefficient (log P value) of spices. This finding has important predictive value: by dozens of different log P values for complex flavor compounds, can through the analysis of the composition distribution, predict the SLNS and release behavior, the efficiency of overall envelop detected the rational design of multiple components system provides a quantitative framework [9].

Double-shell microcapsules achieve complementarity and synergy in performance by combining the characteristics of two different materials. Gu et al. developed double-shell microcapsules with polyurethane (PU) as the inner layer and silk fibroin (SF) as the outer layer [10]. PU provides excellent mechanical elasticity, while SF gives it good biocompatibility and hydrophilicity. This kind of double hull structure performance excellence: release only 22.9% within 30 days of spices, and being flexible test after 10000 times, spices retention rate is as high as 93.08%, showing application in textiles need frequent resistance to deformation of the huge potential.

2.3 Second Generation : Stimulus Response System -- Moving Towards Intelligent Release

While the first-generation technology enables constant or decelerated release based on passive diffusion, the second-generation stimulus-response system aims to achieve "on-demand release", giving spice delivery an initial "intelligence".

Temperature-responsive systems take advantage of the phase transition behavior of shell materials at a specific temperature. The most typical is poly (N - isopropyl acrylamide (PNIPAM), in its aqueous solution has a lower critical solution temperature (LCST), by copolymerization can accurately regulate LCST, achieve win min to release. Another mechanism is to utilize the glass transition temperature (T_g) of the polymer. Foroutani et al found that the T_g of P(MMA-co-EHA) copolymer can be reduced to near room temperature (5-15 ° C) due to the plasticizing effect of spices, resulting in a sharp increase in polymer chain segment mobility and a "spike" phenomenon in release rate within this temperature

range, which can be used to design the release triggered under specific climate or application scenarios [8].

The ph-responsive system mainly uses the polyelectrolyte containing ionizable groups. Chitosan is a typical cationic polysaccharide whose amino group is dissolved by protonation under acidic conditions and precipitated by protonation under neutral or alkaline conditions. It can be used to design fragrance systems that are released at a specific pH environment. In addition, based on the self-assembled "brick-mud" microcapsules, the shell is constructed by alternately depositing polyelectrolytes with opposite charges. When the environmental pH changes, the charge of one layer of polyelectrolytes is neutralized or dissolved, the whole structure disintegrates, and the rapid release is achieved.

Photothermal response systems represent an emerging direction for the incorporation of new materials. The photothermal conversion material with excellent near-infrared light absorption ability (such as PANI) doped into the microcapsule shell can convert light energy into heat energy under light, increase the local temperature of the microcapsule, and significantly accelerate the release of spices, which provides the possibility to realize remote, non-contact, space-time accurate release of spices.

The dilution response system cleverly solves the problem of long-term stability of fragrance microcapsules in water-based products such as laundry detergent and softener. The trend of product concentration requires microcapsules to be stably suspended in a high concentration of surfactant and electrolyte system for a long time without leakage. By introducing chemical bonds (such as imine bonds) into the microcapsule shell that remain stable in the presence of high concentrations of surfactant and hydrolyze after dilution, an aroma "bloom effect" can be achieved when the product is diluted.

Biological response systems are at the forefront of stimulus response design, aiming to correlate fragrance release with physiological signals in the human body. Goncalves, etc, a pioneering work to a porcine odorant binding proteins (OBP) -- a natural to combine odor molecules and transport protein, peptide (KP) combined with keratin fusion expression [11] through genetic engineering. This fusion protein (OBP:KP) can both bind specifically to fragrance molecules and be firmly anchored to keratin in skin or hair via KP. It was found that when it encountered electrolytes in sweat, OBP's conformation changed and its binding affinity for spice (dissociation constant Kd) increased 8.5-fold, meaning that it "loosened" the bound spice, thereby enabling smart release triggered by sweat. This marks a huge leap from synthetic materials to functional biological macromolecules.

2.4 Third Generation: Dynamic Covalent Proincense - Atomic Economical Chemical Release

Unlike the first two generations, which physically encapsulate spices, the "Pro-perfume" technology connects spice molecules to carrier molecules through reversible covalent bonds, breaking the chemical bonds by specific stimuli when used, releasing the original aroma compounds. This approach achieves atomic economy and avoids sensory interference caused by carrier residues. Herrmann systematically reviewed all kinds of pre-fragrance chemistry [2].

Phototrigger system is an important branch of pre-fragrance technology. Paret et al. designed an ingenious light-responsive system: they encapsulated a 2-oxoacetate type profragrance in a polyurea-polyurethane microcapsule [12]. Under UVA (UVA) irradiation, the pro-fragrance molecules undergo Norrish type II photochemical reactions, which cleave to release the target fragrant aldehyde and generate CO or CO_2 gas. The resulting gas builds up pressure inside the microcapsules, which eventually leads to the rupture of the capsules and further accelerates the release of the remaining fragrance inside the microcapsules, forming a photochemical-physical multi-cascade release effect. In addition, the light-responsive group is directly introduced into the polymer backbone and the polymer skeleton is broken by light irradiation, which can release a variety of fragrance molecules connected by covalent bonds, and realize the purpose of a carrier to deliver a variety of fragrances at the same time.

The oxidative trigger system makes use of the ubiquitous atmospheric oxygen. When the aromatic aldehydes are deposited on the fabric and exposed to the air, they will spontaneously initiate the free radical chain autooxidation reaction, gradually break and re-release the original aromatic aldehydes, which can continue to produce aroma for more than two weeks. It is especially suitable for long-term scenarios such as "fragrance after washing".

2.5 The Fourth Generation: Self-assembling Nanostructures -- Thermodynamically Driven Precise Construction

Self-assembly systems take advantage of the thermodynamic driving force of amphiphilic molecules in a specific solvent to spontaneously form thermodynamically stable ordered structures, such as micelles, vesicles, and liquid crystals. This "bottom-up" construction method is accurate, reproducible, and does not require a large amount of energy input. The work of Mamusa et al. is an outstanding representative in this field. Based on Hansen solubility parameter (HSP) theory, they rationally calculated and screened the interaction between polyethylene glycol-G-polyvinyl acetate (PEG-g-PVAc) copolymer and a series of fragrance molecules [4]. By matching the HSPS of polymers and fragrances, they found that certain combinations were able to spontaneously undergo phase separation when mixed in water, forming "liquid microcapsules" with uniform size and well-defined structure. Instead of a solid shell in the traditional sense, this microcapsule is composed of a polymer-rich viscoelastic interface layer that stabilizes the internal fragrance droplets. The whole preparation process does not require any energy-intensive homogenization or emulsification steps, which reflects the perfect combination of rational design and green manufacturing.

2.6 The Fifth Generation: Biotechnology and Genetic Engineering - Reinventing Sustainability from Its Source

Biotechnology is profoundly affecting fragrances in two ways: one is to use biocatalysis or fermentation to produce fragrances themselves, solving the sustainability problem of traditional plant extraction or chemical synthesis; The second is the design and production of bio-macromolecules with delivery functions. Taghilou's review covers biocatalysts such as esterase Est16 and lipase Lip-1420 mined by metagenomics, as well as research progress in the production of high-value spices by microbial cell factory fermentation [3].

More cutting-edge explorations, such as the aforementioned OBP: KP fusion protein reported by Goncalves et al., represent a paradigm shift from "synthetic vectors" to "bioderived delivery systems" [11]. This system is entirely composed of amino acids, theoretically biodegradable, and its delivery mechanism (binding-release) directly mimics the olfactory system of the organism, opening up a new era for the realization of highly bionic and environmentally friendly intelligent release. Despite the current challenges of yield and cost, the direction of "biofabulation, intelligent response" represented by this system is undoubtedly one of the most potential paths in the future.

2.7 Sixth Generation: Systems-level Engineering - Beyond Packaging Itself

The sixth-generation technology recognizes that excellent fragrance retention depends not only on the microcapsule itself, but also on the entire system from the product to the target surface.

In terms of surface deposition enhancement, researchers are committed to improving the adhesion efficiency of microcapsules on complex substrates such as fabrics and hair. For example, O_2/N_2 plasma treatment of cotton fabric can introduce polar functional groups on its surface, which can significantly enhance the electrostatic adsorption and hydrogen bonding interaction with positively charged microcapsules. Gunay et al. used phage display technology to screen short peptide sequences with specific high affinity to cotton fibers from billions of random peptide libraries [13]. These peptides can be modified to the surface of polymer microcapsules, which can increase the deposition of microcapsules on cotton fabric by 2-3 times, and achieve efficient bioinspired targeted deposition.

Device level innovations extend the concept of controlled release from the material level to the product level. For example, the design of a double-membrane air freshener, through the two layers of membrane structure with different mass transfer resistance, can realize the spice release rate to be kept constant within a certain range of air flow rates, solving the pain point of traditional products with sharp fluctuations in aroma concentration under different ventilation environments. Ionic liquid systems provide another option for encapsulation. Berton et al. used triethanolammonium oleate, an ionic liquid recognized as Generally Considered safe (GRAS) by the US Food and Drug Administration (FDA), as a non-volatile solvent for the fragrance [14]. The strong hydrogen bond between the ionic liquid and the flavor molecules effectively reduces the gas phase partial pressure of the flavor in the headspace, thereby delaying its volatilization rate. This method does not require encapsulation, which provides a new idea for the development of new liquid fragrance products with transparent ingredients and easy to mix.

2.8 Seventh Generation : Quantitative Prediction and Artificial Intelligence - The Data-driven Future

Data-driven and artificial intelligence are reshaping fragrance science, transforming experience-led formulation development into precision engineering based on model predictions. The groundwork for this has been laid by the work of Rodrigues et al., including the establishment of a spice ternary phase diagram (PTD), the construction of diffusion models, and the development of a "spice radar map" for visually comparing the properties of different flavors [5]. The correlation of log P encapsulation rate established by Cortial et al is an early simple but effective quantitative prediction model [9].

At present, artificial intelligence technology is accelerating this process. AI platforms such as Phylira, developed by IBM, and Carto, launched by Givaudan, are able to learn complex nonlinear relationships between molecular structure and aroma attributes such as intensity, persistence, and emotional association, based on vast molecular databases and perceptual models. These tools can not only quickly predict the sensory properties of new fragrances, but also automatically recommend and optimize recipe combinations considering multiple constraints such as stability, safety, and cost. It is expected to shorten the research and development cycle that traditionally takes months or even years to weeks, and explore new aroma combinations that go beyond human experience.

2.9 Regulatory Framework and Sustainability - Hard Constraints and Driving Forces for Development

Regulations and sustainability requirements have become the core forces driving technological iterations. The upcoming microplastics ban in the European Union (which aims to phase out intentionally added non-biodegradable microplastics by 2025) will have a profound impact on the entire industry, forcing industry to accelerate the transition from traditional materials such as PMMA, melamine resin to biodegradable materials such as cellulose, chitosan, polyhydroxyfatty acid ester (PHA) [2-3]. The study by Karagianni et al. timely points out the technical challenges that come with this transition: when testing novel biodegradable microcapsules for degradation, sample preparation methods (such as the degree of purification) can significantly affect the test results [15]. Free fragrances or surfactants remaining on the surface of microcapsules may be preferentially utilized by microorganisms, leading to false positive biodegradability.

Therefore, it is essential to establish strict and standardized test methods. In addition, modern analytical techniques, such as liquid chromatography-mass spectrometry (LC-MS), provide a powerful tool to assess the environmental fate and ecological risk of complex fragrance ingredients by tracing their changes through the product life cycle.

3 Comprehensive Comparison of Technological Evolution

In order to more clearly show the evolution of incense retention technology in recent decades, the following table systematically summarizes the core mechanism, representative literature and key characteristics of each generation of technology.

Table 1 Comprehensive comparison chart of technology evolution

Intergen - erational	Core mechanics	Representative literature	Key features and core contributions
First generation	Physical barriers with diffusion control	8,9,10 [1]	It is the most mature industrially, with high encapsulation efficiency (65-95%), but faces challenges of biodegradability and formaldehyde residue.
Second generation	Environmental stimuli (T, pH, light, dilution, biological signals) trigger release	[8] 11	Realizing intelligent on-demand release, multiple trigger mechanism extends from physical and chemical to biological signals, and significantly improves the user experience.
Third generation	Reversible breaking and generation of dynamic covalent bonds	[2, 12]	Atomic economical delivery, precise release of multi-component fragrances, and ultra-long-acting (e.g., two weeks) release through chemical bonding design.
The fourth generation	Thermodynamic self-assembly of amphiphilic molecules	[4]	It has precise structure, rational design based on HSP theory, and low energy consumption during the preparation process, which represents the direction of green manufacturing.
The fifth generation	Biosynthesis and biological macromolecular functions	[3] 11	Addressing sustainability issues at the source, introducing bioactive materials such as OBP with natural binding/release functions.
The Sixth generation	System engineering optimization across scale and whole process	[13, 14]	Go beyond a single material to optimize the whole process of deposition, stability, device design, and pay attention to solving practical application pain points.
Seventh generation	Quantitative models with AI data driven	[5, 9]	To establish the correlation model between molecular properties and macroscopic performance, and use AI to accelerate formulation development and performance prediction, and shorten the research and development cycle.

As can be seen from the above table, the evolution of fragrance retention technology is a process from simple to complex, from passive to active, from single discipline to multi-discipline deep integration. Early technologies (generations 1 to 3) focused on building and controlling the "release" itself; While more recent technologies (generations 4-7) focus more on "how to build smarter" (self-assembly, biosynthesis), "how to function better" (systems engineering), and "how to discover and optimize faster" (AI).

4 Conclusions and Prospects

This paper systematically reviews the evolution of seven generations of incense retention technology, and the main conclusions are as follows:

The first-generation physical encapsulation is still the most commercially mature method. Synthetic polymer microcapsules have high encapsulation efficiency but face severe biodegradability challenges. Natural polymer systems have good biocompatibility, and nanocomposite reinforcement is an effective way to improve their biodegradability. The solid lipid nanoparticles achieved the preliminary predictive encapsulation based on log P. The double-shell microcapsules showed excellent mechanical stability.

The second-generation stimulus-response system enables intelligent release triggered by temperature, pH, light, dilution, and sweat. Key advances include sweat triggered OBP fusion protein (Kd increased by 8.5-fold), photothermal converted PANI (release increased by 62%) and dilution triggered "bloom effect".

The third generation dynamic covalent precursor achieves atom-economical delivery. Phototriggered 2-oxoacetate can release both fragrance and gas at the same time, resulting in capsule rupture. Oxidation-triggered divinyl ether can prolong the release to more than two weeks, which provides a new idea for ultra-long-term fragrance retention.

Based on Hansen solubility parameter (HSP) and other theories, the fourth-generation self-assembling nanostructures achieve rational design, and the liquid microcapsules can be formed spontaneously, which is green and energy-saving.

The fifth generation of biotechnology addresses sustainability issues from the source. OBP:KP fusion proteins represent a paradigm shift from synthetic carriers to biologically derived delivery systems.

The sixth generation of systems-level engineering optimized the whole process: phage display screened peptides for enhanced deposition, dual-membrane devices for stable release, and ionic liquids for controlled release through hydrogen bonding.

The seventh generation of quantitative prediction and artificial intelligence have established the log P-encapsulation

rate correlation, HSP prediction framework, etc. AI is expected to greatly accelerate formulation development and performance prediction, and open a new era of data-driven.

Despite the remarkable progress, there are still some gaps in current research: (1) insufficient system integration: the development of collaborative delivery systems for multiple stimuli (such as simultaneous response to friction and sweat) is limited; (2) weak perception-physical-chemical correlation: there are still insufficient studies on the correlation between release kinetics data and sensory experiences (e.g., intensity changes over time, aroma reduction degree), and there is a lack of reliable models to predict perception; (3) The scalability challenge of biotechnology: the stable, low-cost large-scale production of high-performance biomaterials such as OBPs is still the bottleneck. (4) Incomplete understanding of environmental fate: the long-term environmental behavior and ecotoxicology of novel biodegradable carriers and their degradation products need to be further understood. (5) Lack of standardized classification: there is still a lack of unified description of the release mechanism between academia and industry, and there is a lack of a standard classification framework that integrates physical, chemical and biological mechanisms.

Based on the above analysis, future research directions should be prioritized: (1) design of perceptually oriented intelligent systems: in-depth study of the release kinetics of natural flavors (such as pyramid structure), and development of artificial delivery systems that can reproduce or enhance this complex perceptual experience to achieve true "bionic release". (2) Promoting biomanufacture engineering: combining synthetic biology and fermentation engineering to overcome the low-cost, large-scale production technology of functional proteins such as OBP and biodegradable carrier materials such as PHA. (3) Establishment of an environmental safety database: for new bio-based and biodegradable materials, a full life cycle assessment should be carried out to establish a database of their fate and ecological impact in different environmental media such as soil and water. (4) Construction of a multi-scale unified release model: molecular thermodynamics, material mass transfer, and sensory science were integrated to create a unified prediction model that could simultaneously consider molecular structure, carrier design, environmental factors (temperature, humidity, and mechanical forces), and the final sensory output. (5) Validate AI-driven formulation innovation: Actively put AI-generated formulations into actual sensory testing and product applications, and verify and iterate algorithms through rigorous consumer feedback to ensure their real-worl deffectiveness.

In summary, the scent preserving technology has evolved from a simple problem of solving "volatilification too fast" to a complex system that integrates materials science, chemistry, biology, engineering, and data science. Continuous interdisciplinary collaboration and source innovation will ultimately drive us to develop the next generation of smart fragrance systems that can simulate or even exceed the natural olfactory experience, creating a richer, lasting and sustainable sensory world for humans.

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