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Review

Microencapsulation as a Route for Obtaining Encapsulated Flavors and Fragrances

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Abstract: Microencapsulation methods for active substances, such as fragrance compounds and aromas, have long been of interest to researchers. Fragrance compositions and aromas are added to cosmetics, household, and food products. This is often because the choice of a particular product is dictated by its fragrance. Fragrance compositions and aromas are, therefore, a very important part of the composition of these items. During production, when a fragrance composition or aroma is introduced into a system, unfavorable conditions often exist. High temperatures and strong mixing have a detrimental effect on some fragrance compounds. The environments of selected products, such as high- or low-pH surfactants, all affect the fragrance, often destructively. The simple storage of fragrances where they are exposed to light, oxygen, or heat also has an adverse effect. The solution to most of these problems may be the encapsulation process, namely surrounding small fragrance droplets with an inert coating that protects them from the external environment, whether during storage, transport or application, until they are in the right conditions to release the fragrance. The aim of this article was to present the possible, available and most commonly used methods for obtaining encapsulated fragrances and aromas, which can then be used in various industries. In addition, the advantages and disadvantages of each method were pointed out, so that the selection of the appropriate technology for the production of encapsulated fragrances and aromas will be simpler.

Keywords: microencapsulation; fragrances; flavors; encapsulation technology

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1. Introduction

Grain, bird eggs, or cells in membranes are examples of encapsulation found in nature, which protect the inside of the material, lengthen and facilitate the process of storage or transportation, and finally, protects the contents from the external environment. The first market application of encapsulation was reported in 1957 [1–3]. Since then, there has been a continuous development of knowledge and an increase in the use of encapsulation in many different industries, such as agriculture (pesticides), dietary supplements (vitamins and fish oil), food (flavorings, essential oils, lipids and dyes) and cosmetics (textiles and the fragrance industry) [4,5]. There are many different encapsulation techniques. Choosing the right technique will depend on several factors, including the size of the encapsulates, the chemical structure of the coating, its biodegradability, availability, and price, the final application, and most importantly, the core material that will be encapsulated. Fragrance compositions and flavors are mixtures of organic fragrance compounds, which can include naturally occurring compounds, such as essential oils or resins, that are synthetically produced, but have an equivalent in nature. They are created by several thousand chemical compounds, belonging to different types of organic compound classes, including alcohols, hydrocarbons, esters, aldehydes, ketones, lactones, terpenes, and others, as well as artificial compounds that are unidentifiable in nature, such as musk fragrances [6,7]. Usually, unstable chemical compounds, which have a high tendency to evaporate and are volatile,

sensitive to light, heat, and the external environment, require additional protection [8,9]. To protect them, as well as enhance the organoleptic sensation in the product, they are applied in the encapsulated form.

The main purposes of encapsulation are as follows: (i) immobilization of the active material by encapsulating it, (ii) protection, including separating the core from the destructive influence of the environment, (iii) controlled release of the core material, so that exposure to the active material is prolonged, (iv) structure change, obtaining a solid from a liquid or gas, and (v) functionality.

Encapsulation of flavors and fragrances has a number of benefits, including the following [10]:

1. Extended shelf life;
2. Improved stability during processing and in the final product, with a change in the structure from liquid to solid; liquidity, dispersibility, and dosage accuracy in the final product are improved;
3. Gradual, controlled release of aroma compounds, prolonging exposure to odor or taste;
4. Masking of taste and odor;
5. Protection from external factors, separation of chemically unstable and highly volatile substances from environmental factors, protection from UV radiation, degradation reactions, from heat, oxidation, and dehydration;
6. Improved safety by reducing the flammability of volatile substances.

2. Morphological Characterization of Capsules

Encapsulation is a process in which a micron-sized particle of the main substance is coated and surrounded by a wall/carrier material that insulates and protects the contents from the external environment [11–13]. The resulting encapsulation product is called a capsule or encapsulate. The capsules can be divided into the following two parts: the outer, inert layer, usually called the shell, the coating, the carrier material, the wall material, the matrix or membrane, and the inner, active layer called the core, the active agent, the fill, the internal phase or the payload phase (Figure 1) [3,14]. The resulting microcapsule can be of various shapes and sizes, can be regular and irregular in shape, can be a small spherical sphere, a crystal, a jagged adsorbent particle, an emulsion, a suspension of solids or a suspension of smaller microcapsules [1]. Most of the resulting encapsulates are of the order of magnitude from a few micrometers to a few millimeters. Depending on the physical state and chemical type of the material (Table 1) to be encapsulated, as well as the subsequent use and the application of the encapsulation, different types of techniques are possible for their manufacture [4,10]. Table 1 shows the type of shell and core materials [11,15,16].

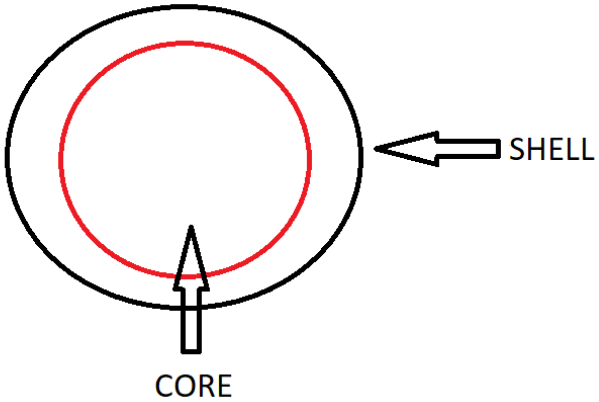

Figure 1. The general structure of the capsule, adapted from [10].

Table 1. Type of shell and core materials [11,15,16].

SHELL MATERIAL	CORE MATERIAL
• Gums: gum arabic, sodium alginate and carrageenan • Carbohydrates: starch, dextran and sucrose • Celluloses: carboxymethylcellulose and methylcellulose • Lipids: beeswax, stearic acid and phospholipids • Proteins: gelatin and albumin Composition of coating material • Inert polymer • Plasticizer • Coloring agent	Gas Liquid Solid Forms: solution, dispersion and emulsion Composition of core material • Flavor and fragrance • Drug or active constituent • Additives such as diluents • Stabilizers • Release rate enhancers

2.1. Effect of the Encapsulation Process on Capsule Size

The main factor that affects the stability and efficiency of the encapsulation process is the average particle size. Compared to small particles, large particles allow better protection, but exhibit low dispersion in a matrix such as food. On the other hand, a very small size can cause difficulties in encapsulation efficiency [9].

Based on size, the capsules can be divided into the following three main groups (Figure 2) [17–19]:

Figure 2. Division of the capsules by size [20].

1. Nanocapsules: <1 µm;
2. Microcapsules: 1 µm–1000 µm;
3. Millicapsules: >1 mm.

The resulting size of the microcapsules (microcapsules are defined as particles between 1 and 1000 µm in size, which contain an active agent—the core—coated with a natural or synthetic shell) depends on the method of obtaining them (the most commonly used methods are described in Section 4.1.), which may include the following: (i) coacervation (2–1200 µm); (ii) emulsion methods (0.5–1000 µm); supercritical liquid precipitation (about 1 µm); melt dispersion (1–50 µm); spray drying (5–5000 µm); coating (5–500 µm); polymerization (0.5–1100 µm); crosslinking (2–20 µm) [21].

2.2. Effect of the Encapsulation Process on Capsule Structure

The structure of the obtained microcapsule is influenced by the type of active ingredient and encapsulating material used in the encapsulation process, as well as the microencapsulation method used. The following structures can be distinguished (Figure 3) [1,4,11,13]:

1. Mononuclear, reservoir type—core/shell capsules and mononuclear encapsulates, in which a single shell is arranged around the core;
2. Polynuclear capsules—contain multiple cores surrounded by the shell;
3. Matrix encapsulation—the core is homogeneously distributed within the shell material. This is currently the most common type of encapsulation used in the pharmaceutical and food industries;
4. Multi-wall—a microcapsule made up of several coatings;
5. Coated matrix type—a combination of the matrix and mononuclear type.

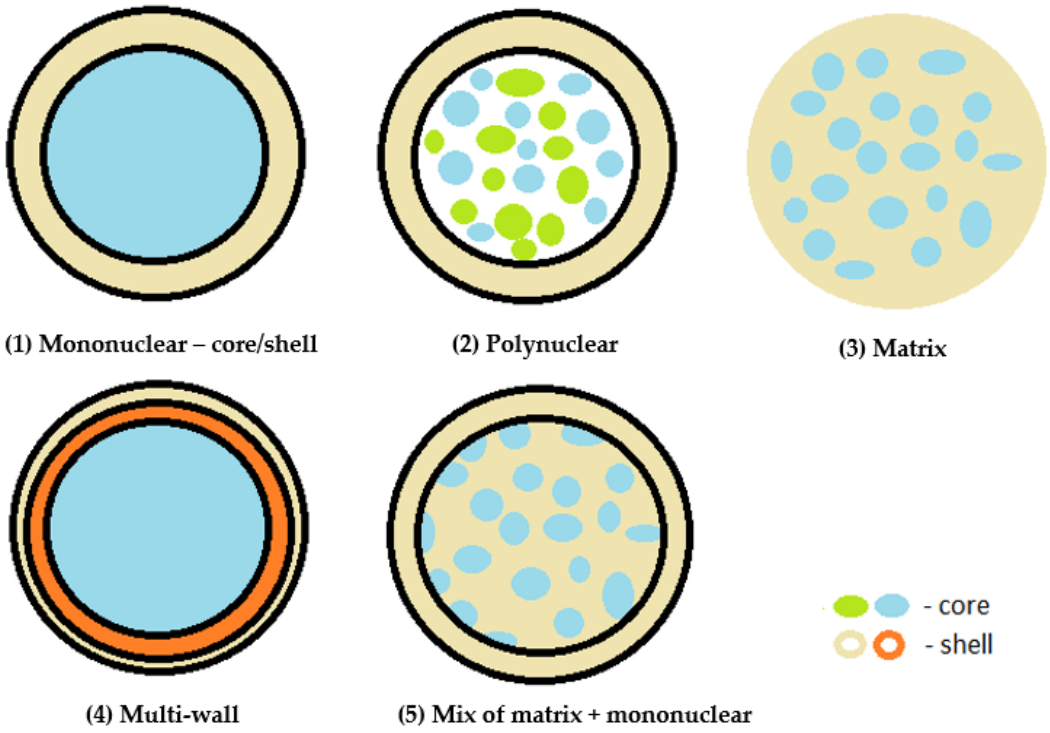

Figure 3. Encapsulate morphology, adapted from, adapted with permission from Ref. [1].

2.3. Selection of the Coating Substance

The determination of the appropriate coating material for the substance to be encapsulated is the most important step in the encapsulation process. The choice of coating material depends on the active ingredient and the desired properties of the final product [22,23]. The structure of the coating material determines the performance of the capsules. An ideal coating material should be characterized by the following: (i) the machinability during encapsulation should be easy; (ii) must display stable emulsifying properties towards active ingredient; (iii) it must not react with the core material during either encapsulation or long-term storage; (iv) it must be able to coat the active substance; and (vi) it must have the ability to be impermeable under processing and long-term storage conditions [2,24].

Due to the chemical properties of the coating, encapsulants are divided into the following categories [15,17]:

1. Polymeric, natural encapsulants, such as gum arabic, alginate, β-glucan, starch, plant protein and gelatin and synthetic encapsulants, e.g., polyesters (poly(lactide-co-glycolide) (PLGA);
2. Inorganic encapsulants, e.g., SiO_2, silica, which is a non-toxic, highly biocompatible, and mechanically stable substance that meets the requirements in pharmacy and biochemistry;
3. Polymers (inorganic).

2.4. Effect of Encapsulation on Prolonging the Aroma Experience

Encapsulation promotes the prolongation of the fragrance experience through the controlled release of fragrance. On the one hand, the volatile chemicals responsible for pleasant fragrances must be protected from evaporation and degradation reactions, ensuring a long shelf life; on the other hand, the key to excellent product performance is the consistent and long-lasting release of fragrance once it is deposited on fabrics or surfaces [25]. A direct consequence of the latter is that less fragrance material can be used in products, which has a positive impact on the environment [26].

Taking into an account the prolonged exposure to taste or odor, and release of aroma compounds under certain conditions, encapsulation can be divided into the following categories:

1. Impermeable sealed encapsulations;
2. Semi-permeable encapsulates;
3. Permeable open encapsulates. The coating on which the material is deposited can be salt or sugar and this process is cheap and sufficient in some cases, but unfavorable when considering the mixture of volatile compounds, as there is no barrier to oxidizing compounds.

3. Physicochemical Characterization of Microcapsules Obtained by Encapsulation Process

When very small capsules are considered, their characterization may concern the resulting powder (the entire mass of the product), rather than a single capsule. In this case, the sample is characterized by determining the (i) particle size distribution, (ii) flow properties, (iii) abrasion resistance, (iv) bulk density and porosity, (v) compressibility, (vi) dust index, and (vii) explosion classification [27]. In turn, the characterization of the resulting particles includes, among others, the size of the obtained capsules, density, abrasion resistance, and efficiency of encapsulation [27].

3.1. Size of the Capsules Obtained in the Encapsulation Process

The size of the capsules affects the various characteristics of the encapsulant, as its size indicates the amount of carrier material from which the encapsulant is constructed. Reactivity, hygroscopicity, and stability are the properties directly related to the size of the encapsulates. Dissolution rate is indirectly related to the size of the obtained capsules, even in combination with its shape and structure. An important characteristic affected by the size of the resulting encapsulates is sedimentation in the dispersion, and in many applications, it is very important to maintain a homogeneous distribution of encapsulates. The smaller the capsule, the greater the viscosity in the dispersion, and thus the slower the movement of the encapsulates in the final sample. In pharmacology, there is a division based on the size of the encapsulates in relation to the speed of delivery of the active material. Smaller capsules are designed for direct, immediate delivery of active ingredients, while larger capsules are designed for delayed delivery. The size of the encapsulates also affects the structure of the food product, and the addition of large particles is not recommended, as they will be felt during eating [16,28].

For spherical encapsulates (Figures 1 and 3), their size is determined from the length of the diameter, while for encapsulation products with irregular shapes, several additional parameters must be considered (Figure 4) [27].

Characteristics of irregular particles:

(1) Weight;
(2) Maximum length;
(3) Minimum length;
(4) Surface area;
(5) Sedimentation velocity;
(6) Siving size;
(7) Projected area;
(8) Volume.

Figure 4. Irregular particle characteristics, adapted from [27].

The size of the encapsulates can be measured by techniques such as laser scattering [29] in the particle size range of 0.1 µm to 3 µm, where light from a coherent light source, such as a laser beam, scattered on particles creates a characteristic intensity distribution that depends on the scattering angle. The exact pattern of this distribution essentially depends on the size of the particle. In this method, the spherical shape of the particle is assumed. The particle size, shape, and morphology image can be obtained using SEM (scanning electron microscopy), where the resulting magnification image of the encapsulates is analyzed using image analysis software. In the case of encapsulates made of a coating of natural polymers, which are not conductive and, in effect, cause image distortion, they are covered with a conductive layer, such as a layer of gold [30]. SEM imaging is designed for encapsulations in the size range of 0.002 µm to 1 µm. In these techniques, precise knowledge of the shape of the encapsulant is needed, and the result is a digit [27]. In order to determine the size percentage of the encapsulates in a given range, mass evaluation techniques should be performed, such as centrifugal sedimentation, giving results in the size range of 0.01 µm to 10 µm, flow classification (gravity, centrifugal and cyclonic), in the range of 0.3 µm to 100 µm, sedimentation from 2 µm to 100 µm and sieving [31] in the range of 5 µm to 4000 µm. However, these techniques are designed for capsules of larger sizes. High-resolution imaging with electron microscopy or confocal laser scanning microscopy (CSLM) is useful for detailed examination of the microcapsule's morphology. CSLM can be combined with staining techniques to gain more insight into the characteristics and distribution of the hydrophobic core and hydrophilic shell [27].

The size distribution of the capsules is a determinant of proper encapsulation. The smaller the range of particle size distribution, the more efficient the process.

3.2. Density

When the presence and location of the pores in the capsules' shell material are taken under consideration, we can distinguish the following three types of density: (i) the true particle density (Equation (1)), which takes into account the open and closed pores in the encapsulating material, (ii) the apparent density (Equation (2)), which only takes into account the open pores of the encapsulating material, and is mostly determined using a pycnometer, where the gas passing through the encapsulant only recognizes the open pores (those that are outside the shell material) and (iii) the effective particle density (Equation (3)), which does not take into account the pores [27] and is also described as bulk density [32–34].

- True particle density

$$\text{True particle density} = \text{Mass of particle (g)}/\text{Volume excluding open and closed pores of particle material (ml)} \quad (1)$$

Equation (1). True particle density equation [27].

- Apparent particle density

$$\text{Apparent particle density} = \text{Mass of particle (g)}/\text{Volume excluding only open pores of particle material (ml)} \quad (2)$$

Equation (2). Apparent density determined using a picnometer [27,34].

- Effective particle density

$$\text{Effective particle density} = \text{Mass of particles (g)}/\text{Volume of particles (ml)} \quad (3)$$

Equation (3). Calculated from the relation of mass (g) and volume (mL) of a known quantity of the sample placed into a graduated cylinder (volume includes the open and closed pores of the particle material) [27,32,34].

- Porosity, €

$$€ = 1 - \frac{\text{Effective partice density}}{\text{Apparent particle density}} \quad (4)$$

Equation (4). Porosity calculations [33].

3.3. Abrasion Resistance

The determination of abrasion resistance is extremely important in the case of transportation, when encapsulates will rub against each other for hundreds of kilometers or when the next stage of production is the mixing of encapsulates, such as with tea. Knowing the abrasion resistance value, one can prevent possible production losses and the destruction of encapsulates. It is also important information for developing appropriate storage or further processing conditions [27].

3.4. Effectiveness and Efficiency of Encapsulation

This can be determined by GC-FID gas chromatography, preceded by extracting the encapsulated aroma compounds. Analysis of the total recovery is based on the chromatographic spectrum, or performed by GC-MS, when analyzing the recovery of individual aroma compounds.

4. Encapsulation Technologies

A number of methods are known for obtaining microcapsules, which include the following: (i) physicochemical methods (coacervation, emulsion methods, precipitation using supercritical fluid and melt dispersion), (ii) physicomechanical methods (spray drying, coextrusion, microfluidization methods, fluidized layer coating and coating in a drag drum), and (iii) chemical methods (polymerization methods and crosslinking method). The selection of the appropriate method for obtaining microcapsules is dictated by the properties of the active substance, the properties of the coating material, and the purpose of the final product. After the encapsulation process, the microcapsules are separated by decantation, filtration and centrifugation, and then washed with distilled water. At the final stage, they are subjected to drying at room temperature or in dryers (freeze-drying). Microcapsules can be used in the form of suspensions or powders. In addition, they can also be coated and used as a substrate for other drug forms [35,36].

As for the technologies used to produce encapsulated flavors and fragrances, the most commercially prevalent technologies are spray drying, extrusion, fluidized bed drying, spray chilling, spray cooling, and coacervation. In each of these technologies, the resulting capsules will vary in size, shape, type of release, or stability. All of these technologies have their own advantages and disadvantages [10].

In simple terms, the encapsulation process can be divided into four main stages, which are summarized in points I–IV.

I. Introduction of a core material, which will then be surrounded by a coating [21]. The active material can be either of the following:
   a. Liquid core:
      - Solution or melt;
      - Emulsion;
      - Suspension.
   b. Solid core (powder).

   Fragrance compositions and flavors are mostly liquids at room temperature.

II. Dispersion stage, where many different technologies are used to produce microcapsules, including the following [1,4,8,11,21]:
   a. Spraying;
   b. Dripping;
   c. Emulsification;
   d. Spray coating;
   e. Formation of suspension coating;
   f. Extrusion.

III. The proper process of encapsulation, which can be divided into the following three groups, in terms of the transformations that take place [13]:
   a. Physical methods:
      - Solidification;
      - Evaporation.
   b. Physical and chemical methods:
      - Gelation;
      - Coacervation.
   c. Chemical methods:
      - Polymerization;
      - Cross-linking.

IV. Scale-up and down processing [21].

4.1. The Most Common Methods Used to Encapsulate Flavors and Fragrances

Physicomechanical methods

4.1.1. Spray Drying

This is an extremely widely used process in the food industry, for example, in the production of milk powder, but also in the pharmaceutical and cosmetic industry [37]. Due to the widespread use of the process and the availability of apparatus, it has found application in encapsulation [12]. The general principle of the spray drying process is to dissolve and emulsify the core substance in an aqueous solution of the carrier material, and then atomize the mixture in a hot chamber, where smaller water molecules evaporate and a coating forms around the active particle. However, highly volatile fragrance compounds evaporate faster than water, so it is important to choose the right carrier to prevent the loss of volatile compounds, while allowing water to evaporate. When selecting a suitable carrier, the following parameters should be considered [1,4,10]:

- Good solubility in water;
- Good emulsifying properties;
- Low viscosity at high concentrations (<500 cps at >45% concentration);
- Low hygroscopicity;
- Taste and/or odor release under the right conditions;
- Low-cost and accessible material;
- Neutrality in taste;
- Stability.

The most commonly used carriers include hydrophilic polysaccharides, such as maltodextrins, chitosan, alginates, and various types of gums, and proteins, such as whey protein. The ratio of the carrier material to the main material is usually 4:1 [38].

The spray drying process can generally be divided into the following three stages (Figure 5) [21,39]:

1. Preparation of an emulsion or slurry from the main material and carrier. The emulsion is usually formed at high mixing speeds or using colloid mills, under high pressure. The product is then processed further by various mechanical means, such as high-pressure homogenization, microfluidization, and ultrasonic emulsification. These methods are used to stabilize the emulsion at least for a certain period of time. The viscosity of the emulsion affects the subsequent drying step and moderate values give the best encapsulation results. Emulsions with too high viscosity can clog the feeder nozzle, or settle on the walls of the chamber [27].
2. Atomization and dispersion of the emulsion. The emulsion is pumped into the drying chamber through an atomizer. Various techniques are used to atomize the emulsion and among the most common are high-pressure nozzles and centrifugal wheels. The atomizer separates the emulsions into small droplets (the size of the droplet depends

on the pore size of the atomizer) and sprays into the hot air in the chamber. The following three methods of atomizer air atomization are possible: co-current, counter-current and mixed. For fragrance compounds, co-current air is commonly used. Then, the moisture evaporates from the emulsion droplets, leading to the entrapment of the main material in the coating [38,40,41].

3. Particle collection. This includes the separation of the resulting encapsulates by a cyclone separator and collection in a receiver. The total duration of the encapsulation process is very short, ranging between a few milliseconds and a few seconds [38,40,41].

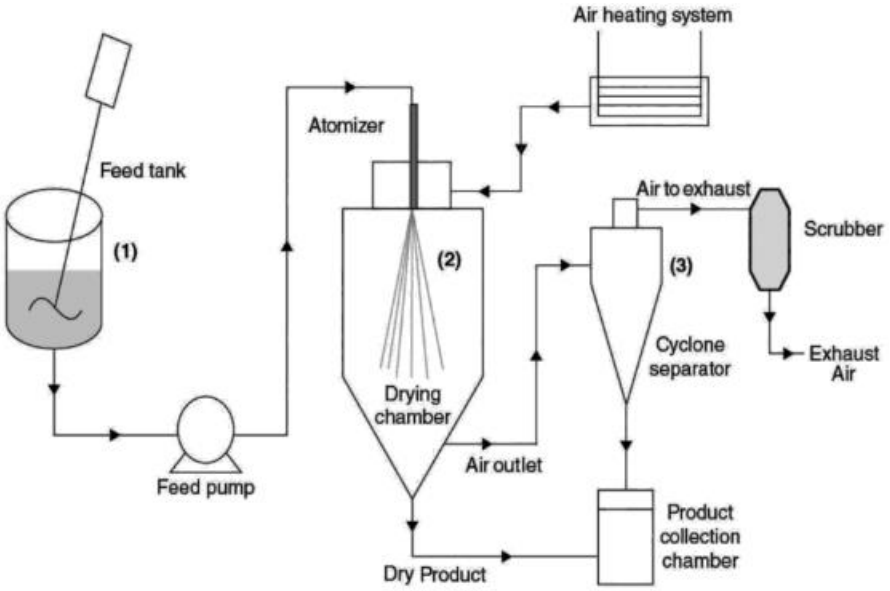

Figure 5. Spray drying technology. (1) Emulsification, (2) atomization and dispersion; (3) particle collection. Adapted from [42].

The finished product of spray drying can contain from 2% to 50% of the active substance. The advantages of spray drying are as follows: the availability of the apparatus, its low cost and good encapsulation results. Complementary to spray drying are product concentration and agglomeration methods. In both cases, the effect is to produce larger encapsulates. In the case of compaction, spray drying products are compressed under high pressure into lumps and crushed into small pieces from 0.7 mm to 3 mm (useful for tea applications). Compaction yields a product with a low porosity. Agglomeration involves fluidizing the spray drying product [41].

4.1.2. Extrusion

Extrusion is a relatively new encapsulation technique, compared to spray drying. Extrusion technology can be divided into the following five types: hot-melt extrusion, melt injection, centrifugal/coextrusion, electrostatic/electrospanning extrusion, and particle from gas saturated solution (PGSS) [43]. Melt injection was the first extrusion technology to be observed and is a vertical screwless extrusion technique, compared to melt extrusion, where, as is the case with most types of extruders, a twin-screw extruder is used [44]. On the other hand, the principle of operation is similar for all of them (Figure 6). The extrusion process involves mixing the active material with the carrier material by melting the carrier material and dispersing it with the core material, then the mixture is pressed through the die into a dehydration fluid, usually isopropyl alcohol, which hardens the coating, encapsulates the core material in the coating, and removes the residue of the active material on the coating. The resulting hard material is broken into small pieces, separated, and dried [45]. The product of extrusion is durable encapsulates that range in size from 250 µm to 2500 µm. The most commonly used carrier material is maltodextrin, simple

sugars, starch and a combination of high- and low-molecular-weight carbohydrates, such as maltodextrin, with sucrose, lactose and maltose. The dosage of flavor or fragrance composition ranges from 7.5% to 40%, but in practice, the dosage amounts are much lower, in the range of 8–12% [46].

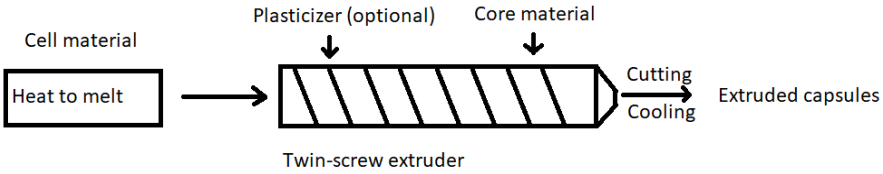

Figure 6. Extrusion process, adapted from [47].

4.1.3. Microfluidization Method

To obtain microcapsules by the aforementioned method, the Microfluidic® device is used, which consists of microchannels that allow the laminar flow of fluids and the production of double emulsions. By using this device, it is possible to obtain uniform sizes of microcapsules and repeatability of the process [48,49].

4.1.4. Pan Coating

Pan coating is used for the microencapsulation of solid particles with a diameter of more than 600 µm. In this apparatus, there are holes on the periphery and a disc that rotates in the direction opposite to the movement of the drum. A solution of the encapsulating material is fed into these holes, while the cores are fed into the center of the disc, from where they are transformed into a layer of the coating substance. If the mass of the core together with the shell reaches a certain value, where the centrifugal force overcomes the cohesion forces of the hole's membrane, the microcapsules are ejected outside the receiver. The solvent used is removed using warm air, while the shells are cured by physical or chemical methods. The advantage of this method is its high production efficiency, in addition to being fast, efficient, and relatively simple [21,50].

Physicochemical methods

4.1.5. Coacervation Method

Coacervation is a physicochemical method and is considered to be the method from which microencapsulation originated. There are two types of coacervation, simple and complex, depending on how the process is carried out [51]. In simple coacervation, there is only one type of polymer, with the addition of a highly hydrophilic agent in the colloidal solution. In complex coacervation, two or more polymers can be used. Complex coacervation relies on the ability of cationic and anionic polymers to interact with each other in water to form a polymer-rich liquid phase, called a complex coacervate. The most common cationic polymer used in this case is gelatin. A variety of natural and synthetic water-soluble polymers react with gelatin to form complex coacervates that are suitable for encapsulation, which occur in equilibrium with a clear liquid on the surface [17,25,52].

The coacervation process involves the separation of phases in a solution of colloids or polymers and the formation of two or more liquid phases as a result of the controlled modification of the solution environment, which includes changing the pH, ionic strength, temperature, solubility and addition of salt, or a countercharged polymer. The coacervation process produces a colloid-rich coacervate phase and an equilibrium phase [53]. The coacervation process (Figure 7) can be divided into the following four steps [54]:

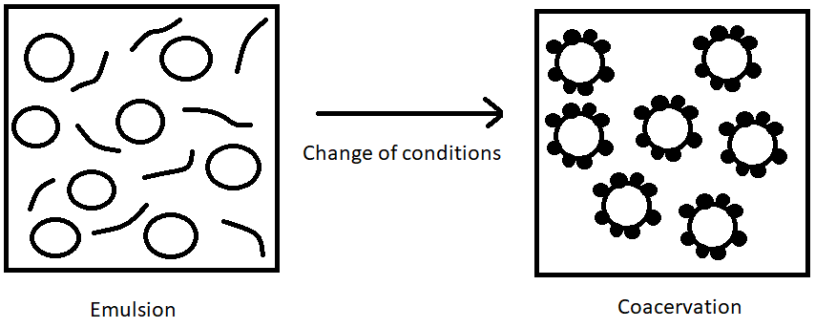

Figure 7. Coacervation scheme, adapted from [54].

1. Preparation of an aqueous solution of two or more polymers;
2. Emulsification of the aqueous phase of the polymers with the hydrophobic phase of the core. The active ingredient should not dissolve in water, as this may lead to losses, reducing the efficiency of the coacervation method (the maximum solubility of the core material in water is 20 mg/mL);
3. Change in the environmental conditions of the solution in order to proceed with coacervation and phase separation. A shell is formed around the core, and the phases are separated into a coacervate phase, with the core material surrounded by a carrier material, and an equilibrium phase (water);
4. Cooling of the system and addition of a cross-linking substance to harden the shell.

In simple coacervation, a single polymer is involved and coacervates are formed by a dehydration or water deficiency mechanism through the addition of salt, or desolvation fluid (coacervating agent or inducer). An example of simple coacervation is the separation of gelatin layers, following the addition of sodium sulfate (Na_2SO_4) or ethanol [53].

Complex coacervation is widely used in the pharmaceutical, food, agriculture, and textile industries. The process is extremely efficient, allowing the formation of 99% encapsulates. Complex coacervation primarily involves the use of two biopolymers with opposite charges, mainly proteins and polysaccharides, which can form a complex shell for the core material. The proteins used can be divided by origin into the following categories [53]:

- Animal: gelatin, whey, egg albumin and silk fibroin;
- Plant-based: soy protein, pea protein, wheat protein, lentil protein and chia protein.

Meanwhile, the most commonly used polysaccharides are alginate, chitosan, gum arabic, pectin, agar, carrageenan and carboxymethylcellulose.

Currently, coacervation is one of the most popular encapsulation methods used in the medicine and food industry.

4.1.6. Emulsion Methods

These methods are among the oldest and most widely used for obtaining microcapsules. They are used to encapsulate compounds that are both soluble and insoluble in water, and substances that are sensitive to external factors, such as temperature. Emulsion methods are based on the formation of single or double emulsions and immiscible liquids that contain an active substance and a polymer. As a result of the solidification of the polymer around the active substance when the solvent is removed, microcapsules are obtained [55–57].

The single-emulsion method involves preparing a solution of the encapsulating polymer in a volatile organic solvent (e.g., methylene chloride, chloroform, and ethyl acetate) that does not mix with water. Polymer solutions may additionally contain the addition of emulsifiers and stabilizers (e.g., Span 80 and Tween 80). The core material (solid form) is then dissolved or dispersed in the prepared polymer solution. The mixture thus obtained is emulsified with the aqueous phase with the addition of emulsifiers and/or stabilizers, resulting in an oil/water-type emulsion. Once the emulsion is obtained, the solvent is

evaporated. During the evaporation of the solvent, the polymer settles around the cores and solidifies [58].

On the other hand, there is a two-step process, which usually involves the formation of water/oil/water or water/oil/oil emulsions. In the first stage, active substances in an aqueous medium are emulsified with a polymer solution in an organic solvent. In the next stage, the formed emulsion is added to the second aqueous phase, then a double emulsion of water/oil/water is formed or added to the other oil phase, leading to the water/oil/oil emulsion. These processes are accompanied by the addition of an emulsifier and/or stabilizers. Once the emulsion is formed, the volatile solvent is removed by evaporation [59].

Chemical methods

4.1.7. Polymerization

One of the chemical methods used to obtain encapsulates is the process of polymerization, including both interfacial and in situ polymerization. The former, interfacial polymerization, involves the formation of encapsulates by polymerizing monomers at the interface between the dispersing (water, glycerol, ethyl alcohol, chloroform and cyclohexane) and dispersed (animal fats, vegetable oils, and synthetic oils) phases [60,61]. The dispersion phase is mixed together with the suspended or dissolved active ingredient and the encapsulating monomer, until an o/w emulsion is obtained. The most commonly used monomers are multifunctional isocyanates and multifunctional organic acid chlorides. In addition to the above, methyl methacrylate, vinyl acetate, and a mixture of styrene and vinylbenzene or diamines are also used [50]. Monomers diffuse between the o/w phase, and the reaction results in a polymeric membrane (polyurea, polynylon, or polyurethane) that is insoluble in oil [55]. On the other hand, in situ polymerization, when compared to interfacial polymerization, occurs without the involvement of additional reactive agents. The polymer that is formed initially exhibits a low molecular weight, which increases with time. Subsequently, this polymer is deposited on the core material and forms a solid capsule shell [50].

The advantages and disadvantages of the above-described encapsulation methods are summarized in Table 2.

Table 2. Advantages and disadvantages of selected encapsulation methods [25,50,60].

Method	Advantages	Disadvantages
Spray drying	- Quick and easy process - High process efficiency - Microencapsulation of temperature-sensitive substances - Possibility to obtain sterile products	- Formation of polymorphic varieties of polymers that cause disintegration of microcapsules - Loss of material due to the adhesion of active substance particles to the drying chamber
Extrusion	- Fast and inexpensive process	- Formation of large-size capsules
Microfluidization method	- Small size of microcapsules - Uniformity of shape and size - Repeatability of process conditions	- Organic solvents and surfactants prevent microencapsulation of living cells
Pan coating	- Fast, effective and simple method	- Formation of large-size microcapsules
Coacervation	- Process is simple, fast and reproducible - Possibility of using low concentrations of surfactants - The process can be run in aqueous and anhydrous environments	- Agglomeration of droplets under the influence of solvent removal - Toxicity of the solvents used
Emulsion methods	- Ability to encapsulate hydrophilic and hydrophobic substances - High process efficiency	- Agglomeration of droplets under the influence of solvent removal - Toxicity of the solvents used
Polymerization	- Able to control the size of microcapsules by adding surfactants in low concentrations	- Reaction difficult to control - Requires significant washing to remove monomers, organic solvents and surfactants

5. Application of Encapsulates Containing Flavors and Fragrances

In recent years, scientific work on the application of encapsulation methods has focused primarily on obtaining encapsulates, which are used in the fields of drug delivery, pesticides, food preservation, and cosmetics [62]. Most of the patents that cover this topic area focus on the use of encapsulates in medical, hygiene, food, and cosmetic products [63–66].

Examples of their use are described below.

Microencapsulation is also used in the textile industry to improve the functional properties of fabrics, such as their antibacterial activity, odor reduction, mosquito repellency, UV protection and thermoregulation [67]. Fragrance can be imparted to the fabric directly by applying microencapsulation, using techniques such as impregnation, spraying, coating, or embossing [1]. This type of scenting is applied to products such as scarves, ties, and underwear, as well as home textiles, such as sofa throws, curtains, and aromatherapy pillows. "Scratch and sniff" T-shirts have also been produced [68]. Fragrance is also imparted by washing fabrics in scented washing powders or rinsing in scented softeners [69]. The fragrance microcapsules of the powder or softener adhere to the fabric, and the fragrance is released gradually under mechanical abrasion. Microcapsules are also used for the thermoregulatory function of fabrics. In this case, the encapsulates perform the function of absorbing or donating heat by melting and solidifying, depending on the temperature; these are called phase-change materials (PCMs), or smart materials [70]. Natural essential oils, which, in addition to their sensory properties, also possess antibacterial, antiviral, antifungal, anti-inflammatory, and insect repellent properties [8], are enjoying increasing consumer interest and are widely applied in the textile industry. Essential oils, trapped in microcapsules on clothing, can, after contact with skin, perform skin care, anti-aging, or odor control effects.

In the food industry, stabilized fragrance compounds are applied to tea, significantly extending the shelf life of the product, while maintaining the aroma profile. For instant coffee, the addition of microcapsules improves the product in terms of intensity. Encapsulated essential oils are also used as antifungal agents, or for fruit preservation.

In the cosmetics and household products industry, the possibilities for the use of encapsulates are numerous; products such as creams, shampoos, gels, soaps, surface washing liquids, and fragrances are hidden everywhere. Consumer demands are increasing and interest in natural fragrances is growing, and once the product is used, the scent is expected to remain for a long time. The use of encapsulated fragrance compounds can be a solution to high consumer expectations. On the other hand, the use of essential oils with antimicrobial properties can replace the use of preservatives, which will also be commercially attractive [71]. For aromatherapy purposes, encapsulates are added to the cellulose pulp to create, for example, scented wallpaper or scented wrapping paper.

Table 3 demonstrates examples of the use of the encapsulation process with respect to the aroma and fragrance compositions.

Table 3. Examples of the preparation of encapsulates with aromas and fragrances.

Core Material	Shell Material	Methodology	Goal	Morphology	Ref.
Vanilla oil	Chitosan/arabic gum	Complex coacervation	Controlled release and thermostability product for spice application in food industries	Spherical shape and smooth surface, 94.2% efficiency (VO/CS 2:1)	[72]
Limonene fragrance	Chitosan/cellulose	Freezing/ thawing/stirring process	A fragrant component widely used in the flavor and fragrance industries; encapsulation prolonged the release of fragrances	Spherical shapes, with an average diameter of 2 mm, 51.3% efficiency	[73]
Fragrances: D-limonene, Claritone, Amarocit, Rose Oxide-High Cis, methyl salicylate, 1-octanal, 1-octanol, hydrocitronitrile, Majantol and ethyl 2-methylbutanoate	Bovine serum albumin and tetramethylrhodamine isothiocyanate-labeled BSA (TRITC-BSA)	Layer-by-layer (LbL)	Fragrance encapsulation. Controlling the release of fragrances; both TA and BSA are relatively cheap and available compounds	The encapsulation efficiency depends on the water solubility; the less water-soluble the ingredient, the smaller its losses upon LbL coating of emulsion in the filtration cell and the higher its relative content in released fragrance	[74]

Table 3. *Cont.*

Core Material	Shell Material	Methodology	Goal	Morphology	Ref.
Pink fruity fragrances and white floral fragrances	Protein, carbohydrate and lipid	Liquid–liquid dispersion	Fabric softener application and long-lasting property in textile applications	Spherical shape with an average size of 100–300 nm, efficiency of 69–75%	[69]
Tuna oil in water emulsion stabilized by lecithin-chitosan membrane, using an electrostatic layer-by-layer deposition process	Maltodextrin	Spray drying of two-layer emulsion	High oil-loaded microcapsules that may lead to a wide range of applications in food products	Spherical particles (except for oil:maltodextrin 1:1), efficiency of 89% for oil:maltodextrin 1:4 ratio and 56% for ratio of 1:1	[75]
Linoleic acid	Gum arabic or maltodextrin	Spray drying	Evaluation of influence of the encapsulation process on the stability of linoleic acid towards oxidation	Particles with an average size of 0.68 µm (gum arabic) or 1.68 µm (maltodextrin), efficiency of 75–99% (gum arabic) and 35–50% (maltodextrin)	[76]
Ascorbic acid	Maltodextrin	Extrusion	Vitamin encapsulation	Crystals with sharp edges, efficiency of 96% and load 19%	[77]
Orange terpenes	Maltodextrin and sucrose	Extrusion	Flavor encapsulation	Partly crystalline samples, about 1 mm particle size, efficiency of 34.5–67.3%; load 4.1%	[44]
Lemon oil	Sago and tapioca starch, gum arabic and stearic acid	Spray drying	Encapsulated agent for food industry	Efficiency of 49–59%	[78]
Orange oil in water emulsion	Lactose and caseinate	Spray drying	Application in different types of food or pharmaceutical products, where maximum protection for flavors or slow release are required	Particle size of 30.9 µm. Efficiency of 44.5%	[79]
Orange essential oil	Octenyl succinic anhydride, modified starch and maltodextrin	Vacuum spray drying	Application of vacuum spray drying	Efficiency of 99.89%	[80]
Gurum seed oil	Gum arabic, maltodextrin, pullulan and whey protein isolate	Spray drying	Evaluating the potential of combining maltodextrin with gum arabic and whey protein isolate	Efficiency of 97.38%	[81]
Citrus sinensil L. (essential oil)	Maltodextrin	Spray drying	Evaluating the factors affecting microencapsulation	Efficiency of 89.94%	[82]
Juniperus communis L. (essential oil)	Gum arabic, maltodextrin, sodium alginate and whey protein concentrate	Spray drying	Food flavoring agent and preservative	Efficiency of 82.66%	[83]
Rosemary (essential oil)	Maltodextrin and whey protein concentrate	Spray drying	The potential of combined emulsification and spray drying procedures to encapsulate polyphenolic components from rosemary	Efficiency of 27.09–42.93%	[84]
Cinnamon (essential oil)	Gum arabic, maltodextrin and whey protein concentrate	Spray drying	Effect of shell materials used on encapsulation efficiency	Efficiency of 13.8–50.1%	[85]
Syzygium Cumin Seed (essential oil)	Gum arabic	Spray drying	Antioxidant	Improvement of water vapor permeability; prolongation of oil oxidation	[86]

6. Conclusions

Although several encapsulated products have been developed and used in the cosmetic, pharmaceutical, and agribusiness industries, the number of studies on encapsulated fragrances and flavors is relatively small. The growing demand for modern cosmetic, pharmaceutical, agrochemical, or functional textiles and food products that contain flavor compounds in their composition contributes to the great interest in the issue of microencapsulation. "The Global Microencapsulation Report" forecasts a CAGR, namely a compound annual growth rate of 12.9% over 2022–2030. The benefits of encapsulation should overcome as many of the negative effects of processing flavor compounds as possible, such as the following:

- Additional costs;
- Increased complexity of the production process and/or supply chain;
- Unwanted consumer information (visual or tactile) about encapsulated foods;
- Stability of capsules during the processing and storage of the food product [12].

This paper presents the main technologies for obtaining encapsulates, in addition to their characteristics, and examples of applications. The challenge facing researchers will be the selection of a suitable method for encapsulate synthesis and its commercialization, since the final product is influenced by many factors, such as capsule size, carrier material, overall performance of the method, and the cost.

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