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minireview

Technological Challenges for Spray Chilling Encapsulation of Functional Food Ingredients

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Summary

Spray chilling technology (also known as spray cooling and spray congealing technology) has been widely studied and used in the pharmaceutical field. In the food industry, this technique is gaining interest and can become useful because functional food formulations can be developed. Spray chilling is a fat-based system, which involves the addition of the component of interest to a molten lipid carrier, and the resulting mixture is fed through an atomiser nozzle. When the nebulised material is put into contact with the environment, which is cooled below the melting point of the matrix material, the vehicle solidifies (due to heat exchange between the molten material and cold air), and solid lipid microparticles are formed at the same time. This technology is fat based, and lipid carriers, such as wax and oil (*e.g.* palm oil, beeswax, cocoa butter, and kernel oil) can be used. This encapsulation technique can potentially change the functionality, reduce the hygroscopicity, mask taste or odour, change solubility, and provide physical protection in addition to allowing the controlled release of these ingredients. This low-cost technology is relatively simple to apply and scale up, and it does not require the use of organic solvents and the application of high temperatures in the process. Therefore, spray chilling encapsulation may facilitate the development and production of functional and enriched foods as it may solve some technological problems associated with the use of certain ingredients, such as those that have high reactivity and low stability.

Key words: spray cooling, spray congealing, nutrient microencapsulation, solid lipid microparticles (SLMs), bioactive compounds

Introduction

The use of encapsulation technology has increased in the last 10 years to address some limitations in the use of medicines, food ingredients, cosmetics, veterinary products and hygiene products. The most popular techniques used to develop microparticles are spray coating, coacervation, solvent evaporation, ionic gelation, liposomes, spray drying and spray chilling (also known as spray cooling and spray congealing).

Spray chilling technology consists of making a solution dispersion or emulsion containing the active ingredient and a molten carrier, which is then atomised into a chamber where cold air or liquid nitrogen is injected. In the case of emulsions, spray chilling technology can be applied to a water-in-fat emulsion in the presence of an emulsifier, such as soy lecithin (*1–4*). Under these conditions, the carrier instantly solidifies, forming spherical particles, and the solubility of these particles depends on the hydrophilic/lipophilic nature of the carriers. These particles can be as small as a microparticle depending mainly on the device used for the atomisation process. To meet food industry requirements, carriers must be food grade. Lipophilic materials that have a melting

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point higher than room temperature are mostly used, thus yielding solid lipid microparticles (SLMs). The most recommended carriers for the development of particles by means of spray chilling are fatty acids, alcohols, triacylglycerols and waxes.

Spray chilling is a convenient technique for the encapsulation of food ingredients because it is a low-cost continuous process, and it is easy to scale up. Moreover, spray chilling does not require organic solvents, such as alcohols or ether, and it does not require application of high temperatures, which should be considered when thermosensitive ingredients, such as ω-3 fatty acids, enzymes and probiotics, need to be encapsulated. However, spray chilling encapsulation presents some technological disadvantages, such as low encapsulation efficiency and the possibility of expulsion of the active ingredient during storage. Another disadvantage of spray chilling encapsulation is related to the hydrophobic character of the particles, which can make some applications difficult.

There is no consensus in the literature regarding the nomenclature of the spray chilling encapsulation technique, which can be considered synonymous with spray cooling (*5*,*6*) and spray congealing (*7–10*). There is often overlapping of terms in technical and scientific literature. The terms spray chilling and spray cooling are normally used when taking into consideration the melting point of the carrier. Herein, spray chilling is the process that uses carriers with a melting point between 32 and 42 °C for the production of spray particles, and spray cooling is the encapsulation process in which the carrier has a melting point between 45 and 122 °C (*11–14*). The term spray congealing is employed indiscriminately when molten carriers are used regardless of their melting temperature (*15*,*16*). However, this impasse or the lack of consensus regarding the right terminology does not obscure the real importance of this encapsulation technology.

Currently, many publications are available in the literature describing the preparation of lipid particles by spray chilling for different purposes as follows: flavour and odour masking, production of ingredients with increased stability, protection and controlled release of probiotics, and encapsulation of bioactive proteins and peptides. However, most studies are from the pharmaceutical field, and there has not been an article published regarding the compilation of such data for the food industry. Based on these considerations, the purpose of the present article is to highlight the importance of spray chilling encapsulation in food fields.

Spray Chilling Technology

In general, the spray chilling process can be divided into two steps. The first step consists of adding the bioactive compound to the carrier material, which is usually a lipophilic compound, to be encapsulated. This incorporation process can be performed by dissolving or mechanically dispersing the material into the encapsulating matrix (the latter is the most widely applied method). The use of an emulsion is also a method to incorporate some hydrophilic bioactive agents (*2–4*). The second step consists of atomisation of the molten material, which is typically carried out by a heated atomising nozzle to maintain a proper temperature, thus avoiding solidification of the feeding material. When this material is atomised in contact with a refrigerated chamber (due to the injection of cold air or liquid N_2), a heat transfer occurs between the molten material and the cold air, thus leading to solidification of the carrier and the formation of particles (*5*).

The residence time of droplets in the spray cooling chamber is relatively short (only a few seconds). The particles are collected in a container located below the cooling chamber, and fine particles are transported by air into a cyclone where they are collected in another container (Fig. 1).

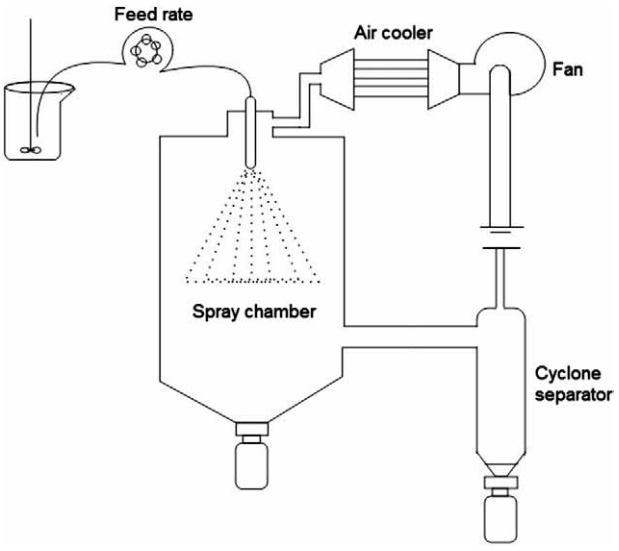

Fig. 1. Scheme of spray chilling equipment

The preparation of particles using this method has been applied in various sectors, including pharmaceutical, cosmetic, agricultural, veterinary and food industry (*1*). The application of spray chilling encapsulation process has also been reported for the production of both food ingredients and several drugs (*17*).

Numerous applications of SLMs obtained by spray chilling have been investigated, such as odour and taste masking (*18*,*19*); protection of the active ingredient against detrimental conditions, such as pH, enzyme activity, moisture, oxygen and light; optimisation of the dissolution of poorly soluble drugs; modulating the release kinetics of active compounds; and improvement of flow properties, handling, appearance and other purposes (*8*).

If lipids are used as the carrier material, the final product is insoluble in water and there is usually controlled release of its contents by utilising the melting point of the carrier (*20*) or by digestion of the carrier in the intestine. The proper selection of the carrier is crucial because the encapsulation process can modify its properties, such as reduce its hygroscopicity and/or increase its chemical/physical stability (*18*).

The atomisation of the molten mixture (carrier plus active ingredient) and the solidification process are con-

sidered critical steps. The atomisation process is related to the disintegration of the molten mixture into small particles (*21*), and the solidification process is associated with the processing of the molten material into a solid (obtained by cooling). From the operational standpoint, insufficient cooling leads to agglomeration of the droplets and/or adhesion of these droplets on the surface of the chamber, thus negatively affecting the morphology, process and other properties of the microparticles.

Spray chilling is similar to spray drying, which is a widely studied and widespread technology. However, there is a difference between these processes in terms of direction of energy flow. With regard to spray drying, the energy is applied to the droplet forcing the solvent evaporation. In spray chilling encapsulation, however, the energy is removed from the droplet forcing the solidification of the carrier (*5*). The spray chilling technique may be considered to be a merger between the hot melt technology (coating or agglomeration) and the spray drying technique. Table 1 (*5,8,10,14,22–25*) presents several differences between these techniques (spray chilling and spray drying).

The initial spray chilling configuration is similar to the spray drying configuration, but there is no solvent evaporation. A conventional spray dryer (when there is input of hot air) can also be used as a spray cooler (when there is input of cold air). The apparatus consists of two main parts: a cooling chamber and an atomiser. For an efficient process, it is recommended that the molten mixture dispersion presents a narrow melting temperature range, so the particles can also be solidified during the spraying process (*26*).

Both spray drying and spray chilling techniques do not produce reservoir-type microcapsules, but instead they produce matrix-type microspheres. However, spray chilling produces dense and massive microspheres, and spray drying generally produces hollow microspheres (Fig. 2).

There are also similarities between the SLMs obtained by the spray chilling technology and by the spray coating technology. These methods can both be fat based, and the physical nature of these processes is similar because they have the same release mechanisms that differ from the spray drying regarding the functionality of par-

Table 1. Differences between spray chilling and spray drying techniques

	Spray drying	Spray chilling	Ref.
Energy flow	energy applied in the droplet forcing evaporation of the medium	energy removed from the droplet forcing the solidification of the medium	(*5*)
Equipment	feeding tubes without heating	heated feeding tubes (to avoid solidification)	(*8*)
Flow in the chamber	hot air	cold air or liquid N_2	(*8*)
Particle size	5–150 μm	20–200 μm	(*14*)
Dissolution mechanism	dissolution	diffusion, heating, lipases and bile salts (GI tract)	(*22*)
Particle morphology	geometric particle with porous and irregular surface due to solvent evaporation	dense, spherical and smooth surface (no effects of solvent evaporation)	(*23*)
Carrier	water-soluble polymers	waxes, fatty acids, water-soluble polymers and water-insoluble monomers	(*24*)
Food ingredients	vitamins, flavours, starter cultures, carotenoids, oils, fats, enzymes and acidulants	iron sulphate, vitamins, minerals, acidulants, enzymes and probiotics	(*25*)
Process steps	dispersing or dissolving the active compound in aqueous solution coating, atomisation, dehydration	dispersing or dissolving the active compound in the molten lipid mixture, atomisation, cooling	(*10*)
Load/%	5–50	10–20	(*10*)

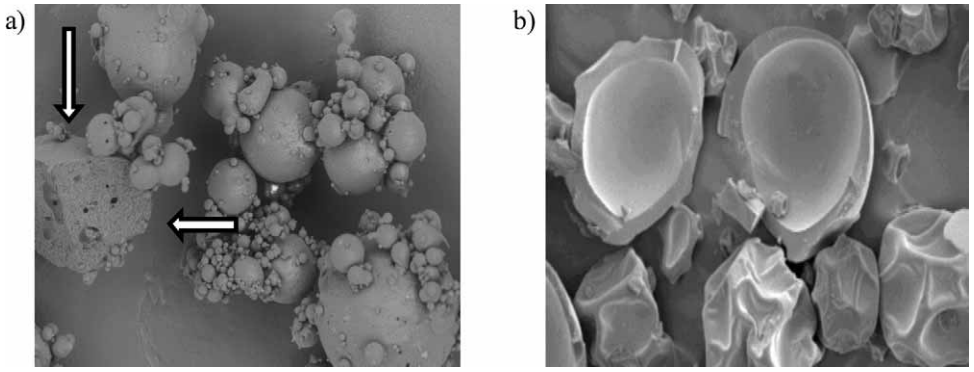

Fig. 2. Differences between particles produced by spray drying and spray chilling: a) micrograph of a fractured particle produced by spray chilling, b) micrograph of a fractured particle produced by spray drying

ticles. Both processes can be used to produce SLMs at low temperatures. However, while spray chilling is used to produce SLMs, spray coating is used for recovering particles. Thus, spray coating has the advantage of producing reservoir-type microcapsules. During the spray coating process, the particles are kept in suspension in a chamber while the coating solution is nebulised over them. When molten lipid matrices are used as coatings, the chamber is kept at low temperatures for promoting solidification of the lipid while recovering the particles.

A wide variety of materials, including lipid material, can be used for the coating of particles by spray coating and spray chilling (*23*). The use of lipids during spray coating leads to SLM features similar to those obtained by spray chilling, including the release mechanism. As the spray coating technique allows the thickness of the coating layer to be adjusted, the control of the release speed of the bioactive compound can be modulated with more accuracy, thus obtaining a desired release profile. Another advantage of the spray coating over the spray chilling technique is the amount of active material that can be used for encapsulation. Currently, spray chillers have a limitation of the maximum amount of active material in the feed. Several spray adaptations were studied using pneumatic atomising nozzle to obtain microspheres with 30 % solid active ingredients and 50 % liquid active ingredients (*10*).

Both the spray chilling and spray coating techniques have the possibility of microencapsulation of thermolabile ingredients, including enzymes and microorganisms, but it is necessary to pay attention to the thermal behaviour of the core and the carrier. Lower melting points and temperatures reached in the cooling chamber result in greater chances of success in obtaining particles without compromising the stability of the active material during the encapsulation process.

Spray coating technology presents some advantages when compared to spray chilling. One of them is the possibility of adjusting the lipid coating thickness by adjusting some process parameters. The spray coating technique also allows multilayer coating with hydrophobic or hydrophilic coats, which enables greater flexibility in the applications of these microparticles in relation to SLMs obtained by spray chilling when incorporated in other products. In addition, there are differences in the structure of the particles obtained by spray chilling and spray coating. In spray chilling, microparticles are matrix-type particles in which the bioactive component is dispersed throughout the volume of the microparticles or microspheres. In spray coating, SLMs are reservoir--type microparticles or microcapsules in which the active ingredient is enclosed by a layer or multiple layers. Thus, in the SLMs produced by spray coating, the bioactive ingredients are better protected compared to the microparticles obtained by spray chilling because in the SLMs produced by spray chilling, part of the bioactive component is dispersed on the surface of the particle.

Process Variables

The following main variables should be strictly monitored in the spray chilling process: melting point of the matrix used, temperature of the molten material during processing, chamber temperature, atomising air temperature, atomising air pressure, and feeding flow of the molten mixture (*8*). Table 2 (*2,8,10,28,29*) summarises several operation conditions of the spray chilling process.

The following conditions are critical to obtain uniform particles: low viscosity and high speed atomisation. Das and Gupta (*30*) studied the micropelletisation technique using modified spray congealing technology with a cross-linked gelatin matrix, and they suggested that the ideal viscosity for this system is approx. 0.024 Pa·s at 55 °C.

The spray chilling performance depends strictly on the spray atomisation efficiency of the molten mixture. The efficiency of the atomisation process, in turn, is directly associated with different types of devices that can perform the atomisation. These devices (atomisers) may be pressure nozzles, centrifugal atomisers, rotary atomisers, dual fluid atomisers or ultrasonic atomisers (*5*). The devices are shown in Fig. 3 (*31*). All available atomisers have similar characteristics, and they all are inadequate when spraying mixtures that have high viscosity. The feeding and pressure conditions employed in the process can exert effects on the homogeneous distribution of the particles. Albertini *et al.* (*10*) described this limiting profile assuming the difficulty of atomising a mixture containing active ingredients in a mass fraction exceeding 30 %. However, these authors suggested that by altering the design of the atomiser, there is a possibility of overcoming the threshold feeding up to 50 % (by mass).

Some studies have been conducted to investigate the effects of process parameters on particle features. Maschke *et al.* (*9*) found that by increasing the atomisation pressure from 5 to 6 bar, there is a decrease in the size of particles obtained by spray chilling. Viscosity is another important variable that influences the particle size, and it can be regulated by monitoring the temperature or by the type and amount of dispersed solids. Low viscosity (due to high temperature) results in smaller particle sizes (*9*), while higher viscosity (for example, due to the addition of solids) results in larger particle sizes (*10*).

The molten mixture, which is the feed stream, presents a characteristic curve of solidification. When the atomised droplets come into contact with the cooled medium, the material cools to a temperature of solidification. Thereafter, the temperature remains constant during the heat release of the product, and stable SLMs are then formed. Some products that have a variety of fatty acids, such as hydrogenated palm kernel oil, do not have a well-defined solidification point. The phase change may occur in a temperature range, or, depending on the condition of the molten material, the product may migrate to an amorphous form without releasing the solidification heat because a non-crystal formation occurs. The atomised material passes through three stages of cooling: liquid cooling, solidification and particle cooling. The temperature of the droplets decreases as they come into contact with cold air. When the solidification temperature is reached, the droplets will gradually solidify. As the freshly solidified particles have a higher temperature than the environment, they continue losing heat until room temperature is reached.

Table 2. Some operating conditions of spray chilling processes

Nozzle	Chamber	Temperature gradient (molten mixture→ air chamber)	Liquid spray rate	Bioactive component/carrier	Air pressure	Ref.
Dual fluid nozzle	15 cm diameter 60 cm length	70 °C → 0–5 °C	6.0 to 23.4 mL/min	Glimepiride/Gelucire 50/13R	1.09 and 1.59 bar	(*8*)
Dual fluid atomiser (0.7 mm)	–	65 °C → 10 °C	force of gravity	α-tocopherol/interesterified fat with fully hydrogenated soybean oil	0.25 MPa	(*28*)
Wide pneumatic nozzle (4.5 mm)	1.80 m in height and 75 cm in diameter	80 to 100 °C* → (25±1) °C	by both the force of gravity and the effect of Venturi	propafenone hydrochloride and vitamin E/carnauba wax, cetearyl and stearyl alcohols	1–3 bar depending on the desired particle size	(*10*)
Ultrasonic atomiser	–	70 °C → room temperature	–	verapamil HCl/ microcrystalline wax, stearyl alcohol and mixtures of the two	–	(*29*)
Dual fluid (0.7 mm)	–	48 °C → 10 °C	force of gravity	*B. lactis* and *L. acidophilus*/ interesterified fat with palm and palm kernel oil	0.98 kPa	(*2*)
Dual fluid	20 cm diameter 54 cm length	60 °C → (13±2) °C	40 mL/min	lycopene/interesterified fats and refined vegetable oils	5 bar	**
Dual fluid	20 cm diameter 54 cm length	60 to 80 °C → (15±2) °C	45 mL/min	soy protein hydrolysate/ interesterified fats and refined vegetable oils	5 bar	**
Dual fluid	20 cm diameter 54 cm length	(52±2) °C → (15±2) °C	60 mL/min	probiotic/interesterified fat with palm and palm kernel oil	5 bar	**
Dual fluid	20 cm diameter 54 cm length	60 °C → (13±2) °C	60 mL/min	ascorbic acid/interesterified fat with palm and palm kernel oil	5 bar	**

*carriers were heated to 10 °C above the melting point
**studies of microencapsulation by spray chilling from our group (unpublished)

Fig. 3. Pressure nozzle and rotary nozzle, respectively. Reprinted from**:** GEA Process Engineering Inc. (*31*)

Another important approach is to control the temperature of the lipid material during solidification because it prevents the polymorphism of fats, which directly impacts the release profile of the active material/bioactive compound. The lipid matrix can crystallise in different ways depending on different reasons related to the cooling process. For example, rapid cooling results in a lipid compound preferably in the unstable α form, and slow cooling tends to lead to the formation of the β form (*32*).

Currently, several studies have investigated the performance and improvement of spray chillers, including the use of pneumatic atomising nozzles (*10*), compact ultrasound atomisers (*33*), and ultrasonic atomisers, to disperse the atomised particles (*34*).

Ilić *et al*. (*8*) studied the influence of several parameters on the obtained particle size when using the spray chilling microencapsulation process. By varying the atomising pressure and liquid feed rate, they reported that the microparticle median sizes ranged from 58 to 278 μm and that the total process yields range from 81 to 96 %. An increased liquid feed rate was found to increase microparticle size, and higher atomising pressures were found to decrease the microparticle size. These authors concluded that the dominant process parameter regarding microparticle size is achieved by monitoring the atomising pressure in relation to the feed rate.

Morphological Characteristics of Particles

Regarding the physical structure, particles produced by spray chilling are typically matrix-type particles, where

the active ingredient is dissolved or dispersed throughout the volume of the particle and not just in its centre. As this is not a reservoir-type system, where the bioactive compound is fully covered by the carrier, these particles cannot be denominated as microcapsules but are instead denominated as microspheres or microparticles (for those produced on the micrometre scale). Fig. 4 shows particles or microspheres produced by the spray chilling technique.

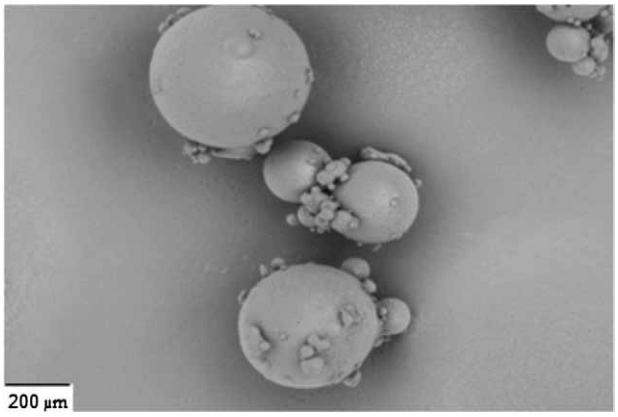

Fig. 4. Micrograph obtained by scanning electron microscopy (SEM) of particles produced by spray chilling using palm oil as a carrier (500× magnification)

In general, the particles produced by spray chilling are micrometric and impermeable to water (but not resistant to water). The particles have a spherical form, which facilitates their flow, and the active ingredient is normally distributed uniformly within the entire particle volume (*8*). These are several essential qualities for a suitable incorporation of the material due to the reduction of surface tension between the microparticle hydrophobic surface and the aqueous environment to which it is added, thus allowing the flow of particles in the food matrix (*26*). Because the process does not involve solvent evaporation, which is commonly observed in other techniques such as spray drying, the particles produced by spray chilling are usually dense and not porous, and they tend to be mechanically resistant and remain intact upon agitation (*26*).

The particle size depends on both formulating and operating parameters. Formulating parameters include the size and concentration of the active ingredient as well as the viscosity of the feed. The operating parameters consist of the configuration of the atomisation device and working conditions (*i.e.* disk configuration, disk rotational speed, air nozzle pressure, and temperature). An additional coating may be applied to microparticles obtained by spray chilling, using spray coating technology, to ensure a complete coverage of the particle and to eliminate undesirable interactions during embedding and storage (*26*).

Regarding the equipment operational configuration, there is a specific height to be used to obtain a suitable fall of particles, and such a parameter depends on the required particle size and melting characteristics of the material (*14*).

Carriers and Additives Used in Food/Feed Encapsulation

Lipids are interesting alternatives to be used as coating materials due to their ability to assume a variety of morphological states, such as emulsions, liposomes and SLMs (*35*). Several characteristics of a suitable carrier should be taken into consideration prior to the process as follows: regulatory acceptability for use in foods; stability under typical process conditions; ease of atomisation; and moderate melting temperature, which is aimed at minimising the degradation of the filling component (*36*). A proper selection of the carrier is important because it may change some properties of the encapsulated material, such as control of the release profile, masking of unpleasant taste, reduction of gastrointestinal irritation, and increased viability in some media (*2,9,37–39*).

The use of surfactants directly impacts the stability of SLMs. The choice of the surfactant agent depends on the compound to be encapsulated and also on the application of the final product. The surfactant should not provide undesirable characteristics to the final product (*40,41*).

The particle release profile can also be influenced by changes in lipid matrices, concentration of surfactant (such as soy lecithin) and by some production parameters (*42*). These surfactants are added to provide greater stability to the capsule as they aid in effective connection between the active ingredient and the matrix. Several researchers have analysed the incorporation of lecithin in the lipid matrix to optimise the incorporation of the active ingredient, and they showed that the encapsulation efficiency increases linearly with increasing concentration of lecithin (up to a certain percentage). This effect was attributed to the development of micelles in the middle of the lipid matrix, which contributes to an additional incorporation of the active ingredient (*43*).

Release Mechanism

Microencapsulation enables controlled release of the active ingredient. In response to specific stimuli, the particle releases its active ingredient in a specific environment. The release mechanisms of bioactive compound from solid lipid microparticles in the intestine can be associated with the presence of digestive enzymes, particularly pancreatic lipase and bile salts (which act as detergents). Another possible release mechanism is through the increase of the temperature above the melting point of the carrier.

The particle type, geometry and composition define the release mechanism of the active ingredient. The processes that employ hydrophilic carriers generally trigger a more rapid release of the active ingredient in relation to those agents that have a lipid carrier (fats and waxes), thus retarding the release of the active compound (*44*). The release of active ingredients from particles obtained by spray chilling can occur *via* erosion and leaching of the matrix. Depending on the type and concentration, some surfactants can dramatically affect the dissolution rate of the matrix.

With regard to SLMs, the release rate can be activated by enzymatic degradation or temperature. When activated by enzymes, lipids are degraded by the action of lipases in the digestive system. When activated by temperature, the active ingredient is released in response to the temperature change affecting the physical state and release rate of the bioactive compound. In this context, two distinct concepts should be noted: (*i*) temperature sensitivity, which is an important characteristic that needs to be taken into consideration for those materials that expand or collapse when a critical temperature is reached, and (*ii*) activation by fusion, which is related to the melting of the carrier, such as those made of modified lipid or wax, in response to the increasing temperature (*45*).

With respect to vitamins, which are potentially oxidisable compounds, the microencapsulation process can also increase their chemical stability and improve the masking of possible off-flavours or odours. In this case, the vitamin must be released after ingestion or directly in the stomach or intestines. For this purpose, hydrophobic materials are employed, although cellulose derivatives and cross-linked proteins can also promote enteral release (*46*).

The concept of release after ingestion, which reflects the actual digestion mechanism of the carrier, is connected with the condition of microparticles containing probiotics. These microorganisms are beneficial to the host's health. However, to be effective, the probiotic strain must remain alive after passing through the stomach because acidic medium and the presence of oxygen are harmful to the microorganism (*47–49*). Therefore, the timing associated with the fact that lipid digestion (carrier) effectively takes place in the intestine where the probiotic strain must act reflects the efficiency and the basic condition to apply this technique.

In the SLM, not the entire bioactive ingredient is effectively encapsulated as it remains on the particle surface. This condition leads to a high initial release of this bioactive compound and is followed by the osmotic diffusion (even if in a small amount) in the matrix. Mechanical breakdowns and melting of the lipid matrix elements also release the bioactive compound (*6*). Additionally, other factors influence the release kinetics, such as osmotic force, slow diffusion rate of water through the imperfections of the capsule, and mechanical disruption (*6*).

Zaky *et al.* (*50*) studied the factors that impact the release kinetics of protein in triglyceride microparticles prepared by spray congealing. These authors investigated the effect of the protein particle size, morphology and distribution in the release of the encapsulated microparticles by confocal laser scanning microscopy (CLSM). The results showed that water entered and dissolved the protein, which led to the formation of water-filled pores allowing the diffusion of the active ingredient in the outside part of the matrix. The penetration of water and release of protein are strongly correlated, showing the importance of more realistic three-dimensional analysis to study the distribution of the protein within the particle.

Furthermore, SLMs have the advantage of lipids being easily digested in the intestine by lipases causing the release of active compounds in the vicinity of their site of action. Thus, it would be interesting to encapsulate probiotics, prebiotics, some proteins, peptides, iron, ω-3 fatty acids, vitamins, antioxidants and many other compounds.

Advantages of the Process

Currently, the use of spray chilling has increased due to the numerous advantages associated with the technological process, such as speed, performance, and relatively low cost. Once this technique does not require the use of water or organic solvents, the elimination of residual solvents will not be required. Moreover, it is a fast, safe and reproducible physical process because it is associated with an easy adjustment of particle size (*51*).

In the last two decades, spray chilling has been considered to be an environmentally friendly technique compared to other procedures, such as spray drying. Spray chilling also reduces energy use and time of operation (*52*). Another positive aspect is the ease to scale up the production because this technique can be operated continuously with the elimination of some manufacturing steps (*53*).

Other advantages of spray chilling include the use of low temperature in the process, thus not requiring heat. Moreover, there is usually controlled release of the SLM contents around the melting point of the carrier and by digestion of the carrier in the intestine. The SLMs are almost perfect spheres, which are structures that give free-flowing powders.

Disadvantages of the Process

Although SLMs present some advantages, some of the technological disadvantages are as follows: low encapsulation capacity of the active material; possibility of expulsion of the active ingredient by the matrix during the particle shelf life; and the degradation of lipid carrier applied for the dispersion development, which may affect the active ingredient material, thereby influencing its stability and/or its release profile (*54*).

Another limitation to be considered is the proper choice of the material to be encapsulated. The encapsulated material should be stable at the carrier melting temperature. For many thermolabile or unstable food ingredients, the degradation temperature is recurrently low, thereby reducing the possibilities of using a suitable carrier. The use of molten mixtures should also be considered, which requires operational attention to avoid agglomeration and solidification of such materials in the production line, thus impairing the efficiency of the atomisation process.

The method has a drawback related to fast cooling rates that sometimes crystallise the lipid matrix into a polymorphic form (unstable α arrangement), which leads to the development of disordered chains and/or undesirable orientation, thus promoting lower barrier properties due to the tendency of more stable arrangements resulting in the release of the active ingredient (*32,53*).

Other possible disadvantages include the effect of particles on food texture, which depends on the particle

size, as well as the possibility of particles to float in liquid systems. In addition, SLMs prepared by spray chilling are insoluble in water due to the lipid carrier, which can limit or not allow some applications. Moreover, the matrix structure of the particles obtained by spray chilling leads to the dispersion of the part of the active material that is not protected on the surface of the particles.

Encapsulation of Functional Ingredients by Spray Chilling

Microencapsulation has been widely used in various industrial applications. However, even though microencapsulation by spray chilling has been thoroughly studied in the last years, especially in the pharmaceutical and veterinary production, it has just a few food-based industrial applications. Although the use of encapsulation is fairly new in the food field, the interest in spray chilling technology has grown significantly. This interest grows as the need arises to develop alternatives for new applications. In this regard, the encapsulation of food ingredients that present functional properties has been studied more vigorously.

A common characteristic of many functional ingredients is that they are subjected to rapid inactivation or degradation (*55*). Thus, the incorporation of bioactive ingredients in foods presents many challenges, especially with respect to their stability during processing and storage and the need to prevent undesirable interactions with the food matrix (*56*). Encapsulation technology arises in this context as a viable alternative to ensure that the functional ingredient may be active in the body, that is, it must be delivered to the site of action at a relevant concentration for a sufficiently long period of time to do its work. Furthermore, by encapsulating the active ingredient into a food matrix, the loss of food quality is avoided, thus ensuring the product's intrinsic characteristics (*57*).

Although rarely studied by food researchers, spray chilling has several advantages that make it an important alternative for encapsulation of functional ingredients. The process is fast, simple and relatively inexpensive. Moreover, spray chilling can be applied to different types of functional ingredients, such as vitamins, minerals, antioxidant compounds, proteins, protein hydrolysates and probiotic microorganisms.

The use of lipid material as a carrier together with the use of low temperatures and the absence of organic solvents during the process are some of the advantages that have encouraged researchers to explore the microencapsulation of food ingredients by means of spray chilling to overcome technological limitations in their use. The process has been tested for different purposes, including the development of controlled release systems and protection of food ingredients against adverse environmental conditions, such as the presence of light, oxygen or unfavourable pH. Furthermore, the process has been tested for taste masking and encapsulation of flavour ingredients, such as peptides and proteins, with therapeutic potential.

Encapsulation of Probiotic Microorganisms

Although the first reports involving studies with probiotics are not recent, the incorporation of these microorganisms in foods is still considered a challenge to the industry. Probiotics represent one of the most important classes of functional ingredients and are defined as live microorganisms that, when administered in adequate amounts, confer a health benefit to the host (*58*).

To be able to present prophylactic or therapeutic functions, probiotic microorganisms need to have their functional structures preserved until they reach their site of action in a high enough quantity to be capable of positively influencing the local microbiota (*55*). However, a number of factors may compromise the functionality and/or the viability of probiotic microorganisms. These factors are related to the technological processes used for food production, the physicochemical characteristics of the food matrix to which the probiotic is added and its passage through the gastrointestinal tract until its arrival in the intestine, which is its primary site of action (*59*).

Microencapsulation is a promising alternative to overcome the problems related to the vulnerability of probiotic cells to adverse factors, including the presence of oxygen and acidic media (*60,61*). Various techniques have been used for the encapsulation of probiotics, and each technique presents particular advantages and disadvantages. Among the available techniques, the following are highlighted: extrusion, atomisation or spray drying, emulsion, coacervation, spray coating and immobilisation in fat or starch granules (*62*).

The spray chilling technique has several advantages in the encapsulation of probiotic microorganisms. The absence of organic solvents during the encapsulation process is one of these advantages. The use of these organic chemicals is necessary in other techniques and may be toxic/lethal to the probiotic strains, which compromises not only the viability but also their functionality.

With spray chilling, it is possible to develop SLMs only by using the lipid matrix and a probiotic culture without using any further substance. Furthermore, spray chilling does not require the application of high temperatures to produce the microparticles that contain probiotics, which differs from the spray drying and extrusion techniques. However, there is the need to melt the lipid matrix. It is possible to use fats with a low melting point, which prevents the microorganism from being affected by the temperature. Another good aspect of the spray chilling technique is related to the release mechanism of the active ingredient. If the matrix is composed of fats, the release of the bacteria occurs directly in the intestine as a result of the action of lipases present in the intestinal lumen.

Pedroso *et al.* (*2*) encapsulated probiotics by spray chilling using interesterified palm fat and palm kernel, and they found that the SLMs provided protection for *Bifidobacterium lactis* and *Lactobacillus acidophilus* against the simulated gastric and intestinal fluids. Moreover, they obtained promising results for viability during storage when refrigerator and freezer storage temperatures were applied.

Pedroso *et al.* (*3*) also reported that the encapsulation of *B. lactis* and *L. acidophilus* in cocoa fat by spray chilling provides protection and cell viability. Although *in vivo* gastrointestinal studies demonstrate protection, the viability during storage of SLMs is low. This study indicated that spray chilling using fat as a carrier is an innovative technology for the protection, application and delivery of probiotics. The use of spray chilling favours the microencapsulation of bacteria mainly because it does not employ high temperatures or toxic solvents in the process.

Encapsulation of Vitamins and Minerals

Vitamins may be added to food products to enhance their nutritional value or to act as additives, such as antioxidants or colouring agents added to cured products. Herein, essential minerals are usually added to foods to enhance their nutritional value. The nutrients in these foods may be hampered by the low stability of vitamins or by their ability to interact with other food compounds, thus promoting undesirable changes to the food chemical and sensory properties. Iron is a classic example because its bioavailability is severely affected by interactions with food compounds (*e.g.* tannins, phytates and polyphenols), and iron may also act as a catalyst for the oxidation of fats and vitamins (*56*). Microencapsulation has been studied as an alternative to add these nutrients to foods. Once more, the choice of the encapsulation technique is a limiting factor because the vitamin content and chemical stability can be negatively affected by several factors related to the microencapsulation processes. Some technological features related to spray chilling, such as not using organic solvents and high temperatures, have stimulated further studies involving the encapsulation of these nutrients.

To develop a stable salt for the fortification of iodine, iron and vitamin A, Zimmermann *et al.* (*63*) applied the spray chilling technique to package potassium iodate, micronised ferric pyrophosphate, and retinyl palmitate into microcapsules (mean particle size of 100 μm) using hydrogenated palm oil as a carrier. The purpose of microencapsulation was to ensure iron bioavailability to prevent colour changes and to ensure the chemical stability of iodine and vitamin A. The microparticles were used in a randomised, double-blind trial that evaluated the bioavailability of these nutrients and the nutritional deficit in the studied population. The results showed that iron, vitamin A and iodate are highly stable when added to salt, and its use for a six-month period is efficacious in reducing the prevalence of iron, iodine, and vitamin A deficiencies in the studied population.

The addition of vitamin E to foods represents an interesting strategy to ensure its adequate daily intake. However, the addition of vitamin E into foods may be difficult due to its chemical instability, that is, it slowly undergoes oxidation by atmospheric oxygen by means of a reaction catalysed by light and heat in the presence of metals. Based on this consideration, Gamboa *et al.* (*28*) used the spray chilling technique to encapsulate α-tocopherol using interesterified fat with no *trans* isomer fatty acids prepared with fully hydrogenated soybean oil and soybean oil in the mass ratio of 70:30. The results showed values for the encapsulation efficiency above 90 %, and high levels of retention of the active agent were attained. Thus, the α-tocopherol encapsulation by spray chilling was successful.

Vitamin C is another important nutrient for food companies. This vitamin is widely used by the industry to supplement foods or to act as an antioxidant agent. However, the stability of vitamin C is considerably reduced when it is added to solutions due to the influence of temperature, pH, and presence of oxygen and metal ions (*64*). In a study performed by our research group (unpublished data), ascorbic acid was encapsulated by spray chilling using interesterified fat and palm kernel oil as a carrier. The results showed that the stability of ascorbic acid was substantially higher compared to free ascorbic acid. By measuring the instrumental colour, it was possible to observe a significant colour change in free ascorbic acid stored at 22 and 37 °C. This effect was associated with the development of compounds resulting from the degradation of ascorbic acid. In contrast, the colour change observed in the encapsulated ascorbic acid stored under similar conditions was considerably smaller, thereby confirming that spray chilling is a useful alternative to enhance the chemical stability of ascorbic acid.

Zoet *et al.* (*65*) patented microencapsulation of fat-soluble vitamins (*e.g.* vitamin D) by a spray cooling/chilling process. The process included making a homogeneous solution of a fat-soluble vitamin with the fatty phase preferably being a hydrogenated vegetable oil, such as palm kernel oil, cotton seed oil, sesame seed oil, palm oil, carnauba wax or beeswax. These authors suggested that the carrier should have a melting point in the range from 45 to 90 °C, and their invention relates to the use of microencapsulated fat-soluble vitamins, which was defined herein before as an ingredient in animal feed.

Encapsulation of Proteins and Protein Hydrolysates

The application of hydrolysed proteins that have a specific biological activity for the development of functional foods is of great interest for industrial purposes. However, the bitterness of hydrolysates and their low structural stability may limit the use of these ingredients in food products. Additionally, some protein hydrolysates are hygroscopic and reactive (*66*). In this case, encapsulation can be useful for masking the bitter taste and preventing the high reactivity of protein hydrolysates, which may cause undesirable changes in food. Although scientific studies are limited in the literature, the spray chilling technique has been mainly studied for masking the taste of antibiotics (*38*).

Taste masking is often cited in the literature as one of the main purposes of drug/food microencapsulation. However, spray chilling has not been explored for this purpose by food companies, and only few studies have explored the application of spray chilling microencapsulation in the production of drugs. Yajima *et al.* (*38*) reported the microencapsulation of clarithromycin, which is a macrolide antibiotic that has a strong bitter flavour.

These authors found that when clarithromycin was microencapsulated using glyceryl monostearate and aminoalkyl methacrylate copolymer, the drug did not dissolve in the mouth but instead was released immediately in the intestinal tract, resulting not only in taste masking but also offering a high bioavailability.

Another application of spray chilling that has not been fully explored is the microencapsulation of proteins and peptides. Although the use of these components in pharmaceutical formulations has experienced rapid growth since 1980 with the advent of recombinant DNA technology, the development of delivery systems is still considered challenging due to difficulties in maintaining the stability of proteins (*67*).

The choice of the technique for encapsulation is limited by the need for maintaining the integrity and bioactivity of the protein, and the use of high temperatures and organic solvents are two factors that can be directly linked with protein denaturation (*68*). Taking this into consideration, the use of spray chilling can be advantageous because the process does not require the use of organic solvents or elevated temperatures. Moreover, carriers that have a high biocompatibility, such as triacylglycerides, are used in formulations for parenteral delivery (*9*). Several publications have addressed the use of microencapsulation for the development of controlled release systems for peptides and proteins of high therapeutic potential, including lactase (*69*), somatostatin (*70*), insulin (*9*), growth hormone (*71*) and casein hydrolysate (*72*). Recently, a study conducted by Maschke *et al.* (*9*) showed positive results regarding the stability of insulin towards the spray chilling microencapsulation process. Using glycerol tripalmitate as the carrier, these authors demonstrated that it is possible to obtain spherical microcapsules with different protein fractions (0.5, 1 and 2 %) with particle sizes between 182.2 and 315 μm, and their experimental results also showed that insulin is released for at least 28 days, indicating a possible release for up to six months.

Di Sabatino *et al.* (*73*) produced lipid-based particulate systems by a spray congealing technique using bovine serum albumin as the model protein, and they reported that the solvent-free drug encapsulation process can be successfully employed to obtain particles with high (10–20 % by mass) protein loading without altering its structure.

Despite the therapeutic potential of some peptides, proteins and protein hydrolysates, their low structural stability (especially for proteins), bitter taste (of some peptides and hydrolysates), hygroscopicity, allergenicity and reactivity represent obstacles for their use with the aim to produce functional foods. In addition, some of these products should not be digested before they reach the gut because digestion can destroy their therapeutic effect. Consequently, the use of these protein hydrolysates is challenging for both food industries and researchers. Microencapsulation by spray chilling technology may be an alternative for reducing these problems. As this process does not require the use of organic or aqueous solvents and high temperatures, it could be suitable for protection and production of protein delivery systems.

Conclusions

Although spray chilling encapsulation is not a new method, it is exploited to a lesser extent compared to spray drying encapsulation, especially in the food science field. In the last 10 years, however, mainly in the veterinary and pharmaceutical fields, there has been a significant increase of published papers involving spray chilling microencapsulation. Many recent research articles have aimed at understanding the process variables of spray chilling encapsulation and to propose changes in the equipment and/or procedures, as well as to propose new carriers in order to improve the process performance in relation to the development of microparticles. It is likely that with the increase of successful reports on spray chilling encapsulation, more researchers will be interested in using this technique, thus enabling further studies and new applications in various technological fields.

This technique represents an excellent alternative for encapsulation of various food ingredients with the possibility of providing controlled release, taste and aroma masking as well as conferring a remarkable stability for different compounds. In addition to the above-mentioned applications, the use of spray chilling technique has a potential to address many other limitations in the food industry, such as the reduction in the volatility of flavourings and the hygroscopicity of powders, without excessively increasing the commercial value of food ingredients once it becomes a low-cost technique.

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