

Aroma Encapsulation in Powder by Spray Drying, and Fluid Bed Agglomeration and Coating

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ABSTRACT

The objective was to produce aroma powder able to replace liquid aroma in sweet paste composition. Three aroma molecules corresponding to top, middle and end notes as ester, aldehyde and lactone, were studied in a mix with a ratio of 96.5, 1, 2.5% w/w respectively as in real aroma. An aqueous flavour emulsion (<5µm) was prepared with water (60% w/w), maltodextrin/acacia gum (ratio 3/2) as carrier (32%) and aroma (8%). The powders were prepared using spray drying and fluid bed processes, with three structures: spray dried powder, agglomerated spray dried powder, and coated agglomerates, keeping constant aroma concentration of 20% w/w in powder. The flavour/carrier emulsion was spray dried with inlet air temperatures 140 or 160°C. Then the thin atomized powders (<30µm) were agglomerated in a fluidized bed with hot air (50-60°C), by top spraying water or flavour/carrier emulsion. Then agglomerates were coated in fluid bed with a dry thin layer of emulsion. The powders properties were compared for size, water content, water activity, wettability, size of reconstituted emulsion and organoleptic behaviour when incorporated in chewing gum paste (0.6% w/w). The different processes led to different median diameters d_{50} : <30 µm for spray dried powders, 100 to 1000µm for agglomerated and coated powders. Water content was inferior to 8g/100g dry matter and water activity inferior to 0.3. Wettability of agglomerates was improved compared to spray dried powders. During the process steps (spraying, heating, drying) aroma losses were low (<15%), keeping global composition. No coalescence of aroma was observed in reconstituted emulsions. Coating process gave particles with smoother surface. The atomized and agglomerated powders gave a global positive answer for both taste and intensity when mixed in chewing gum paste compared to liquid aroma. The properties of the tested coating must be improved.

Keywords: encapsulation; flavour; spray-drying; fluidized bed; powders.

INTRODUCTION

Aromas in food products bring typical characteristics named taste and flavour. The aroma compositions (named « aroma ») are complex mixtures of different molecules, more or less stable with temperature, light, oxygen, soluble or not in water. They are often very strong if they are pure, which imposes to dilute them in a neutral support. They must often be protected against environment till their utilization in specific conditions, where their final release must be controlled.

In this study the objective was to replace liquid aroma by aroma powder in sweets, chewing gum. These products must deliver the right balanced flavour in specific conditions of temperature, humidity, with a persistent intensity. The global perception during chewing will depend on the composition in various molecules, on their release when the humid neutral saliva reaches the active component. These properties may be built by using flavour containing microcapsules, able to release core material progressively through mechanical stress and diffusion/dissolution.

Microencapsulation has become an attractive approach to convert liquid food flavorings into a dry and free flowing powder form, easy to handle and to incorporate into a dry system^[1, 2, 3, 4]. The powder properties depend closely on the initial formulation and on the production process: fraction of aroma in the matrix; integrity of shell; practical conditions about the initial formulation as viscosity, aroma dispersion, temperature required^[5]. The dry final powder grains (µm to mm) will be made of aroma dispersed in wall material, with a controlled size distribution. Different sizes may give different time release. Microcapsules must ensure protection of aroma against oxygen, humidity especially during storage. The main encapsulation techniques, more or less expensive, are spray drying, spray cooling, drum drying, extrusion, coacervation. Fluidised bed process is used to modify the solid particles properties as size, solubility, stability, by agglomeration and additional coating keeping in mind the difficulty of coating very thin particles (<100µm)^[5, 6, 7, 8, 9].

Spray drying remains the dominant method to produce a dry aroma emulsion^[10]. This continuous process uses the spraying of emulsions in small drops with high surface exchange for heat and mass transfer when in contact with hot air. Solvent evaporation from initial drops is very fast and the temperature reached by the product will be low, consequently minimizing alteration and loss of volatile aroma components able to modify the final aroma profile in powder. Volatile losses may occur mainly during the early stages of formation and drying of drops, before the formation of a dry surface layer retaining flavour molecules^[11,12]. Besides the composition of wall materials, the main loss factors for aroma molecules are related to molecular weight, relative volatility, polarity type, and the ratio aroma/wall. Optimal conditions will depend on spraying mode and drop fast initial drying in relation with the feed emulsion properties as high total solid content and small size ($\sim 1\mu\text{m}$)^[10,13,14]. The nature and quantity of the food grade shell materials are important for an optimal dosage, and a good protection during storage against humidity and oxygen, insuring long shelf life. They will participate, even in small proportion, in composition, texture and taste of the final product. The mechanisms of release, mainly disruption, diffusion, will be considered with the global final product composition and structure. In the case of powders added to a formulated paste, the solid particles must be limited in size not to be perceived in the mouth. The properties of wall materials differ by molecular weight and stability with temperature, including phase transitions. They may be hydrophilic, hydrophobic, some with emulsifying properties. The main materials are polysaccharides, celluloses, proteins, gums, fats, waxes, glyceride fatty esters, modified starches, alone or in combination. Studies are mainly reported on pilot or laboratory equipments, for model oil molecules^[6,13]. The encapsulation efficiency is usually characterized by the percentage of oil encapsulated, the fraction on surface of particles, and the stability against oxidation during storage with variable humidity and time^[15,16,17]. For example the rate of release and oxidation of encapsulated D-limonene in carbohydrates was investigated with structural changes and glass transition temperature of the capsule matrixes in relation with relative humidity^[13]. In parallel, single drops drying (in constant conditions) was studied to understand the drying mechanisms^[18]. To control the aroma release, multi-stage encapsulation was proposed with different aroma molecules distributed between the core (solid, paste, liquid) and the solid external layer often made of sugars and polyols. In this layer, aroma is usually protected to avoid aroma and water migration, using various encapsulation methods and supports. That provides two levels of release^[19,20,21,22,23,24]. For example, an oil-in-water emulsion with coffee aroma was sprayed on coffee powder to form a dry coating layer enriched in aroma, with release in hot water^[25]. Spray dried powders have small diameter, in the range 10-30 μm , resulting in poor properties for reconstitution and handling. The aggregation of these small particles into large size porous agglomerates leads to powders with improved properties as wettability, dispersibility in liquid. Agglomeration is possible by creating contacts and links between particles. The main parameters are the surface state of particles, the contact duration and intensity, and the adhesion mechanisms to form strong bridges. Those bridges may be made of the same material as the basic powders or of a different material. In the case of flavour powders with wall materials able to be rehydrated and to form inter-particle bridges, the fluid bed process will be favoured using wet growth agglomeration. Water or aqueous binder solution is sprayed on the surface of the moving fluidised particles. If humid sticky particles collide, liquid bridges will form at the points of contact, bridges being subsequently dried (*agglomeration*). If the sprayed solution dries on the particle surface before collision the surface will be coated progressively with binder deposit (*coating*). In agglomeration, it is important to avoid collapse of carriers, this phenomenon being influenced by glass transition temperature, water activity, process temperature, spraying rate^[26].

The present study focused on the preparation of dry emulsion powders with several shapes and structures, keeping a constant chemical composition for carriers and aroma (20% w/w). Powders were first prepared by spray drying, then agglomerated in a fluid bed with different binder solutions, and finally coated with dry aroma emulsion. The powders obtained with the different processes were compared for their physicochemical properties and evaluated by incorporation in chewing gum base paste.

MATERIALS & METHODS

Products

The support matrix was made of Maltodextrin (MD) (Glucidex DE12, Roquette, Fr) and Acacia Gum (AG) (Instant Gum AA, CNI, Fr), with bulk densities 0.47 and 0.3 g/cm^3 respectively. Both soluble in water, they were used with a weight ratio MD/AG = 3/2^[27].

The pure aroma molecules were: T, top note, ester, water soluble; C, middle note, aldehyde; F, end note, lactone (Fruitafloor, Fr), all with boiling point $> 130^\circ\text{C}$, and a molecular weight in the order $T < M < F$. M1 was a mix: (T) 96.5%, (C) 0.93%, (F) 2.5% (% w/w) representing $\sim 30\%$ of the real aroma composition M2.

Emulsion MD/AG/M1 (ou M2) + water (40% w/w)

Aroma was added to the carrier aqueous solution MD/AG (3/2) (30°C ; stored 12h at 4°C). At ambient temperature, the aroma (pure, M1, M2) representing 20% of the total solid content (MD/AG: aroma = 4:1) was incorporated under mechanical agitation. The final emulsion was obtained by agitation with Polytron (PT-3000,

Kinematica); 4000 rpm, 25min). The emulsion size was inferior to 5 μ m. Volumes of 4.5L were prepared for each spray drying trial. The emulsion AG/M2 (1.6/1, 67.5% water) was prepared with the same procedure. Analyses performed on emulsions and powders and the techniques used are described in Table 1.

Table 1. Analyses and techniques

Analyses	Techniques
Viscosity of emulsion	Viscosimeter co-axial cylinders (Rheomat 180, Lamy, Fr)
Size	Laser granulometer (MS2000, Malvern, Fr) : wet mode for emulsion (initial or reconstituted); dry mode for powders (<100 μ m)
Distribution of size	Sieving for powders > 100 μ m (14 sieves; ϕ 53mm; 100 μ m to 3150 μ m; 5g)
Water content of powder	Oven 105 $^{\circ}$ C - 24h
Water activity/Sorption isotherm	Aw-meter (Thermoconstanter, Novasina; GBX FA-st lab)
True, packed, bulk density, porosity of powder	Air pycnometer (Accupyc 1330, Micromeritics, Fr) Graduated test tube with/without tapping
Wettability of powder	Contact time with water surface before disappearance (5g, 100ml, 20 $^{\circ}$ C)
Observation of solid particles	Optical microscopy (BX60, Olympus) & SEM (JSM-5200, Jeol)
Chromatographic aroma analysis	Hewlett Packard 5890, capillary column +IFD (after extraction)
Sensory evaluation	Chewing gum paste with 0.6% w/w aroma

Spray drying

The flavour emulsion was spray dried in a co-current spray dryer (Niro Minor, Niro, Dk; 0.5m³, h = 1.2m) equipped with a centrifugal wheel atomizer (Figure 1). The process conditions were: air inlet temperatures of 140, 160 $^{\circ}$ C, with constant feed rate respectively 46, 28g/min; rotational speed of atomizer 25,000rpm (5bars); air flow rate 110kg/h. Temperatures for inlet/outlet air, for liquid feed and final powder (<50 $^{\circ}$ C) were measured. The powder was separated from air in a cyclone and stored either in plastic bags or glass containers. The process powder yield was defined as the ratio between the total collected powder and theoretical powder quantity from the sprayed emulsion.

Fluidised bed agglomeration and coating

In a batch fluidizer (Uni-Glatt, Ge) solid particles (~500g) were fluidised by hot air (70 $^{\circ}$ C) and humidified on surface by spraying binder solution to get finally, after drying, either bigger agglomerates or coated individual particles (Figure 1). The air bed temperature was maintained between 47 and 60 $^{\circ}$ C. All the parameters were adapted to prevent any collapse of the bed. After initial stabilization of temperatures, the sprayed liquid flow rate was progressively increased (i.e. 2.9 till 16 g/min). The air flow rate was adjusted to compensate the increased weight of particles (i.e. 80 to 199 kg/h) keeping a constant bed height under the nozzle. Duration was fixed by liquid quantity to spray and/or particle size to reach. The process powder yield was the ratio between the final powder mass and the theoretical mass of solids.

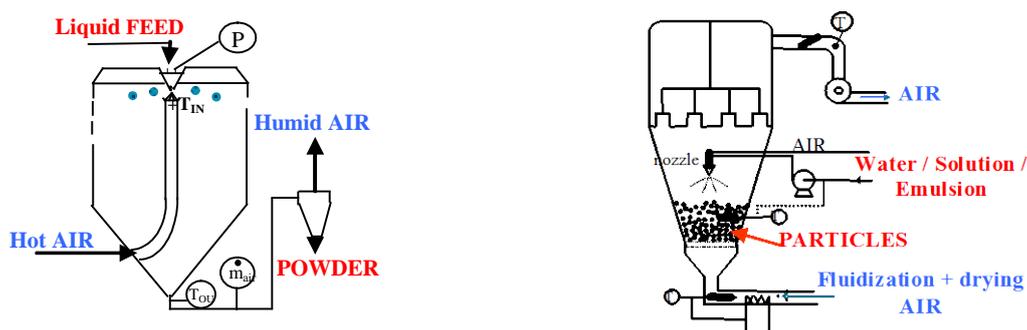


Figure 1 - Spray dryer Niro Minor and Fluidised bed Uni Glatt.

RESULTS & DISCUSSION

Aroma emulsion was first spray dried in thin powder. Then agglomerates were produced in fluidised bed in three ways by top spraying binder: water or aroma/carriers emulsion on fluidised atomized aroma powders; acacia gum/aroma emulsion on fluidised maltodextrin powder.

Coating was realised by spraying aroma/carriers emulsion on agglomerated particles (>200 μ m), with two spraying modes, top or bottom (with internal Wurster tube). Coating is a slow process with a layer built progressively with successive deposits of emulsion drops and drying.

All these conditions finally led to flavour powders keeping the same total chemical composition, but with different structures, the aroma being present inside the particles (atomized, agglomeration with water), and/or in the bridges of agglomerates and in the coating layer (Figure 3).

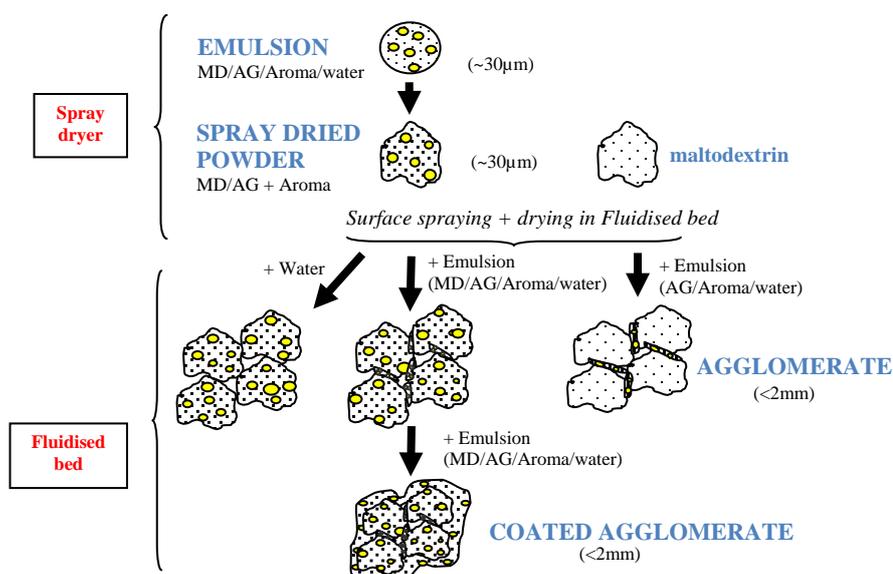


Figure 3 - Production of spray dried powder, agglomerates and coated agglomerates.

Spray dried aroma powders

Spray drying conditions were determined along preliminary trials using emulsions with the pure aromas alone and mixed. Then emulsion MD/AG/M1 was spray dried with two inlet air temperatures 160 and 140°C, at constant air flow rate, leading to powders SD160 and SD140. The liquid feed flow rate (respectively 45.8 g/min and 27.6 g/min) was adapted to keep the outlet temperature between 70 and 90°C, to be able to collect powder without sticking. The powder yield was superior to 75%. Drying was more efficient for 160°C than for 140°C and faster (respectively 109 and 232 min to spray dry 4600g emulsion). For the lower inlet air temperature the initial drying of the drop was less efficient, leading to slower drying along the chamber. Compared to SD140, SD160 powder corresponded to lower mean powder water content (3.8 and 4.5 g/100gDM), water activity (0.1 and 0.13), and bulk density (0.49 and 0.51g/cm³). Similar results were observed in literature^[6].

In both cases, the mean particle size was low, 20-30 µm, with a narrow size distribution, powder wettability was bad (>15min). The reconstituted emulsions showed no coalescence of aroma drops.

Total aroma content in powders was 14% (SD140) and 16.7% (SD160) (retention 70% and 83%). The relative composition in T,C,F was preserved, with a little decrease for F. During sensory tests the three molecules were perceived distinctly, strongly for powder SD160 and satisfactory for SD140.

One trial with the emulsion MD/AG/M2 with an equipment of bigger size (x 6) was done in similar conditions (air 160°C). The resulting powder gave encouraging results concerning the aroma retention and perception.

Agglomerated spray dried powders in fluidized bed

The final objectives were to have aroma encapsulation, not only inside the particles but also more accessible on their surface as in a coating layer. It was necessary to increase the atomized powder size by agglomeration before coating in fluidised bed. For the agglomeration it was possible to spray either water to induce solid links between sticky made surfaces, or the binder emulsion to find it finally also into the solid interparticles bridges^[8, 28].

The powder MD/AG/M2 obtained with the bigger spray dryer was agglomerated by spraying water on the fluidised particles. The T, C, F aroma content was decreased (5.3% instead of 6.4%) keeping a good relative proportion of T,C,F. Air flow and temperature in the fluidized bed during the agglomeration process probably stripped part of the oil remaining on surface. Size distribution was very large, and powder properties similar to the other trials we shall describe further.

In another trial, fluidised maltodextrin powder (240g) was agglomerated by spraying the aqueous emulsion AG/M2 (800g). The conditions were applied to get a final powder with the same composition MD/AG/aroma (20% w/w aroma in powder, with 6.4% of T,C,F for M2). The size increase was rapid till 160µm and then slower, leading to a narrow final size distribution around 450µm. The final concentration in molecules T,C,F was low (1.4 instead of 6.4% with M2), but the proportion of each molecule was preserved. In that case the

maltodextrin was the support and the whole aroma was associated with acacia gum in the bridges between particles, with not enough protection during the long coating process (93min), which may explain losses.

Powders SD140 and SD160 were agglomerated by spraying MD/AG/M1 emulsion on 500g of fluidised SD powder. In that case aroma was distributed in grains and in bridges.

For agglomeration of SD160, the powder yield was good, 88%. The size distribution was large, with a median diameter d_{50} of 608 μm : size till 1800 μm and 46% <450 μm . After spraying 340g emulsion on 500g powder (58min) the aroma concentration was 17.6% with a good ratio T,M,F.

For SD140 agglomeration, two trials (SD140A, SD140B) were compared by spraying different quantity of emulsion. With a lower quantity sprayed (450g, 53min), the powder size distribution of SD140A was bimodal: 133 μm (14.6% with $d < 200\mu\text{m}$) and 486 μm (85% with $d > 200\mu\text{m}$), with a high mean powder water content (7.3g/100g DM) and yield of 85%. Some coalescence of aroma was observed in the reconstituted emulsion.

For SD140B with more emulsion sprayed (600g, 70min) with slower feed flow rate increase, the size distribution was large till 2000 μm . A high fraction (36%) inferior to 200 μm may be due to no agglomeration of small particles or appearance of new particles by breaking of some agglomerates. Lower mean powder water content 5.5g/100g DM was observed, with a high powder yield 93%. The total aroma content was 16% instead of 17% for SD140A, probably due to longer heating.

For the three agglomerated powders SD160A, SD140A, SD140B, the mean preservation of aroma molecules T,C was 80%, and 60% for the end note F. More specifically, preservation was better for shorter processes and losses higher for fine particles. The sensory perception was strong for SD160A and with separation of the three notes for the others, SD140A being globally too strong and SD140B too short in persistence.

All the agglomerated powders have low bulk density (0.40kg.m⁻³) and improved wettability (<7min) due to their porous structure compared to the atomized powder. The wettability was not equal for all the particles fractions, the big agglomerates being more rapidly wetted.

For atomized and agglomerated powders, the aspect of surface was not smooth, with numerous asperities.

Coated agglomerates

The three types of agglomerates SD160A, SD140A and SD140B, were fluidized for coating by top spraying the emulsion MD/AG/M1 (no modification of composition). The sprayed quantity of emulsion was calculated to cover regularly the surface of particles (mean diameter) with a coating layer of 10 μm . No significant increase of size was expected.

For SD160A (346g emulsion sprayed on 484g powder, 32min) the coating yield was 97%. Powder size distribution gave one peak with d_{50} of 750 μm , (disappearance of fine particles 20% < 315 μm), showing some agglomeration occurring together with coating.

For powders SD140A, SD140B only agglomerates larger than 200 μm were coated. For SD140A, even with a slow pulverization rate (< 9g/min on 400g powder) it was necessary to stop to clean the nozzle. Total spraying time was 62min. The resulting size was high ($d_{50} = 1090\mu\text{m}$) corresponding to agglomeration with some coating. For SD140B two modes of coating were tested by spraying in top mode (similar to SD140A) and in bottom mode (particles circulation through internal Wurster tube). In both cases the initial mass of agglomerates to coat was reduced to 150g with 160g emulsion sprayed (384g on 400g powder for 140A). In top mode, the final powder had a narrow mono-modal size distribution ($d_{50} = 572\mu\text{m}$). In that case the initial bigger agglomerates had disappeared. For bottom mode we collected a thin homogeneous powder ($d_{50} = 452\mu\text{m}$) but big dry agglomerates were stuck on the inner tube.

The coated powders SD140A,B had a regular shape and smooth surface, characteristic of coated particles. Wettability was inferior to 4min and water content inferior to 6 g/100gDM. During these coating trials the total aroma content was decreased to 15% and 10% for bottom mode. For the tested conditions, the coating process led to more aroma losses than during spray drying and agglomeration, probably because of the prolonged heating of the dry thin aroma emulsion deposit on fluidised particles. To improve this behaviour it should be necessary to modify the composition of coating and aroma.

CONCLUSION

In this study, aroma powders were produced using three processes, spray drying, fluid bed for agglomeration of solid particles and then coating. The carriers for aroma protection were maltodextrin and acacia gum. The studied aroma composition was made of three types of molecules, with proportion as in real aroma formula.

The atomized powders (140-160°C) and their agglomeration with aroma emulsion gave a global positive answer considering the production of aroma powder (~20% w/w) and the global taste and intensity in chewing gum paste compared to the use of liquid aroma. The agglomeration process led to large distribution of powder sizes in the

studied conditions. Optimized conditions could be found for controlled aroma release. No coalescence of aroma oil was observed during the different processes (spraying, heating, drying) and the final total aroma content was superior to 17% for all powders.

The coating process must be improved by considering a more stable composition for the coating layer, leading to modified total composition. More generally, the operating conditions will have to be optimised in function of required objectives: wall materials composition, powder yield at different process scales, percentage of encapsulated aroma, efficiency encapsulation for specific release conditions.

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