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Particle Structures of Encapsulated Milkfat Powders

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Emulsions of milkfat (cream or anhydrous butteroil), carbohydrate (sucrose, modified starch or all-purpose flour), nonfat dry milk and mono- and diglycerides as emulsifying agents were prepared and spray-dried, yielding free-flowing powders containing 400, 500 or 600 g/kg milkfat. Structural features of the powders were evaluated by optical and scanning electron microscopy. Powder particles were generally spherical in shape, with particle size distributions ranging from 20 to 120 μm; surface morphology of the particles could be related to the encapsulant used. Butteroil was more readily encapsulated than cream in all cases; butteroil-containing powders contained less extractable fat than did those made with heavy cream. Milkfat was distributed in the walls of the powder particles when modified starch was the encapsulant; these particles contained occluded air in their cores, which could be detrimental to long-term storage. Powders with all-purpose flour as the encapsulant showed loose matrices enclosing large fat droplets, demonstrating structural weakness. Fat droplets were enclosed without central voids within the capsules when sucrose was the encapsulant; with less than 6 g/100g extractable fat, sucrose encapsulated milkfat powders were structurally stable, suggesting good resistance to air/oxygen diffusion during storage.

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Introduction

Reduction of fat in the diet in the U.S.A. and Western Europe has led to a surplus butter supply, despite the desirable flavour and textural properties that butter provides. As this trend is expected to continue, a more efficient means of long-term storage for butter is needed (1). Milkfat is readily susceptible to autoxidation and the development of rancidity. Butter is stable at room temperature for about 4 weeks, and for about 6 months at low refrigeration temperature. Salted butter maintains good storage stability when stored frozen for up to 3 years, but freezer storage is expensive. Dried butter powder can be stored for 12 to 24 months at ambient temperatures (2,3). Transforming surplus butter into a dried form has the potential for increasing the use of milkfat.

Modified starch has been successfully used for encapsulating oils; it forms stable emulsions and provides good oil retention properties (4). Spray-drying oils or fats with functional encapsulants such as carbohydrates and milk proteins has been shown to enhance oxidative stability and improve functionality (5,6).

Encapsulated powders can be readily rehydrated and easily incorporated into existing food processing systems as functional ingredients. Dried high-milkfat powders, having 'shortening' properties, make excellent food ingredients (7,8). The objective of this study was to examine the internal structure and surface morphology of spray-dried powders containing 400, 500 or 600 g/kg milkfat encapsulated in three different types of carbohydrate matrix; success of encapsulation was established by correlation of quantity and location of extractable fat to observed structural features of the various powders.

Materials and Methods

Anhydrous butteroil (0.05 g/100 g moisture) was purchased from Land-O'-Lakes (Minneapolis, MN). Heavy cream (400 g/kg milkfat) was purchased from a local distributor (Longacre's Modern Dairy, Inc., Barto, PA). Materials used for encapsulants were sucrose (Domino Sugar Corp., NY, NY), all-purpose flour (N-starch) (ADM Milling Co., Kansas City, MO) and modified starch (M-starch) (Capsul, National Starch and Chemical Co., Bridgewater, NJ). An emulsifying agent, composed of mono- and diglycerides (52 and 47 g/100 g, respectively) (BFP 75A, American Ingredients Co., Kansas City, MO), was also added. Nonfat dry milk served as an added protein source.

A full 2 × 3 × 3 factorial design, completely randomized and replicated, was used. Two forms of milkfat, anhydrous butteroil and cream (400 g/kg fat), were compared at three fat levels, 400, 500 or 600 g/kg, with three encapsulants, sucrose, N-starch and M-starch. Results were analysed for trends with the Statistical Analysis system of General Linear Models (SAS User's Guide: Basics, Version 6.03.1991. SAS Institute, Inc., Cary, NC).

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Table 1 Ingredient composition and sample formulation for encapsulated powders

Ingredients	Moisture (g/100g)	Amount needed for powder with 400g/kg milkfat (g/kg)
Anhydrous butteroil	0.05	191
Cream (400g/kg milkfat)	—	436
Sucrose	0.02	246
All-purpose flour	14.0	284
Modified starch[a]	5.0	255
Nonfat dry milk	3.0–4.0	23
Mono-+ Diglyceride	0.05	9
Water	—	627

[a]Capsul® (National Starch and Chemical Co., Bridgewater, NJ).

Sample preparation

Encapsulated powders were formulated to have 400, 500 or 600 g/kg milkfat, 20 to 30 g/kg emulsifier, 100 g/kg nonfat dry milk and the remainder, carbohydrate, after drying (9). An ingredient list and formulations for powders containing 400 g/kg milkfat are presented in **Table 1**. The processing sequence was as follows. The encapsulant of choice was mixed with dry nonfat milk solids and dispersed in water to make a slurry with approximately 300 g/kg total solids. The anhydrous butteroil or cream, and the emulsifier, were heated to 24 °C. The two blends were combined and made to 400 g/kg total solids, and mixed by hand for 5 min with a stainless steel milk stirrer, after which the slurry was slowly raised to 63 °C with constant stirring, and homogenized at 17.2 MPa with a Manton-Gaulin Model 100 DJF3 855 × Triplex homogenizer (APV Gaulin, Inc., Everett, MA). The homogenized slurry was spray-dried in a compact dryer (APV Crepaco, Inc., Attleboro Falls, MA) at an inlet temperature of 193 to 196 °C and an outlet temperature of 82 to 88 °C. The slurry was atomized in a centrifugal atomizer at 25,000 rpm. Powders were produced batch-wise; residual powder was swept from the dryer wall every 30 min. Powders were stored at 4 °C until used. When N-starch (all-purpose flour) was used as the encapsulant, it was necessary to homogenize at 10.3 MPa and 54 °C to accommodate its pasting properties; at higher homogenization pressure, the slurry thickened into a paste. The milk protein content ranged from 20 g/kg for butteroil powders to 40 g/kg for cream powders. Emulsifier concentration was based on results showing that higher levels improved baking performance of butter powders (10).

Retained fat

The ability of the carbohydrate matrices to retain milkfat was determined by dispersing 10 g powder in 50 mL carbon tetrachloride (0.002 g/100 g moisture) and shaking for 15 min with an orbital shaker (Labline Instruments, Melrose Park, IL) at 200 rpm (11,12). (Extraction was carried out at ambient temperature in a fume hood with appropriate safety precautions being taken, as carbon tetrachloride is a known carcinogen.) The supernatant was decanted and filtered and the filtrate evaporated, leaving the extracted milkfat. The efficiency of encapsulation was inversely proportional to the percentage of milkfat extracted: the lower the amount of milkfat removed, the more complete the encapsulation.

Optical microscopy

Samples of powders were sprinkled on 1 cm-long segments of Double Stick Tape (3M Company, St. Paul, MN) on microscope slides. Excess powder was removed from the surface of the tape using a jet of pressurized air. A drop of immersion oil was added to the layer of adherent powder particles, and a coverslip was applied. Optical magnifications were calibrated using a slide micrometer with an Olympus BH2 phase contrast optical microscope (Olympus Corp., Lake Success, NY).

Selective staining of lipid was carried out by suspending dry particles in a saturated solution of Oil Red O in isopropyl alcohol for 5 min. Particles were separated from the solution by centrifugation, followed by washing in isopropyl alcohol for 5 min. After further centrifugation, particles were suspended in glycerol and mounted on glass slides for bright-field optical microscopy using a narrow-pass interference filter (550 nm).

Particle size distribution

Particle size distributions were calculated from sets of optical photomicrographs as follows. High-contrast outlines of the circumferences of 431 to 585 particles from each type of powder were traced onto transparent overlays of photographic prints and digitized. Particle diameters were then calculated from projections of the digitized images of the integrated particle areas with a digital image analysis system (Dapple Microsystems, Sunnyvale, CA) running ImagePlus software. The diameter was estimated as the best fit of an integrated circular area over an equivalent area of an irregular powder particle.

Scanning electron microscopy

Samples of powders were sprinkled on aluminum specimen stubs coated with Spot-o-glue labels (Avery, Azusa, CA). Excess, nonadherent particles were removed by exposing the surface of the sample stub to a jet of pressurized air. Some particles in the samples were dry-fractured by applying a second aluminum specimen stub with glue to the particle layer, followed by separation of the stubs. Specimen stubs were coated with a thin layer of gold in a DC cold sputtering module in an E306A vacuum evaporator (Edwards High Vacuum, Inc., Grand Island, NY). Observations and images were made in the secondary electron imaging mode of a JEOL Model 840A electron microscope (JEOL, USA, Peabody, MA).

Results and Discussion

The amount of fat retained in the capsule was dependent on the milkfat content and the type of encapsulating agent used. The relative efficiency of milkfat retention within the carbohydrate matrix for anhydrous butteroil (**Fig. 1**) or heavy cream (**Fig. 2**) shows significant differences ($P < 0.05$) when the source of milkfat is considered. Anhydrous butteroil was more readily encapsulated than heavy cream, since it consistently retained more fat (**Fig. 1**). The type of carbohydrate encapsulant was equally important in determining the amount of fat retained. Best encapsulation of both milkfat sources was with sucrose at all fat levels; however, all extracted powders retained at least 40 g/100 g of the total fat even at high fat content (600 g/kg). The poorest fat retention was shown by all-purpose flour (N-starch); this encapsulant tended to form a viscous pasty emulsion after homogenization that was difficult to atomize. At low fat (400 g/kg) content, N-starch retained 65 g/100 g of the encapsulated fat, M-starch retained 85 g/100 g, and sucrose retained 94 g/100 g when heavy cream was the milkfat source (**Fig. 2**). Retention efficiency declined in all cases as the fat content increased from 400 to 600 g/kg. A similar trend was reported for anhydrous milkfat encapsulated in cheese whey protein (5).

Particle size distributions of the encapsulated milkfat powder particles with 400 g/kg milkfat, (**Figs 3** and **4**), measured from optical micrographs, show normal distribution patterns ranging from 20 to 120 μm, the greatest number of particles being around 40 to 55 μm in diameter. Size distributions for 400 g/kg fat-containing powders made from anhydrous butteroil and cream were similar when sucrose or M-starch was used as the encapsulant. When N-starch was used as the encapsulant, smaller particle sizes were obtained for powders made with cream as the fat source (**Fig. 4**) than for those made with butteroil (**Fig. 3**). There is a greater

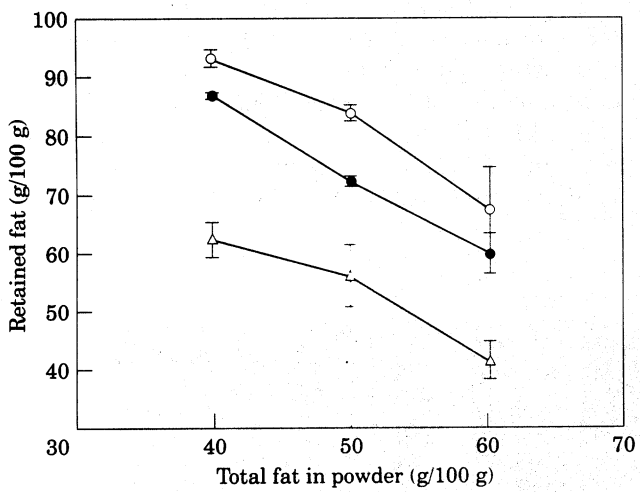

Fig. 1 Amount of milkfat retained in powder particles prepared with three different carbohydrate encapsulating agents after solvent extraction. Source of milkfat: anhydrous butteroil. (-△-) N-starch; (-●-) M-starch; (-○-) sucrose

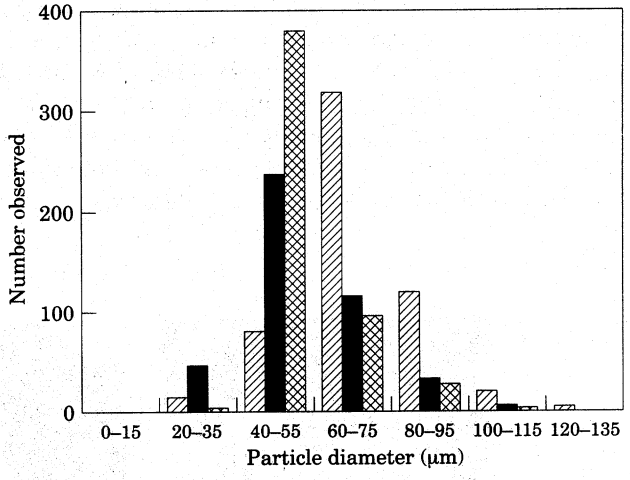

Fig. 3 Frequency distributions of powder particles prepared with three different carbohydrate encapsulating agents and containing 400 g/kg anhydrous butteroil as the source of milkfat. (▨) N-starch; (■) M-starch; (▩) sucrose

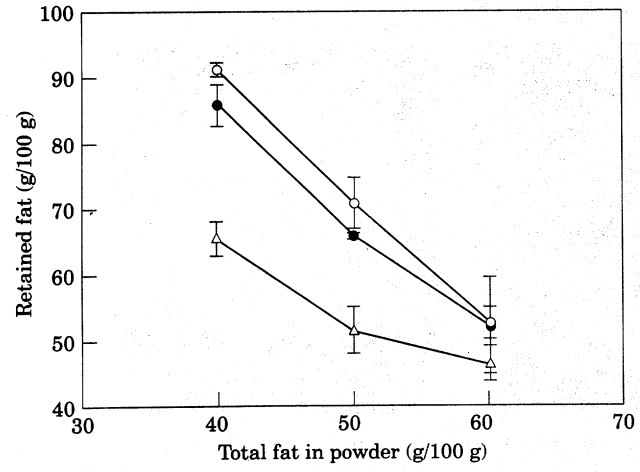

Fig. 2 Amount of milkfat retained in powder particles prepared with three different carbohydrate encapsulating agents after solvent extraction. Source of milkfat: cream. (-△-) N-starch; (-●-) M-starch; (-○-) sucrose

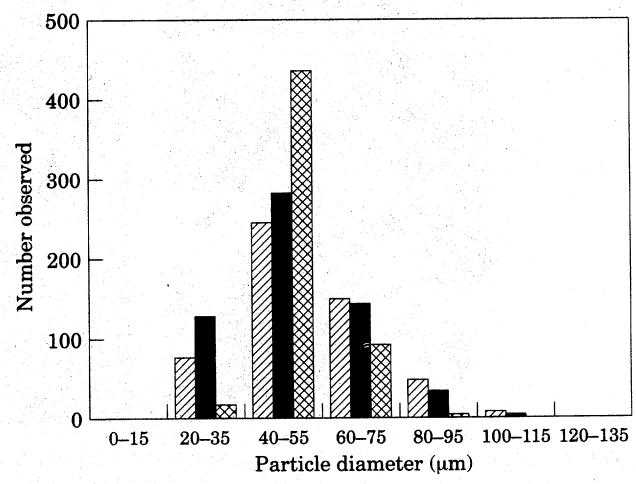

Fig. 4 Frequency distributions of powder particles prepared with three different encapsulating agents and containing cream with 400 g/kg milkfat as the source of milkfat. (▨) N-starch; (■) M-starch; (▩) sucrose

number of particles of 40 to 55 μm in diameter and a lower number of particles of 60 to 75 μm in diameter for powders made with cream (**Fig. 4**) compared with powders made with butteroil (**Fig. 3**). Differences may be attributable to the source of milkfat or to difficulties in homogenization and subsequent atomization of the emulsified mixture. Uniformly sized particles with a relatively narrow size range are advantageous in that more consistent functional behaviour in terms of rehydration characteristics is attained. Larger particles, although more easily reconstituted, may be less shelf-stable if air diffuses into the voids present. More porous structures allow increased oxygen diffusion, increased extractrable fat and lower physical strength and durability of the particle structure (13,14). Optical images of powders made from heavy cream or anhydrous butteroil to contain 400 g/kg fat, with M-starch, sucrose or N-starch as the encapsulating agent, reveal differences in structural features of the constituent particles related to the type of carbohydrate matrix formed by spray-drying, but not to the source of milkfat (**Fig. 5**). M-starch particles with butteroil or cream were roughly spherical in shape and contained one or more internal cavities of low refractive index (**Fig. 5A** and **D**, respectively). The cavities ranged in diameter from around 5 to 20 μm. The wall and matrix surrounding the cavities contained fine, granular material. The powder particles with sucrose as the encapsulant were smooth spheres, containing optically bright, homogeneous granular material (**Fig. 5B** and **E**, butteroil and cream, respectively). Powder particles containing N-starch as the encapsulant were large, roughly spherical and contained an admixture of coarse granular material, which often included several whole starch granules (**Fig. 5C** and **F**, butteroil and cream, respectively). In each case, particles with each type of encapsulant, containing 400 g/kg milkfat as butteroil or cream, were comparable in size and structure, indicating that stable powders could be made from both sources of milkfat. However, the production of encapsulated powders with anhydrous butteroil as the milkfat source offers the advantages of increased fat retention and better flow properties (15).

The effect of increasing milkfat content on powder structure was examined also by optical microscopy (**Fig. 6**). Powder particles with M-starch as the encapsulant and with 400 or 500 g/kg milkfat were composed of a phase-bright matrix surrounding one or more cavities (**Fig. 6A** and **B**), but at 600 g/kg fat content, the cavities nearly matched the optical density of the matrix (**Fig. 6C**). Powder particles with sucrose as the encapsulant were either singular or aggregated into small groups containing 5 to 10 particles (**Fig. 6D**, **E** and **F**). At 400 and 600 g/kg milkfat content, the particles were uniformly bright and contained a homogeneous fine granular material (**Fig. 6D** and **E**). At 600 g/kg fat, some

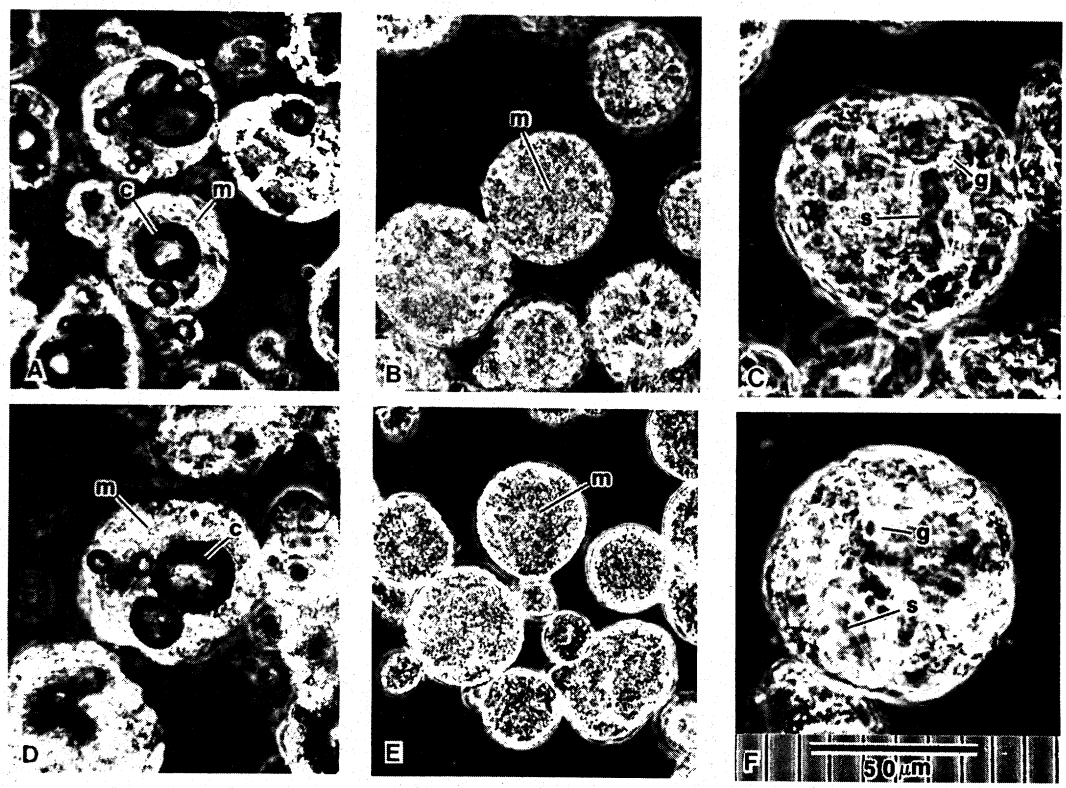

Fig. 5 Optical phase contrast micrographs of powders prepared with three different carbohydrate encapsulating agents and containing 400 g/kg anhydrous butteroil or cream as the fat source. (A) M-starch with butteroil showing hollow cavities (c) and surrounding matrix (m). (B) Sucrose with butteroil showing uniform matrix (m). (C) N-starch with butteroil showing coarse granules (g) and portions of whole starch granules (s). (D) M-starch with cream showing cavities (c) and matrix (m). (E) Sucrose with cream in a homogeneous matrix (m). (F) N-starch with cream showing comparable coarse granules (g) and starch (s), and slide micrometer with 50 μm bar

particles contained a centrally located cavity (**Fig. 6F**). Powders with N-starch as the encapsulant were comparable in size and structure (**Fig. 6G, H** and **I**). Whole starch granules and other large granular materials were features of these particles. External granular material around and between the particles comparable to that found in the particles was abundant at 500 g/kg (**Fig. 6H**) and especially at 600 g/kg milkfat content (**Fig. 6I**). M-starch encapsulant formed true barriers around the milkfat (**Fig. 6A, B** and **C**). Powder particles with sucrose as the encapsulant were relatively free-flowing and nonsticky; best properties were found in those powders containing 400 g/kg milkfat (**Fig. 6D, E** and **F**). Milkfat globules were encapsulated within the core of the powder particles at all fat levels. Powders made with N-starch as encapsulant showed fatty streaks, the amount increasing with increasing fat content (**Fig. 6G, H** and **I**). The tendency for powder particles to cluster together was influenced to some extent by the nature of the encapsulant, the amount of encapsulated fat and the mode of encapsulation.

M-starch functions as a good encapsulant for fats (4); however, in our study, the capsules formed contained central voids that could permit diffusion of air; this might pose long-term storage problems when milkfat is encapsulated. Occluded air and vacuoles may arise from air incorporation into the emulsified concentrate before spray drying (16). Powders with sucrose as the

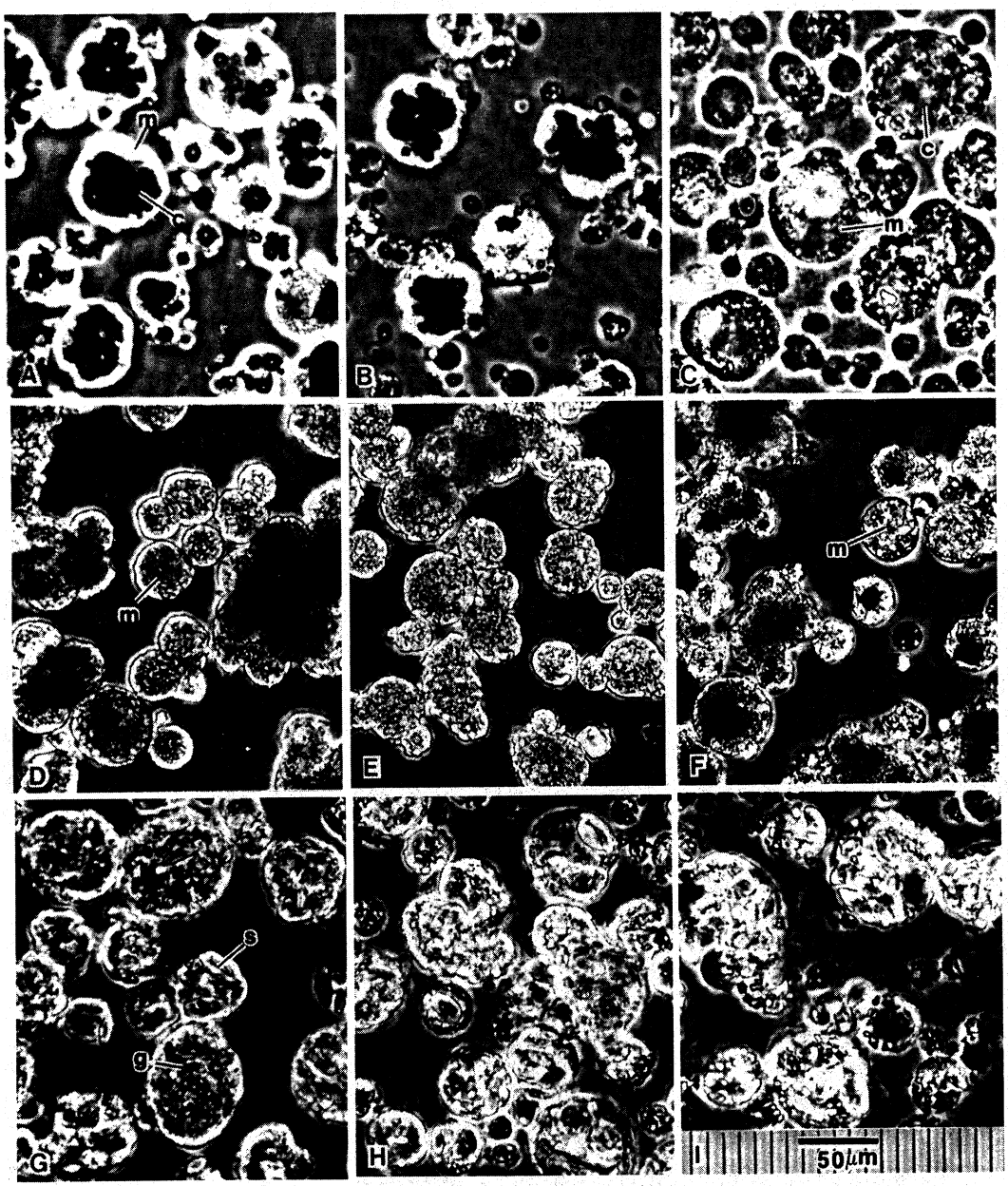

Fig. 6 Optical phase contrast micrographs of powders prepared with three different carbohydrate encapsulating agents and containing 400, 500 or 600 g/kg milkfat. (A) 400 g/kg butteroil in M-starch. (B) 500 g/kg butteroil in M-starch. (C) 600 g/kg butteroil in M-starch showing cavities (c) and matrices (m). (D) 400 g/kg butteroil in sucrose. (E) 500 g/kg butteroil in sucrose. (F) 600 g/kg butteroil in sucrose showing uniform matrix (m). (G) 400 g/kg butteroil in N-starch. (H) 500 g/kg butteroil in N-starch. (I) 600 g/kg butteroil in N-starch, showing whole starch granules (s) and coarse granules (g), with slide micrometer with 50 µm bar

encapsulant tended to agglomerate; the amount was dependent on the 'stick-point' temperature and moisture content, which are functions of the drying process (17). Powders made with N-starch as the encapsulant were lumpy at high fat levels, probably because of significant amounts of extractable fat (see **Figs 1** and **2**), some of which was on the particle surface. Surface fat acted as a binder for the particles, causing them to form lumps. The topographical features of powder particles containing 400 g/kg milkfat from either anhydrous butteroil or cream were examined by scanning electron microscopy (**Fig. 7**). The surfaces of most powder particles with M-starch as the encapsulant were smooth but indented at multiple sites, with a concave radius of curvature that appeared to match the shape of the convex surfaces of many smaller particles (**Fig. 7A** and **D**). In some cases, small particles were impinged on the surfaces of larger particles. Individual powder particles with sucrose as the encapsulant were circular in profile and slightly flattened; their surfaces were lumpy or rippled with small semicircular projections (**Fig. 7B** and **E**). Powder particles with N-starch as the encapsulant were also slightly flattened and roughly circular in profile, with contours of shallow ripples and lumps forming the irregular surfaces (**Fig. 7C** and **F**). Surface structures of the powder particles show distinctly different wrinkling and dimpling for different encapsulants. Powder particles with M-starch as the encapsulant showed surface indentations similar to those described for spray-dried nonfat milk but without the surface wrinkling (16). It has been suggested that the dimpling was caused by implosion during the final stages of drying or by cooling of particles containing relatively large vacuoles, such as those we observed (see **Fig. 5A** and **D**) (18). Rosenberg *et al.* (19), in their study of the microstructure of microcapsules, attributed particle dimpling to rapid water loss from the drying droplet. In a more recent study, Rosenberg and Young (20) were able to eliminate or to severely limit shallow surface dents in whey protein isolate- or whey protein concentrate-based, spray-dried microcapsules containing anhydrous milkfat by increasing the drying temperature.

Butteroil-containing powder particles with sucrose as the encapsulant had a rough-textured surface with many protrusions, whereas particles containing cream as the fat source showed a smoother surface texture with fewer protrusions (**Fig. 7B** and **E**). Powders made with cream as the fat source contained twice as much milk protein (4 g/100 g) as those prepared with butteroil, and also contained milk phospholipids; emulsion formation and stability may have been improved in this case, resulting in droplets that dried more evenly with less shrinkage, yielding a smoother surface.

The structures of powder particles made with N-starch as the encapsulant show that surface morphology did not appear to be dependent on the source of fat (**Fig. 7C** and **F**). In contrast to the other powders, the surfaces of these powders were highly convoluted, with clearly identifiable pores in the particle walls. Buma

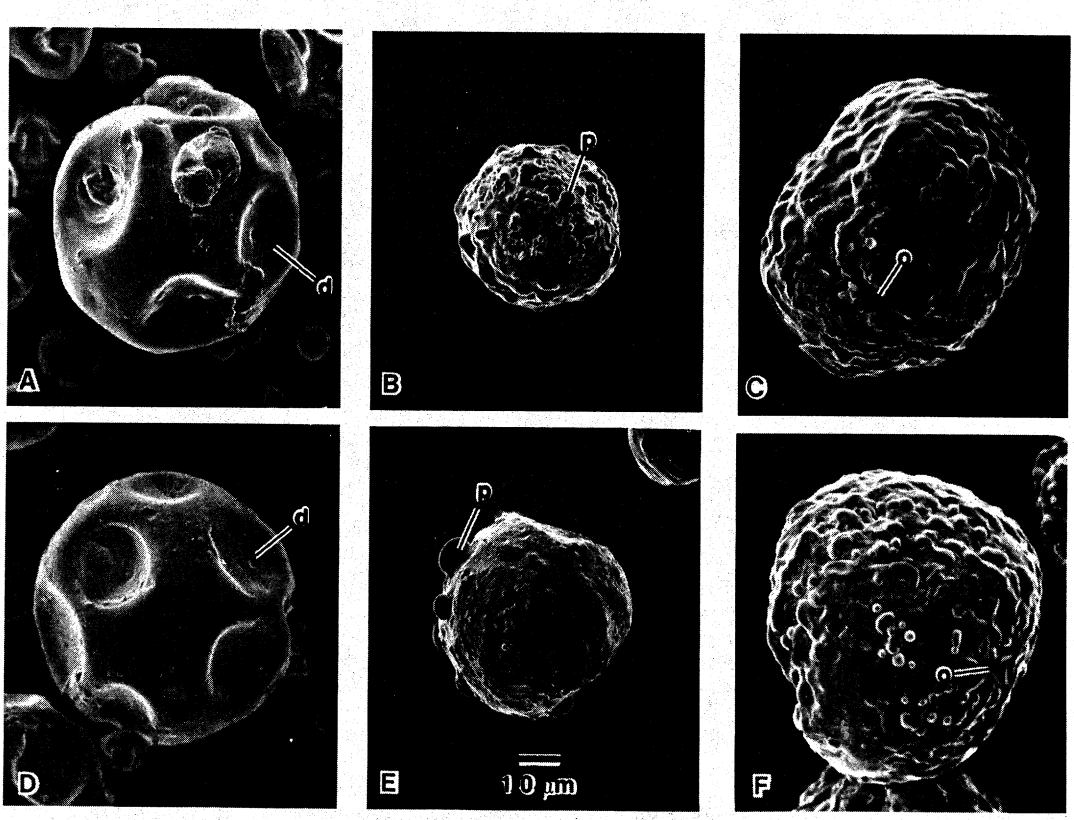

Fig. 7 Scanning electron micrographs of powder particles containing 400 g/kg fat from either anhydrous butteroil or cream. (A) M-starch with butteroil showing regular indentations (d). (B) Sucrose with butteroil showing superficial protrusions (p). (C) N-starch with rippled surface and open pores (o). (D) M-starch with cream showing indentations (d). (E) Sucrose with protrusions (p) and bar measuring 10 µm. (F) N-starch with butteroil showing rippled surface and open pores (o)

(21) demonstrated that homogenization of whole milk concentrates before spray-drying decreased both particle porosity and free-fat content regardless of whether the concentrate was spray-dried by a pressure nozzle or a spinning disc. The reduced homogenization pressure and lower homogenization temperature required to form the emulsion prior to spray-drying when N-starch was used as the encapsulant are undoubtedly partially responsible for the observed structural morphology of the dehydrated powder particles. Increased surface area brought about by surface convolutions coupled with particle porosity enhances air penetration and may reduce oxidative stability, especially when milkfat is encapsulated.

Obvious differences in the chemical composition of the encapsulants may further explain differences in the topographical features observed. M-starch, a waxy low amylose chemically modified corn starch, was highly acidic (pH 2 to 3) and cross-linked, which improves resilience. N-starch was an all-purpose high-protein wheat flour; sucrose, a disaccharide, was the least complex of the encapsulants studied.

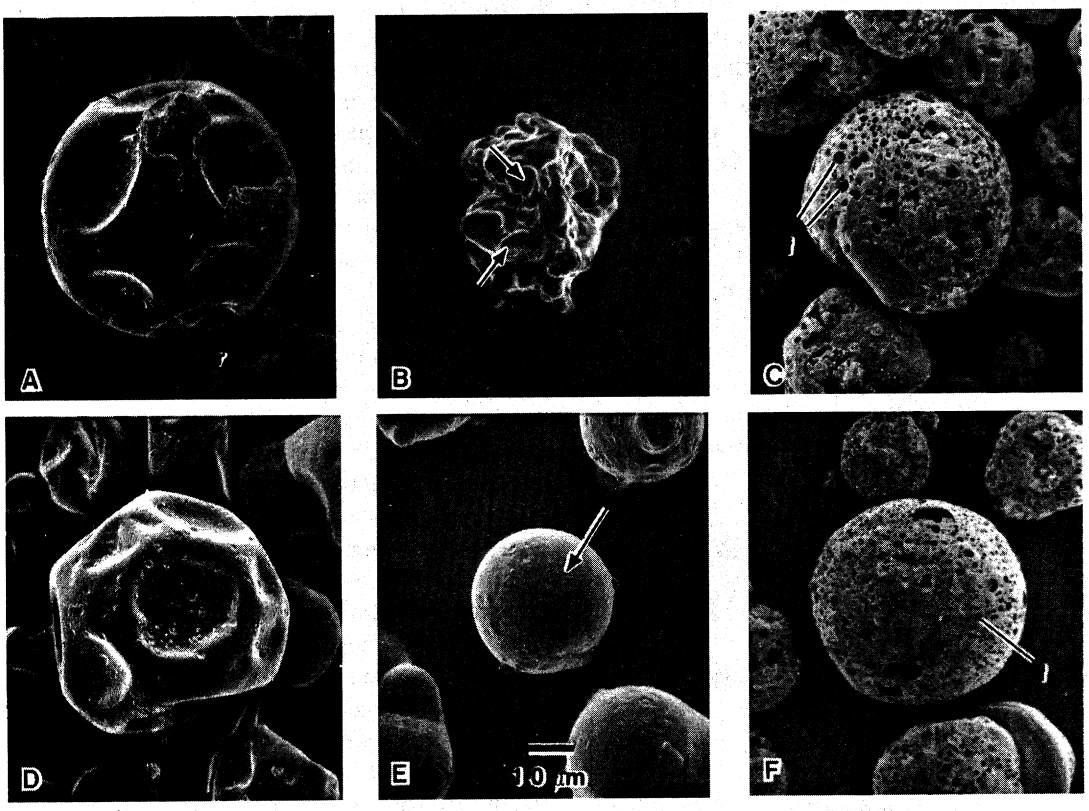

Fig. 8 Scanning electron micrographs of powder particles containing 400 g/kg of either anhydrous butteroil or cream after extraction for 15 min with carbon tetrachloride. (A) M-starch with butteroil is unchanged. (B) Sucrose with butteroil contains lumps (arrows). (C) N-starch with butteroil is porous and the surface contains many fenestrations (f). (D) M-starch with cream is unchanged. (E) Sucrose with cream has a smooth spherical surface (arrow) except for a few residual protrusions; 10 μm bar. (F) N-starch with cream has a uniform porous surface with many small fenestrations (f)

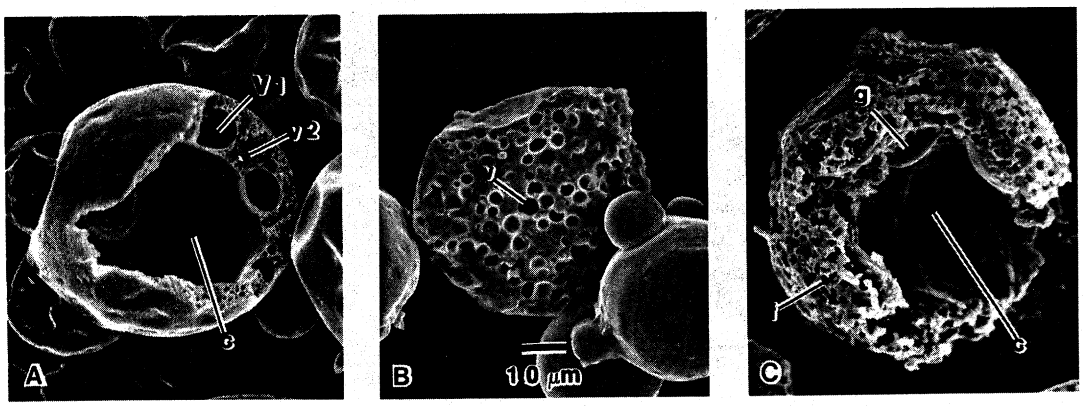

Fig. 9 Scanning electron micrographs of dry fractured particles with 400 g/kg butteroil. (A) M-starch particle showing central cavity (c) and a few large (VI) and many very small (v2) vesicles in the matrix wall. (B) Sucrose particle with a uniform distribution of small vesicles (v) and a 10 μm bar. (C) N-starch particle showing cavity (c), portion of granule (g) and fenestrations in the matrix wall (f)

The effects of the removal of extractable fat on the superficial structure of the particles in each type of powder with 400 g/kg fat were examined (**Fig. 8**). The structure of powder particles made with M-starch as the encapsulant was apparently unaffected by 15 min contact with the solvent. However, some milkfat, possibly surface fat, was available for extraction (see **Figs 1** and **2**).

An anomalous particle structure was observed for extracted powder particles containing 400 g/kg fat from anhydrous butteroil with sucrose as the encapsulant (**Fig. 8B**). Distortion of the particle structure could be caused by crystallization of the sucrose in the presence of trace moisture in the solvent. Sucrose-encapsulated structures may also sorb moisture from the air, leading to crystallization, followed by distortion and/or structural collapse (22). Particle structure of extracted powders with 400 g/kg milkfat from cream (**Fig. 8E**) appeared to be unaffected by the exposure to the solvent. This powder had slightly more extractable fat as shown by data of **Fig. 2** than did the powder containing 400 g/kg of its milkfat from butteroil, but more than 90 g/100 g of the milkfat was retained within the capsule after solvent treatment.

Surface appearance of powder particles made with N-starch as the encapsulant was significantly altered by solvent extraction regardless of fat source (**Fig. 8C** and **F**). The largest extracted particles were 10 to 20% smaller than their unextracted counterparts; in contrast to the convoluted surfaces of the unextracted powders (see **Fig. 7C** and **F**), their outer surfaces were uniform and perforated by numerous small pores (see **Fig. 8C** and **E**). The solvent was able to penetrate into the powder particles to remove fat from the particle interiors; even so, more than 60 g/100 g of the fat was retained within the particles initially containing 400 g/kg milkfat regardless of fat source (see **Figs 1** and **2**) under the experimental conditions used.

Dry-fractured particles of each type of powder made with 400 g/kg anhydrous butteroil were examined to resolve internal structures (**Fig. 9**). Each fractured particle with M-starch as the encapsulant contained a large empty vacuole surrounded by a thick, somewhat porous, wall. The wall contained a few large and many small vesicles, giving this structure the appearance of a foam (**Fig. 9A**). Evidently, the milkfat droplets are entrapped in the wall matrix in such a manner that they are not readily extractable when the milkfat content is only 400 g/kg. Fractured powder particles with sucrose as the encapsulating agent had no large central cavities but contained a uniform distribution of vesicles with a narrow size range, around 200 to 300 nm in diameter, embedded in a solid matrix (**Fig. 9B**). Milkfat is apparently encapsulated in small pockets within the matrix; small droplets are located around the edges of the particle, and small holes, possibly containing fat, are scattered throughout the particle. Projections on the surface of these particles appeared to be smaller particles stuck to the 'sticky' surfaces of the hot powder during the drying process. Fractured powder particles with N-starch as the encapsulant contained a hollow central cavity lined with the surfaces of large smooth granules and surrounded by a thick porous wall (**Fig. 9C**). These powder particles had a loose matrix of

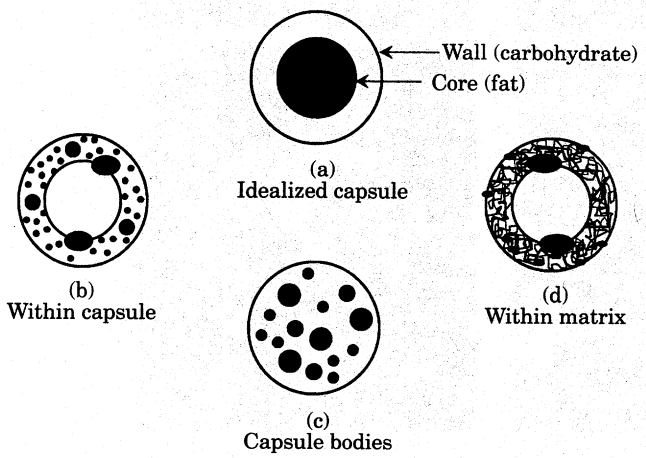

Fig. 10 Illustration of mode of encapsulation of milkfat in various carbohydrate matrices. (A) Idealized capsule; (B) M-starch capsule; (C) sucrose capsule; (D) N-starch capsule. Dark spots or bodies represent fat globules

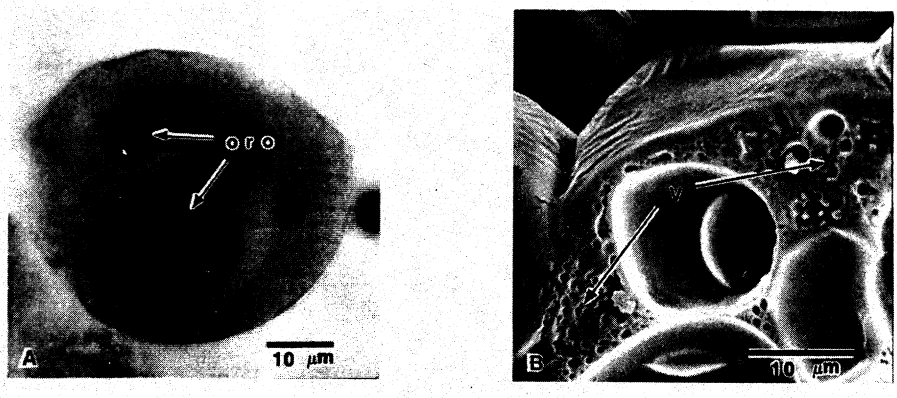

Fig. 11 Micrographs related to location of milkfat in particles. (A) Optical bright-field image of M-starch particle (400 g/kg fat) with several spheres stained with Oil Red O (oro) in cavities surrounded by matrix that contained numerous, small, stained specks, possibly related to fat; 10 μm bar. (B) Scanning electron micrograph of large fracture-face of a sucrose particle (400 g/kg fat) aggregate, showing areas of the matrix with very small vesicles (v) between large vesicles; 10 μm bar

carbohydrate and protein surrounding large fat globules. Possible modes of encapsulation of milkfat are schematically illustrated in **Fig. 10**. (a) represents one idealized capsule form with a birefringent wall material enveloping milkfat in the core. Our encapsulation techniques did not achieve this idealized structure as it is improbable that such a structure could be achieved through spray-drying. Structure (b) represents capsules prepared with M-starch as the encapsulant. These particles were true capsules, hollow in the middle, with finely dispersed milkfat globules entrapped within the wall matrix. Capsules prepared with sucrose as the encapsulant are represented by structure (c); these particles had a filled core with vesicles (dark) dispersed uniformly throughout the matrix. Powders prepared with N-starch as the encapsulant are represented by structure (d); these particles were also hollow and showed large interior bodies with smaller strands interlaced through the loosely structured particle wall. These particles were not true capsules; consequently, much of the fat content was extractable under our experimental conditions. M-starch formed capsular shells that entrapped the fat within the wall. Sucrose was the most efficient encapsulant. The interspatial fat bodies were not easily accessible even to the solvents used. Differential staining of the microparticles with Sudan III and Hematoxylin showed that the birefringent outer capsule contained virtually all of the carbohydrate (blue stain), whereas the core bodies contained the milkfat (red stain).

M-starch particles stained with Oil Red O commonly revealed several bright red spherical droplets located within the cavities. Optical images of the stained particles also showed many small stained areas in the matrix (**Fig. 11A**). Further evidence for the location of the fat was found in particles with large fracture faces; many small vesicles were located in the matrix, around and between large vesicles (**Fig. 11B**, particle with 400 g/kg milkfat from anhydrous butteroil, encapsulated in sucrose). Similar structures have been reported for milkfat encapsulated in whey protein matrices (23). The milkfat is located in 'microcapsules' within the capsules.

Conclusions

The structures of milkfat-containing powders encapsulated in carbohydrate matrices show distinct features. Powders with M-starch as the encapsulant entrapped milkfat in the wall matrix; however, powder particles contained large voids, suggesting reduced stability to oxidative deterioration if air/oxygen diffusion occurs. Powders with N-starch showed a loose matrix, without tight capsule formation, enclosing large milkfat droplets; this structural weakness may permit easy air diffusion into the interior cavity. Milkfat globules were successfully encapsulated when sucrose was used as the encapsulating agent; no voids or large vacuoles were present. With minimal extractable fat, powders with sucrose were structurally stable, suggesting good milkfat retention. Storage stability of these powders will be the subject of a future study.

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Mention of brand or firm names does not constitute an endorsement by the U.S. Department of Agriculture over others of a similar nature not mentioned.

References

1 U.S. Department of Agriculture, *Agricultural Statistics.* Washington, DC: USDA, pp. 301–335 (1991)

2 Kehagis, C. and Radema, L. Storage of butteroil under various conditions. *Netherlands Milk and Dairy Journal*, **27,** 379–398 (1973)

3 Lampert, L.M. *Modern Dairy Products.* New York: Chemical Publishing Company, pp. 273–294 (1970)

4 King, W., Trubiano, P. and Perry, P. Modified starch encapsulating agents offer superior emulsification, film forming and low surface oil. *Food Product Development*, **10**(10), 54, 56–57 (1976)

5 Young, S.L., Sarda, X. , Shoemaker, C. and Rosenberg, M. Microencapsulation of anhydrous milkfat by whey proteins. Paper D13. *Journal of Dairy Science*, **75**(suppl. 1), 93 (1992)

6 Bangs, W.E. and Reineccius, G.A. Characterization of selected materials for lemon oil encapsulation by spray drying. *Journal of Food Science*, **55,** 1356–1358 (1990)

7 Claypool, L.L. Functional role of components of dairy products in processed cereal products. In: Vetter, J.L. (Ed), *Dairy Products for the Cereal Processing Industry.* St Paul, MN: American Association of Cereal Chemists, pp. 83–99 (1984)

8 Deman, J.M. Butter as an ingredient for processed cereal products. In: Vetter, J.L. (Ed), *Dairy Products for the Cereal Processing Industry.* St Paul, MN: American Association of Cereal Chemists, pp. 155–165 (1984)

9 Hansen, P.M.T. The baking performance of butter powder. *Australian Journal of Dairy Technology*, **18,** 86–91 (1963)

10 Snow, N.S., Townsend, F.R., Bready, P.J. and Shimmin, P.D. 2. The effect of manufacturing conditions on baking performance. *Australian Journal of Dairy Technology*, **22,** 125–133 (1967)

11 Anonymous. *Analytical Methods for Dry Milk Products.* Copenhagan, Denmark: Niro Atomizer Company, pp. 46–47 (1978)

12 Tamsma, A., Edmondson, L.F. and Vettel, H.E. Free fat in foam-dried whole milk. *Journal of Dairy Science*, **42,** 240–250 (1959)

13 Chang, Y.I., Scire, J. and Jacobs, B. Effect of particle size and microstructure properties on encapsulated orange oil. In: Risch, S.J. and Reineccius, G.A. (Eds), *Flavor Encapsulation.* Washington, DC: American Chemical Society, pp. 87–102 (1988)

14 Daemen, A.L.H. The estimation of the mean particle density, the vacuole volume and the porosity of spray-dried porous powders. *Netherlands Milk and Dairy Journal*, **36,** 53–64 (1982)

15 Onwulata, C.I., Smith, P.W., Craig, J.C. Jr and Holsinger, V.H. Physical properties of encapsulated milkfat powders. *Journal of Food Science*, **59,** 316–320 (1994)

16 CARIC, M. AND KALAB, M. Effects of drying techniques on milk powders' quality and microstructure: a review. *Food Microstructure*, **6,** 171–180 (1987)
17 DOWNTON, G. E., FLORE-LUNA, J. L. AND KING, C. J. Mechanism of stickiness in hygroscopic, amorphous powders. *Industrial and Engineering Chemistry Fundamentals*, **21,** 447–451 (1982)
18 BUMA, T. J. AND HENSTRA, S. Particle structure of spray-dried milk products as observed by a scanning electron microscope. *Netherlands Milk and Dairy Journal*, **25,** 75–80 (1971)
19 ROSENBERG, M., TALORON, Y. AND KOPELMAN, I. J. Microstructure of spray dried microcapsules. *Food Microstructure*, **7,** 15–24 (1988)
20 ROSENBERG, M. AND YOUNG, S. L. Whey proteins as microencapsulating agents. Microencapsulation of anhydrous milkfat-structure evaluation. *Food Microstructure*, **12,** 31–41 (1993)
21 BUMA, T. J. Free fat in spray-dried whole milk. 8. the relation between free-fat content and particle porosity of spray-dried whole milk. *Netherlands Milk and Dairy Journal*, **25,** 123–140 (1971)
22 WHITE, G. W. AND CAKEBREAD, S. H. The glassy-state in certain sugar-containing food products. *Journal of Food Technology*, **1,** 73–82 (1966)
23 YOUNG, S. L., SARDA, X. AND ROSENBERG, M. Microencapsulating properties of whey proteins. I. Microencapsulation of anhydrous milkfat. *Journal of Dairy Science*, **76,** 2868–2877 (1993)