Fat Encapsulation in Spray-Dried Food Powders

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ABSTRACT: The surface composition of spray-dried sodium caseinate/lactose emulsions having different oil phases were estimated using electron spectroscopy for chemical analysis (ESCA), and the particle structure was studied using scanning electron microscopy (SEM) both before and after storage under humid conditions. After spray-drying, powders in which the oil phases consisting of fats with intermediate melting points, such as hardened coconut oil and butter fat, had the highest surface coverage of fat, approximately 34%. The powder with soybean oil as the oil phase had a surface coverage of fat of approximately 15%. The high-melting hardened rapeseed oil was almost completely encapsulated after spray-drying. After storage in a humid atmosphere, fat was released onto all the powder surfaces (surface fat after storage, between 50–65%) except for those with hardened rapeseed oil in which the fat remained encapsulated. These observations are consistent with the powder structure observed by SEM. The surface composition estimated by ESCA for spray-dried sodium caseinate/lactose-containing emulsions with different amounts of soybean oil and a constant lactose/sodium caseinate ratio showed an almost completely encapsulated oil-phase after drying. Storage of these powders in a humid atmosphere leads to a release of fat onto the powder surface even if the soybean oil content is low (1% of the dry weight). Powders made from soybean oil emulsions with sodium caseinate alone exhibit a much lower degree of encapsulation than in the system where lactose is present.
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KEY WORDS: Emulsion, ESCA, fat encapsulation, fat properties, food powder, lactose, lactose crystallization, scanning electron microscopy, sodium caseinate, spray drying.

Spray-drying is a widely used technique to facilitate storage and handling of food products. In a spray-dried food powder, the fat is in many cases the component which is most sensitive to oxidation. It is therefore often desirable to encapsulate the fat to protect it from oxidation. In addition, the presence of fat on a powder surface reduces the wettability and the dispersibility of the powder in water.

Spray-drying is also used to encapsulate sensitive material like flavoring agents (1–5), which in many cases appears as an oil phase. Encapsulation, using different encapsulating materials like gum arabic (6,7) maltodextrin (6) and whey protein isolate (8,9), has been thoroughly investigated by scanning electron microscopy (SEM).

If the dried powder contains crystallizing saccharides such as lactose, it is very sensitive to moisture during storage. The increase in moisture content due to water uptake lowers the glass-transition temperature of lactose and induced crystallization during storage of amorphous lactose formed during the rapid drying (10–13). Storage of lactose-containing powders in a humid atmosphere induces an almost complete release of the encapsulated fat onto the powder surface, as previously shown by electron spectroscopy for chemical analysis (ESCA) (14).

The stability of an emulsion is strongly affected by the presence of crystalline fat in the oil phase (15,16). Emulsions containing oil phases with different melting points will therefore probably give different degrees of fat encapsulation.

A new method using ESCA to estimate the percent coverage of a powder surface by component has been developed at the Institute for Surface Chemistry (Stockholm, Sweden) (17). This technique makes it possible to investigate directly the chemical composition of the powder surface after the drying and storage of food powders.

In this paper, the drying of emulsions with different fat phases has been investigated to study the influence of the oil phase on the fat encapsulation during spray-drying of emulsions containing sodium caseinate and lactose. The surface composition of the dried powders was estimated by ESCA, and the structure of the powders was studied by SEM before and after storage under humid conditions. In addition, the influence of lactose on the fat encapsulation in spray-dried sodium caseinate-stabilized emulsions having different fat contents was investigated.

EXPERIMENTAL PROCEDURES

Materials. Sodium caseinate, Miprodan 31, protein content 94.9%, was obtained from MD Foods (Viby, Denmark) lactose monohydrate, pro analysis from BDH Lab Supplies (Poole, England) and sodium hydroxide, pro analysis from Merck (Darmstadt, Germany). Refined soybean oil, hardened

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*To whom correspondence should be addressed at Institute for Surface Chemistry, P.O. Box 5607, S-114 86 Stockholm, Sweden coconut oil and hardened rapeseed oil, Akodur R, were obtained from Karlshamn AB (Karlshamn, Sweden). Butterfat was obtained from Panova Partner AB (Stockholm, Sweden). The water used in the experiments was double-distilled.

Methods: Preparation of emulsions. Two experimental series were used and are described in detail below. In the first series sodium caseinate–lactose emulsions with differing oil phases were spray-dried. To have the high-melting oil phase melted during homogenization, the solutions were heated to 70°C. In order to exclude eventual effects of the heat treatment of protein, all emulsions were prepared at 70°C.

In the second experimental series, sodium caseinate stabilized emulsions having different soybean oil content were spray-dried with and without lactose present. In this case, emulsification was made at room temperature. Before spray-drying, the emulsions were cooled down to room temperature.

Emulsions with different oil phases. The dry solids content in all emulsions was 10%. The dry matter contained 30% oil. The oil phases were soybean oil (m.p. ~ –20°C), hardened coconut oil (m.p. ~ 33°C), butterfat (m.p. ~ 38°C) and hardened rapeseed oil (m.p. ~ 59°C). The remaining solid phase consisted of sodium caseinate and lactose at a ratio of 0.67:1 by weight. To avoid precipitation of the protein, the pH of the lactose solution was raised to pH 7 by the addition of NaOH before mixing with sodium caseinate and soybean oil. All solutions were heated to *ca.* 70°C prior to prehomogenization. Before homogenization, the emulsions were prehomogenized in a high-speed colloid mill, Ultraturrax IKA (Staufen i. Br., Germany) for 2 min. The emulsions were then homogenized in a high-pressure homogenizer, Microfluidizer, TM 110 (Microfluidics, Inc., Newton, MA) at 1000 bar. The homogenization was done at 70°C. Each emulsion was recycled approximately eight times to achieve a narrow particle size distribution. The volume average particle size, d(4.3) for emulsions prepared as described above, are normally around 0.3–0.4 µm.

Emulsions with different soybean oil concentrations with and without lactose present. The dry solids content in all the emulsions was 10%. The soybean oil concentration varied between 40 and 0.1% of the dry weight. The remains of the solid phase consisted of sodium caseinate and lactose at a ratio of 0.67:1 by weight or sodium caseinate alone.

The pH of the lactose solution was raised to 7 by the addition of NaOH before it was mixed with the sodium caseinate and soybean oil. Homogenization was performed as described above excepting room temperature instead of 70°C.

Spray-drying of the samples. The samples were spray-dried in a laboratory spray-dryer constructed in-house. The dimensions of the drying chamber are 0.5 × 0.15 m. The dryer operates co-currently and has a spray nozzle with an orifice having a diameter of 1 mm. The inlet gas temperature was 180°C for all the samples investigated. The outlet gas temperature was held at 80–90°C. The flow of drying air was *ca.* 0.8 m^3/min, and liquid feed to the dryer was *ca.* 11 mL/min.

Storage of the powders. Part of each powder was stored over dry silica gel, and ESCA analysis was performed the following day.

An additional part of each of the lactose-containing powder samples was spread on microscopic slides and stored in a closed container having an atmosphere of relative humidity of 75% created by saturated NaCl solution (18). Analysis was made after 4 d of storage in humid atmosphere to secure lactose crystallization.

ESCA analysis. Electron spectroscopy is a well established technique for the analysis of solid surfaces. By ESCA, it is possible to identify elements present at the surface of the specimen. Relative atomic concentrations of elements can be quantified using appropriate sensitivity factors (supplied by Kratos Analytical, Manchester, Great Britain). Electrons emitted from the sample originate from the near-surface region for most solids (~10 nm). When a curved surface is analyzed, the depth of information is diminished even further (17). In this particular case, atomic concentration of carbon, oxygen and nitrogen in the surface of the samples is analyzed. Elemental ratios in the sample are assumed to be a linear combination of the elemental ratios of the pure components making up the sample. By using this relation in a matrix formula, described in detail elsewhere (17), the percent coverage of the different components on the powder surface can be calculated.

The ESCA measurements were performed using an AXIS HS photoelectron spectrometer (Kratos Analytical). The instrument uses monochromatic Al K_α X-ray source. The take-off angle of the photelectrons was perpendicular to the sample. The analyzer operated with a pass-energy of 80 eV. The step size was 0.1 eV. Spectrum acquisition time depended upon the peak area. Analyzed area was a circular region with a diameter of 1.3 mm, and pressure in the vacuum chamber during the analysis was less than 10^{-7} torr.

SEM. Samples were mounted on SEM stubs secured by double-sided adhesive tape. Samples were covered by gold/palladium using a sputter, Balzers SCD 050 (Balzer Union AG, Balzers, Lichtenstein) and were examined with a Philips SEM 515 (Philips Export BV, Eindhoven, the Netherlands) operating at 15 kV.

RESULTS

Emulsions with different oil phases. The surface compositions of the dried powders having different oil phases are shown in Figure 1. Powders were made from emulsions having 30% of the dry weight as oil phase, and the remaining dry weight was lactose and sodium caseinate at a constant ratio. The triangular diagram shows the composition of the emulsion, the composition of the dried powder and the surface composition of the powder after storage in humid atmosphere. Sodium caseinate is over-represented on the powder surface after drying relative to the composition of the emulsion. This result is consistent with previous observations from the drying of lactose/protein solutions (19) and emulsions (14). Fat encapsu-

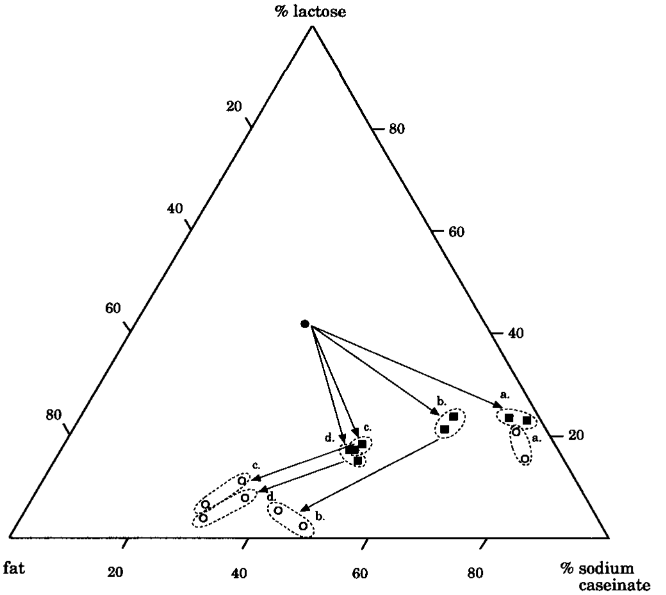

FIG. 1. Emulsions with different oil phases and a constant ratio of lactose to sodium caseinate: ●, the composition of the emulsion (% of dry weight) before spray-drying; ■, the surface composition of the spray-dried powder before storage; ○, the surface composition of the powder after storage in a humid atmosphere. a) Hardened rapeseed oil; b) soybean oil; c) hardened coconut oil; d) butterfat.

lation after drying depends on the oil phase used, Figure 1. The powders made from emulsions with the high-melting rapeseed oil were almost completely encapsulated by the fat phase. Surface coverage by fat was only about 3%. The powder made from emulsion with soybean oil had a fat coverage of about 15%. Powders made from emulsion with hardened coconut butter and butterfat had higher surface coverage by fat after spray-drying, ~34%, compared to those with the other two oil phases investigated (Fig. 1).

After storage in a humid atmosphere, a release of encapsulated fat onto the powder surface was observed for all powders with the exception of the powder containing the high-melting rapeseed oil. The fat phase in this powder remained well encapsulated even after storage in a humid atmosphere. The fine powder made from the emulsion with hardened rapeseed oil was still rather soft after storage in a humid atmosphere, whereas the powders made from the other emulsions appeared to be rather firm after storage under humid conditions.

SEM micrographs of the spray-dried emulsions are shown in Figure 2 (a–f). The figure shows that the powders made from all the different oil phases, except rapeseed oil, appear rather similar after drying. The powders consisted of smooth round particles mixed with particles having dents. The dents on these powders were rather shallow (Fig. 2 [b–d]). The powder particles made from hardened rapeseed oil had deeper dents, and the frequency of round particles was much lower than in the other dry samples (Fig. 2a).

SEM micrographs of the powders after storage under humid conditions are presented in Figure 2 (e–h). A significant change in structure is observed for the powders made from all the different fat phases, excepting that containing hardened rapeseed oil. The particles were highly agglomerated, though the particle structure is still distinguishable, especially in the powders made from soybean oil (Fig. 2h).

The powder from rapeseed oil starts to agglomerate slightly even though the particle structure remained clear. Some crystals started to appear on the surface of these powders after storage in humid atmosphere (Fig. 2e).

Emulsions with different oil contents with and without lactose present. The surface composition of powders with differing soybean oil content, but with a constant ratio of sodium caseinate to lactose (0.67 to 1), is shown in Figure 3. At all the soybean oil concentrations, fat was well encapsulated after drying. As before, sodium caseinate was over-represented on the powder surface compared to the composition of the emulsion. The surface coverage of fat in these experiments was low compared to that in the experiments made with the soybean oil emulsions described in Figure 1. One explanation for the poorer encapsulation of soybean oil in those experiments may be that the sodium caseinate was heat-treated during the emulsification. The sodium caseinate seems to lose some of its encapsulating ability on heat treatment.

As previously reported, there was a significant release of fat onto the powder surface after storage in a humid atmosphere (14). Even at a fat concentration as low as 5% of the dry weight, there was a rather significant release of fat (Fig. 3). The appearance of the lactose-containing powders studied by SEM is shown for two examples in Figure 4. Directly after spray-drying, all the powders appeared similar, a few round smooth particles mixed with particles having dents (Fig. 4a); when the soybean oil content was lowered, only particles with dents were observed. After storage in humid atmosphere, the particles became slightly agglomerated, and a few crystals were observed on the powder surface (Figure 4b).

The surface coverage of fat on powders having different fat content with and without lactose present is presented in Figure 5. Results show that the fat coverage after spray-drying was remarkably high on the powders without lactose, even when the fat content remained the same in the two experimental series.

It was previously observed that powders made from emulsions containing only sodium caseinate and soybean oil exhibit no change in surface composition after storage in humid atmosphere (75% RH, 4 d) (14), so the effect of storage under humid conditions was not investigated with these particular samples.

DISCUSSION

In earlier investigations (14,19), we have suggested that a powder containing protein, lactose and fat can be considered to be a solid solution of lactose and protein with protein film covering the powder surface more or less. In this solid solution, fat droplets are dispersed, and the powder surface is partly covered by fat leaked out from the fat droplets inside the powder particles. The fat phase inside the powder parti-

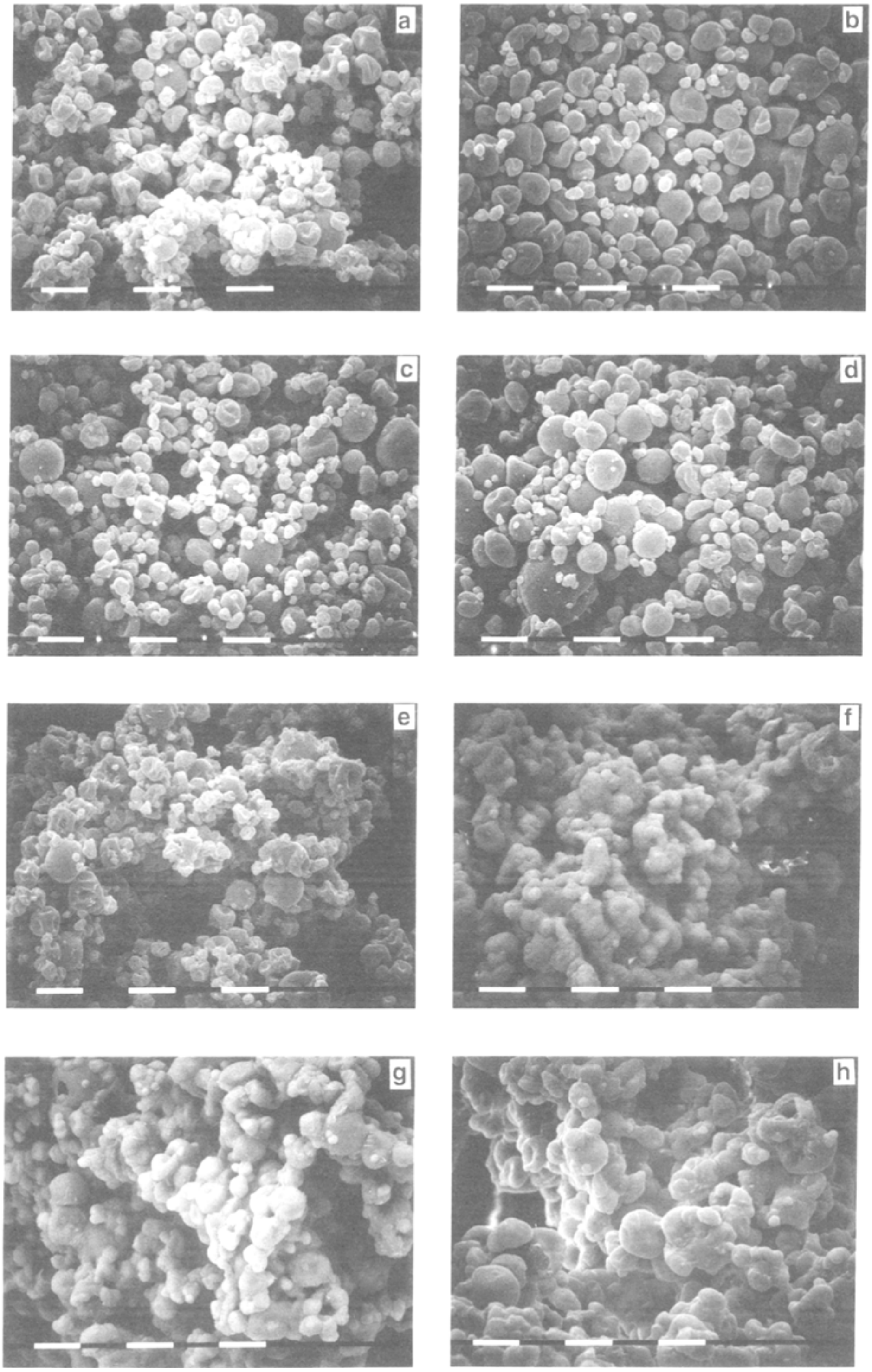

FIG. 2. Scanning electron micrographs of spray-dried powders made from emulsions with different oil phases and a constant ratio of lactose to sodium caseinate, bar = 10 μm: a, hardened rapeseed oil, before storage; b, butterfat, before storage; c, hardened coconut oil, before storage; d, soybean oil, before storage; e, as a, but after storage in a humid atmosphere (4 d, RH 75%); f, as b, but after storage in a humid atmosphere (4 d, RH 75%); g, as c, but after storage in a humid atmosphere (4 d, RH 75); h, as d, but after storage in a humid atmosphere (4 d, RH 75%).

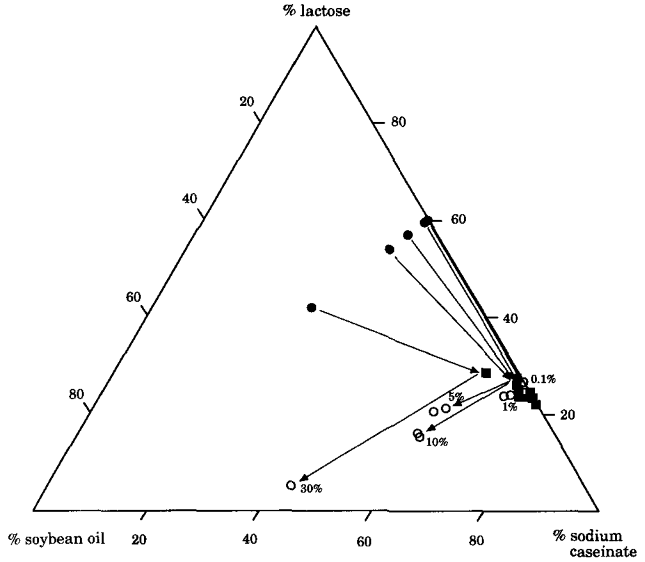

FIG. 3. Emulsions with different soybean oil contents and a constant ratio of lactose to sodium caseinate: ●, the composition of the emulsion (% of dry weight) before spray-drying; ■, the surface composition of the spray-dried powder before storage; ○, the surface composition of the powder after storage in a humid atmosphere.

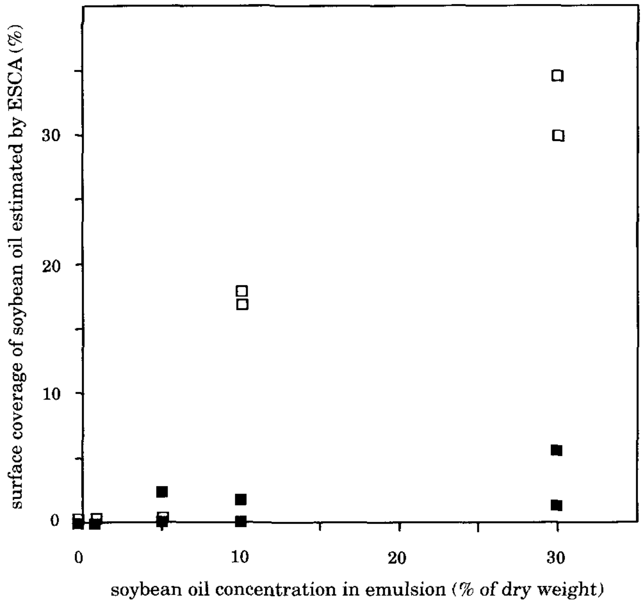

FIG. 5. The surface coverage of fat estimated by electron spectroscopy for chemical analysis (%) as a function of soybean oil concentration (% of dry weight): □, emulsions with only sodium caseinate; ■, emulsions with sodium caseinate and lactose in a proportion of 0.67 to 1 (dry weight).

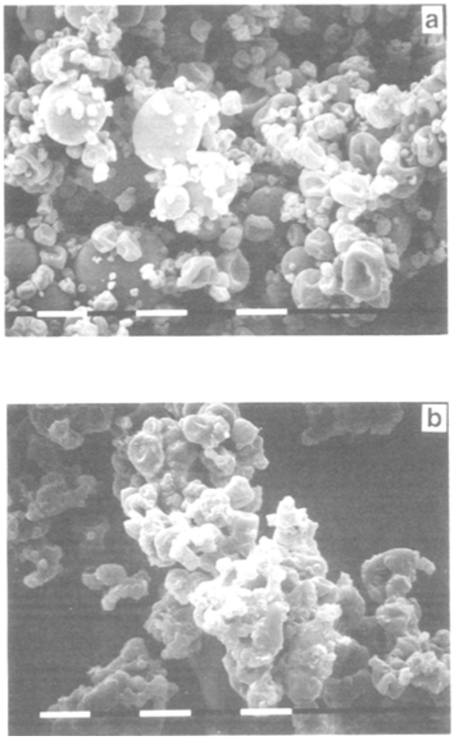

FIG. 4. Scanning electron micrographs of spray-dried powder with 10% soybean oil (% of dry weight) and a constant ratio of sodium caseinate to lactose of 0.67 to 1 in the dry weight, bar = 10 μm: a, 10% soybean oil, before storage; b, 10% of soybean oil, after storage under humid conditions.

cles may leak onto the powder surface during the drying and after lactose crystallization following storage in a humid atmosphere. In powders containing oil phases with intermediate melting points, such as butter fat and hardened coconut oil, the fat is not as well encapsulated as in the powders containing soybean oil and hardened rapeseed oil. One possible explanation is that the presence of fat crystals in the oil droplets of an oil/water emulsion induces coalescence of emulsion droplets during the drying process and reduces the stability of the emulsion (15,16). Above a certain amount of solid fat in the emulsion, the rate of coalescence again decreases (16). Since the emulsion droplets are dispersed in the solid solution of lactose and sodium caseinate, an instability of the oil droplets increases the ability of fat to leak onto the powder surface during the drying process. Earlier investigations show that in the absence of lactose there is no significant difference in surface coverage by fat for low-melting point and intermediate-melting point oil phases (17).

Previously, it has been observed that fat is released onto the powder surface when lactose-containing powders are stored in humid atmosphere (14). It was suggested that the release of fat was caused by a phase separation between lactose and sodium caseinate inside the powder particles (14). An increase in moisture content lowers the glass-transition temperature of lactose (10–13), and the lactose starts to crystallize. The phase separation causes stresses on the oil droplets inside the powder particles and forces the fat to spread onto the powder surfaces (14). These investigations performed earlier were made on powders from emulsions with soybean oil as the oil phase. In addition, the recrystallization of lactose induces a change in porosity of the powder which may also contribute to the release of fat onto the powder surface.

Results obtained in this investigation show that the release of fat onto a powder surface after storage in a humid atmosphere is prevented when the oil phase consists of a high melting point fat such as hardened rapeseed oil, having a melting point of approximately 59°C.

Our hypothesis is that the stresses arising inside the powder particles after moisture uptake because of the phase separation between lactose and protein are not strong enough to force the solid rapeseed oil onto the powder surface. It should be noted that the hardened coconut oil is forced out onto the powder surface even though it is solid at room temperature. The low melting point soybean oil is released after storage in a humid atmosphere onto the powder surface even when the concentration is low.

An interesting observation in this study is that the presence of lactose is important in obtaining complete encapsulation of the fat after spray-drying of sodium–caseinate-stabilized soybean oil emulsions. The following explanation is suggested. In the emulsion before drying, the protein is the most surface-active component present and accumulates at the air–water interface of the drying droplets (19). However, the protein in the surface film in the emulsion is completely hydrated, and the loss of water during drying would cause shrinkage of the film. If the emulsion contains lactose, the lactose may replace the water to some extent and keep the protein solubilized after drying and thereby reduce the shrinkage. This increases the stability of the sodium caseinate film on the powder surface, and less fat leaks out onto the powder surface during the drying process. However, the presence of lactose in a powder requires that the powder is well protected from humidity since lactose is the component responsible for the release of fat onto the powder surface when lactose-containing powders are stored in a humid atmosphere (14).

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