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REVIEW

Influence of Lactose on the Physicochemical Properties and Stability of Infant Formula Powders: A Review

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ABSTRACT
Different stages of infant formula (IF) products differ in their composition in terms of carbohydrate, protein and fat content, which subsequently influences their stability during storage. IF composition and processing conditions influence their physico-chemical properties which can further impact caking, Maillard browning and release of surface free fat. A good understanding of interaction of macronutrients in IF powders during storage is critical to optimize formulations that result in products with a prolonged shelf-life and enhanced bio- accessibility. This review focusses on the physical state of carbohydrates in IF powders and its influence on molecular and macroscopic changes as well as phase transitions during storage.

KEYWORDS
lactose crystallisation; Infant formula; Maillard browning; caking; Surface free fat

Introduction

According to the Codex Alimentarius Commission, infant formula (IF) is defined as a complete or partial substitute for human milk designed for use by infants as the sole source, if not the major source, of nutrition during infancy.[1] The vulnerability of the consumer makes IF products unique in comparison to all other processed foods primarily because unlike in mixed diets, nutritional inadequacies in IF cannot be compensated for by nutrients in other foods in the diet of the newborn/ toddlers. Furthermore, nutritional deficiencies in infancy may lead to development of serious and irreversible adverse effects. Hence, the standards and regulations for IF production are much stricter than other areas of food manufacture.[2] Commercially, IF products are available in the form of powders, concentrated liquids and ready-to-feed products.[3] The cost of manufacture as well as the comparative shelf life of the products makes IF powders dominate the world market.[4]

The ingredients in IF are the macro-nutrients, i.e., proteins, fats and carbohydrates, and the micro-nutrients, such as vitamins and minerals, which are carefully incorporated to result in a product that mimics human milk composition as closely as possible.[5] The composition of IF powders varies according to the stage of infant growth. These differences in IF composition can influence the physical stability of the powders during production, storage and distribution.[6–8] Lactose, and phase transitions therein during handling and storage, can have a large influence on physicochemical properties and storage stability of IF powders. This review aims to provide a detailed account of the role of lactose, and its interaction with other constituents, such as fat and protein, on the physico-chemical stability of IF powders during storage.

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Infant formula composition and ingredient interactions

The composition of IF powders is designed to meet the nutritional needs of the growing infant. Stage 1 of IF products (intended for infants aged 0–6 months) generally contains the highest amount of carbohydrates (mainly lactose) while stage 3 products (intended for 1–3 year old children) contain more protein and less carbohydrate than stage 1 products.[9] The variation in the macro-composition of commercial IF powders, as well as in the main sources of protein and carbohydrates, is shown in (Table 1). Fat sources in most cases are blends of vegetable oils. Stage 1 IF powders generally contain lactose as the main carbohydrate fraction and it makes up approximately 55–60% of the dry matter. In some cases, lactose may be (partially) replaced by maltodextrin, whereas galacto-oligosaccharides (GOS) or fructo-oligosaccharides (FOS) may also be added to IF products. In some cases, (part of) the lactose may also be replaced by maltodextrin. Stage 2 IF products typically contain similar carbohydrate sources as stage 1 IF products.[9,16–18] The protein content in the IF powders is generally between 10–15% of the total composition and may vary in casein: whey protein ratio, as shown in (Table 1).[9,16,19] Variations may also be observed based on the degree of hydrolysis of whey proteins, when dealing with IF powders containing partially or extensively hydrolyzed whey protein.[18,20–22] The fat content in IF powders is in the range of 20–26% of the total composition, [9,12,19] and decreases with increasing stage of IF nutrition. Furthermore, vitamins and minerals are vital to meet the specific nutritional requirements of infants during growth and development. Their presence can also positively or negatively influence the storage stability of IF powders. This variation in the composition affects moisture sorption in IF powders which influences the degree of lactose crystallization further influencing the non-enzymatic browning reactions as well as fat migration. Tham et al.[13] and Thomas et al.[23] suggested that the moisture sorption properties of the IF powders is dependent on the properties of their individual components in the composition, as well as their interactions.

Various ingredient interactions can be distinguished in infant formula products. The fat exists in emulsion droplets, which are largely stabilized by the proteins present in the wet mix.[24,25] Some polar lipids, such as phospholipids may also be present on the emulsion droplet interface. The protein can be found in various forms. Some protein is present on the emulsion droplet surface, but caseins and whey proteins are also in suspension in the reconstituted product. Salts are partially present in a dissolved state in the reconstituted products, however some salts are also associated with the protein, either as counterions for charged amino acid residues, or in the form of colloidal calcium phosphate nanoclusters present in casein micelles.[26] In reconstituted products, lactose and other carbohydrates are found in a dissolved state and show little interaction with other constituents. However, in dried products, lactose and the other carbohydrates typically form an anhydrous matrix in which the other constituents are dispersed. The nature of the interactions between constituents can be either non-covalent or covalent. Covalent interactions include the disulphide bonds between denatured proteins[27] and the interaction of lactose, or other reducing carbohydrates, with the ε-amino group of lysine residues in the early stage of the Maillard reaction.[28] Non-covalent bonds are responsible for other protein-protein, protein-fat, protein-ion and ion-ion interactions. The interactions between lactose in either an anhydrous or crystalline matrix are also non-covalent.[29,30]

Carbohydrates in infant formula powders: physical state and phase transitions

Physical state of carbohydrates in infant formula

Carbohydrates, especially lactose, are an essential component of IF products. Apart from the nutritional aspects, carbohydrates assist in drying and contribute to better physico-chemical and handling properties of dairy powders containing protein and fat. Lactose in IF powders can be present in two forms, i.e., the amorphous form or crystalline form, with the former being predominant. During spray drying of IF, the rapid removal of moisture causes lactose to dry under its saturation point, thus leading to a rapid increase in its viscosity and hence, forming amorphous lactose.[31] Amorphous lactose is hygroscopic in nature, which makes it prone to plasticization by water.

Table 1. Concentrations of protein, carbohydrates and fat; and protein and carbohydrate sources listed for commercial infant formula powders. Where known, the casein: whey protein ratio is included in brackets after the protein content.

Publication	Protein and carbohydrate sources listed	Protein (%)	Carbohydrate (%)	Fat (%)
Chávez-Servín et al.[10]	Skim milk powder, demineralized whey, lactose	12 (40:60)	58	26
Rodrıguez-Alcala et al.[11]	Skim milk, demineralized whey, lactose	10.2	55.2	29
	Skim milk, demineralized whey, lactose, maltodextrin	15	53.7	25
Chávez-Servín et al.[12]	Demineralized whey, skim milk, maltodextrin	12 (40:60)	58.7	24
	Demineralised whey, skim milk	12.5	58.6	24
	Demineralised whey, skim milk	12.5	58.6	24
	Demineralised whey, milk powder (partially demineralised), lactose	11.7	58.6	24.4
	Demineralised whey, skim milk powder, lactose,	11.6 (40:60)	56.0	28
	Demineralised whey, starch, skim milk, corn syrup,	12.5	55.6	24
	Skim milk, demineralized whey, maltose, maltodextrin	11.0 (60:40)	59.7	24.5
	Skim milk, lactose, starch, demineralized whey	12.8	58.3	23.1
	Skim milk, lactose, maltodextrin, milk proteins,	9.5	58	26
	Skim milk, lactose, fractionated milk protein (a-lactalbumin)	11 (40:60)	56	28
	Skim milk, whole milk, lactose, milk proteins	11.5	55	27
	Skim milk, lactose, glucose syrup	13.9 (70:30)	52.4	27.5
	Lactose, skim milk, maltodextrin, whey protein	10.7 (40:60)	58.3	26
	Lactose, skim milk, whey milk protein concentrate,	11.0	56	29
	Lactose, skim milk, whey milk protein concentrate,	11	56	29
	Demineralized hydrolysed whey milk protein, corn syrup	11.5	57.7	26
	Demineralized hydrolysed whey milk protein, lactose, maltodextrin	11.5	56.4	26.4
	Whey powder, skimmed milk, lactose	11.3	60.5	28.2
	Milk protein, skim milk, whey protein, lactose, carob flour, glucose syrup	11.2 (60:40)	60.0	23.9
Tham et al.[7,13,14]	Skim milk powder,	12	56	29
	Skim milk powder, demineralized whey protein	11.8	56.6	28.3
	Skim milk powder, whey protein concentrate, α-lactalbumin,	11	58	29
Contreras-Calderón et al.[15]	Whey, skim milk, milk proteins, lactose	9.5	57.9	
	Whey, skimmed milk, lactose	12	62.1	
	Whey, skimmed milk, lactose	12	54.5	
	Milk proteins, lactose	10.4	55.5	
	Milk proteins, lactose, maltodextrin	11.6	53.4	
	Skim milk, lactose	12.5	55.9	
	Skim milk, lactose, maltodextrin	12	57.4	
	Skim milk, lactose, maltodextrin	14.6	56.5	
	Partially hydrolyzed whey protein milk, lactose, maltodextrin	11.5	57.7	
	Partially hydrolyzed whey protein milk, lactose, maltodextrin, starch	11.5	55	
	Partially hydrolyzed whey protein milk, lactose, maltodextrin, starch	12.1	54.3	
	Partially hydrolyzed whey protein milk, maltose, maltodextrin	12.4	54.2	
	Soy protein isolate, maltodextrin	14.2	52	
Saxena et al.[8,9]	Milk, hydrolyzed whey protein	10	59	26
	Milk, hydrolyzed whey protein, maltodextrin	9.5–11.5	59–62.5	20–23
	Milk, maltodextrin, protein	10.5 (40:60)	56	26
	Milk, maltodextrin, protein, galacto-oligosaccharides	14	56	25
	Milk, maltodextrin, protein, galacto-oligosaccharides, glucose syrup solids	15	59	14

While most IF products contain predominantly amorphous lactose, high levels of crystalline lactose have also been found in some commercial IF powders.[9] It is perceivable that these levels were the result of pre-crystallization of lactose prior to spray drying.[9] Lactose pre-crystallization is primarily applied to reduce the hygroscopicity of amorphous lactose and is mostly used commercially in whey powder manufacture. Pre-crystallization of lactose is generally a two-step process that involves nucleation e.g., by shock cooling the product which is subsequently followed by slow cooling to allow growth of the crystals.[32,33] Approximately 28–32% of the lactose was reported to be in crystalline form in commercial IF powders also containing hydrolyzed whey protein.[9] This level is lower than the 50–75% of crystalline lactose observed in whey powders in a commercial setting.[34] Lactose pre-crystallization has also been explored previously in other dairy powders where stickiness was reduced and flow properties were improved.[35,36]

Phase transitions of carbohydrates in IF

Glass transition behaviour

The glass transition temperature (T_g) is the key factor influencing the physical stability of commercial IF powders. The glass transition occurs over a temperature range, where amorphous lactose undergoes transformation from 'glassy' (solid-like) to a 'rubbery' (fluid-like) state.[6,37,38] When stored at temperatures below the T_g, the powders remain stable without any significant visual changes in the product. However, storage at temperatures above the T_g increases molecular mobility, thus decreasing the viscosity of the amorphous lactose matrix, which can result in sticking and caking. Enhanced mobility in the (highly concentrated) lactose matrix can also promote the development of ordered crystalline structures, with simultaneous liberation of moisture, [6,37] which is described further in section 3.2.3. In general, T_g is well above the normal storage temperature for most commercial IF powders[9,33]; however, water sorption can drastically reduce the T_g to values below the storage temperature of powders.

T_g is also strongly affected by product composition. The presence of low molecular weight compounds or shorter chain length carbohydrates reduce the overall T_g of the powders,[39] whereas high molecular weight carbohydrates and proteins have a higher T_g.[40–43] Hence, the presence of higher molecular weight compounds such as maltodextrin in powders can increase their T_g,[37] and can enhance product stability on exposure to high temperatures and relative humidity (RH). The effect of maltodextrin on the T_g of lactose-maltodextrin systems has been well established.[19,42–46] In a recent study, Masum et al.[44] showed that replacing 15 or 30% of lactose by maltodextrin in a model IF powder increased T_g by ~3 or ~8°C, respectively. The use of maltodextrin in IF powders is primarily to retard crystallization and reduce hygroscopicity of powders in addition to assist drying.[47,48] The T_g of maltodextrin is influenced by its dextrose equivalent (DE) value, which also determines its moisture sorption behaviour.[49] Monosaccharides like glucose and fructose reduce the T_g of a system even when mixed in a 1:1 ratio with casein.[50]

It has also been demonstrated that amorphous carbohydrates undergo changes in the mechanical moduli and dielectric properties around their T_g, which reflects the spontaneous approach of the material towards equilibrium.[51,52] Several studies have been conducted to understand the dielectric and mechanical relaxation in systems containing amorphous solids.[53,54] Enthalpy relaxations occur as the mobility of the molecules increases in the system as a result of increasing storage temperature and water activity. Relaxation times are longer in glassy systems, but rapid decreases are observed in systems above their T_g.[41,53] Furthermore, higher T_g and α-relaxation temperatures are observed in amorphous lactose-protein systems than in absolute amorphous lactose systems which is further impacted by a_w.[40,53,54]

The higher T_g observed in lactose-protein systems is related to the association of lactose with proteins via hydrogen bonding, which limits their mobility in the powders. This effect, however, is dependent on the water content of the system because at higher a_w, the lactose molecules show higher affinity towards association with water via hydrogen bonding.[41,54,55] On the other hand, some other studies have reported no significant effect of the presence of protein on the T_g of powders containing lactose. For instance, Shrestha et al.[56] and McCarthy et al., [6] showed that decreasing the protein:lactose ratio in skim milk powder systems reduced T_g, but increasing the protein:lactose ratio did not significantly increase the T_g. However, at low protein:lactose ratio in IF products, protein could affect T_g notably.

The effect of hydrolyzed protein on T_g was studied by Zhou et al.,[57] who concluded that the moisture-induced reductions in T_g were higher in model powders containing hydrolyzed whey protein than in powders containing intact whey protein; this is most likely due to greater mobility of shorter peptide chains compared to the intact protein molecules. However, Mounsey et al.[58] demonstrated no significant effect on the T_g of model powders regardless of the degree of hydrolyzed casein. Similarly, Kelly et al.[21] showed no effect of intact or hydrolyzed whey protein on the T_g of the model IF powders, indicating that lactose is the primary factor that governs the T_g of the powder.[59] The influence of individual components as well their interaction on T_g is therefore critical to understand the glass transition behaviour of IF powders.

Moisture sorption and lactose crystallization

Lactose influences the moisture sorption characteristics of IF powders, which plays a crucial role in their storage stability. In general, amorphous lactose is highly hygroscopic and readily absorbs moisture, which can result in the formation of liquid bridges between particles resulting in the formation of lumps. Thereafter, subsequent exposure of powders to low RH causes moisture desorption which triggers lactose crystallization.[60]

In addition to lactose, protein also tends to affect the moisture sorption properties of IF powders, thereby influencing their T_g during storage. For instance, McCarthy et al.[6] showed that model IF powders with a higher protein: lactose ratio (~0.23) had delayed moisture sorption compared to the powders with a lower protein: lactose ratio (~0.12). This delayed moisture sorption at higher protein:lactose ratio was also shown by Murrieta-Pazos et al.[61] for whole milk powder and Hogan et al.[35] for skim milk powder. This delayed moisture sorption due to the presence of protein can also delay, or even inhibit, lactose crystallization in various dairy powders.[7,13,62–64] Studies have been conducted to investigate the effect of protein type on moisture sorption in powders. Hogan and O'Callaghan[65] observed that whey proteins delayed the onset of lactose crystallization in systems containing whey protein and lactose at various ratios. In a system containing both whey protein and casein, Li et al.[66] concluded that model milk powders with lower casein content (even in a mix containing higher protein:lactose ratio) could possibly result in early lactose crystallization. Tham et al.[13] showed that despite lower moisture sorption prior to lactose crystallization in powders containing lower protein levels, a similar onset of lactose crystallization was observed irrespective of the type of protein i.e. skim milk powder vs skim milk powder and demineralized whey protein vs a mixture of skim milk powder with whey protein concentrate and α-lactalbumin. Zhou and Roos[67] showed a greater delay in crystallization in carbohydrate-casein systems compared to carbohydrates-soy protein isolate systems, emphasizing the influence of the type of protein on sugar crystallization.

The presence of protein hydrolyzates also impacts the moisture sorption in IF powders. The effect of these hydrolyzates on moisture sorption of the powders is influenced by the protein source from which the hydrolyzate is derived (i.e., casein or whey protein) as well as the degree of hydrolysis. Hogan and O'Callaghan[65] reported an increase in hygroscopicity in a whey protein-lactose mixture with increasing the degree of whey protein hydrolysis. The hydrolysis of whey protein influenced the relaxation behaviour as well as the rate of lactose crystallization.[65]

In addition, moisture sorption in powders is also influenced by the presence of minerals, which can bind notable amounts of water.[68] This was also shown by Omar and Roos,[69] where lactose bivalent salt mixtures showed higher moisture sorption relative to lactose monovalent salt mixtures. There could possibly be an additional effect of lower mineral content in the powders containing lower protein levels.[69,70]

Impact of phase transitions of carbohydrates on physicochemical properties of IF powders

As a result of phase transitions of lactose, several changes in IF powders can occur. These changes mostly revolve around the changes in the amorphous solids of the powder during storage which affects their ordered structure (Fig. 1).

Caking of IF powders

Caking is a deleterious phenomenon by which a powder is first transformed into lumps, then into an agglomerated solid compromising its functional properties .[7] Crystallization of the carbohydrate in the liquid bridges formed as a result of particle interaction results in the formation of solidified bridges that can lead to loss of powder structure. Caking in most dairy powders can occur due to changes in the T_g of amorphous carbohydrates as well as moisture sorption after exposure to a high RH environment whereby the moisture adsorbs to the particle surface leading to the formation of liquid bridges between particles that further solidify when exposed to lower RH.[71] In some cases, the presence of surface fat may also influence caking in powders. Higher temperatures during storage may cause melting of fat leading to the formation of liquid bridges which may subsequently solidify on exposure to a low temperature environment, thus resulting in caking.[14] Therefore, storage conditions are critical in understanding the caking behaviour of IF powders.

Masum et al.[72] studied the effect of elevated temperature and RH on the storage stability of model IF powders. At RH 11 and 23%, no caking was observed in the product during storage for up to 180 days at 22 and 40°C. However, when RH during storage was increased to 54%, the products were observed to become lumpy after 30 days at 22°C and caked after 60 days at this temperature. When storage temperature at RH 54% was increased to 40°C, caking was already observed for some samples after 7 days storage and for all other samples after 15 days storage. These findings highlight that caking is particularly a risk in cases where high RH and storage temperature are combined. This is primarily the case after opening cans of IF in humid environments with elevated temperature. Similar results were observed by Saxena et al.[8] where storage at RH 54% resulted in caking in all IF powders after 4 weeks at 45°C and 6 weeks at 25°C. Tham et al.[7] investigated the effect temperature on the caking strength of IF powders during storage at 25–60°C for 6 weeks and demonstrated that while powders

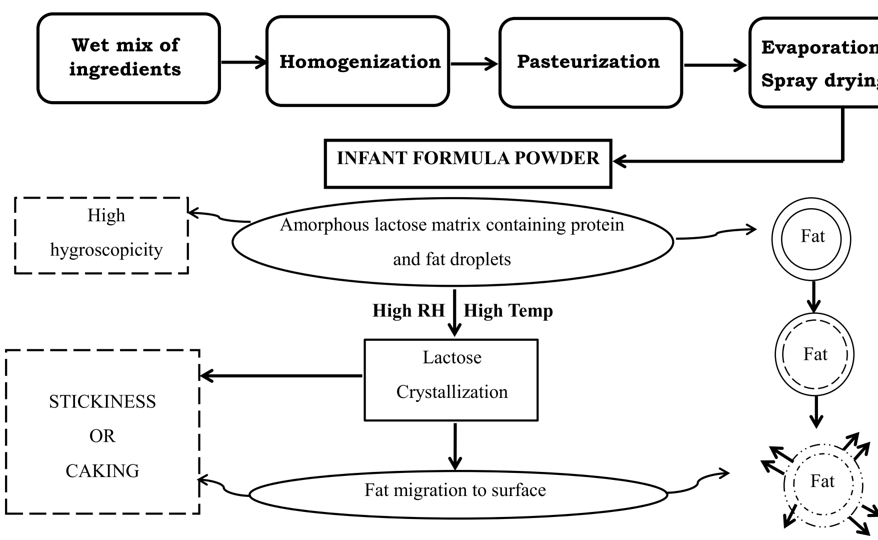

Figure 1. Schematic representation of changes in infant formula (IF) powders during storage and distribution.

stored at 25°C showed little difference in caking strength to unaged powders after 6 weeks of storage, increase in temperature from 45–60°C significantly increased the caking strength of powders after every 2 weeks. In addition, the caking strength of powders stored at 60°C for 2 weeks was comparable to those stored at 45°C for 6 weeks. In another study by Phosanam et al.,[73] the effect of both temperature and RH was studied on the caking strength of powders stored at RH 11–85% and 25–45°C, and the highest caking strength was reported for powders stored at RH 44% and 45°C after 21 days of storage.

Caking strength largely depends on the powder packing efficiency, composition and hygroscopicity. Masum et al.[72] observed that powders containing maltodextrin (DE = 20) formed only loose agglomerates after storage at 54% RH and 22°C for 60 days as opposed to caking that was observed in IF powders containing only lactose in their composition. In addition, no effect of increasing maltodextrin in the composition was observed at elevated temperatures. The presence of high molecular weight oligosaccharides has been shown to influence the storage behaviour of dairy powders.[49] While the presence of maltodextrin delays the onset of caking in IF powders by increasing T_g and reducing moisture sorption, the DE value of the maltodextrin plays a critical role in determining the extent of these effects. An increase in DE value of maltodextrin will increase the hygroscopicity of the maltodextrin.[49,74] However, Castro et al.[48] showed that beyond a point, moisture sorption was seemingly independent of the DE value of maltodextrin which was also confirmed previously by Wang and Wang.[49] This indicates that the DE alone may not be sufficient in predicting the plasticization effect of water on maltodextrin and that the molecular weight distribution of the maltodextrin is critical to the behaviour of maltodextrin. Compositional variation may also influence the degree of caking in IF powders via viscosity of concentrates, which influences powder particle size, because smaller particles cake more easily than the bigger ones.[75,76]

Changes in surface free fat

Surface free fat is the fat that migrates from the particle core to the surface of the particle during drying or storage conditions depending on the conditions employed. The presence of surface free fat in fresh powders is influenced by both the composition of powders and processing conditions. Drying causes preferential migration of components towards the core of the powder particle. The order of the migration of these components depends on their molecular size such that larger the particle size, slower its diffusivity. Hence the order of diffusion in decreasing trend is: lactose and salts> whey proteins> casein micelles> fat globules.[77] Therefore, the slow diffusivity of fat towards the centre of the particle core results in its over representation on the particle surface[9,61] which can lead to compromised storage stability as well as increased stickiness in powders during drying .[62] In general, higher surface free fat is observed in powders containing a higher fat content.[62] Homogenization prior to drying strongly reduced surface free fat.[78] During storage, however, surface free fat can increase further, as is discussed later in this section.

The presence of surface fat is influenced by drying conditions, emulsion stability (which further depends on the protein content as well as the type of protein (hydrolyzed/intact), drying rate, solids content, outlet temperature.[21,56,78–80] McCarthy et al.[70] studied the effect of varying protein: fat ratio on the emulsion stability and fat globule size and reported that increasing the protein: fat ratio increased the viscosity of the emulsion while simultaneously reducing the fat globule size, thus resulting in lower surface free fat. This was further confirmed by McCarthy et al.[6] who showed that lowering the protein content increased the surface free fat content in model IF powders. As far as the effect of whey protein: casein ratio on the surface free fat is concerned, Masum et al.[81] observed no effect of varying whey: casein ratios (60:40, 50:50, 40:60) on the surface free content of IF powders. However, hydrolysis of proteins has been shown to result in increased surface free fat in IF powders.[9,21] Similarly, varying the lactose: maltodextrin ratios did not impact the surface free fat content of fresh IF powders.[44] The amount of surface free fat increase is also dependent on the composition of the fat used in the IF formulation which further impacts their drying behaviour. The melting point of the fat blends used in IF powders ranges between −10 to 40°C[14] and increase in the unsaturated fatty acids will result in higher representation on the particle surface post drying.[8,71,82] The composition of fat reported in IF powders is variable depending on their source and both the amount and the type of fat influence the keeping quality of IF powders.[83,84] In practice, a higher unsaturated fatty acid content results in a lower melting point which pose a risk of oxidation as well as caking due to fat bridging.[14] Inclusion of lecithin in formulations can help reduce lipid oxidation.[84]

Surface free fat can also increase further during storage of powders, which is often related to the crystallization of lactose. Progression of lactose crystallization results in the disruption of powder particle structure that allows the movement of melted fat from the core to the surface of the particle, thus causing an increase in the surface free fat.[8,14,33] Although few studies have reported the influence of lactose crystallization on surface free fat, Saxena et al.[8] reported a linear and exponential increase in surface free fat with increasing degree of lactose crystallization in IF containing hydrolyzed and non-hydrolyzed whey protein, respectively, during storage at 25 and 45°C and 11–54% RH. The influence of pre-crystallized lactose on surface free fat was confirmed by Saxena et al.[33] where the presence 18% pre-crystallized lactose in IF powders containing hydrolyzed whey protein resulted in a surface free fat of ~2% after 6 weeks of storage at RH 54% and 45°C relative to control samples that showed ~8% surface free fat under the same storage conditions and a distinct surface morphology (Fig. 2).

The presence of fat on the particle surface influences the moisture sorption of powders during storage which further influences changes in glass transition and crystallization of lactose.[56,61] It was demonstrated that fat interfered with the moisture sorption of powders during storage at high RH particularly due to its hydrophobic nature that reduced the diffusion of hydrophilic components, thus affecting crystallization of lactose. These results were confirmed in IF powders by Tham et al.[7,13,14] as well as in spray dried milk protein concentrates by Kelly et al.[64]

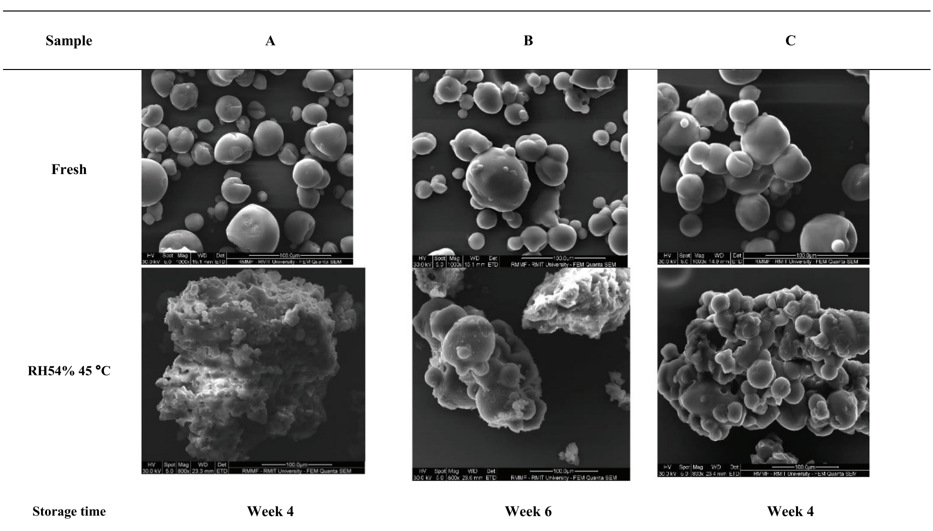

Figure 2. SEM images of fresh and aged (4 or 6 week at 54% RH and 45°C) infant formula powders containing 0.5 (A), 18 (B), and 45 (C) % pre-crystallized lactose (adapted from Saxena et al.[33]).

Chemical changes in carbohydrates in infant formula

Protein-carbohydrate interactions

IF composition (in terms of reducing sugar content, lysine-rich proteins, ferrous content, Vitamin A), processing conditions and storage influence the degree of Maillard reaction in the product.[17,85,86] Maillard reactions are sugar-amine reactions where reaction between most reactive lysine group (followed by arginine residue and N-terminal α-amino acid residue[87] and a reducing end of a saccharide results in protein glycation which in the advanced stages is also accompanied by the formation of furfurals.[85] The formation of furfurals in IF powders can be a result of the heat exposure during processing[16,86,87] or the storage conditions after manufacture .[17] For instance, in a recent study Pires et al.[86] showed lower formation of HMF in reconstituted IF powders processed by ohmic heating as compared to IF powders processed by conventional heating.

The formation of furfural compounds can occur as a result of lactose isomerization or Amadori rearrangement[16] which is dependent on the pH of the medium. For instance, in an acidic medium, Maillard reactions may occur as a result of dehydration of carbohydrates as well as formation of Amadori compounds and hence, HMF may be formed as a consequence of both.[88] Chàvez-Servin et al.[12] studied the changes in pH of different IF powders during storage at room temperature for 0–70 days and showed no significant changes in the pH of samples from day 0, suggesting that IF powder were most likely to undergo formation of furfurals as a result of Amadori rearrangement. The potential HMF has been reported to be within the range of 400–2200 μg/100 g sample for different IF powders[10,12,16,17] and the variation is a consequence of the difference in the composition of IF powders as well as their storage conditions.

The extent of HMF formation is largely dependent on the presence of proteins and lactose in the composition of IF powders. Higher the lactose and protein content in the powders, higher the propensity for Maillard browning. This was confirmed by Ferrer et al., [16] who showed that increases in HMF content during storage at 20 and 37°C were faster in a formula containing lactose as the only carbohydrate source compared to one containing both lactose and maltodextrin. Similar results have also been reported by Contreras-Calderon et al.[15] in their work on the progression of Maillard reaction in IF powders available in the Spanish market. Furthermore, both Gonzales et al.[89] and Contreras-Calderon et al.[15] showed that soy protein isolate containing IF powders showed limited formation of Maillard reaction products due to lower lysine availability than in IF powders containing dairy proteins. The presence of hydrolysed protein in IF powders demonstrate a higher propensity to browning than those containing intact dairy proteins[90–93] owing to the relatively high exposure of amino groups for reaction with reducing sugars[93] which in turn is dependent on the degree of hydrolysis of the protein chain and the type of enzyme used for hydrolysis.[94]

Similarly, the presence of monosaccharides, which occur e.g., after enzymatic hydrolysis of lactose[95] increase the rate of Maillard reaction due to a higher proportion of the open chain available for reaction with lysine. In general, the reactivity of monosaccharides decreases as follows: pentoses>hexoses>disaccharides>maltodextrins, [96,97] where pentoses like glucose are most reactive while oligosaccharides like maltodextrins are the least reactive. In the formulations that contain a mix of sugars, the extent of the Maillard reaction is hard to predict, [98–100] since there is also competitive inhibition among the sugars. Other factors that influence the Maillard reaction in IF powders include the presence of iron and carbonylated substances that may be derived from lactose, vitamin C degradation products or oxidized polyunsaturated fatty acids.[101]

The progression of the Maillard reaction can influence changes in the secondary structure of proteins which may subsequently affect the functional characteristics of powders.[102,103] A possible explanation for reduced α-helical structures during progression of browning could be the formation of hydrogen bonds between sugar and whey protein/peptides that weaken the intermolecular interaction between hydrogen atoms of amide and oxygen atoms of carbonyl leading to reduced α-helical structures, as suggested by Srivastava et al.[104] and Sun et al.[105] Furthermore, advanced Maillard reaction products like glyoxal and methylglyoxal may promote cross-linking in proteins by reacting with lysine or arginine residues.[106] In addition, protein modifications may also occur because of cross-linking reactions involving the formation of lysinoalanine, histidinoalanine and lanthioalanine[107] as well as the interaction of proteins with carbonyls formed via lipid oxidation reactions[108] that cause changes in non-covalent interactions of proteins during storage.

Color changes due to Maillard reaction

Crystallization of lactose can result in the release of water from the anhydrous lactose matrix, which can further increase the susceptibility to formation of brown-coloured complexes by Maillard reactions.[109] Progression of Maillard reactions is marked by the changes in color.[28,108] The intensity of color development is an indicator of the progression of Maillard reactions stages which varies between pale yellow to dark brown depending on the degree of advancement of Maillard reactions in the product which is in turn dependent on the composition as well as the storage conditions.[11] Browning in IF powders may result from lactose caramelization or Maillard reactions, depending on the pH of the powder during storage (as discussed previously), hence color change cannot be regarded as the only indicator of Maillard reactions in dairy powders.

Color changes in IF powders during storage have been reported previously by Nasirpour et al.,[83] Rufián-Henares et al.[97] and Ferrer et al.,[17] all highlighting that the degree of color change is dependent on the degree of heat damage to the proteins such that higher temperatures cause increased rate of HMF reaction leading to increased browning in the powders. In addition to browning, the changes in protein structure are translated as compromised functional properties such as solubility of powders[28] suggesting the possibility of hydrophobic cross-linked protein network on powder surface that prevents powder hydration. Since IF powders are complex mixtures, the changes in solubility during their storage are a combined result of the effect of surface free fat, lactose crystallization as well as changes in the structural conformation of proteins.

Conclusion

In conclusion, the overall composition of IF powders has a profound impact on their physico-chemical properties during processing as well as storage under adverse conditions of temperature and RH. This review highlights the changes in lactose during storage and its concomitant effect on the physico-chemical stability of IF powders. In instances where the handling and storage of IF cans are not always optimum, knowledge of ingredient behaviour under such conditions can provide sufficient understanding of the product and may help predict their shelf life. The physical state and phase transitions of carbohydrates in IF powders influence their moisture sorption behaviour which is accompanied by changes in lactose crystallization during storage. The degradative changes following lactose crystallization include the increased presence of surface free fat as well as the progression of Maillard browning. Understanding the influence of physical form of lactose and its interaction with proteins on moisture sorption is essential to predict the changes in IF powders during storage. In addition, the understanding of the fat composition and their migration to particle surface is equally critical. Exploring areas of compositional modifications such as lactose pre-crystallization in order to result in powders with better keeping quality has opened avenues for exploration. Further research is essential to investigate its influence in variable IF composition as well as their interaction with the micronutrients.

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Declaration of Interests

No conflict of interest to declare.

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