

Chapter 19

Polymers for Structure Design of Dairy Foods



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Abstract Dairy foods ranging from liquids, semi-solids to solids are considered as complex viscoelastic materials. Maintaining the physical stability over the shelf life and delivering superior mouthfeel of foods after series of treatments have always been challenging for the dairy industry. During the manufacturing process, both high shear stress and temperature changing history can significantly affect the macro- and microstructure of dairy food systems. Therefore, their physical stability and sensory attributes are altered as consequences. Food polymers as stabilizer and texturizer are used in different dairy systems for eliminating negative impacts of intensive processing treatments and for manipulating texture for meeting the specific sensory preference for a targeted group of consumers. As kinetically metastable systems, the optimum structure of dairy foods may be engineered by following a universal two steps principle: (1) Apply the proper mix of food polymers in the dairy food formulation; (2) Process the formula with dedicated parameters and procedures. Although the principle is simple, the implementation is complicated. Such existence of challenge is due to the detailed interaction mechanisms between non-dairy polymers and dairy components in various physicochemical environments are not entirely understood. In this chapter, the nondairy polymers induced destabilization/stabilization of dairy systems are explained, the technical challenges of stabilization of dairy systems are discussed. It focuses on three major topics regarding dairy food structure design: (1) Formulation strategy of thickening dairy matrices; (2) Formulation strategy of increasing perception of the creaminess of dairy matrices; (3) The current updates about the synergetic functionality of food polymers.

Keywords Food hydrocolloids · Dairy Foods · Stability · Texture

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19.1 Overview

Specific polymers as food hydrocolloids are often found in the ingredient list of dairy food products such as starch, pectin, carrageenan, gellan gum, β -glucan, inulin, or gelatin. The presence of these items in dairy systems is not expected from consumers' perspective, and most of the consumers have little knowledge about the significance of using these non-dairy materials in dairy foods. These materials are traditionally recognized as stabilizers which are responsible for maintaining the physical stability of dairy foods, preventing the dairy dispersion particles or gel systems are visibly separated from the aqueous phase. The US Food and Drug Administration (FDA) and European Food Safety Authority (EFSA) classified such food hydrocolloids as emulsifiers, stabilizers, gelling agents and thickeners as part of the larger family of food additives. FDA provides a Food Additives Status List which includes short notations on use limitations for each additive. The related regulations of usage of the additives are promulgated under the Federal Food, Drug, and Cosmetic Act (FD&C Act) under section 401 (food standards) and 409 (food additives) (FDA 2017). The detailed application guidelines of the most stabilizers can be found from the FDA Code of Federal Regulations Title 21, part 172—food additives permitted for direct addition to food for human consumption. According to EFSA, all stabilizers as food additives are identified by specific E numbers (EFSA 2017b). The European Union is carrying out a re-evaluation programme accessing the safety usage of all food additives. By 2020, EFSA's Expert Panel on Food Additives and Nutrient Sources Added to Food (ANS) is going to provide updated conclusions on the safety of the intended uses of the food additive for the consumers (EFSA 2017a).

The classical reason for applying food polymers in dairy products are: (1) constructing physically stable dairy liquid/gel systems preventing phase separation over the shelf-life or preventing the growth of ice crystals caused by temperature abuse. (2) Establishing and maintaining the texture of dairy foods for delivering body and specific mouthfeel for attracting consumers (Hansen 1993). In light of the development of food technology and the demand of nutritional foods, food polymers are also used as wall materials for encapsulating sensitive nutrients or probiotics in dairy material based food systems (Li et al. 2017; Anal and Singh 2007). Microencapsulation can reduce the reactivity of the core material with environmental reactants due to the low permeability of the barrier wall. Therefore, for instance, undesirable oil oxidation may be prevented or slowed down to an acceptable oxidation rate according to the shelf life; the release of specific nutrients or probiotics can be engineered for the targeted delivery; the unfavorable taste or flavor of the beneficial health materials are masked during consumption (Shahidi and Han 1993).

All in all, the aforementioned three areas for food polymer application in dairy foods can be summarized as the dairy structure and texture design. Based on the three core purposes of application of food polymers in dairy foods, this chapter is aiming to provide technical knowledge to dairy food technologists who use food polymers to formulate different dairy foods for achieving physical stability, and desired mouthfeel. One can use this chapter for acquiring knowledge about the working mechanisms of different food polymers in different types of dairy matrices

and use it as guidelines for constructing ideal structure and texture of dairy foods which meet the various markets' needs. The term of "polymer" is extensively used the following texts, it represents non-dairy polymer molecules distinguishing from dairy proteins.

19.2 Milk System and Polymer-Protein Interactions

19.2.1 Milk Components

Milk is a complex oil-in-water (o/w) emulsion system containing oil droplets, dissolved whey protein, colloidal casein protein micelles, lactose and a small amount of minerals. In raw milk system, the milk fat presents as globular shape droplets named as milk fat globule (MFG). The size of MFGs ranges from 0.2 to 15 μm with a volume weighted mean diameter (d_{43}) between 3.5 and 5.3 μm . The MFG is stabilized by a tri-layer membrane system namely the milk fat globule membrane (MFGM) which composed of phospholipids and larger molecular weight proteins (Huppertz and Kelly 2006)(Zheng et al. 2014). The MFGM system stabilizes the native MFGs against aggregation and flocculation. However, the MFGM cannot prevent creaming. Casein as the major type protein in milk accounts for around 80% protein proportion; it is a mixture of four proteins: α_{s1} -, α_{s2} -, β -, and κ -casein have molecular masses around 20 kDa (Dalgleish 1997; Walstra et al. 2006). Casein presents in milk aqueous phase in different states from dissolved macromolecules to stable colloidal larger proteinaceous particle namely casein micelle (Dalgleish 1997). The typical volume-average radius of casein micelle is of ~ 100 nm (Kruif and Holt 2003). About 20% bovine milk protein proteins are whey/serum proteins including α -lactalbumin ($\sim 19\%$ w/w of total whey protein), β -lactoglobulin ($\sim 52\%$ w/w of total whey protein), bovine serum albumin (BSA, $\sim 6\%$ w/w of total whey protein), Immunoglobulins ($\sim 13\%$ w/w of total whey protein), proteose peptone ($\sim 13\%$ w/w of total whey protein). Most of the serum proteins are globular proteins; unlike casein micelles, whey proteins present in a dissolved form in bovine milk. Both casein and whey proteins are hydrophobic having hydrophobic side groups ranging from 22 to 29%; they are all negatively charged at neutral pH of bovine milk ($\sim \text{pH } 6.5$) (Walstra 2006). To be able to engineer dairy structure and texture using nondairy food polymers three types of interaction mechanisms need to be elaborated (see 19.2.2).

19.2.2 Nondairy Polymers and Dairy Protein Interactions

Nondairy polymers together with whey protein and casein micelles in the aqueous phase of milk form a pseudoternary "protein-polysaccharide-water" polyelectrolyte dispersion system. Such ternary dispersion system is highly complicated due to the presences of polyelectrolytes and irregular distribution of charged groups along the

nondairy polymer/dairy protein (Syrbe et al. 1998). For instance, α_{S1} -, α_{S2} -, β -, and κ -caseins have an uneven distribution of charges; such complexity indicates site-specific interactions among different nondairy polymers. Moreover, several different nondairy polymers (>3 types) are often mixed forming a “stabilizer mix” and applied in dairy systems. Therefore, the commercial dairy foods are more than a ternary system. For better understanding the nature of the pseudoternary system, it is important to point out that the physical behavior of the system is controlled by enthalpic effects (Syrbe et al. 1998).

Syrbe and co-authors (1998) summarized three equilibrium conditions of polymer-protein-solvent ternary systems; these conditions may apply to dairy matrices containing multi-polymers: (1) Incompatibility: the different polymers (dairy protein and polysaccharide) are concentrated in separated domains in the system, which is also termed as “segregative phase separation.” For instance, an increase of the concentration of high methoxyl pectin in skim milk at pH 6.7 induces depletion flocculation of casein micelles forming casein-rich domains (Acero-Lopez et al. 2010). Such interaction mechanism is explained and understood as deletion (Tuinier et al. 2003); (2) Complex formation (coacervation): such interaction among polymers are initiated by electrostatic attraction, hydrogen binding and hydrophobic attraction, the protein-polymer complex results in a polymer-rich phase in the food matrix (Corredig et al. 2011; Thies 2003). The formation of protein-polymer complex induces a series of changes such as modification of a rheological property of the system (Wang et al. 2007) and cause of precipitation of protein (Niederauer and Glatz 1994). At relative lower pH (pH 5.3) both high and low methoxyl pectin interact with casein micelles via electrostatic force driven bridging flocculation mechanism and can form a physically stable system (Marozieni and de Kruif 2000). Using quasi-elastic light scattering and fluorescence spectroscopy techniques, it was found that globular whey protein (human serum albumin) can interact with polyethylene glycol (used antifoaming agent or plasticizers in aqueous film coatings) forming an intrapolymer complex via hydrogen bonding (Azegami et al. 1999). Polyelectrolytes may regulate protein/peptide drug delivery (Vasir et al. 2003), and it was found that both the concentration of soluble whey protein (free BSA) and the hydrophobicity of polyacrylates control the formation of the soluble protein-polymer complex (Porcar et al. 1999); (3) Miscibility: in such scenario, different polymer species are homogeneously co-distributed in a system via only physical contact without chemical and physicochemical interactions. The true interaction mechanism between dairy protein and starch has been extensively studied, but it is not yet fully understood. The interactions between dairy protein ingredients and gelatinized starch paste (Kumar et al. 2017, 2018) and the interactions between starch granules and different dairy food systems (Considine et al. 2011) are studied and reviewed. In these studies, the authors demonstrated that although protein-polymer complexes are formed, protein/protein aggregates may be homogeneously miscible in starch paste system (Fig. 19.1) or gelatinized starch granules may be homogeneously distributed in acidified dairy protein gel systems (as shown in Fig. 19.2). The miscible system may become incompatible system under specific physical stress in a certain timescale. The miscible system is considered as a stable

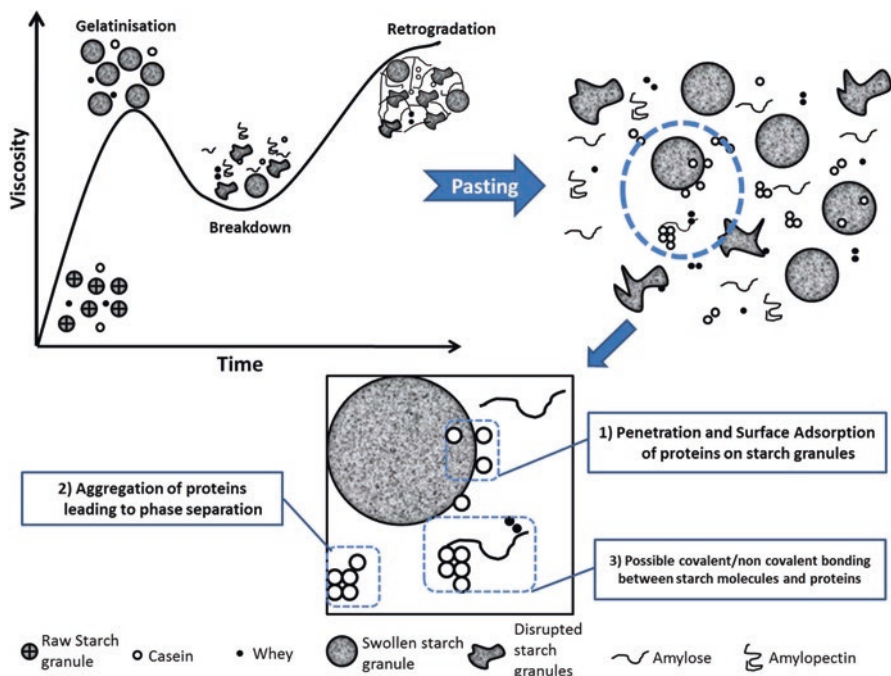


Fig. 19.1 Starch-milk proteins interactions during heat treatment under continuous shear (Kumar et al. 2017)

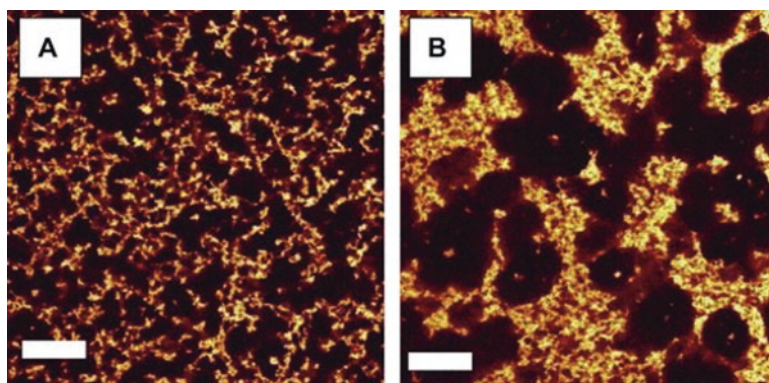


Fig. 19.2 Confocal laser scanning micrographs show homogenous distribution of gelatinized starch granules in the acidified milk gels containing (b) 2% w/w unmodified rice starch granules. Figure (a) is the control sample containing 0% w/w starch. Protein network appears in yellow, and dark spherical regions in (b) are the swollen starch granules. White scale bar: 20 μ m (Considine et al. 2011)

system. However, such stability is only a relative condition subjected to mechanic treatment and time. One can utilize different polymers to increase the relative stability and therefore, improve the shelf life stability.

19.3 Destabilization/Stabilization Mechanisms

It is important to the point out that the complex formation does not necessarily result in a stable or unstable system. Moreover, physical stability is one of the primary concerns in dairy food manufacturing. Therefore, for the food technology research works investigated rheological behavior of protein-polymer mixtures (Lizarraga et al. 2006; Rodd et al. 2000), the physical stability and structural features must need to be reported (e.g., both rheological properties and stability phase diagrams are reported (Langendorff et al. 2000)). Rheological behavior is time, structure and shear force-dependent, without elaborating the stability and structure features, the published information of rheological parameters have little significance in terms of guiding the real-life application of food formulation design for the food industry.

Although the functionality of food polymer in multiphasic systems is not fully clear, the basic destabilization mechanisms of protein-polymer containing model food systems were illustrated in several studies (Corredig et al. 2011; Doublier et al. 2000; Syrbe et al. 1998). Entropic effect is greater than the enthalpic effect in polymer mixture systems. The fate of multiphasic system is separation. The physical stable food matrix means the homogenous distribution of incompatible materials cross the whole food system and nonappearance of undesirable (visible) phase separation. Microscale phase separation is negligible if non-negative impacts on the appearance and mouthfeel of the product are detected. Based on the affinities between protein, polymer, and solvent, two types phase separation mechanisms are introduced: (1) segregative phase separation (for describing non-adsorbing polymers containing systems); (2) associative phase separation (for describing adsorbing polymers containing systems). In summary, both types of phase separation depend on absolute concentrations of dairy protein and nondairy polymer; relative ratio of concentration between dairy protein and nondairy polymer, species of added polymer, pH and ionic strength of the mixture system.

19.3.1 *Segregative Phase Separation (Non-adsorptive Stabilization)*

Milk proteins are negatively charged at native milk pH (~pH 6.5–6.7). Neutral or anionic polymer/polysaccharide is incompatible with milk proteins at the natural pH range of milk, thus, pH and ionic strength determine the phase separation. Such polymers are recognized as non-adsorbing or non-interacting polymers. In a dairy

protein dispersion system containing non-adsorbing polymers, its stability depends on the concentration of the free polymer in the aqueous phase (Corredig et al. 2011; Syrbe et al. 1998). (a) At extremely low concentration (0–1.5% w/w) of non-adsorbing polymer (amylopectin), the dairy protein (casein) dispersion system is stable (De Bont et al. 2002); (b) At relatively higher concentration level of non-adsorbing polymer (>0.3% w/w, κ -carrageenan) the casein dispersion system (0–5% w/w casein content) becomes unstable (Schorsch et al. 2000). This is due to the increased concentration of polymer induced an osmotic pressure gradient, consequently caused the depletion flocculation of the casein micelles (Suresh et al. 2006); (c) Keep increasing the non-adsorbing polymer in liquid milk system may overcome the depletion flocculation effect and result a stable milk-polymer mixture system (Fig. 19.3 a3). Konjac glucomannan (KGM) is a natural polysaccharide extracted from konjac which may be used as thickening and gelling agent in dairy food systems. Dai and co-workers (2017) found that KGM and milk components in a mixture system follow the segregative phase separation mechanism; also, the authors used binodal curve and showed that the KGM-milk mixtures may be stabilised from phase separation by using either low or high dosage of KGM. For instance, in diluted milk system (70% liquid milk), adding <0.2% or >0.7% KGM may result stable polymer-milk mixture systems; however, phase separation is induced by adding 0.25–0.6% KGM. In the same research work, the authors also observed the formation of aggregate structures when the concentration of KGM is higher than 0.5%. Such observations suggested that at higher concentration levels of KGM in milk system, the KGM polymers form non-adsorption self-packing structure/network at the aqueous phase, therefore, increase the viscosity of the continuous phase in the mixture system. The network of polymer (including gelation) in aqueous phase results increase of system viscosity (Hemar et al. 2001b). According to the Stokes's law, the viscosity of the aqueous phase of dispersion system determines the phase separation rate (Huppertz and Kelly 2006). Therefore, the relatively higher volume of non-adsorbing polymer (higher than the critical concentration of depletion effect) may be considered in dairy food formulation for improving physical stability and thickness (Fig. 19.3a).

19.3.2 Associative Phase Separation (Adsorptive Stabilization)

Electrostatic attraction is the driving force of association between dairy protein and polysaccharides. Most of the formation of protein-polymer complex happens under the pI of dairy proteins due to both protein and polysaccharides for food applications are negatively charged at native pH of milk. If the protein-polymer complex is needed for stabilizing a system from phase separation, for the benefit of protein-polymer complex formation, the polysaccharides are expected to have relatively lower pI comparing with dairy proteins ($pI_{\text{casein micelle}}$: pH 4.6; $pI_{\beta\text{-Lactoglobulin}}$: pH 5.2; $pI_{\alpha\text{-Lactalbumin}}$: pH ~4.3) so that they remain negatively charged at acidic pH when dairy proteins have zero or positive net charges. Consequently, polysaccharides can

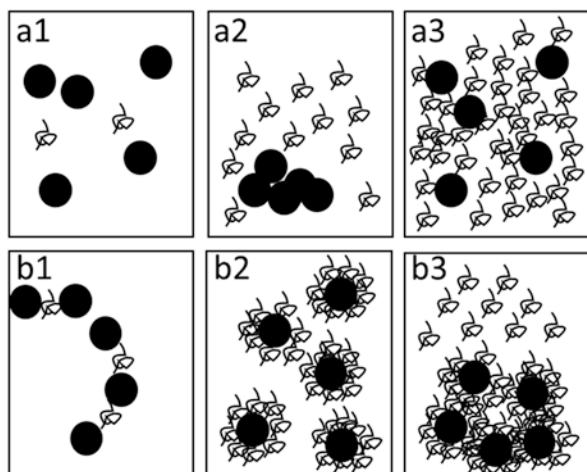


Fig. 19.3 (a) Non-adsorbing polymers in dairy protein dispersions. (a1) Stable. (a2) Segregative phase separation (depletion flocculation). (a3) Stable (polymer networks increase viscosity of aqueous phase). (b) Adsorbing polymer in dairy protein dispersions. (b1) Bridging flocculation. (b2) Adsorptive stabilization. (b3) Depletion flocculation. *Linear lines* nondairy polymers. *Solid dots* dairy (micellar) proteins. The figure is re-drawn according to (Dickinson 1998)

adsorb onto dairy proteins via electrostatic attraction. Moreover, weak coulombic complex formation between whey protein and polymer (pectin) was observed at region of pH 4–5 when the protein concentration is relatively low where pectin is negatively charged (Cape et al. 1974) and whey proteins have nearly zero net charge (Zaleska et al. 2000; Bystrický et al. 1990). This phenomenon suggests that not only counter charges initiate coulombic attraction, zero net charge molecules may also induce protein-polymer interaction. In general, under acidic condition, upon increasing the concentration of anionic polymer in dairy protein dispersion/solution system, it undergoes three stages transition. (a) Bridging flocculation, this is an unstable state in which polymers bridge dairy proteins forming flocculated complex particles (Gancz et al. 2006; Everett and Mcleod 2005; Langendorff et al. 1999). Bridging flocculation of milk proteins (e.g., casein micelles) are caused by insufficient addition volume of counter charged polymer. (b) Optimum adsorption, this is a stable state in which the amount of polymer molecules are just enough to encapsulate the individual milk protein molecules or micelles (Dickinson 1998; Syrbe et al. 1998). The newly formed protein-polymer complex particles have identical surface charge and being repulsive to each other. The adsorbing polymer on the surface of colloidal protein particles are saturated, and steric repulsion effect is generated between the outer layers formed by nondairy polymers (soybean & soluble polysaccharide) (Nakamura et al. 2006; Nobuhara et al. 2014). The presence of small quantity of free polymer molecules does not necessarily disrupt the stabilized system (Syrbe et al. 1998). (c) Depletion flocculation, if excessive amount of polymers are added into stable protein-polymer colloidal system where nondairy polymer has been

already saturated on dairy protein particles, at a certain concentration, the free, non-adsorbed polymers induce depletion flocculation of protein-polymer complexes (Repin et al. 2012; Mession et al. 2012; Rohart and Michon 2014) (Fig. 19.3b).

19.3.3 Challenges of Applying Theories of Stabilization Mechanism

Even though the general destabilization/stabilization mechanisms of a polymer containing dairy system are understood, strong uncertainty exists when choosing proper polysaccharides and applying them for stabilizing a specific dairy colloidal/emulsion system. For example, carrageenan is one of the special stabilizers being used in dairy systems as its stabilization mechanism is still not fully clear. It is still controversial that whether carrageenan interacts with micellar casein forming a polymer-protein complex in milk systems or carrageenan only forms a self-supporting gel which holds up other colloidal particles responsible for the stabilization. Carrageenans are linear, negatively charged (at neutral pH), sulphated polysaccharides containing D-galactose and 3,6-anhydro-D-galactose extracted from red seaweed (*Rhodophyceae*). Three major types of carrageenan as kappa (κ), iota (ι), and lambda (λ)-carrageenans are widely applied in dairy systems (Spagnuolo et al. 2005; Lynch and Mulvihill 1994; Lin and Hansen 1970; Bayarri et al. 2010; Camacho et al. 1998); and they differ in number/position of sulphate groups and the content of 3,6-anhydrogalactosyl ring per disaccharide (Damodaran et al. 2007). κ and ι -carrageenans are able to form self-supported gel network in the presence of cations (Drohan et al. 1997; Langendorff et al. 1997), their sulfate groups and the 3,6-anhydro-D-galactopyranosyl ring may undergo coil (disordered) to helix (ordered) transition as the response to temperature change; nevertheless, due to lack of 3,6-anhydro-D-galactopyranosyl ring, λ -carrageenan is not able to gel (Rees et al. 1969). Spagnuolo et al. (2005) summarized two theories which explain the stabilization of micellar casein using κ -carrageenan. In the first theory, researchers believe negatively charged κ -carrageenan may adsorb onto casein micelles via the interaction with a positively charged region of κ -casein (residues 97–112) (Dalglish and Morris 1988). However, the second theory states κ -carrageenan forms self-supporting gel system with the presence of cations (e.g., Ca^{2+}) and hold up casein micelles or dairy protein stabilized emulsion droplets without phase separation rather than to interact with casein proteins (Drohan et al. 1997; Vega et al. 2005).

A nondairy polymer can be dairy protein non-adsorbing and protein adsorbing at different pH levels. Therefore, different stabilizing mechanisms need to be considered when pH, temperature, protein composition and concentration are all formulation variables even the same type of polysaccharide is used for stabilizing a dairy food (Gu et al. 2005; Langendorff et al. 1999; Corredig et al. 2011). For instance, in β -lactoglobulin stabilized emulsion system, at pH 3 (below the pI of whey protein), ι - and λ -carrageenans as adsorbing agents at different concentration levels caused

both stabilization (at concentration range: $0\% < x < 0.08\%$ w/w) and creaming effects (at concentration range: $0.08\% < x < 0.15\%$ w/w) to the same emulsion system, moreover, ι -carrageenan as non-adsorptive agent is able to stabilize the same emulsion system from creaming at pH 6 (above the pI of whey protein) at concentration range 0–0.15% w/w (Gu et al. 2005).

The authors believed that at pH 3, the aggregation of oil droplets is induced by the droplet-carrageenan-droplet bridging effect moreover, they observed a greater change of ζ -potential in the system containing a higher level of carrageenan comparing the emulsion systems containing a lower concentration of carrageenan (Gu et al. 2005). However, according to the aforementioned mechanism of “optimum adsorption” in adsorptive stabilization and the gelling mechanism of carrageenan, it is reasonable to re-interpret the experimental results from this research using an alternative reaction mechanism which is explained as follows. At pH 3, at lower concentration levels of carrageenan ($0\% < x < 0.08\%$ w/w), weak gel is formed by carrageenan–carrageenan cross-linkages (carrageenan is able to form gel at concentration as low as 0.018% w/w) (Drohan et al. 1997). The weak gel hinders oil droplets interaction and the relatively high surface charge results in electrostatic repulsion between oil droplets. The emulsion system is thus stable. At higher concentration levels of carrageenan ($0.08\% < x < 0.15\%$ w/w), the carrageenan molecules adsorb onto the surface of emulsion droplets via the interaction with the emulsifying agent, β -lactoglobulin, therefore neutralizing the surface charge of oil droplets. The excessive free carrageenan interacts free β -lactoglobulin forming polymer-protein complex and induces depletion flocculation of oil droplets. Consequently, the emulsion system is destabilized. The interaction mechanism between the nondairy polymer and dairy protein or dairy protein stabilized emulsion droplets is complicated. It is just shown in this paragraph that different speculative mechanisms may be used to explain the rationale of the same set results. Such uncertainties in stabilizing dairy food systems are the driving force for initiating more systemic research so that one can draw a clearer picture of the functionality of a nondairy polymer in a colloidal/emulsion systems containing milk proteins or milk protein stabilized oil droplets.

The significant suggestion to dairy technologists is that one should hypothesize a stabilization mechanism of the targeted product before formulating and processing the dairy matrix. Subsequently, the microstructure and stability can be engineered by selecting proper nondairy polymers, applying them at proper concentrations, adjusting system temperature, pH, and ionic strength, etc. for achieving the non-adsorptive or adsorptive stabilization. Sometimes, probably most cases of commercial product applications, both non-adsorptive and adsorptive reactions may occur in one system for achieving the physical stability. For instance, in the stabilized system containing gelatinized starch granules, leached starch polymers (amylose and amylopectin) and dairy proteins (whey and casein), the starch-casein association presents in the dairy protein dispersion system. Moreover, starch self-associated networks also present in the dairy protein dispersion system. These starch-protein and starch-starch networks are responsible for increasing complex viscosity (Kumar et al. 2017).

19.4 Texture Establishment

Besides stabilization, nondairy polymers are applied as texturizers to manipulate the rheological, tribological properties of dairy foods so that the market preferred sensory attributes may be achieved (Foegeding et al. 2010; Marshall and Rawson 1999; Foegeding 2007; Van den Berg et al. 2007). The heterogeneous group of long-chain polymers mainly polysaccharides and proteins/polypeptides are able to engage water molecules forming viscous dispersion system in the aqueous phase of dairy dispersion/emulsion/gel systems. The engagement of water molecules is due to the presence of large quantity of hydroxyl groups in the nondairy polymers. The capability of forming higher viscous dispersion system makes the hydrocolloid as a “thickening agent” which is used for providing body and increasing creaminess in dairy food products. A gel as tangled and interconnected molecular network may also be formed in the aqueous phase of dairy food system through crosslinking of polymers (Oakenfull and Glicksman 1987). The gel system may provide mechanical rigidity, and it may be flowable liquid-like (injectable, low yield stress) gel or non-flowable (highly elastic, high yield stress) solid-like gel (Piron and Tholin 2001; Le et al. 2017; Brenner et al. 2015). In general, the polymer-polymer and polymer-dairy protein interactions defer to the aforementioned of non-adsorptive and adsorptive interaction mechanisms.

19.4.1 Thickening Dairy Matrixes

Polymers as thickeners in dispersion system presented as polymer random coils; its concentration determines the physical feature of the dispersion system (Daoud et al. 1975). Three concentration domains are defined as separated chains, overlapping chains, and concentrated solution regime (Berry et al. 1979). The thickening effect derives from polymer-solvent interactions, and it is strongly correlated with the restricted freedom of movement of individual chains of the polymer which is determined by the degree of overlapping chain (Morris 1994). Therefore, it is logical to state that an ideal concentration of polymer is needed for initiating functional thickening effect. The ideal concentration is named as “critical coil overlap concentration” (c^*), the polymer chain starts overlapping and entangling between each other. Consequently, the viscosity of the dispersion/emulsion systems significantly increases (Baines and Morris 1987). Therefore, for thickening and improving mouth feel of dairy food using nondairy hydrocolloids, one needs to know the c^* of the polymer candidate. Although the physical features and perception of taste intensity of polysaccharides fortified solution systems have been studied, more research through engineering approaches is required for constructing mathematical models containing c^* as one of the key predictors which may be used in predictions of sensory attributes.

c^* is the critical parameter which determines the transition of rheology nature of dispersion system between the Newtonian fluid and non-Newtonian fluid (Phillips and Williams 2009). The c^* may be measured and calculated via using experimental approaches (Morris et al. 1981; Cook et al. 2002), and may also be estimated from prediction models (Ying and Chu 1987). In the experimental approach, zero-shear specific viscosity (η_0) and intrinsic viscosity (η) are measured and estimated using Huggins and Kraemer models (Eqs. (19.1) and (19.2), η : intrinsic viscosity, η_{sp} : specific viscosity, c : concentration). Then c^* is calculated using the model $\log(c \times \eta)$ (x-axis)- $\log(\eta_0)$ (y-axis), c^* is the c which results the intersect point on the log-log model (Morris et al. 1981, Cook et al. 2002). In a recent study, the author stated intrinsic viscosity is used for estimating c^* using the equation $c^* = 1/\eta$ (Van der Sman 2015); intrinsic viscosity can be expressed by Fiery-Fox relationship (Flory 1953).

$$\frac{\eta_{sp}}{c} = \eta + k'\eta^2 c \quad (19.1)$$

$$\frac{\ln \eta_{rel}}{c} = \eta + k''\eta^2 c \quad (19.2)$$

Starch as its native or modified forms has been widely used in dairy food formulations as a thickening agent (Gutiérrez et al. 2017; Gutiérrez 2018). This may due to its relatively lower cost comparing with other gum stabilizers and relatively clean taste (Saha and Bhattacharya 2010). In dairy pudding dessert system containing milk proteins, carrageenan, and unmodified starch granules, it was found that the gelatinized starch granules do not contribute the gel structure formation (Verbeken et al. 2004). Continuously increasing the concentration of starch results exclusion effect. This effect causes a concentration of gelling agent in the aqueous phase of the dairy food system, therefore, strengthens the gel structure. Exclusion effect dominates the starch enriched dairy food system containing gelling agents, and such effect is more pronounced when the effects caused from dairy protein and gelling polysaccharide (carrageenan) (Verbeken et al. 2004). Such starch induced exclusion effect may be utilized in the dairy formulation strategy when one attempts to enhance the dairy gel structure without using additional relatively expensive dairy proteins and gelling gums.

Xanthan gum as a thickening agent is used for increasing viscosity of the different dairy liquid or semi-solid systems at different pH levels (Hemar et al. 2001a, b; El-Sayed et al. 2002). Its viscosity dominated the viscosity of protein dispersion systems made from different dairy protein ingredients at neutral pH (incl. skim milk powder, milk protein concentrate and sodium caseinate) (Hemar et al. 2001b). However, phase separation appeared in the dispersion systems when xanthan gum is mixed with skim milk powder and milk protein concentrate (Hemar et al. 2001b). The author attributed the phase separation to depletion flocculation. Low concentration of xanthan gum ($\leq 0.2\%$ w/w) induced visual creaming of milk protein stabi-

lized emulsion droplets at neutral pH (Hemar et al. 2001a). In yogurt (acidified dairy protein gel system), the addition of a small volume of xanthan gum (at concentrations of 0.01% and 0.05%, w/w) resulted in a dramatic increase of gel curd tension and a significant decrease of syneresis rate (El-Sayed et al. 2002). Xanthan gum and locust bean gum (LBG) are co-used in food gel systems for improving sensory and rheological attributes due to the proven synergetic effects (Juszczak et al. 2003). However, such synergy does not necessarily appear in an emulsion system; it found that xanthan gum-LBG combination did not result in higher viscosity in a mayonnaise-like emulsion comparing the system containing the same amount of sole type of polymer (Dolz et al. 2007).

Other polysaccharides such as carboxymethyl cellulose (CMC), methyl cellulose (MC), hydroxypropylmethyl cellulose (HPMC), gum Arabic (GA), guar gum (GG), tara gum (TG), konjac mannan (KM), gum tragacanth (GT) are all know as thickening agents in dairy food systems at different pH levels (Bayarri et al. 2009; Zhao et al. 2009; Arboleya and Wilde 2005; Ibanoglu 2002; Mudgil et al. 2014; Bourriot et al. 1999; Saha and Bhattacharya 2010; Tobin et al. 2011; Azarikia and Abbasi 2010). Moreover, some of these aforementioned polymers may also be used as emulsifiers for stabilizing emulsion oil droplets, for instance, MC, HPMC, GA (Arboleya and Wilde 2005; Mcnamee et al. 1998). Bayarri et al. (2009) found that when a higher concentration of CMC (>1%, w/w) is applied in skim milk, a weak gel rheology feature is identified. Therefore, it is important to point out that the concepts about thickening agent, gelling agent, emulsifying agent are an empirical description of the functionality of polymers rather than absolute definition. Dairy technologists need to know the broad functionalities of individual polysaccharides in different dairy systems rather than simply group a polymer as a thickening agent or a gelling agent.

19.4.2 Smoothing Dairy Matrixes

Creaminess is one of the key sensory attributes of dairy foods which strongly correlated with consumers' hedonic response (Folkenberg and Martens 2003). It is generally understood that creaminess perception in dairy products are determined by both flavor and texture, and the fat content plays a crucial role (Mela 1988). Reducing fat content from dairy matrix results in undesirable sensory properties and less consumer acceptance (Cardello 1994; Tuorila et al. 1994). Polysaccharides are used as fat replacers in fat-reduced dairy foods for compensating thickness, rheological behavior and creaminess perception (Bayarri et al. 2010). However, it is important to point out that the detailed mechanism of creaminess perception is not fully understood. It might be combined sensations of flavor, texture, and psychology (Drake 1989; Antmann et al. 2011; Frøst and Janhøj 2007; Kilcast and Clegg 2002; Elmore et al. 1999). The general agreement according to a series of research states that structure, texture, and smoothness are highly correlated with creaminess perception (Akhtar et al. 2006; Elmore et al. 1999; Kilcast and Clegg 2002). Extensive

knowledge and research findings are available for manipulating structure and texture (mainly rheological features) properties of dairy foods. However, techniques about how to engineer dairy food structure using non-dairy polymers with enhanced smoothness are relatively scarce. Clearly, creaminess improvement is far beyond manipulating rheological properties. Classically, viscosity at 50 s^{-1} shear rate is used as an indication of mouthfeel perception (Wood 1968). However, it was noted that such parameter is insufficient for describing perceived thickness or creaminess of dairy-based emulsion systems containing nondairy polymers used as texturizer (Akhtar et al. 2006). Recently, tribology techniques are developed for quantitatively characterizing the smoothness of dairy systems (Sonne et al. 2014; Nguyen et al. 2016; Meyer et al. 2011b; Laguna et al. 2017; Dresselhuis et al. 2008). Such technical developments open a gate for screening tribological functionalities of food polymers in dairy food matrices.

Regarding modifying creaminess perception, maltodextrin (13.5% w/w) and xanthan gum (0.17%), respectively, are able to enhance creaminess perception of dairy protein stabilized emulsion system, and maltodextrin is relatively more functional comparing with xanthan gum in terms of elevation of creaminess level (Akhtar et al. 2006). The authors controlled the viscosities of the emulsion systems which contain the two different polymers at the same level at a reference shear rate (50 s^{-1}). They noted that the sensory creaminess perception might be altered by nondairy polymer without changing viscosity (Akhtar et al. 2006). The same research group also found that low methoxyl pectin is more functional than xanthan gum for increasing creaminess of dairy emulsion at the relatively thinner system (viscosity $50\text{ mPa}\cdot\text{s}$ at 50 s^{-1}). Such discrimination of functionality vanished at the thicker system (viscosity $100\text{ mPa}\cdot\text{s}$ at 50 s^{-1}) (Akhtar et al. 2005). It is important to reveal that although pectin and xanthan gum were able to increase creaminess perception, their capability of creaminess enhancement is not comparable with fat content. It is interesting to note that at the same viscosity level ($50\text{ mPa}\cdot\text{s}$ at 50 s^{-1}), the creaminess perception of pectin fortified emulsion is still lower than the plain emulsion system, even though the fat content of the pectin-containing emulsion (22% vol/vol) is slightly higher than the plain emulsion (20% vol/vol) (Akhtar et al. 2005). Therefore such polymers cannot be used as fat replacers for maintaining the intrinsic creaminess.

In the dairy protein gel structured system, nondairy polymers may be used for improving creaminess. λ -carrageenan as a non-gelling agent only increases the viscosity of aqueous phase in milk or dairy gel system; it was reported that λ -carrageenan at 0.06% (w/w) increased creaminess of gelled dairy dessert (Tarrega and Costell 2006). Application of long-chain chicory inulin (4% w/w) in no fat yogurt was found to be able to mimic the rheological features of full-fat yogurt. However, no sensory data is available in that research (Paseephol et al. 2008). Mimicking the rheological parameters does not guarantee matching of creaminess (Szczeniak 2002). 8% (w/w) of long-chain inulin was recommended to be added to skim milk for compensating the creaminess of whole fat milk (Villegas et al. 2007). Meyer and co-authors (2011a) elaborated the working mechanism of inulin in dairy products as texture modifier for enhancing creaminess, it was noticed that the smoothing effect

of long-chain inulin is hindered by higher concentration of starch (4% w/w). For increasing the creaminess of low fat yogurt, inulin must be applied before fermentation and be part of the protein gel structure (Kip et al. 2003). Inulin can interfere with the extracellular polysaccharides, therefore, reduce the “brush friction” resulting elevating creaminess level (Kip et al. 2006; Marle et al. 1999).

19.5 Conclusion

Application of nondairy polymers offers dairy technologists a great opportunity to construct sophisticated dispersion/gel structures which are essential for maintaining the physical stability of innovative; value-added dairy products. The nondairy polymers are capable enough to modify the texture of dairy foods resulting desirable changes of sensory attributes. The general interaction mechanisms of different type of polymers in liquid/gel based dairy systems are explained in this chapter. Such information as a powerful guideline provides strategic approaches for future formulation of dairy foods, for instance, one would know how to prevent segregative phase separation and associative phase separation when thickening agents and gelling agents are needed in what type of dairy system. However, the technical boundaries of the functionality of individual polymers and detailed mechanism of polymer-dairy system interaction are not fully clear. For the interest of specific application, further systemic research is needed for mapping the functionalities of individual polymers in different dairy systems.

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